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STUDIES IN THE LIQUID-DROP THEORY OF NUCLEAR FISSION

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STUDIES IN THE LIQUID-DROP THEORY OF NUCLEAR FISSION

James Rayford Nix

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ABSTRACT

In connection with nuclear fission we study the division of an idealized charged drop, using a simplified version of the liquid-drop model. The degrees of freedom essential to a discussion of the division of a charged drop and the separation of the fragments to infinity are taken into account: a fragment-separation coordinate, a mass-asymmetry coordinate, a deformation coordinate for each fragment, and rotational coordinates for each fragment. To specify fragment deformation, the fragments are represented by spheroids; a nucleus prior to division is represented by two overlapping spheroids. The Hamiltonian for the idealized system consists of a sum of surface, Coulomb, and kinetic energies. A study of the saddle-point energies and shapes calculated in this two-spheroid approximation indicates that the approximation is most useful for discussing the fission of elements lighter than about radium. On the basis of this model, we calculate probability distributions for certain observable characteristics of fission fragments at infinity—their total translational kinetic energy, mass, individual excitation energies, and individual angular momenta. This is done by applying standard static, dynamical, and statistical methods to the Hamiltonian for the system. The present treatment, for the most part, is classical; quantum mechanics is considered only in the statistical-mechanics discussion of the behavior of the system near the saddle point.

The predictions of the model are compared with existing experimental data for distributions in fragment mass and total translational

kinetic energy, for nuclei lighter than radium. The comparisons are made without the use of any adjustable parameters. The theory is capable of accounting for the magnitudes of the most probable values and widths of the experimental distributions, as well as some, but not all, finer details of the distributions. The dependence of the experimental distributions upon nuclear temperature, and the dependence of the experimental most probable kinetic energies upon fissionability parameter are also approximately reproduced by the calculations.

I. INTRODUCTION

Nuclear fission was discovered by Hahn and Strassmann[1,2] in 1938, and yet, in the two and a half decades since its discovery, an adequate theory of the fission process has still not emerged. Numerous papers have been written dealing with various aspects of the theory, but in no instance has a definite model with a well-defined Hamiltonian been chosen and the implications of this model systematically worked out in detail. Such an approach would require that one select a model characterized by definite degrees of freedom; apply standard static, dynamical, and statistical methods to the resulting Hamiltonian; and compare the predictions of the model with experiment. In the present study we attempt to do this for a simplified version of the liquid-drop model. [A preliminary account of this work (for a restricted case) is given in reference 3.]

Shortly after the discovery of nuclear fission, Meitner and Frisch emphasized the analogy between the fission process and the division of a charged drop of liquid.[4] On the basis of the liquid-drop model, Bohr and Wheeler showed that a satisfactory account could be given of some of the phenomena observed in fission.[5] These early successes of the liquid-drop model and its conceptual simplicity firmly established its popularity for discussing fission.

The major effort in the development of liquid-drop theory has been concerned with the static problem of calculating the potential energy of a deformed charged drop. The coordinates normally used to describe a deformed drop are the coefficients in the expansion of the drop's radius vector in spherical harmonics. The energy of a nearly spherical drop can then be calculated in terms of these coordinates as an expansion about a sphere.[5-13] A similar technique, appropriate for shapes not far removed from spheroids (ellipsoids of revolution), is the expansion about a spheroid.[8-13] For highly distorted shapes the deformation energy has to be calculated numerically.[14-18] The potential energy of charged drops is now fairly well understood, particularly with regard to the variation of fission barriers throughout the periodic table.

Although statics has been extensively studied, dynamics, on the other hand, has not been so exhaustively treated. A fragmentary study of the dynamical aspects of fission was performed by Hill, and by Hill and Wheeler in connection with the question of mass asymmetry.[19-21] For a few special cases, the division of a charged drop was traced out numerically to a short distance before the actual division of the drop into two fragments (scission). However, no relationship between initial conditions and final results was established. Some limited aspects of dynamics have also been considered by other authors.[8, 22-25]

The statistical mechanics of fission has been investigated in some detail. Bohr and Wheeler formulated the calculation of fission probabilities by applying statistical transition-state methods at the saddle point.[5] (See in particular the recent article by Wheeler.[26]) The application of statistical mechanics in a somewhat different manner was discussed by Fong, whose starting point was the assumption of statistical equilibrium at the scission point.[27-30] This theory, however, suffers from the fact that the nuclear configuration at the scission point is not defined (until one has performed a dynamical calculation). Statistical aspects of fission have also been examined by several other authors.[31-37]

We see that statics, dynamics, and statistical mechanics have all been considered to some extent in previous studies of fission. Never, however, have they been treated systematically for one and the same Hamiltonian.

Here an attempt is made to study each of these steps for a simplified version of the liquid-drop model. The outline of our program is as follows:

(1) Statics: After the coordinates specifying the system have been selected, the potential energy of the system (for a given nucleus) is mapped as a function of its coordinates, and the saddle point is located and its properties studied.

(2) Dynamics: The kinetic energy of the system is calculated as a function of the coordinates and their conjugate momenta. This makes it possible, by solving Hamilton's classical equations of motion for the system, to discuss the division of the nucleus and the separation of the fragments, from some given initial configuration to infinity.

(3) Statistical mechanics: In analogy to the method employed in discussing chemical reaction rates,[38] attention is focused on the system at the saddle point (transition state), and statistical equilibrium is assumed to hold there. In the case of chemical reactions, this provides information regarding the rate of reaction, and it was for the calculation of rates that Bohr and Wheeler used the transition-state method.[5] In our case, however, we use the transition-state method to calculate the probability for finding the system in a given state of motion close to the saddle point. These probability distributions are then combined in the sense of initial conditions with the dynamical calculations [step (2)] to trace out the separation of the fragments to infinity. This converts the probability distributions of the states of motion around the saddle point into the probability distributions of observable characteristics of fission fragments: their kinetic energies, excitations, and angular momenta.

Since the difficulty of solving dynamical equations of motion increases with the number of coordinates, it is desirable from the calculational point of view to have present in one's model as few degrees of freedom as possible. Let us therefore see what degrees of freedom it is essential to consider. Although we will be concerned with the liquid-drop model, the discussion that we are about to give for the degrees of freedom essential to fission is quite general and does not depend upon the assumption of the liquid-drop model.

A single isolated fission fragment has three rotational degrees of freedom and three degrees of freedom associated with its center-of-mass motion. Thus for a system consisting of two fission fragments a total of 12 coordinates is required to specify the orientation in space and the position of the center of mass of each fragment.

These 12 coordinates may be distributed as follows: three spatial coordinates for the center of mass of the entire system; two angles specifying the direction in space of the line connecting the centers of mass of the two fragments; the distance between the fragments' centers of mass; and, for each fragment, three angles (e.g., the Euler angles) indicating its orientation in space.

In addition to the above 12 coordinates, further coordinates are required to describe the intrinsic shapes of the fragments. The specification of a fragment's extension in three dimensions requires three lengths (say three orthogonal axes a, b, c), which means, in general, two dimensionless numbers (for example, the β and γ deformations of the collective model, or related parameters[39]). This means that four numbers are needed to specify the intrinsic shapes of the two fragments. Finally, one coordinate is needed to specify the relative sizes of the fragments. This brings to 17 the number of coordinates required to describe a system consisting of two separated fission fragments (see Fig. 1).

It is necessary in a model of fission that one be able to describe in a continuous manner the sequence of shapes of a fissioning nucleus from the original sphere, through the saddle point and scission configuration, to the two fragments at infinity. In order for the number of degrees of freedom not to change suddenly in the course of the division, it follows that the number of degrees of freedom specifying fission shapes before division into separate fragments must also, in general, be 17 or more.

One could in principle continue introducing additional degrees of freedom to describe finer details of the dividing nucleus, including ultimately single-particle structure, until the number of degrees of freedom equaled the sum of the degrees of freedom of all the individual nucleons. We will consider explicitly, however, only the 17 basic degrees of freedom enumerated above. Although the remaining degrees of freedom will never be treated explicitly, they will, on the other hand, not be disregarded entirely. Their presence will be recognized implicitly when we consider the statistical mechanics of fission,

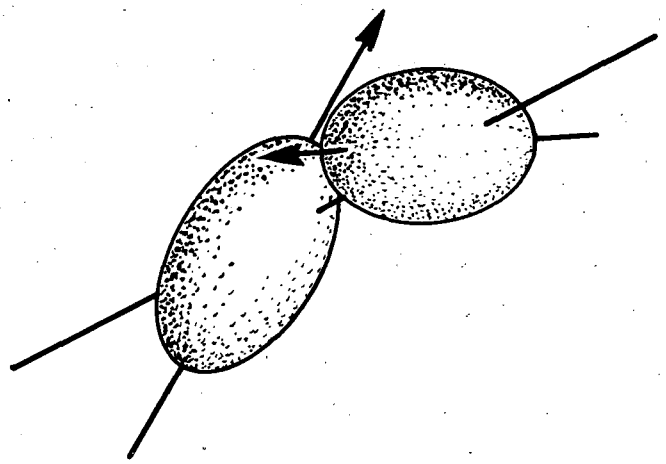

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Fig. 1. A two-fragment configuration described by 17 degrees of freedom. Three (Euler) angles describe the orientation in space of one fragment, three of the other, and two angles describe the orientation of the line joining their centers. One number specifies the relative sizes of the fragments, two specify the intrinsic shape of one fragment, two of the other, and one their separation. Three degrees of freedom describe the location in space of the common center of mass. Total: 3 + 3 + 2 + 1 + 2 + 2 + 1 + 3 = 17.

when they will be assigned their statistical share of the total energy.

The question arises of how to choose the deformation coordinates for each fragment. We shall not be concerned here with effects arising from deviations of the fragments from axial symmetry. We will therefore restrict ourselves from the beginning to fragments that are axially symmetric, thus reducing the number of degrees of freedom to 15. Because spheroids can represent any desired degree of prolate and oblate elongation, and because their surface and Coulomb energies can be easily calculated, we will use spheroids to represent the fission fragments. Following a suggestion by Wladyslaw J. Swiatecki (Lawrence Radiation Laboratory, Berkeley), we will represent a nucleus prior to division by two overlapping spheroids, with the interior surface of each simply "erased." With this parametrization, one is able to describe continuously in an approximate way the sequence of shapes of a fissioning nucleus from the original sphere to the two fragments at infinity.

Of the 15 degrees of freedom required for specifying a system composed of two axially symmetric fragments, 3 are trivially eliminated by working in the center-of-mass system. Three more may be eliminated if, as will be done in the present work, one restricts the discussion to a system with zero total angular momentum.

This, then, leaves a system possessing nine degrees of freedom. In the neighborhood of the saddle point, these nine degrees of freedom correspond to certain characteristic motions of the system. The most important of these is the motion in the fission direction—an over-all separation of the system leading to its eventual division into fission fragments. The potential energy in this direction is of the form of a potential-energy barrier. For division of the drop to occur, the system must pass over this barrier.

Most of the remaining motions near the saddle point (motions in the non-fission direction) consist, in general, of bounded small oscillations, resulting from the potential energy increasing with deviations from the saddle point in these directions. These oscillations involve the relative sizes of the fragments, their eccentricities,

and their orientations in space. The various modes of oscillation that occur correspond roughly to (a) a change in the relative sizes of the fragments, (b) an in-phase or out-of-phase stretching and contraction of the left-hand and right-hand halves of the saddle-point configuration, and (c) a bending and "wriggling" of the saddle-point configuration. In the treatment of these oscillations, the approximation that they are small will be made, although there will be some discussion of higher-order effects.

The separation of the fragments from the saddle point to infinity will be traced out dynamically. This converts the possible states of motion near the saddle point into states of motion of two fragments at infinity. The motion of the fragments at infinity consists of a translation of their centers of mass, and rotations and vibrations about their centers of mass.

The states of motion of the fragments at infinity correspond directly to observable properties of fission fragments: (a) The speed of separation of the fragments' centers of mass determines their translational kinetic energies. (b) The relative sizes of the fragments are directly observable. (c) The vibrations of the fragments are associated with their excitation (vibrational or deformation) energies.* (d) The rotations of the fragments are associated with their angular momenta.

We will find that for each of the above quantities we are able to discuss not only its most probable value, but also the distribution about its most probable value. In particular, our theory predicts the probability distribution $P(E, U, X_1, X_2, \vec{L}_1, \vec{L}_2)$ of simultaneously observing the two fragments at infinity with total translational kinetic energy E, fractional mass U, individual excitation energies X_1 and

*When used in this context, the term "excitation energy" refers to the energy of the collective vibrations and deformations of the fragment; it does not include the internal (excitation) energy of the individual nucleons.

X_2, and individual angular momenta $\vec{L}_1$ and $\vec{L}_2$. (An arrow above a symbol denotes a vector quantity.) It proves convenient in practice to obtain from this distribution, by integrating over the quantities not of immediate interest, probability distributions involving a smaller number of observable quantities. Three such distributions are $P(E, U)$, $P(X_1, X_2)$, and $P(\vec{L}_1, \vec{L}_2)$.*

Our theory will also predict how these distributions should vary with the (internal) excitation energy of the compound nucleus undergoing fission, as well as with its charge and mass.

The immediate test of the theory will be the comparison of predictions with experiment for distributions in mass and total translational kinetic energy $[P(E, U)]$.† It is hoped that these comparisons, as well as those to be made when more experimental information becomes available, will yield a more accurate idea of the relevance of the liquid-drop model for discussing fission phenomena. Indeed, the entire purpose of undertaking this study was to trace out in detail the characteristics of the division of an idealized droplet whose size, surface tension, and charge are those of a nucleus, and to compare the results with what is observed experimentally in the fission of real nuclei. Stated in this way, there are <u>no adjustable parameters</u>

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*The same symbol P is used throughout this paper to denote each of several probability distributions; the argument or subscript will indicate which explicit function is being referred to.

†Experimental information regarding the other distributions is at present not available for the fission of nuclei lighter than radium. We will see later that it is for these nuclei that our model is most applicable.

in the problem.* Thus, when the comparison is made with experiment, we shall learn unambiguously to what extent an idealized liquid-drop model is capable of representing the characteristics of fission, as regards both over-all order-of-magnitude agreement and also more detailed aspects of the process.

Concerning the question of the validity of the liquid-drop model for discussing fission, we believe that the above clear-cut program of an unambiguous comparison of nuclear fission with the division of an idealized drop should be carried through irrespective of how good the liquid-drop model is thought to be. Nevertheless it is of interest to form some a priori judgment as to how well an idealized drop might be expected to represent nuclear fission. Some comments on this subject are made in Appendix A, where we indicate that the surface and Coulomb energies may be regarded as the two leading shape-dependent terms in an expansion of the potential energy in increasing powers of the small dimensionless number characterizing the nuclear problem—the ratio of the range of the nuclear force to the nuclear radius. The general conclusion is that the effects arising from the discreteness of nucleons (single-particle shell structure) are of a lower order in this dimensionless number, and, barring accidental cancellations of the leading terms, should in general be smaller than the effects of the surface and Coulomb energies. In the region of the heavy elements the changes in the Coulomb and surface energies do indeed tend to cancel, and, especially in the case of fission at low excitation energy, the single-particle effects may then be essential for discussing certain aspects of the process. On the other hand, for nuclei lighter than about radium, where the cancellation of the

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*We have taken the constants of the Bethe-Weizsäcker semi-empirical mass formula from Green's analysis of ground-state masses.[40] The nuclear temperature at the saddle point is determined from the excitation energy and fission barrier of the compound nucleus. All other quantities are calculated directly from the model.

changes in the Coulomb and surface energies is not so close, single-particle effects should not have the same relative importance, particularly at higher excitation energies.* A logical approach would seem to be to work out the details of the theory first, considering only the surface and Coulomb energies, and later to incorporate single-particle effects. Even though the liquid-drop theory of fission would not be expected to be accurate for a particular nucleus, it can be expected to yield the correct average behavior of nuclei throughout the periodic table, in analogy to the way the liquid-drop semiempirical mass formula reproduces the correct trends in the masses of nuclei, apart from oscillations due to shell structure.

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*We will see later that other properties of the model we introduce also make it most suitable for discussing the fission of elements lighter than radium.

II. STATICS

We shall be concerned in this section with the static properties of our model—with mapping the potential energy of the system as a function of its coordinates, and with studying the properties of the saddle-point configuration. Before proceeding with this study, however, it will be advantageous for us to take care of some preliminaries.

A. Units, Notation, and Coordinates

For discussing fission within the framework of the liquid-drop model, it is convenient to use a system of units based on the original spherical drop, rather than the conventional units of length, mass, and time. The three "natural" units to choose are the radius, mass, and surface energy of the original drop. Time is then no longer a fundamental unit, but is expressed in terms of a length, a mass, and an energy. This is analogous to the situation in elementary-particle physics, for example, where the speed of light, Planck's constant divided by 2π, and the pion mass are chosen as units.

In actual applications it is necessary to convert these liquid-drop units to conventional units. For the comparison of the theoretical and experimental results discussed here, this conversion is made with Green's values for the constants in the semiempirical mass formula.[40*] We denote by A the number of nucleons in the compound nucleus undergoing fission. Then the radius of the original drop is

$$R_0 = r_0 A^{1/3}, \quad \text{with } r_0 = 1.216\times10^{-13} \text{ cm}, \tag{1a}$$

and its surface energy is

$$E_S^{(0)} = a_S A^{2/3}, \quad \text{with } a_S = 17.80 \text{ MeV}. \tag{1b}$$

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*When future determinations of these constants are made, the new values may be used since all theoretical quantities are given here in liquid-drop units.

The mass of the original drop, to an accuracy within one part per thousand, is equal to the mass unit times the number of nucleons; thus,

$$M_0 = m_0 A, \tag{1c}$$

with

$$m_0 = 1.660\times10^{-24}\ \text{g} = 931\ \text{MeV}/c^2 \text{ (reference 41).}$$

In this discussion of units the symbol c denotes the speed of light.

The units for other familiar quantities may be readily derived from these three fundamental ones. For example, the unit of time is

$$T_0 = [M_0/E_S^{(0)}]^{1/2} R_0 = (m_0/a_S)^{1/2} r_0 A^{1/2}$$

$$= (2.933\times10^{-23}\ \text{sec})A^{1/2}, \tag{2}$$

the unit of linear momentum is

$$P_0 = [M_0 E_S^{(0)}]^{1/2} = (m_0 a_S)^{1/2} A^{5/6} = (128.8\ \text{MeV}/c)A^{5/6},$$

and the unit of angular momentum is

$$L_0 = [M_0 E_S^{(0)}]^{1/2} R_0 = (m_0 a_S)^{1/2} r_0 A^{7/6} = (0.793\,\hbar)A^{7/6}, \tag{3}$$

where $\hbar$ is Planck's constant divided by 2π. The unit of frequency is

$$\Omega_0 = 1/T_0 = (3.409\times10^{22}/\text{sec})/A^{1/2}; \tag{4a}$$

when multiplied by $\hbar$ this becomes

$$\hbar\Omega_0 = 22.44\ \text{MeV}/A^{1/2} = 1.261\ E_S^{(0)}/A^{7/6}. \tag{4b}$$

Although the unit of charge is formally $[R_0 E_S^{(0)}]^{1/2}$, the charge on the drop is more conveniently specified through the dimensionless fissionability parameter x, defined by[5]

$$x = \frac{(\text{charge})^2}{10(\text{volume})\ (\text{surface tension})} . \tag{5a}$$

(The surface tension multiplied by $4\pi r_0^2$ is equal to a_S.) For a uniformly charged drop with a sharp surface,

$$x = E_C^{(0)}/\left[2E_S^{(0)}\right], \tag{5b}$$

where $E_C^{(0)}$ is the Coulomb energy of the original drop, given by

$$E_C^{(0)} = 3Z^2e^2/(5R_0) = a_C Z^2/A^{1/3} ,$$

with Z the number of protons, e the electronic charge, and the constant $a_C = 0.710$ MeV.[40] A third conventional form for x is

$$x = (Z^2/A)/(Z^2/A)_{crit} , \tag{5c}$$

where

$$(Z^2/A)_{crit} = 2\, a_S/a_C = 50.13.$$

When these liquid-drop units are used, the results of our calculations can be displayed as functions of a single parameter (e.g., x) rather than as functions of two parameters (e.g., Z and A).

The notation of this paper will adhere, insofar as is practicable, to the following convention: Quantities referring to the left-hand fragment will be distinguished by the subscript 1, and to the right-hand fragment by the subscript 2. Furthermore, unless otherwise noted, any quantity that is the sum of two quantities, each referring to an individual fragment, will be designated by the same symbol but without subscripts; e.g., the total excitation energy of both fragments $(X_1 + X_2)$ will be denoted by X.

For specifying the angles involved in our problem, we will adopt the following scheme (see the lower part of Fig. 2): We denote by Θ and Φ the two angles specifying the direction in space of the line connecting the centers of the two fragments. We define a right-handed coordinate system whose origin is at the center of fragment 1,

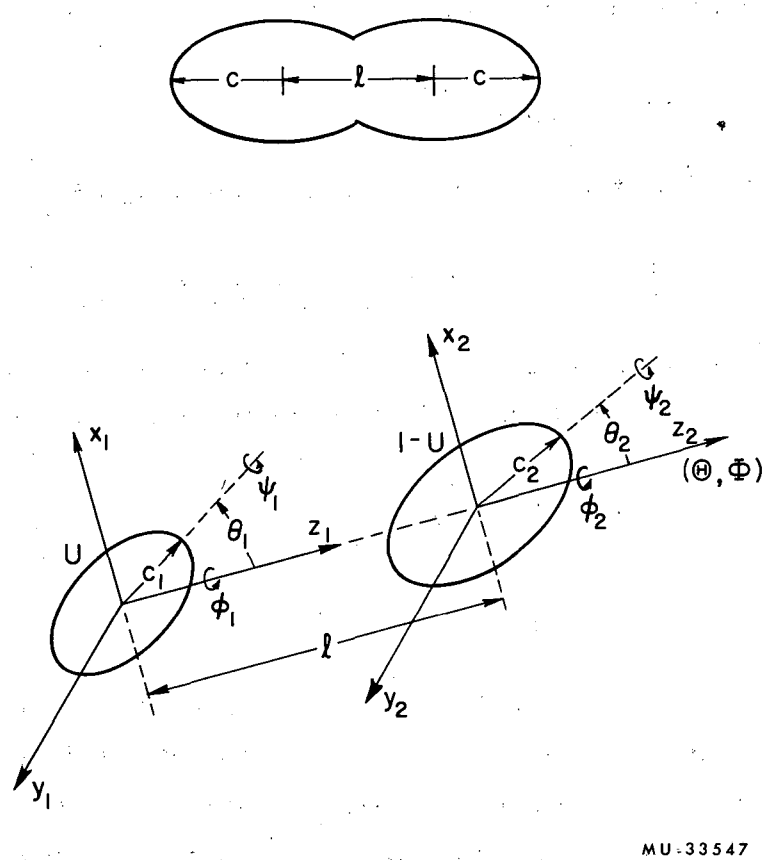

Fig. 2. Coordinates used for describing a two-spheroid configuration. The three Euler angles θ_1, ϕ_1, ψ_1 describe the orientation in space of the left-hand spheroid (see description in text), and θ_2, ϕ_2, ψ_2 the right-hand spheroid; the two angles Θ and Φ describe the orientation in space of the line connecting their centers. The fractional mass (fractional volume) of the left-hand spheroid is denoted by U. The semisymmetry axis of the left-hand spheroid is denoted by c_1, and that of the right-hand one by c_2. The distance between their centers is ℓ. Illustrated in the upper part of the figure for overlapping spheroids are the two coordinates ℓ and $c \equiv c_1 = c_2$, used for discussing the restricted case of completely symmetrical fragments.

whose z_1 axis is along the line connecting the spheroid centers (positive in the direction fragment 2), and whose x_1 and y_1 axes are perpendicular to one another and to the z_1 axis. We define a second coordinate system whose origin is at the center of fragment 2, and whose x_2, y_2, and z_2 axes are parallel (with the same sense of direction) to the x_1, y_1, and z_1 axes, respectively. Then the orientation in space of fragment 1 is specified by the three Euler angles θ_1, ϕ_1, and ψ_1 (as defined, for example, by Goldstein[42]), and the orientation of fragment 2 by θ_2, ϕ_2, and ψ_2. These angles are defined as the (counterclockwise) angles of rotation about three mutually inclined axes necessary to bring a fragment from a position with its symmetry axis along the z_1 (or z_2) axis to its final position. For fragment 1, say, these rotations are: one through the angle ϕ_1 about the z_1 axis, one through the angle θ_1 about the new x_1 axis, and one through the angle ψ_1 about the new z_1 axis, made in that order. The angle θ_1 is the polar angle of spherical coordinates usually denoted by the same symbol: the angle between the z_1 axis and the symmetry axis of fragment 1. The angle ϕ_1 is related to the usual azimuthal angle of polar coordinates by an additive constant.

Because of the symmetry of the system, the potential energy is independent of all but the following three of the above angles: θ_1, θ_2, and the difference $\phi = \phi_1 - \phi_2$ between the azimuthal angles of the fragments' symmetry axes.

We choose the four remaining coordinates required for specifying a two-spheroid configuration (in the center-of-mass system) as follows: (a) the distance ℓ between the centers of the two spheroids, (b) the fractional mass (fractional volume) U of the left-hand fragment,* and (c) the semiaxes c_1 and c_2 of each fragment along its line of symmetry† (see again the lower part of Fig. 2).

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*The fractional mass of the right-hand fragment is 1-U.

†For example, if spheroid 1 is prolate, then c_1 is its semimajor axis; its semiminor axis is determined by volume conservation.

Prior to division into separate fragments, the nucleus is represented by two overlapping spheroids. When the spheroids are overlapping, both the potential energy and the kinetic energy are much more difficult to calculate than when they are separated, and we then consider only the restricted case of symmetrical fragments, in which $U = 1/2$, $c_1 = c_2 \equiv c$, and $\theta_1 = \theta_2 = 0$. (The potential energy is for this case independent of ϕ.) For this restricted case, the two coordinates ℓ and c completely specify the system (see the upper part of Fig. 2).

B. Potential Energy of the System

The potential energy of the system is simply the sum of the surface energy E_S and the Coulomb energy E_C. The potential energy $\mathcal{V}$ of a deformed drop, relative to the original drop, is then

$$\begin{aligned}\mathcal{V} &= E_S - E_S^{(0)} + E_C - E_C^{(0)} \\ &= (B_S - 1)\, E_S^{(0)} + (B_C - 1) E_C^{(0)} \\ &= [\,(B_S - 1) + 2 \times (B_C - 1)]\, E_S^{(0)} \,. \end{aligned} \tag{6a}$$

The function B_S is the total surface energy of the system in units of the surface energy $E_S^{(0)}$ of the original drop, and B_C is the total Coulomb energy of the system in units of the Coulomb energy $E_C^{(0)}$ of the original drop. The definition (5b) is used in going from step 2 to step 3. When the system consists of two separated fragments, the potential energy is conveniently written as

$$\mathcal{V} = \left\{ [\, B_S^{(1)} + B_S^{(2)} - 1] + 2x\, [\, B_C^{(1)} + B_C^{(2)} + B_I - 1] \right\} E_S^{(0)} \,. \tag{6b}$$

The function $B_S^{(1)}$, for example, is the surface energy of fragment 1 in units of $E_S^{(0)}$, $B_C^{(1)}$ is the Coulomb self-energy of fragment 1 in units of $E_C^{(0)}$, and B_I is the Coulomb interaction energy between

fragments 1 and 2 in units of $E_C^{(0)}$. In terms of these appropriate units, the various energies denoted by B depend upon neither the value of A nor of Z of the original nucleus; they are functions only of the shape of the system.

Different formulae for calculating the energies are used, depending upon whether the system consists of separated spheroids or overlapping spheroids. The case in which the spheroids are separated is the simpler. The surface energies and Coulomb self-energies are then expressible in a closed form, and the interaction energy may be calculated by performing a triple multipole summation. Formulae for calculating $\mathcal{V}$ as a function of the coordinates specifying the system are given in Appendix B.1. For the restricted case of overlapping spheroids, specified by the two coordinates ℓ and c, the surface energy is again expressible in a closed form, but the Coulomb energy must now be calculated by numerically evaluating a double integral. The formulae given in Appendix B.2 for the symmetric overlapping case could be readily generalized to the case where $U \neq 1/2$ and $c_1 \neq c_2$, but not to the non-axially-symmetric case.

Let us now examine the appearance of the potential-energy surfaces calculated with these formulae. Since the results can be displayed easily in at most two dimensions, we present maps of the potential energy as functions of the two coordinates specifying symmetrical fragments: the separation coordinate ℓ and the deformation coordinate c. An examination of the potential energy for this restricted case will tell us many things of interest, including the location of the symmetric saddle point. The potential energy is mapped in Fig. 3 for values of the fissionability parameter x between 0.05 and 1.0, at intervals of 0.05. Shown also in this figure is a map of the surface energy B_S and the Coulomb energy B_C, as well as an illustration of the configurations of the system for various values of the coordinates.

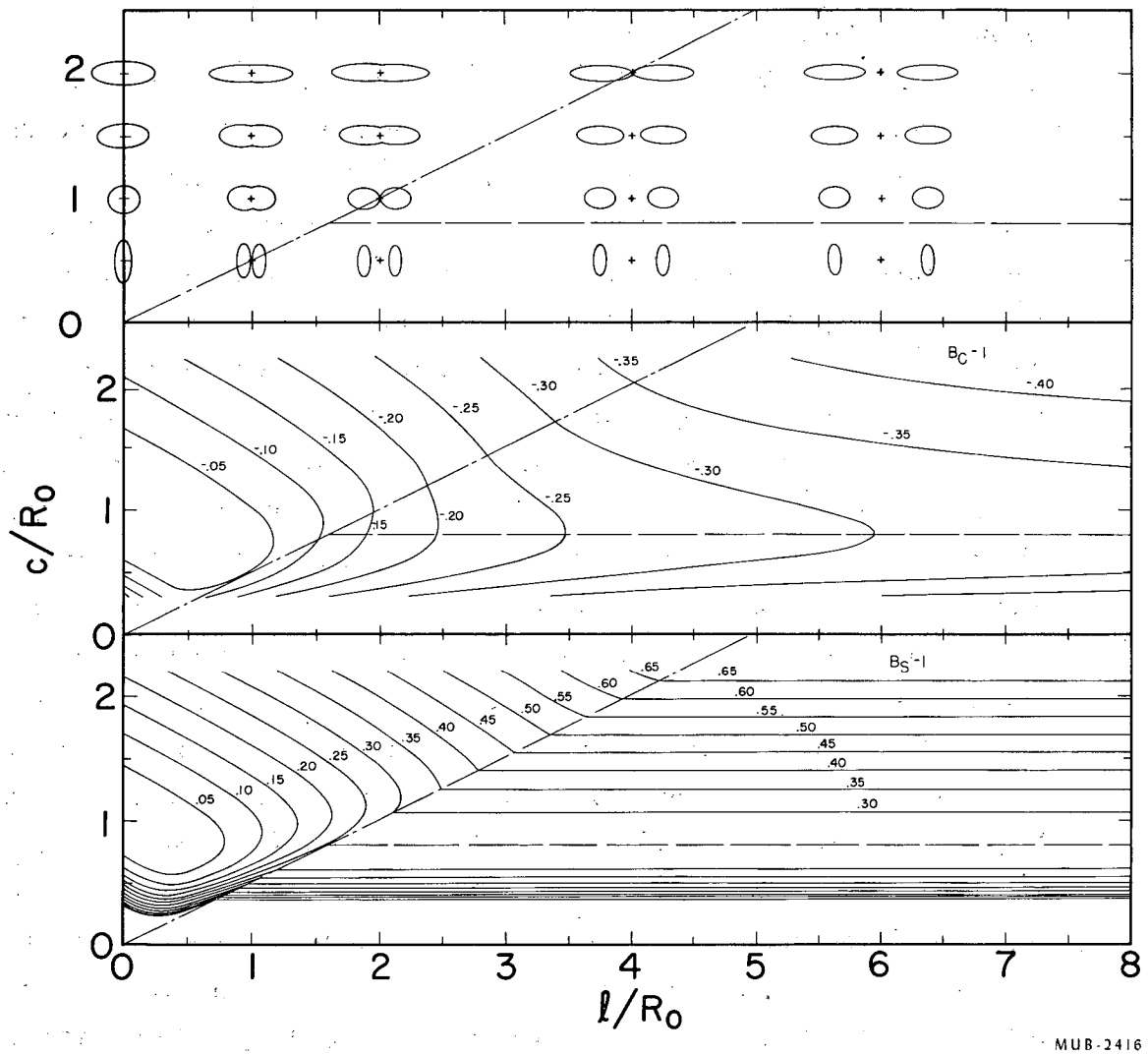

Fig. 3a. Symmetric two-spheroid configurations for selected coordinates (marked by +), Coulomb energy B_C, and surface energy B_S. The function B_C is the Coulomb energy in units of $E_C^{(0)}$, and B_S is the surface energy in units of $E_S^{(0)}$. The dot-dashed line in each case corresponds to the scission configuration of tangent spheroids ($\ell = 2c$); the long-dashed line corresponds to the configuration of two separated spheres ($c/R_0 = 1/2^{1/3}$).

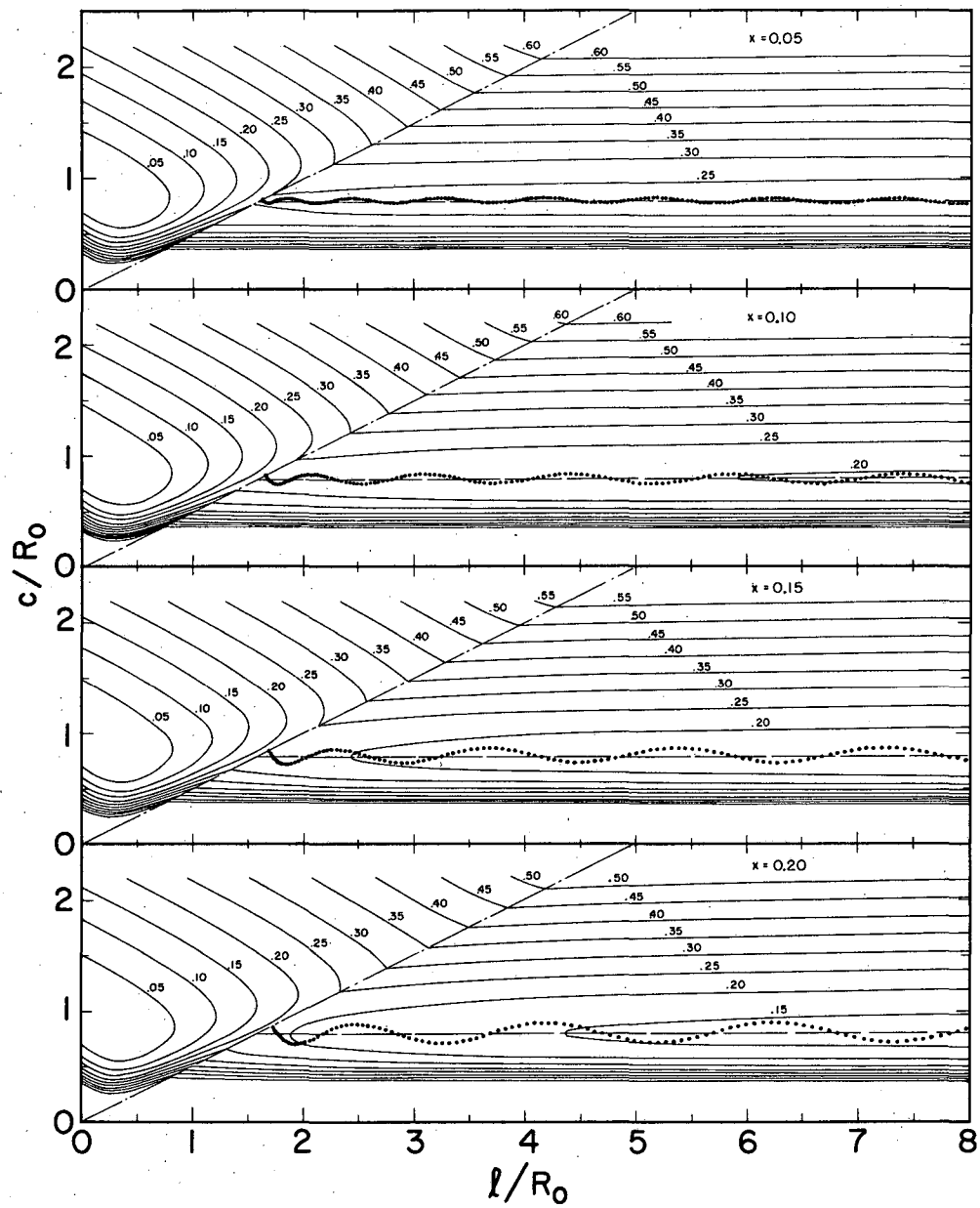

Fig. 3b. Maps of potential energy $\mathcal{V}$ for x = 0.05, 0.10, 0.15, and 0.20. The function $\mathcal{V}$ is in units of $E_S^{(0)}$. The solutions of the equations of motion corresponding to starting from rest at the saddle points are indicated by the points, which are equally spaced in time at intervals of 0.1 T_0 [see Eq. (2) for value of T_0].

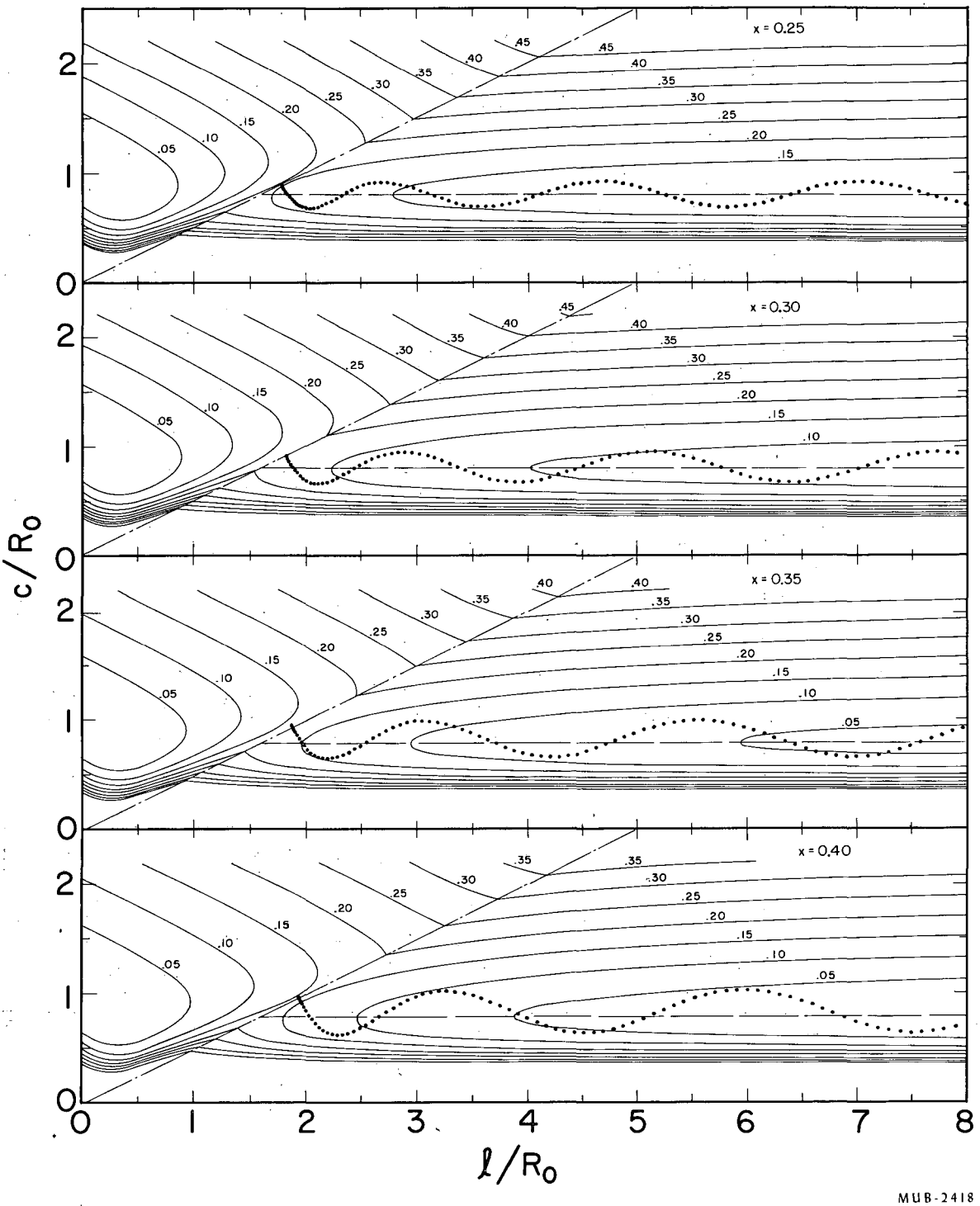

Fig. 3c. Maps of potential energy $\mathcal{V}$ for x = 0.25, 0.30, 0.35, and 0.40. The function $\mathcal{V}$ is in units of $\mathbf{E}_S^{(0)}$. The solutions of the equations of motion corresponding to starting from rest at the saddle points are indicated by the points, which are equally spaced in time at intervals of 0.1 T_0 [see Eq. (2) for value of T_0].

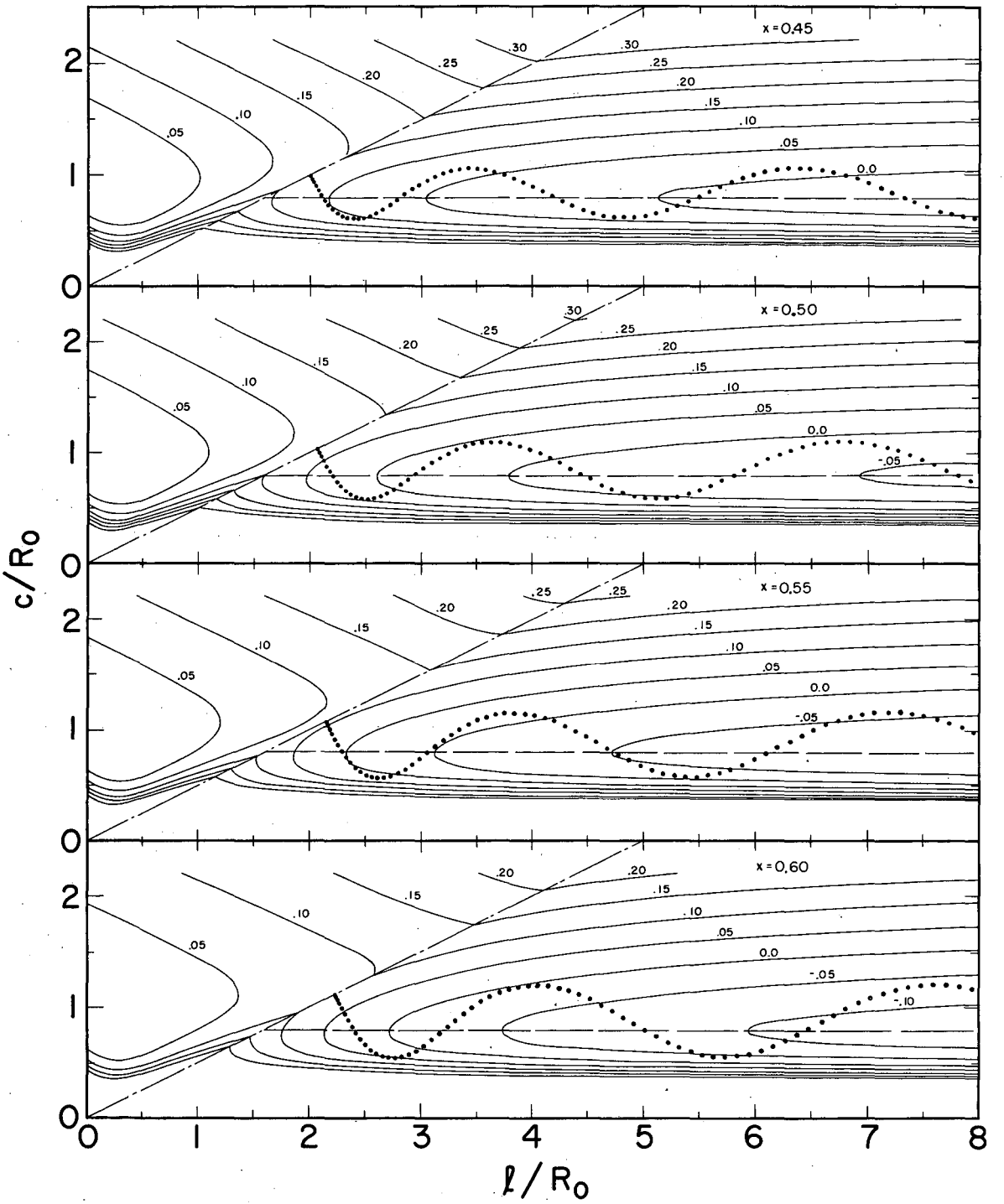

Fig. 3d. Maps of potential energy $\mathcal{V}$ for x = 0.45, 0.50, 0.55, and 0.60. The function $\mathcal{V}$ is in units of $E_S^{(0)}$. The solutions of the equations of motion corresponding to starting from rest at the saddle points are indicated by the points, which are equally spaced in time at intervals of $0.1\,T_0$ [see Eq. (2) for value of T_0].

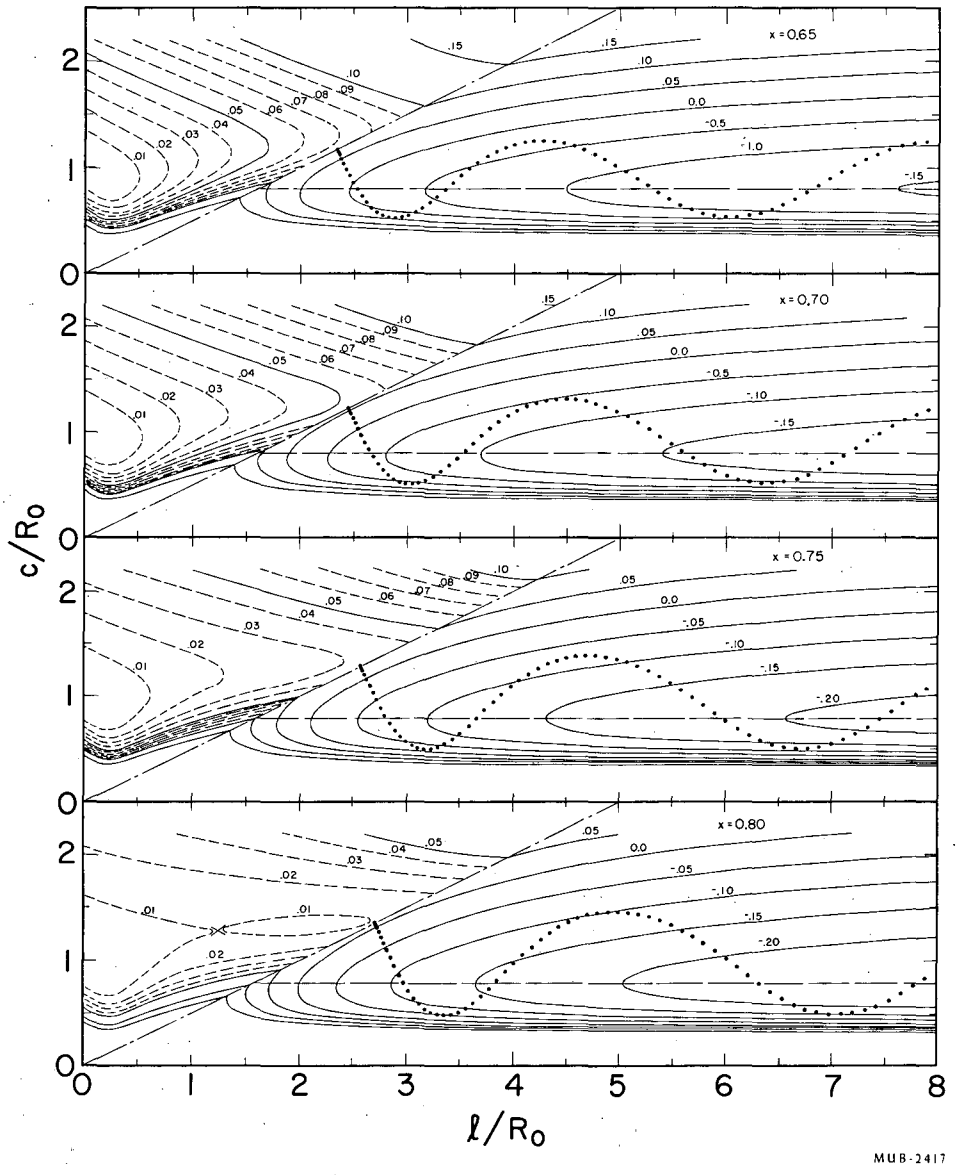

Fig. 3e. Maps of potential energy $\mathcal{V}$ for x = 0.65, 0.70, 0.75, and 0.80. The location of the overlapping saddle point for x = 0.80 is shown by the cross mark; for this value of x there are two saddles and a stable minimum point (apart from the original sphere). The function $\mathcal{V}$ is in units of $E_S^{(0)}$. The solutions of the equations of motion corresponding to starting from rest at the saddle points are indicated by the points, which are equally spaced in time at intervals of $0.1\,T_0$ [see Eq. (2) for value of T_0].

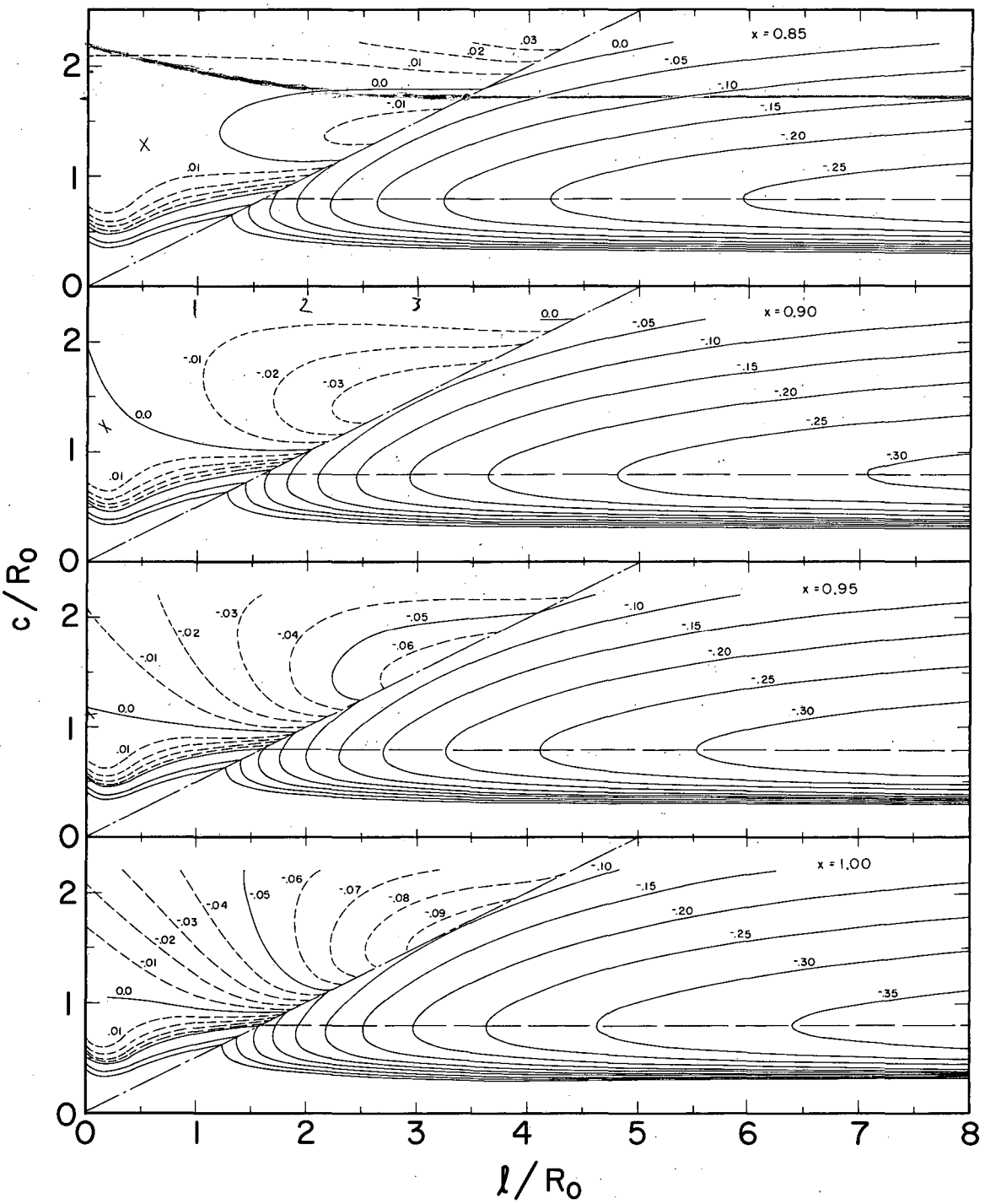

Fig. 3f. Maps of potential energy $\mathcal{V}$ for x = 0.85, 0.90, 0.95, and 1.00. The locations of the saddle points for x = 0.85, 0.90, and 0.95 are indicated by cross marks.

An expansion of the potential energy about the sphere indicates that for small distortions the contours are portions of ellipses whose major axes are tilted at angles of about 30.5° from the horizontal (see Appendix B.2). As the drop distorts in the direction of the saddle point, the potential energy increases until the saddle point is reached and then begins to decrease. For large values of x the potential-energy surface is much flatter in the overlapping region than in the separated region; the opposite is true for small values of x. The potential-energy surface in the separated region is a "trough" that drops off approximately as $1/\ell$ along the separation coordinate. At the scission line $\ell = 2c$ dividing the two regions there occurs a cusp in the energy surface.* This results from the discontinuous rate of change of the surface energy in passing from the region of overlapping spheroids, where the surface energy increases with separation, to the region of separated spheroids, where the surface energy is independent of separation.

The results of electronic computer studies of equilibrium configurations of idealized charged drops have recently become available.[15-18] It is possible, then, to compare with these essentially exact results various properties of equilibrium configurations calculated in the two-spheroid approximation. This will provide us with some idea of the

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*When all coordinates are considered, the cusp occurs at those values of the coordinates that correspond to the configuration of touching spheroids.

limitations of our model. The following properties of equilibrium configurations are compared as functions of fissionability parameter: the total potential energy $\mathcal{V}$, in Fig. 4; the individual surface and Coulomb energies B_S and B_C, in Fig. 5; the saddle-point shape of the drop, in Fig. 6; and the drop's maximum and minimum radii R_{max} and R_{min}, in Fig. 7.

There are several observations that may be made. One concerns the general appearance of the saddle-point shapes. A striking discrepancy between the true saddle-point shapes and those calculated in the two-spheroid approximation occurs in the central region of the drop (the neck). The restriction that the fragments maintain spheroidal shapes allows the drop very little freedom at its neck. In particular, a discontinuity is introduced in the surface of the drop, and the radius of the neck (R_{min}) in its dependence on x is reproduced very poorly. The two-spheroid model is thus not suitable for discussing phenomena that depend upon the properties of the neck.

In the two-spheroid approximation, when the fissionability parameter x is between 0 and 0.80, the saddle point lies on the scission line, possessing the shape of two tangent spheroids. For values of x between 0.80 and 1.0, the saddle point occurs for some value of $\ell < 2c$; i.e., the saddle-point shape is the configuration of two overlapping spheroids. The results of the exact calculations indicate that the saddle-point shapes change from dumbbell-like (approximated by two tangent spheroids) for $x \lesssim 0.67$ to cylinder-like (approximated by two overlapping spheroids) for $x \gtrsim 0.67$.[15] Thus the transition at $x \approx 0.80$ in the two-spheroid model, although qualitatively correct, occurs at a value of x that is somewhat too high.

In the short interval $0.79 \lesssim x \lesssim 0.85$ there are in the two-spheroid model three equilibrium configurations (apart from the original sphere): two saddle points and one stable minimum point.*

*This is clearly seen in Fig. 3 in the map for $x = 0.80$ but is not discernible for $x = 0.85$. The transition point is at $x = 0.80$, since it is for this value of x that the two saddles have the same energy.

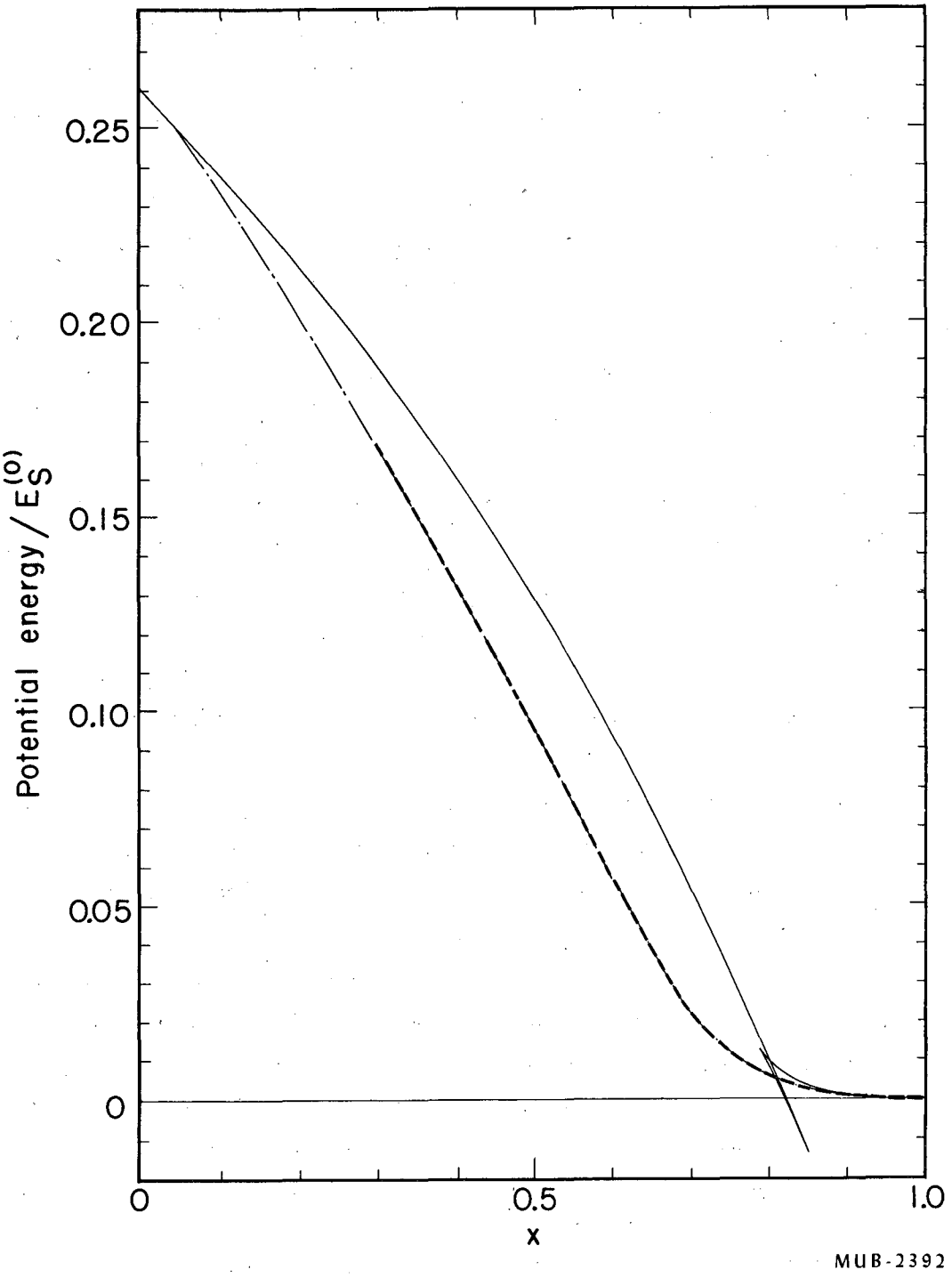

Fig. 4. The potential energy of equilibrium configurations, as a function of fissionability parameter x. The result calculated in the two-spheroid approximation is given by the solid line, the result of Cohen and Swiatecki[15] by the short-dashed line, and the result of Strutinskiĭ[17] by the dot-dashed line.

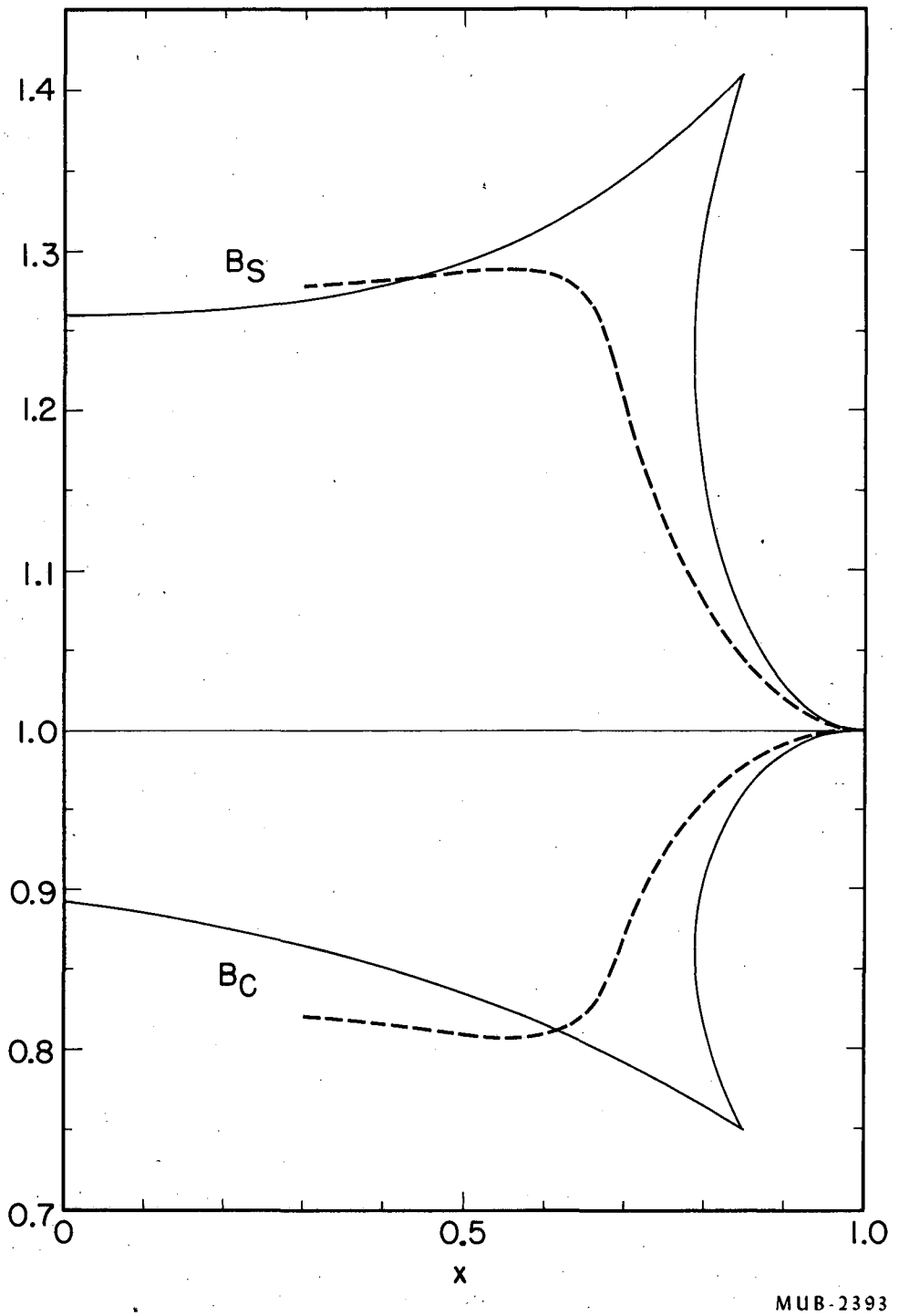

Fig. 5. The surface energy B_S and Coulomb energy B_C of equilibrium configurations, as functions of fissionability parameter x. The function B_S is the surface energy in units of $E_S^{(0)}$, and B_C is the Coulomb energy in units of $E_C^{(0)}$. The results calculated in the two-spheroid approximation are given by the solid lines, and the results of Cohen and Swiatecki[15] by the dashed lines.

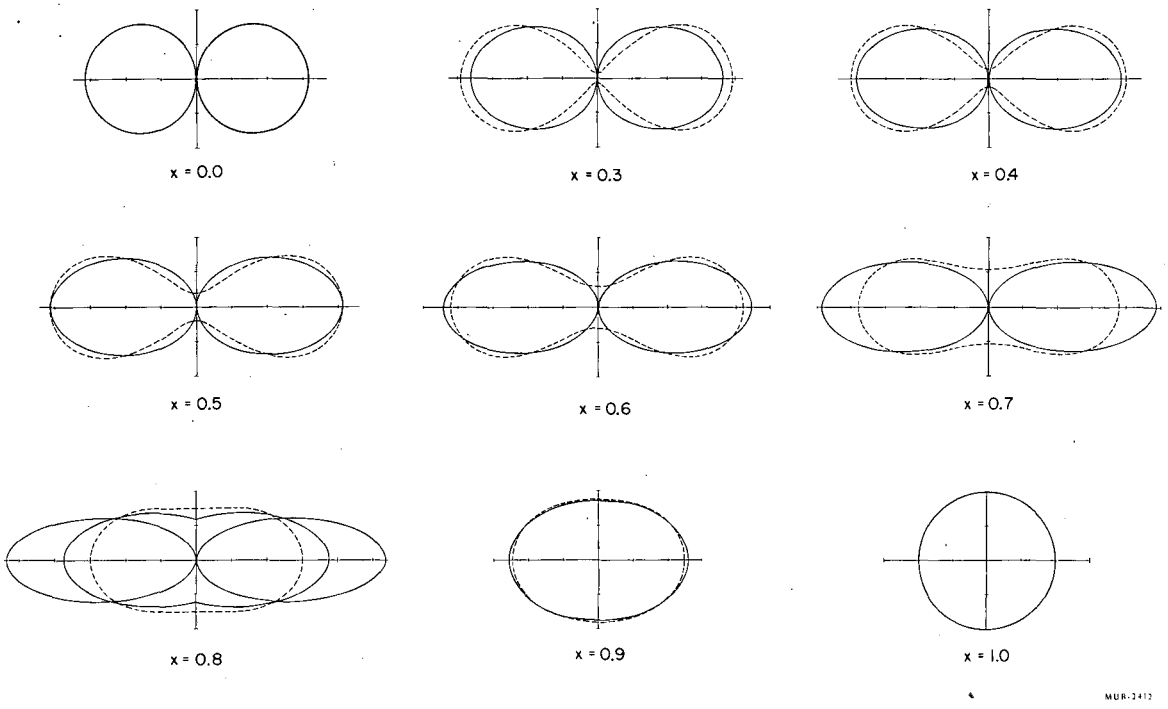

Fig. 6. Saddle-point shapes for various values of the fissionability parameter x. Shapes calculated in the two-spheroid approximation are given by the solid lines, and those calculated by Cohen and Swiatecki[15] by the dashed lines. For x = 0.8 the two saddle-point shapes occurring in the two-spheroid approximation are shown.

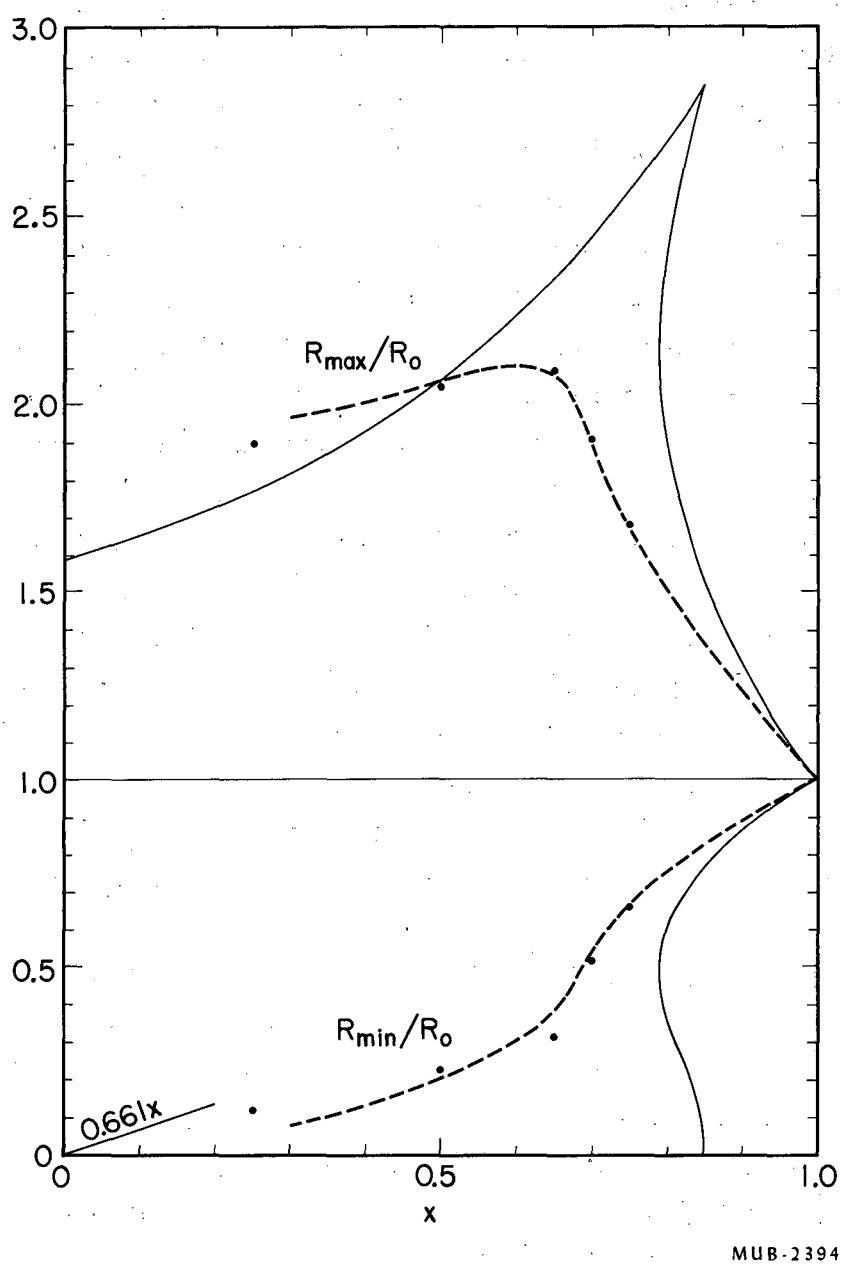

Fig. 7. Maximum and minimum radii of equilibrium configurations, as functions of fissionability parameter x. The results calculated in the two-spheroid approximation are given by the solid lines, the results of Cohen and Swiatecki[15] by the dashed lines, and the results of Strutinskiĭ[17] by the solid points. Note that the minimum radius calculated by Strutinskiĭ for x = 0.65 is appreciably lower than that of Cohen and Swiatecki. The known limiting form of R_{min}/R_0 for $x \to 0$ is indicated by the straight line.

The occurrence of three configurations of equilibrium is associated with a "dip" in the potential energy as a function of distortion. For x close to the respective transition point in both the two-spheroid approximation and in the exact calculations, the potential energy becomes extremely flat. A very slight dip occurs in the two-spheroid model, resulting in three equilibrium configurations, but does not occur in the exact calculations.*

The dependence on x of the remaining properties calculated in the two-spheroid approximation is qualitatively correct. The two-spheroid model in addition reproduces the exact results both for x close to 1.0, where the saddle-point shape is a single spheroid (a single sphere at $x = 1.0$), and at $x = 0$, where the saddle-point shape is a pair of tangent spheres. However, it is clear from the figures that for the range of x roughly between 0.67 and 0.85 the two-spheroid model represents very poorly the true saddle-point properties. Thus, for $0.67 \lesssim x \lesssim 0.85$ this model should not be expected to accurately describe phenomena associated with saddle-point shapes.† Nevertheless, the model is still useful for this range of x for discussing phenomena not related to the saddle point, such as the separation of the fragments after scission.

For $x \lesssim 0.67$ there is fair agreement between the exact saddle-point properties and those calculated in the two-spheroid approximation. The following major discrepancies should be noted. The fission barrier (saddle-point potential energy) calculated in the two-spheroid approximation is higher than the true liquid-drop barrier. This difference becomes as large as about 23 MeV, where the two-spheroid

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*Historically, however, it was for a time believed that three equilibrium configurations possibly did exist for a short range of x close to 0.7.[15,43]

†Unfortunately, most experimental information on fission is for nuclei that lie in this range of x. Some data exist, however, on the fission of nuclei where $x \lesssim 0.67$, and more are becoming available as time progresses.

barrier is roughly twice as high as the exact one. This means that the division of the total energy of the system at the saddle point into potential energy and internal excitation energy will be incorrect. The potential energy of deformation will be greater than it should be, and the internal excitation energy less by a corresponding amount.

Recall that for the lower values of x under consideration ($x \lesssim 0.67$) the saddle point lies on the scission line, where there is a cusp in the potential energy. In the direction of motion across the saddle point, then, there is a discontinuity in the rate of change of the potential energy. In particular, the second derivative, representing the (negative) "fission" stiffness constant, is not defined. Thus both the magnitude of the potential energy and the associated stiffness constant in the fission direction are not well reproduced by the two-spheroid model.

The failure of the model in these two respects (as well as its other failures, including the more drastic ones for the region $0.67 \lesssim x \lesssim 0.85$) is due to the lack of freedom of the drop at its neck. At the suggestion of Wladyslaw J. Swiatecki, we generalized the two-spheroid model by introducing a third conicoid of revolution (a hyperboloid of revolution of one or two sheets, or a spheroid) to connect the two end spheroids.* By including one additional coordinate to explicitly describe the neck degree of freedom, we found that all the saddle-point properties of Figs. 4-7 are reproduced with amazing accuracy. For example, over the entire range of x from 0 to 1.0, the fission barriers calculated in this generalization are accurate to within one-half an MeV. This is illustrated by the comparisons of saddle-point properties made in Figs. 8-11.

One would expect the two-spheroid model (without the conicoidal neck) to adequately describe those saddle-point properties not dependent upon details of the neck. The theory we develop depends in

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*Formulae for calculating the potential energy of such a system are given in Appendix B.3.

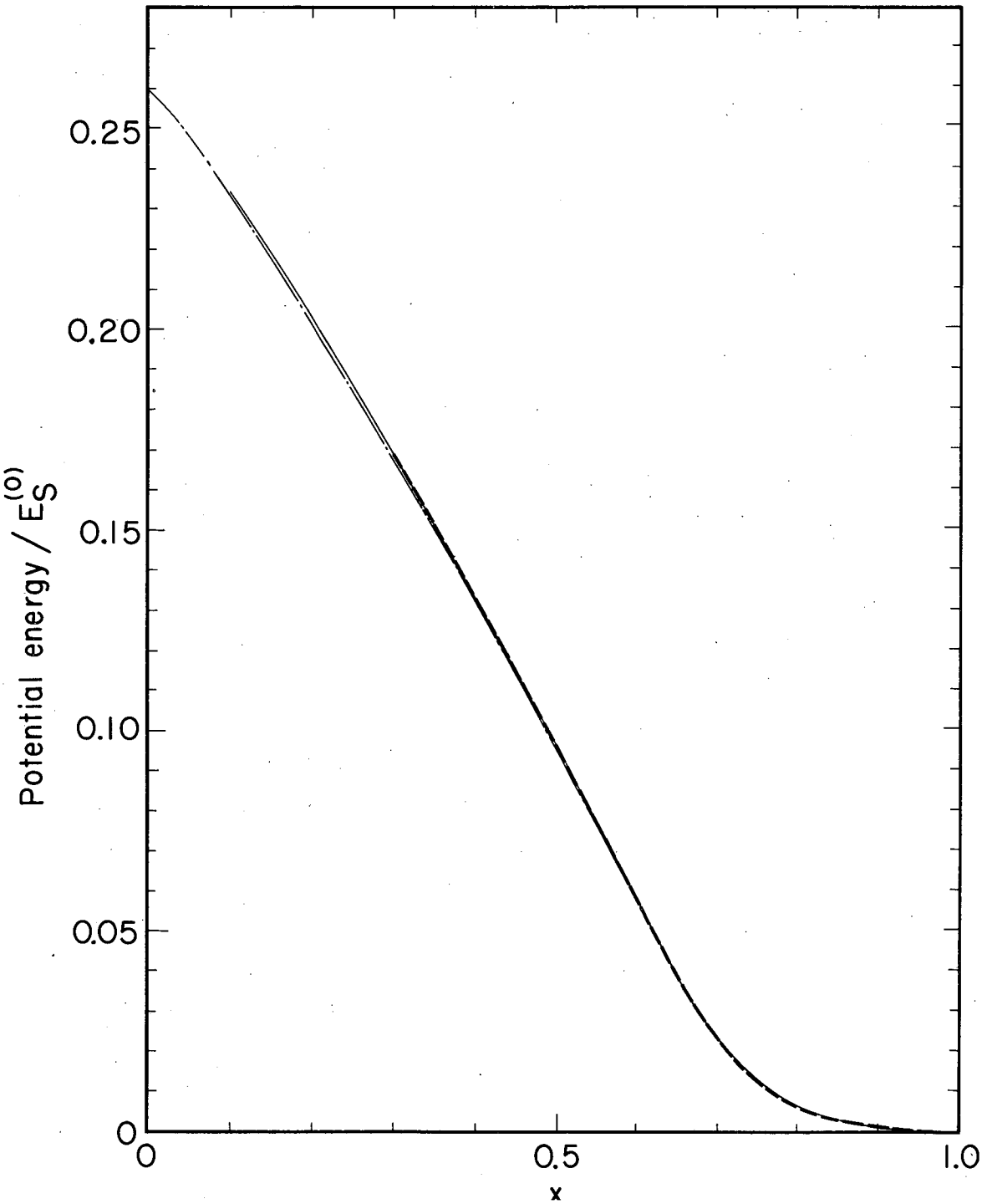

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Fig. 8. The potential energy of saddle-point shapes, as a function of fissionability parameter x. The result calculated by using two spheroids connected by a conicoid of revolution is given by the solid line, the result of Cohen and Swiatecki[15] by the short-dashed line, and the result of Strutinskiĭ[17] by the dot-dashed line.

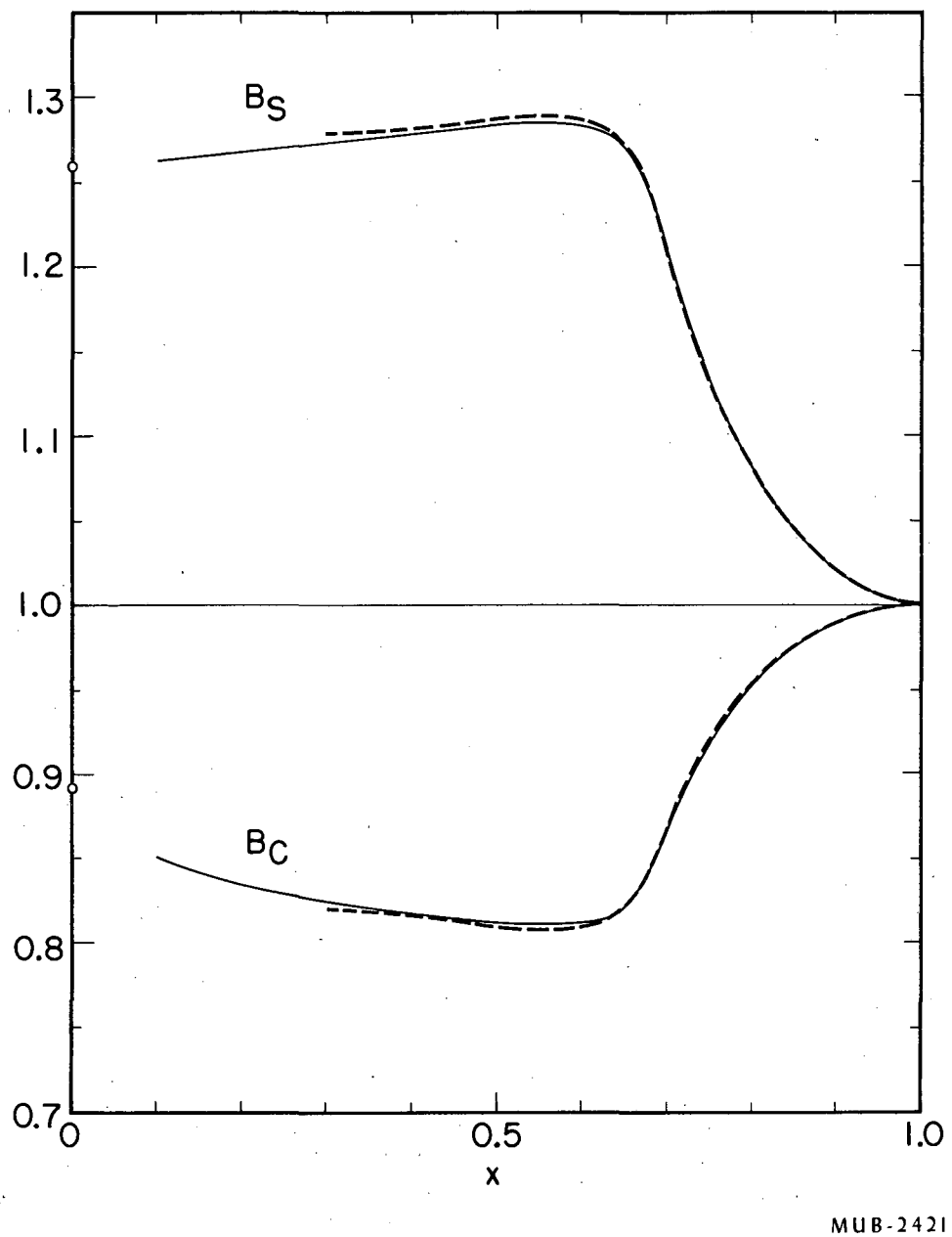

Fig. 9. The surface energy B_S and Coulomb energy B_C of saddle-point shapes, as functions of fissionability parameter x. The function B_S is the surface energy in units of $E_S^{(0)}$, and B_C is the Coulomb energy in units of $E_C^{(0)}$. The results calculated by using two spheroids connected by a conicoid of revolution are given by the solid lines, and the results of Cohen and Swiatecki[15] by the dashed lines. The known values of B_S and B_C for $x = 0$ are indicated by open circles.

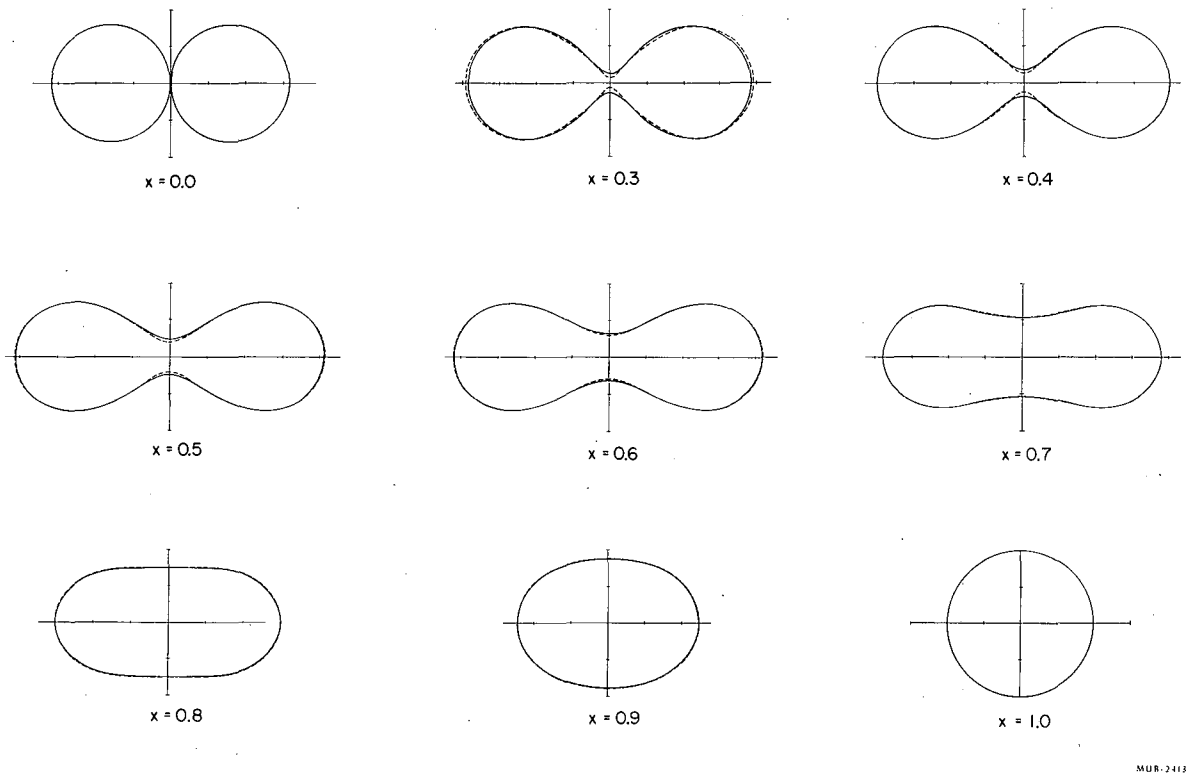

Fig. 10. Saddle-point shapes for various values of the fissionability parameter x. Shapes calculated by using two spheroids connected by a conicoid of revolution are given by the solid lines, and those calculated by Cohen and Swiatecki[15] by the dashed lines.

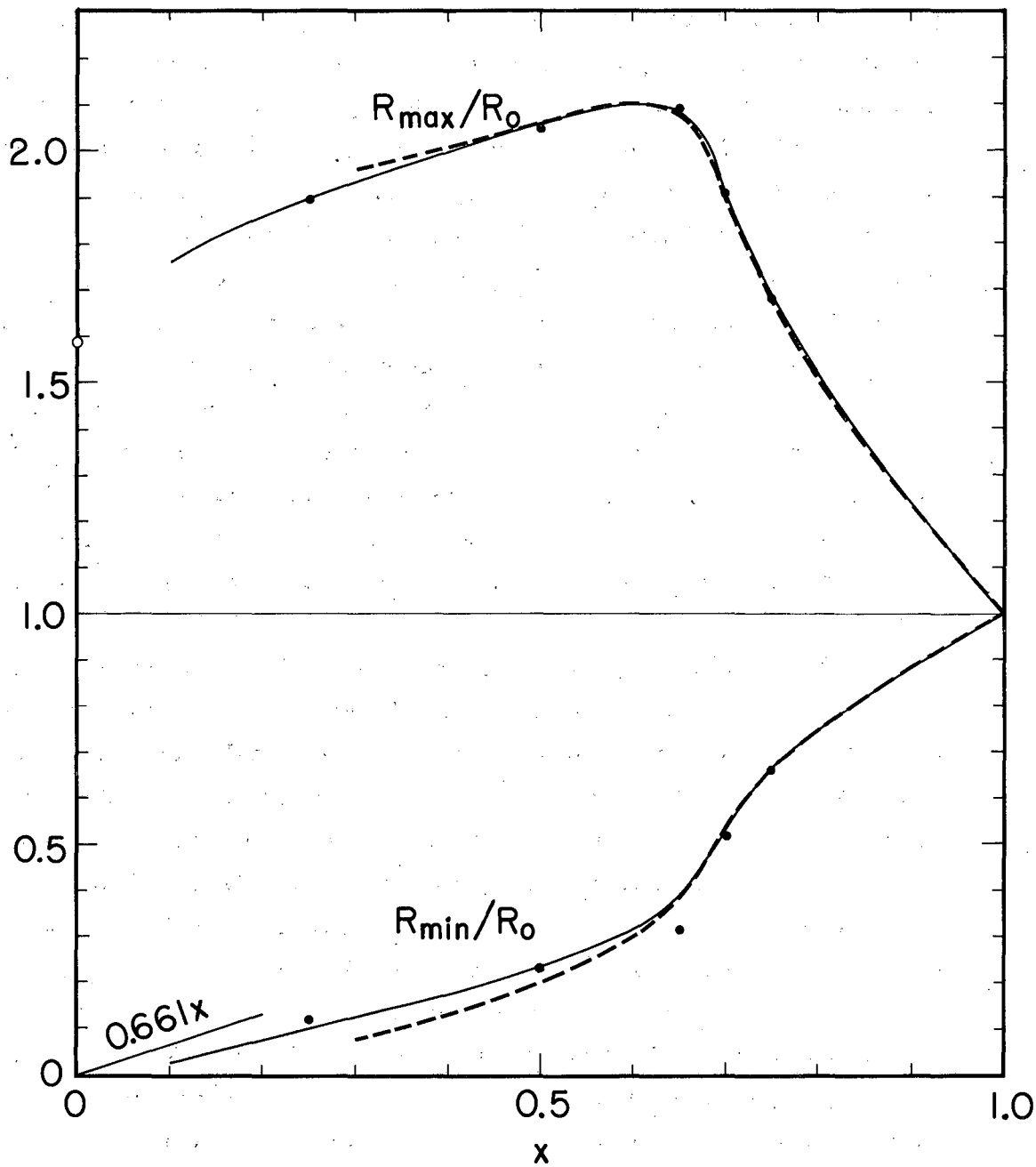

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Fig. 11. Maximum and minimum radii of saddle-point shapes, as functions of fissionability parameter x. The results calculated by using two spheroids connected by a conicoid of revolution are given by the solid line, the results of Cohen and Swiatecki[15] by the dashed lines, and the results of Strutinskiĭ[17] by the solid points. Note that the minimum radius calculated by Strutinskiĭ for $x = 0.65$ is appreciably lower than the other two results. The known limiting form of R_{min}/R_0 for $x \to 0$ is indicated by the straight line, and the value of R_{max}/R_0 for $x = 0$ by an open circle.

part upon the variation of the potential energy in the neighborhood of the saddle point—in particular, upon the second derivatives of the potential energy evaluated at the saddle point (the stiffness constants). The increase in potential energy arising from changes in the fractional mass and in the length of the fragments' axes is affected little by the presence or absence of a neck. We would thus expect the phenomena associated with the second derivatives with respect to fractional mass and semisymmetry axes—the distributions in mass, total translational kinetic energy, and excitation energies—to be described moderately well by the two-spheroid model. On the other hand, the crudeness of the neck makes the reliability of the stiffness constants for the angular coordinates less certain. We therefore do not have a clear idea of how well we should expect the distributions in fragment angular momenta (associated with the angular-coordinate stiffness constants) to be described by the model.

To summarize, we see that the two-spheroid model is inadequate in two important respects: First of all, it cannot be reliably used for $x \gtrsim 0.67$, where most of the experimental information on fission lies. Secondly, even for $x \lesssim 0.67$, it does not reproduce well those saddle-point properties associated with the neck—in particular, the fission barrier and the fission stiffness constant. It is thus unsuitable even in this range of x for discussing quantities dependent upon these properties, such as the probability of fission. On the other hand, the two-spheroid model is expected to be useful for estimating the distributions in mass, total translational kinetic energy, excitation energies, and angular momenta of the fragments at infinity. The accuracy of the estimated distributions in angular momenta is uncertain. As regards the remaining distributions, we would expect the estimates to be moderately good—better than order of magnitude—and yet certainly not exact.

III. DYNAMICS

We considered in the last section one part of the Hamiltonian for the system: the potential energy; we now turn our attention to the other part: the kinetic energy. Although the potential energy of a liquid drop is uniquely determined as soon as the drop's boundary is given, the kinetic energy, on the other hand, is not uniquely determined by specifying the time rate of change of the boundary. One must in addition make some statement regarding the nature of the hydrodynamic flow of the fluid inside the drop, i. e., the curl (rotation or vorticity) in the system must be specified. A thorough treatment of the liquid-drop model would thus require that one study the system for various types of hydrodynamic flow, consistent with one and the same motion of the boundary.

We work out the details of the theory for the case of an incompressible fluid in which vibrations of a fragment are treated as irrotational, whereas rotations are treated as if the fragment were a rigid body. This means that the hydrodynamic flow within each fragment is taken to be a superposition of an irrotational flow and a flow corresponding to a uniform rotation; i. e., the curl within each fragment is equal to twice the angular-velocity vector (which is constant throughout the fragment). (For a brief discussion of such composite types of flow, see Lamb.[44])

The type of hydrodynamic flow that we have chosen to consider is, of course, only one of several types for which the theory should be worked out. For example, the case in which the flow is completely irrotational (as regards both vibrations and rotations), and the case in which the drop is very viscous should also be considered.* The actual situation in nuclei is presumably intermediate between these limiting cases.

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*When we discuss the solutions to the equations of motion we will, whenever possible, also indicate the result for the case in which the fragments are infinitely viscous, and would therefore separate to infinity without oscillating.

It is of interest to have some a priori opinion as to how well the type of flow we are considering represents real nuclei. The deduction of vibrational inertial parameters and moments of inertia for nuclei in their ground states has been extensively considered. (See, for example, reference 45.) Estimates of the vibrational inertial parameters corresponding to quadrupole vibrations of even-even nuclei about spherical equilibrium shapes indicate that the actual values are considerably greater than the values corresponding to irrotational flow.[45] These values, however, are strongly influenced by the effects of single-particle shell structure on ground-state nuclei. For the vibrations of interest in fission—small vibrations of the system about a deformed saddle-point shape, and relatively large vibrations of the fragments about their centers following scission—the effects of single particles would be expected to be less important.

Estimates of moments of inertia corresponding to rotations of deformed even-even nuclei indicate that the actual values are greater than the values corresponding to irrotational flow, and less than the values corresponding to rigid-body rotation. As the deformation of the nuclear equilibrium configuration increases, the ratio of the observed moment of inertia to the rigid-body value increases.[45] This would imply that for the relatively large fragment deformations encountered in fission the fragment moments of inertia should not be too far from their rigid-body values.

In addition, the fragments in fission will, in general, possess some internal excitation, and, as we will discover, are typically rotating with several $\hbar$ units of angular momentum. One would expect each of these effects to increase the moment of inertia. For example, since the Coriolis force associated with rotations counteracts the pairing correlations (that are responsible for the reduction of the moment of inertia), the moment of inertia should increase with increasing angular momentum.[46] When the angular momentum of a nucleus exceeds a critical value ($\approx 12\,\hbar$ for $A \approx 180$, and $\approx 18\,\hbar$ for $A \approx 238$), the moment of inertia is predicted to equal the rigid-body value.[46]

In our dynamical study we shall be concerned with setting up and solving the classical equations of motion of the system.* This will make it possible to trace out dynamically the motion of the system from some given initial configuration near the saddle point to the final configuration of two fragments at infinity. The state of motion at infinity will correspond directly to observable characteristics of fission fragments—the speed of separation of fragment centers will correspond to their translational kinetic energies, the vibrations about their centers of mass will correspond to their excitation energies, and the rotations about their centers of mass will correspond to their angular momenta. Out of the dynamical study will come, then, the relationship between the initial configuration of the system near the saddle point and the observable quantities of interest at infinity. This relationship becomes extremely simple when the deviations of the initial configuration from the saddle point are small. (In practice, this turns out to be a good approximation.)

For the case in which the saddle point consists of two tangent spheroids ($x \lesssim 0.80$), we will find a simple equation that expresses very accurately the total translational kinetic energy E of the two fragments at infinity in terms of the initial coordinates and momenta.

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*The use of classical equations of motion for discussing the separation of the fragments to infinity may be partially justified on the grounds that a short distance from the saddle point the de Broglie wavelength for translational motion has become relatively small, that the vibrations about each fragment's center of mass involve several quanta of energy, and that the angular momentum of each fragment is typically several $\hbar$ units in magnitude.

The value of E will be seen to depend primarily upon the initial distance between the fragment centers and the product of their charges.

Since the fractional mass does not change after division into two fragments, the fractional mass U at infinity will equal its original value at the saddle point.

The excitation energy of an individual fragment at infinity will be seen to depend primarily upon its initial elongation. We will content ourselves with establishing an equation for the excitation energy at infinity that is valid to first order in the small deviations of the configuration from its saddle-point shape.

We will find that the angular momentum of an individual fragment at infinity depends primarily upon the initial angular momentum it possessed near the saddle point. Fragments that are infinitely viscous and therefore separate without oscillating are capable of acquiring some additional angular momentum by virtue of the torque exerted by one fragment on the other through the electrostatic interaction.* The amount of such induced angular momentum depends primarily upon the fragment's initial angle θ_1 and upon its initial angular momentum p_{θ_1}. Fragments that oscillate with irrotational flow as they separate acquire very little angular momentum by this mechanism because the torque decreases as the symmetry axis shortens, and even changes sign as the spheroid changes from prolate to oblate. We will establish an equation for the angular momentum at infinity that is valid to first order in the initial coordinates and momenta, similar to what we did for the excitation energy.

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*The angular momentum acquired by a rigid spheroid moving in the electrostatic field of a sphere has been discussed by Hoffman.[47]

A. Kinetic Energy of the System

We shall in this subsection concern ourselves with the kinetic energy of the system. Before launching into this undertaking, let us consider another simplification arising from the smallness of the initial deviations from the saddle point.

Because—as regards angular momenta—we are working only to first order in the initial coordinates and momenta, it is sufficient when discussing the separation of the fragments to infinity to consider explicitly only configurations in which the symmetry axes are coplanar, and in which the spheroids are not spinning about their symmetry axes. This is true because of the symmetries of the saddle-point shape. Consider, say, the x component of angular momentum of a fragment at infinity. From symmetry, it will have no linear dependence upon an initial rotation of either fragment about its y or z axis. Similarly, it will not depend linearly upon the initial y or z components of angular momentum of either fragment. Analogous statements hold for the fragment's y and z components of angular momentum at infinity. Indeed, to first order, the fragment's z component of angular momentum at infinity is equal to its initial z component of angular momentum (near the saddle point). This means, then, that to first order in the initial coordinates and momenta, the result for the general noncoplanar configuration with rotations about the spheroids' symmetry axes is simply a superposition of the results for two special cases: (a) coplanar symmetry axes with no rotations about them, and (b) rotations about collinear symmetry axes, for which we know the result.

For the sake of clarity the exposition throughout this subsection will be as if the symmetry axes of the spheroids were coplanar and there were no rotations about them. Thus the difference $\phi = \phi_1 - \phi_2$ between the azimuthal angles of the symmetry axes (see Fig. 2) is taken to be zero. The results obtained by considering coplanar spheroids with no rotations about their symmetry axes will be subsequently generalized to the original case of two-spheroid configurations

described by nine degrees of freedom (in a center-of-mass system with zero total angular momentum). It must be borne in mind, however, that if one desired to discuss the solutions to a higher order than linear in the initial angles and their conjugate momenta, then it would be necessary to set up and solve the equations of motion for the general noncoplanar configuration with rotations about the spheroids' symmetry axes.

As we found to be true with the potential energy, the kinetic energy of the system is simpler when the fragments are separated than when they are overlapping. We consider first the case in which the fragments are separated.

The total kinetic energy of the system is equal to the kinetic energy of the centers of mass of the two fragments, plus the kinetic energy of motion of each fragment relative to its own center of mass. We will discuss the terms contributing to the kinetic energy one by one. We denote by $\dot{\Theta}$ the time rate of change of the angular coordinate specifying the orientation of the line connecting the spheroid centers in the plane formed by the spheroids' symmetry axes.* The total kinetic energy of motion of the centers of mass of the two fragments is then

$$(1/2)M_\ell \dot{\ell}^2 + (1/2)M_\ell \ell^2 \dot{\Theta}^2 ,$$

where M_ℓ is the reduced mass of the two fragments, given by

$$M_\ell = U(1 - U)M_0 , \tag{7a}$$

and $\dot{\ell}$ is the derivative with respect to time of ℓ. (We shall consistently use a dot above a coordinate to denote a time derivative.)

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*The angular velocity $\dot{\Theta}$ will be expressed in terms of $\dot{\theta}_1$ and $\dot{\theta}_2$ through the requirement that the total angular momentum be zero. Each of the three angles is measured positively in the counterclockwise direction.

The kinetic energy of fragment 1 relative to its center of mass is equal to an integral over its volume of one-half the mass density times the square of the local fluid velocity (relative to its center of mass). For the type of flow we are considering, the integration can be performed exactly (see Appendix C.1), yielding

$$(1/2)M_{c_1}\dot{c}_1^{\,2} + (1/2)M_{\theta_1}(\dot{\theta}_1 + \dot{\Theta})^2 .$$

The quantity M_{c_1} is the vibrational effective mass of fragment 1, given by[44]

$$M_{c_1} = \frac{1}{5}\left(1 + \frac{1}{2}\frac{a_1^2}{c_1^2}\right)UM_0 = \frac{1}{5}\left(1 + \frac{U}{2}\frac{R_0^3}{c_1^3}\right)UM_0 , \quad (7b)$$

and M_{θ_1} is the rigid-body moment of inertia of fragment 1 for rotations about an axis through its center perpendicular to its symmetry axis, given by

$$M_{\theta_1} = \frac{1}{5}(c_1^2 + a_1^2)\,UM_0 = \frac{1}{5}\left(c_1^2 + \frac{UR_0^3}{c_1}\right)UM_0 . \quad (7c)$$

The semiaxis of fragment 1 perpendicular to its symmetry axis is denoted by a_1. The kinetic energy of fragment 1 relative to its center of mass is thus equal to the kinetic energy of vibration of the fragment plus the kinetic energy of rotation about its center of mass (with total angular velocity $\dot{\theta}_1 + \dot{\Theta}$). Because of the spheroid's symmetry, there is present no cross term involving $\dot{c}_1\dot{\theta}_1$. We note that M_{c_1} and M_{θ_1} are functions of the coordinate c_1 as well as U. Results analogous to these hold for fragment 2.

The total kinetic energy $\mathcal{T}$ of the system for the separated case in which the spheroids do not rotate about their symmetry axes, and the symmetry axes are coplanar, is then

$$\mathcal{T} = \frac{1}{2}\Big[M_\ell\dot{\ell}^2 + M_{c_1}\dot{c}_1^2 + M_{c_2}\dot{c}_2^2 + M_\ell\ell^2\dot{\Theta}^2 + M_{\theta_1}(\dot{\theta}_1 + \dot{\Theta})^2 + M_{\theta_2}(\dot{\theta}_2 + \dot{\Theta})^2\Big]. \quad (8)$$

The quantity $\dot{\Theta}$ is determined from the condition that the total angular momentum be zero. The total angular momentum L of the system is equal to the angular momentum of the centers of mass of the two fragments, plus the angular momentum of motion about the centers of mass:

$$L = M_\ell \ell^2 \dot{\Theta} + M_{\theta_1} (\dot{\theta}_1 + \dot{\Theta}) + M_{\theta_2} (\dot{\theta}_2 + \dot{\Theta}) .$$

If we substitute in (8) the result for $\dot{\Theta}$ obtained by setting $L = 0$, we find

$$\mathcal{T} = \frac{1}{2} \left[M_\ell \dot{\ell}^2 + M_{c_1} \dot{c}_1^2 + M_{c_2} \dot{c}_2^2 + M_{\theta_1} \dot{\theta}_1^2 + M_{\theta_2} \dot{\theta}_2^2 - \frac{(M_{\theta_1} \dot{\theta}_1 + M_{\theta_2} \dot{\theta}_2)^2}{M_\ell \ell^2 + M_{\theta_1} + M_{\theta_2}} \right] . \tag{9}$$

We note that for the separated case there is no term involving $\dot{U}$ in the kinetic energy. This is because once the drop has split into two fragments, the fractional volume U no longer changes with time.

From the Lagrangian for the system,

$$\mathcal{L} = \mathcal{T} - \mathcal{V} ,$$

we find that the momenta conjugate to the coordinates are

$$p_\ell = M_\ell \dot{\ell} ,$$

$$p_{c_1} = M_{c_1} \dot{c}_1 ,$$

$$p_{c_2} = M_{c_2} \dot{c}_2 ,$$

$$p_{\theta_1} = M_{\theta_1} \dot{\theta}_1 - \frac{M_{\theta_1} (M_{\theta_1} \dot{\theta}_1 + M_{\theta_2} \dot{\theta}_2)}{M_\ell \ell^2 + M_{\theta_1} + M_{\theta_2}} ,$$

$$p_{\theta_2} = M_{\theta_2} \dot{\theta}_2 - \frac{M_{\theta_2} (M_{\theta_1} \dot{\theta}_1 + M_{\theta_2} \dot{\theta}_2)}{M_\ell \ell^2 + M_{\theta_1} + M_{\theta_2}} .$$

The Hamiltonian for the system is then

$$\mathcal{H} = \mathcal{T} + \mathcal{V} , \tag{10}$$

where $\mathcal{V}$ is given by (6b), and

$$\mathcal{T} = \frac{1}{2} \left[\frac{p_\ell^2}{M_\ell} + \frac{p_{c_1}^2}{M_{c_1}} + \frac{p_{c_2}^2}{M_{c_2}} + \frac{p_{\theta_1}^2}{M_{\theta_1}} + \frac{p_{\theta_2}^2}{M_{\theta_2}} + \frac{(p_{\theta_1} + p_{\theta_2})^2}{M_\ell \, \ell^2} \right] . \tag{11}$$

When the fragments overlap, the kinetic energy of the system is much more difficult to calculate. We then specialize, as we did for the potential energy, to the restricted case of fragments that are symmetrical. The kinetic energy of the system can then be calculated approximately by using the method of Werner and Wheeler (see Appendix C.2).[48] A closed expression is obtained for the kinetic energy as a function of the coordinates ℓ and c, and their time derivatives $\dot{\ell}$ and $\dot{c}$. This method yields the exact result for two limiting cases: (1) when the system consists of two separated spheroids, and (2) for spheroidal distortions when the system consists of a single spheroid. The accuracy of the method for the general overlapping case is not known. However, for the lower values of x, where the saddle point consists of two tangent spheroids, this approximate method is never used, since we focus attention on the system only from the saddle point to infinity.

B. Transformation to Normal Coordinates

A fundamental aspect of the dynamical discussion of any physical system is the question of small oscillations about the positions

of equilibrium. In the normal fission process there are three equilibrium configurations: (1) the initial sphere, (2) the saddle point, and (3) the two spherical fragments at infinity. Both the initial sphere and the two spherical fragments at infinity are positions of stable equilibrium. The small oscillations about each of these spherical shapes can be readily discussed by expanding the drop's radius vector in spherical harmonics: The motion of the drop is a superposition of simple harmonic oscillations of different frequencies corresponding to the various spherical-harmonic distortions. The question of small oscillations about the remaining equilibrium position—the unstable configuration at the saddle point—will now be considered.

The first step in the discussion of the small oscillations about the saddle point is to transform from the original coordinate system to a system of normal coordinates.* In terms of the normal coordinates and their conjugate momenta, both the potential energy and the kinetic energy are simultaneously diagonalized at the saddle point; i.e., there appear through second order no cross terms of the form $c_1 c_2$, for example. The Hamiltonian for the system thus separates into a sum of terms, each involving only a single coordinate and its conjugate momentum. This mean physically that the motion of the system in the vicinity of the saddle point separates into several independent modes that may be discussed separately.

We consider first the case in which the saddle point consists of two tangent spheroids. The normal-coordinate transformation for this case is not completely straightforward because of the cusp in the energy surface at the saddle point. The transformation to normal coordinates can nevertheless be performed by regarding the cusp as the limiting case of a regular (rounded) barrier. As the barrier becomes infinitely sharp, all but one of the normal coordinates come to lie in the subspace

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*See the discussion of normal coordinates in any classical-mechanics textbook, for example, Goldstein.[49]

of touching spheroids. In this subspace, the potential energy has no irregularities. The remaining normal mode of motion—the fission mode—retains a well-defined direction. The physical meaning of this direction is that it represents the initial mode of motion that would be acquired by the system if started with an arbitrary infinitesimal displacement from the saddle point.

For the discussion of the normal modes we again treat the more general case in which we consider the three angular degrees of freedom of each spheroid; i.e., the spheroids are permitted to rotate about their symmetry axes, and the symmetry axes are not required to lie in the same plane. (We are still considering a system with zero total angular momentum; this will introduce one condition on these six angular coordinates.) It is convenient in discussing small-angle oscillations to lowest order to use a set of angular coordinates defined more symmetrically with respect to the coordinate axes than the Euler angles. We denote by θ_{1x} and θ_{1y} the angles of rotation of fragment 1 away from the z_1 axis, the rotations being about the x_1 and y_1 axes, respectively. The angle of rotation of spheroid 1 about the z_1 axis is denoted by θ_{1z}. The angles θ_{2x}, θ_{2y}, and θ_{2z} are defined in an analogous manner with respect to coordinate system 2.

We expand the potential energy in the subspace of touching spheroids about the saddle point, retaining terms through second order in the coordinates. Let us denote by $K_{c_1c_2}$, for example, the second partial derivative in this subspace of the potential energy with respect to c_1 and c_2, evaluated at the saddle point. (Since the saddle point is a position of equilibrium, all first derivatives are zero.) In addition to the usual equalities between the stiffness constants (the K's) obtained by interchanging the order of differentiation, symmetry considerations at the saddle point yield the following equalities between the nonzero stiffness constants:

$$K_{c_2 c_2} = K_{c_1 c_1} ,$$

$$K_{\theta_{2x}\theta_{2x}} = K_{\theta_{2y}\theta_{2y}} = K_{\theta_{1x}\theta_{1x}} = K_{\theta_{1y}\theta_{1y}} \equiv K_{\theta_1\theta_1} ,$$

$$K_{\theta_{1x}\theta_{2x}} = K_{\theta_{1y}\theta_{2y}} \equiv K_{\theta_1\theta_2} ,$$

$$K_{Uc_1} = - K_{Uc_2} .$$

In addition, with the exception of K_{UU} and $K_{c_1 c_2}$, all the remaining stiffness constants are seen from symmetry to be zero. We are left then with only six independent stiffness constants:

$$K_{UU}, \; K_{c_1 c_1}, \; K_{\theta_1\theta_1}, \; K_{Uc_2}, \; K_{c_1 c_2}, \text{ and } K_{\theta_1\theta_2} .$$

We denote the value of a coordinate at the saddle point by a superscript 0, and the difference between a coordinate and its saddle-point value as follows:

$$\delta\ell = \ell - \ell^0 = \ell - 2c_1^0 ,$$

$$\delta U = U - U^0 = U - \frac{1}{2} ,$$

$$\delta c_1 = c_1 - c_1^0 ,$$

$$\delta c_2 = c_2 - c_2^0 = c_2 - c_1^0 ,$$

$$\delta\theta_{1x} = \theta_{1x} - \theta_{1x}^0 = \theta_{1x} ,$$

$$\delta\theta_{2x} = \theta_{2x} - \theta_{2x}^{0} = \theta_{2x} ,$$

$$\delta\theta_{1y} = \theta_{1y} - \theta_{1y}^{0} = \theta_{1y} ,$$

$$\delta\theta_{2y} = \theta_{2y} - \theta_{2y}^{0} = \theta_{2y} .$$

The coordinate ℓ, which in this subspace is the distance between the centers of the touching spheroids, is chosen as the dependent variable; it is thus expressed in terms of the other coordinates (see Appendix D.1). To second order, the potential energy of the system in the vicinity of the saddle point in this subspace can be written in matrix form as

$$\mathcal{V} - \mathcal{V}^{0} = \frac{1}{2} \left(\delta U \;\; \delta c_1 \;\; \delta c_2 \;\; \delta\theta_{1x} \;\; \delta\theta_{2x} \;\; \delta\theta_{1y} \;\; \delta\theta_{2y}\right)$$

$$\times \begin{pmatrix} K_{UU} & -K_{Uc_2} & K_{Uc_2} & 0 & 0 & 0 & 0 \\ -K_{Uc_2} & K_{c_1c_1} & K_{c_1c_2} & 0 & 0 & 0 & 0 \\ K_{Uc_2} & K_{c_1c_2} & K_{c_1c_1} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & K_{\theta_1\theta_1} & K_{\theta_1\theta_2} & 0 & 0 \\ 0 & 0 & 0 & K_{\theta_1\theta_2} & K_{\theta_1\theta_1} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & K_{\theta_1\theta_1} & K_{\theta_1\theta_2} \\ 0 & 0 & 0 & 0 & 0 & K_{\theta_1\theta_2} & K_{\theta_1\theta_1} \end{pmatrix} \begin{pmatrix} \delta U \\ \delta c_1 \\ \delta c_2 \\ \delta\theta_{1x} \\ \delta\theta_{2x} \\ \delta\theta_{1y} \\ \delta\theta_{2y} \end{pmatrix} .$$

The kinetic energy in the neighborhood of the saddle point, to second order in the coordinates and velocities (or momenta), is given by*

$$\mathcal{T} = \frac{1}{2}\left\{M_\ell^0\,\dot{\ell}^2 + M_{c_1}^0\left(\dot{c}_1^2 + \dot{c}_2^2\right) + M_{\theta_1}^0\left(\dot{\theta}_{1x}^2 + \dot{\theta}_{2x}^2 + \dot{\theta}_{1y}^2 + \dot{\theta}_{2y}^2\right)\right.$$

$$\left. - \frac{\left(M_{\theta_1}^0\right)^2\left[\left(\dot{\theta}_{1x} + \dot{\theta}_{2x}\right)^2 + \left(\dot{\theta}_{1y} + \dot{\theta}_{2y}\right)^2\right]}{M_\ell^0\,(2c_1^0)^2 + 2M_{\theta_1}^0} + M_{\theta_{1z}}^0\left(\dot{\theta}_{1z}^2 + \dot{\theta}_{2z}^2\right)\right\},$$

$$= \frac{1}{2}\left[\frac{p_\ell^2}{M_\ell^0} + \frac{p_{c_1}^2 + p_{c_2}^2}{M_{c_1}^0}\right.$$

$$+ \frac{p_{\theta_{1x}}^2 + p_{\theta_{2x}}^2 + p_{\theta_{1y}}^2 + p_{\theta_{2y}}^2}{M_{\theta_1}^0}$$

$$+ \frac{\left(p_{\theta_{1x}} + p_{\theta_{2x}}\right)^2 + \left(p_{\theta_{1y}} + p_{\theta_{2y}}\right)^2}{M_\ell^0\,(2c_1^0)^2}$$

$$\left. + \frac{p_{\theta_{1z}}^2 + p_{\theta_{2z}}^2}{M_{\theta_{1z}}^0}\right],$$

where the effective masses are now constant:

$$M_\ell^0 = \frac{1}{4}\,M_0\,,$$

$$M_{c_1}^0 = \frac{1}{10}\left[1 + \frac{1}{4}\left(\frac{R_0}{c_1^0}\right)^3\right]M_0\,,$$

---

*When discussing the kinetic energy, we are not restricted to the subspace of touching spheroids.

$$M_{\theta_1}^{0} = \frac{1}{10}\left[(c_1^0)^2 + \frac{R_0^3}{2c_1^0}\right] M_0 ,$$

$$M_{\theta_{1z}}^{0} = \frac{1}{5}(a_1^0)^2 M_0 = \frac{1}{10}\left(\frac{R_0^3}{c_1^0}\right) M_0 .$$

This result is obtained by evaluating at the saddle point the effective masses (7) of our previous expressions for $\mathcal{T}$ [Eqs. (9) and (11)], and generalizing the results to include general rotations of the spheroids about the x, y, and z axes (rather than in a plane). The requirement that the total angular momentum be zero introduces the condition that $\dot{\theta}_{2z} = -\dot{\theta}_{1z}$ (or $p_{\theta_{2z}} = -p_{\theta_{1z}}$).

Before listing the normal-coordinate transformation formulae, let us describe physically the normal modes of oscillation about the saddle-point configuration. We have tried to descriptively name each mode; the corresponding normal coordinate is designated by the first letter of the name. Figure 12 has been prepared to aid in visualizing the modes, which we now enumerate:

(a) Fission: This normal mode is distinguished from the others in that it is always unstable—the potential energy decreases rather than increases as we move away from the saddle point. Because of the cusp in the potential energy the stiffness constant for this mode is not defined; it would be negative for a saddle point in which there is no cusp. The motion of the system is a simultaneous separation of the centers of the spheroids and a decrease in their elongations, or vice versa.

(b) Mass-asymmetry: As will be discussed later, this normal mode is stable for x greater than x_{BG} (equal to 0.47 in the two-spheroid approximation), and unstable for x less than x_{BG}. The motion here is an increase in both the mass (volume) and elongation of one spheroid and a decrease in the mass and elongation of the other

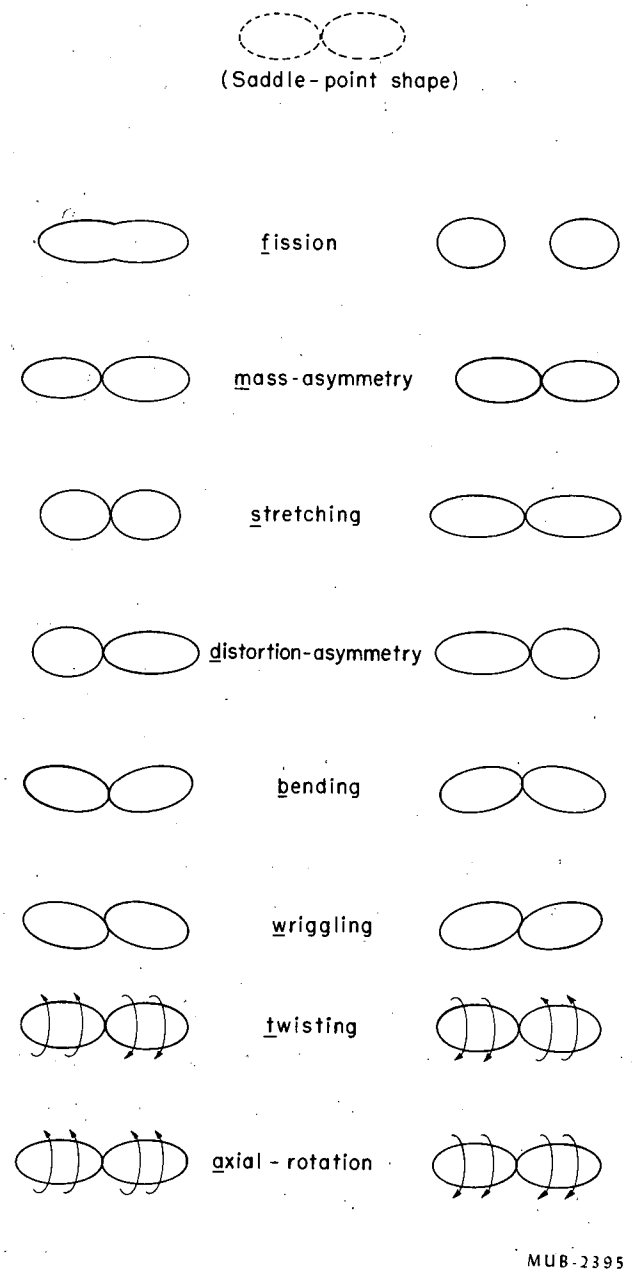

Fig. 12. Illustration of normal modes of oscillation about the saddle-point shape (when the saddle point is represented by two tangent spheroids). The fission mode is always unstable; the mass asymmetry mode is unstable for $x < x_{BG}$, and stable for $x > x_{BG}$. The bending and wriggling modes are each doubly degenerate, corresponding to rotations in two perpendicular planes. The twisting and axial-rotation modes consist of uniform (rather than oscillatory) rotation; for a system with zero total angular momentum the axial-rotation mode is not excited.

spheroid, such that the distance between spheroid centers (as well as the total length of the drop) is constant.

(c) Stretching: The elongation of each spheroid increases (or decreases) by the same amount.

(d) Distortion-asymmetry: The elongation of one spheroid increases, and the elongation of the other spheroid decreases; the distance between spheroid centers (as well as the total length of the drop) remains constant.

(e) Bending: This mode is doubly degenerate (occurs twice), corresponding to rotations in the x-z plane and the y-z plane. For a given plane, one of the spheroids rotates clockwise about an axis through its center perpendicular to the plane, and the other counterclockwise through the same angle; the spheroids remain touching.

(f) Wriggling: This mode is also doubly degenerate, corresponding to rotations in the x-z and y-z planes. For a given plane, both spheroids rotate through the same angle either clockwise or counterclockwise about axes through their centers perpendicular to the plane; they remain touching. The entire system rotates in the opposite direction, ensuring conservation of the x and y components of total angular momentum.

(g) Twisting: One of the spheroids rotates about its symmetry axis clockwise and the other counterclockwise through the same angle. The restoring force (stiffness constant) for this mode is zero, resulting in a uniform, rather than oscillatory, rotation.

(h) Axial-rotation: This mode would correspond to a uniform rotation of the spheroids about their symmetry axes through the same angle in the same direction. Conservation of the z component of total angular momentum means that this mode is not excited.

The linear transformation that takes us from the original coordinates to the normal coordinates may be written as

$$f = \frac{\delta\ell - (\delta c_1 + \delta c_2)}{\sqrt{2}\,[1 + (2M_{c_1}^{0}/M_0)]} ,$$

$$m = \delta U ,$$

$$s = \frac{\delta\ell + 2(M_{c_1}^{0}/M_0)(\delta c_1 + \delta c_2)}{\sqrt{2}\,[1 + (2M_{c_1}^{0}/M_0)]} ,$$

$$d = (1/\sqrt{2})(-B\,\delta U + \delta c_1 - \delta c_2) ,$$

$$b_x = (1/\sqrt{2})(\delta\theta_{1x} - \delta\theta_{2x}) ,$$

$$w_x = (1/\sqrt{2})(\delta\theta_{1x} + \delta\theta_{2x}) ,$$

$$b_y = (1/\sqrt{2})(\delta\theta_{1y} - \delta\theta_{2y}) ,$$

$$w_y = (1/\sqrt{2})(\delta\theta_{1y} + \delta\theta_{2y}) ,$$

$$t = (1/\sqrt{2})(\delta\theta_{1z} - \delta\theta_{2z}) ,$$

$$a = (1/\sqrt{2})(\delta\theta_{1z} + \delta\theta_{2z}) ,$$

where the quantity B is defined by*

$$B = 2K_{Uc_2}/(K_{c_1c_1} - K_{c_1c_2}) .$$

---

*We will see later (Fig. 13) that K_{Uc_2} is always positive and that $K_{c_1c_1}$ is always greater than $K_{c_1c_2}$; hence, B is always positive.

These formulae may be verified by explicitly demonstrating that both $\mathcal{V}$ and $\mathcal{T}$ are diagonalized in terms of the new coordinates and their time derivatives (or conjugate momenta).* The normal coordinates are all identically zero at the saddle point. The inverse transformation is given by

$$\delta\ell = \sqrt{2}\ [\ 2(M_{c_1}^{0} / M_0)\ f + s\ ]\ ,$$

$$\delta U = m\ ,$$

$$\delta c_1 = (1/2)\ Bm + (1/\sqrt{2})\ (-f + s + d)\ ,$$

$$\delta c_2 = -\ (1/2)\ Bm + (1/\sqrt{2})\ (-f + s - d)\ ,$$

$$\delta\theta_{1x} = (1/\sqrt{2})\ (b_x + w_x)\ ,$$

$$\delta\theta_{2x} = (1/\sqrt{2})\ (-b_x + w_x)\ ,$$

$$\delta\theta_{1y} = (1/\sqrt{2})\ (b_y + w_y)\ ,$$

$$\delta\theta_{2y} = (1/\sqrt{2})\ (-b_y + w_y)\ ,$$

$$\delta\theta_{1z} = (1/\sqrt{2})\ (t + a)\ ,$$

$$\delta\theta_{2z} = (1/\sqrt{2})\ (-t + a)\ .$$

---

*In verifying that $\mathcal{T}$ is diagonal in terms of the time derivatives of the normal coordinates, note that $\dot{U}$ is zero at the saddle point.

In terms of the normal coordinates, the potential energy in the subspace of touching spheroids is, to second order,

$$\mathcal{V} - \mathcal{V}^0 = \frac{1}{2}\left[K_m\, m^2 + K_s\, s^2 + K_d\, d^2 + K_b\,(b_x^2 + b_y^2) + K_w\,(w_x^2 + w_y^2)\right],$$

where the normal-coordinate stiffness constants are expressed in terms of the original stiffness constants by

$$K_m = K_{UU} - BK_{Uc_2},$$

$$K_s = K_{c_1c_1} + K_{c_1c_2},$$

$$K_d = K_{c_1c_1} - K_{c_1c_2},$$

$$K_b = K_{\theta_1\theta_1} - K_{\theta_1\theta_2},$$

$$K_w = K_{\theta_1\theta_1} + K_{\theta_1\theta_2}.$$

The kinetic energy is similarly given to second order by

$$\mathcal{T} = \frac{1}{2}\left[M_f\,\dot{f}^2 + M_s\,\dot{s}^2 + M_d\,\dot{d}^2 + M_b\,(\dot{b}_x^2 + \dot{b}_y^2) + M_w\,(\dot{w}_x^2 + \dot{w}_y^2) + M_t\,\dot{t}^2 + M_a\,\dot{a}^2\right]$$

$$= \frac{1}{2}\left(\frac{p_f^2}{M_f} + \frac{p_s^2}{M_s} + \frac{p_d^2}{M_d} + \frac{p_{b_x}^2 + p_{b_y}^2}{M_b} + \frac{p_{w_x}^2 + p_{w_y}^2}{M_w} + \frac{p_t^2}{M_t} + \frac{p_a^2}{M_a}\right),$$

where the normal-coordinate effective masses are related to the original ones by

$$M_f = \left(1 + \frac{2M_{c_1}^{0}}{M_0}\right) M_{c_1}^{0} ,$$

$$M_s = \frac{1}{2}\left(1 + \frac{2M_{c_1}^{0}}{M_0}\right) M_0 ,$$

$$M_d = M_{c_1}^{0} ,$$

$$M_b = M_{\theta_1}^{0} ,$$

$$M_w = \left[1 - \frac{2M_{\theta_1}^{0}}{M_0 (c_1^0)^2 + 2M_{\theta_1}^{0}}\right] M_{\theta_1}^{0}$$

$$= \left[\frac{5}{6 + \frac{R_0^3}{2(c_1^0)^3}}\right] M_{\theta_1}^{0} ,$$

$$M_t = M_{\theta_{1z}}^{0} ,$$

$$M_a = M_{\theta_{1z}}^{0} ,$$

and the momenta conjugate to the normal coordinates are

$$p_f = M_f \dot{f} ,$$

$$p_s = M_s \dot{s} ,$$

$$p_d = M_d \dot{d} ,$$

$$p_{b_x} = M_b \dot{b}_x ,$$

$$p_{b_y} = M_b \dot{b}_y ,$$

$$p_{w_x} = M_w \dot{w}_x ,$$

$$p_{w_y} = M_w \dot{w}_y ,$$

$$p_t = M_t \dot{t} ,$$

$$p_a = M_a \dot{a} .$$

The Hamiltonian (with respect to the saddle point) in the subspace of touching spheroids is then given to second order by (the axial-rotation kinetic-energy term has now been dropped)

$$\mathcal{H} = \frac{1}{2} \left\{ K_m m^2 + \left(K_s s^2 + \frac{p_s^2}{M_s} \right) + \left(K_d d^2 + \frac{p_d^2}{M_d} \right) + \left[K_b (b_x^2 + b_y^2) + \frac{p_{b_x}^2 + p_{b_y}^2}{M_b} \right] + \left[K_w (w_x^2 + w_y^2) + \frac{p_{w_x}^2 + p_{w_y}^2}{M_w} \right] + \frac{p_t^2}{M_t} \right\} . \tag{12}$$

The frequency for a particular normal-mode oscillation is given by the square root of the appropriate stiffness constant divided by the appropriate mass. There are four well-defined nonzero frequencies (the bending and wriggling frequencies are, of course, doubly degenerate):

$$\omega_s = (K_s/M_s)^{1/2} , \tag{13a}$$

$$\omega_d = (K_d/M_d)^{1/2} , \tag{13b}$$

$$\omega_b = (K_b/M_b)^{1/2} , \tag{13c}$$

$$\omega_w = (K_w/M_w)^{1/2} . \tag{13d}$$

Since the (negative) fission stiffness constant K_f is not defined, the (imaginary) frequency ω_f is also not defined. It is shown in Appendix D.2 that, because the drop's minimum radius is zero, the mass-asymmetry effective mass M_m is infinite.* The corresponding frequency ω_m is therefore zero.† The twisting frequency ω_t is also zero, because the stiffness constant K_t is zero.

A straightforward numerical method was used to locate the saddle point and calculate the stiffness constants (see Appendix G). The results are presented as functions of fissionability parameter from 0 to 0.80 in a series of graphs. The value of c_1^0 defining the saddle-point shape (when the saddle point consists of two tangent spheroids) can be obtained from the R_{max} curve in Fig. 7 ($R_{max} = 2\ c_1^0$). The stiffness constants for the original coordinate system are given in Fig. 13.

The normal-coordinate stiffness constants are shown in Fig. 14. Four of these constants are always positive; the mass-asymmetry stiffness constant K_m changes sign at $x = x_{BG} \approx 0.47$. This value of x is the two-spheroid approximation to the true value of 0.39_4, where the Businaro-Gallone family of asymmetric equilibrium shapes bifurcates from the family of symmetric equilibrium shapes.[15,43]‡

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*This result states physically that the kinetic energy associated with a finite flux of matter through an aperature of infinitesimal radius is infinite. This follows from the fact that the velocity of flow through the aperture is infinite, and that the kinetic energy depends upon the product of the amount of matter and the square of the velocity.

†The reason for the vanishing of ω_m is that in the two-spheroid model the neck radius of the saddle-point shape is zero. For the exact saddle point the neck radius is not zero but is small, and ω_m is not zero but is small in comparison with the other frequencies.

‡The two-spheroid model thus predicts this bifurcation point at a value of x that is somewhat too high, in analogy to its prediction of a transition region at $x \approx 0.80$, when the true transition region occurs at $x \approx 0.67$.

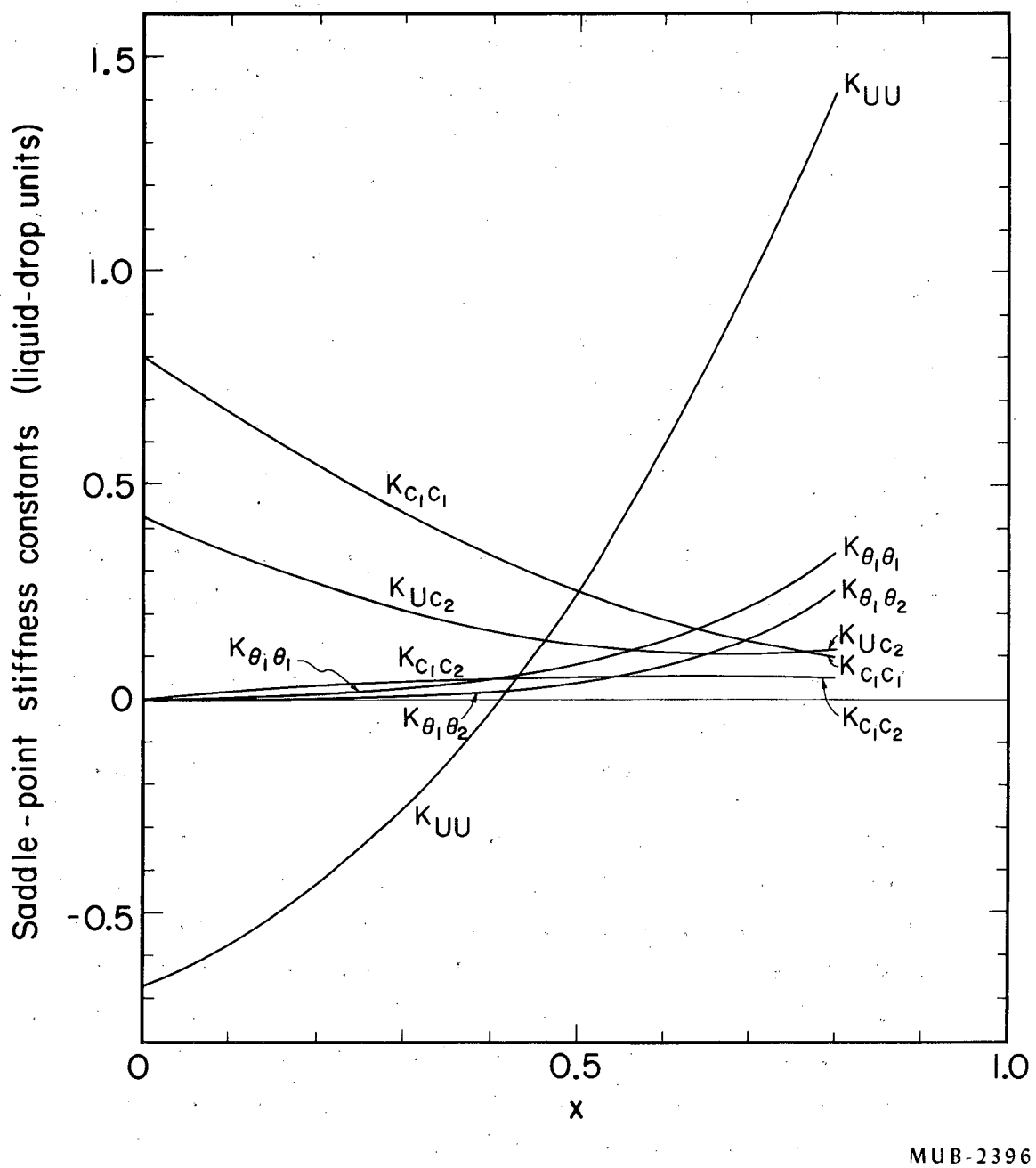

Fig. 13. Saddle-point stiffness constants (second derivatives of the potential energy, evaluated at the saddle point) for the original coordinate system, as functions of fissionability parameter x. The constant K_{UU} is in units of $E_S^{(0)}$, $K_{c_1c_1}$ and $K_{c_1c_2}$ are in units of $E_S^{(0)}/R_0^2$, K_{Uc_2} is in units of $E_S^{(0)}/R_0$, and $K_{\theta_1\theta_1}$ and $K_{\theta_1\theta_2}$ are in units of $E_S^{(0)}/\mathrm{rad}^2$.

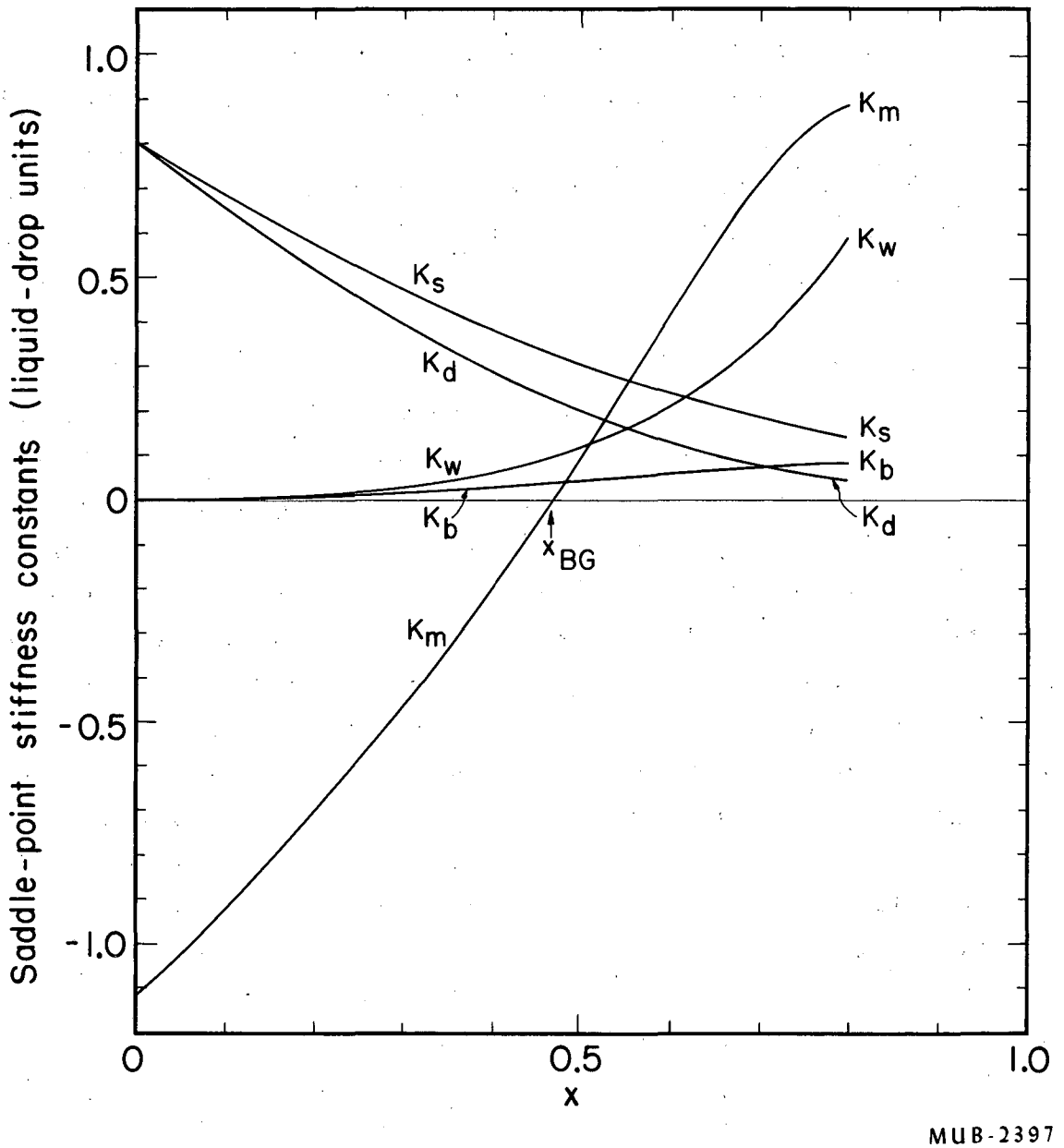

Fig. 14. Saddle-point stiffness constants (second derivatives of the potential energy, evaluated at the saddle point) for the normal-coordinate system, as functions of fissionability parameter x. The constant K_m is in units of $E_S^{(0)}$, K_s and K_d are in units of $E_S^{(0)}/R_0^2$, and K_b and K_w are in units of $E_S^{(0)}/\text{rad}^2$. The two-spheroid approximation to the Businaro-Gallone value of x is indicated by the arrow.

For $x > x_{BG}$ the symmetric equilibrium shapes are stable against changes in asymmetry, and for $x < x_{BG}$ they are unstable. Thus, for $x > x_{BG}$ the symmetric equilibrium shapes are unstable with respect to displacements along only one normal coordinate (the fission coordinate), and for $x < x_{BG}$ they are unstable with respect to two (the fission coordinate and the mass-asymmetry coordinate). This means physically that for $x > x_{BG}$ the equilibrium configuration defines a barrier energy, whereas for $x < x_{BG}$ the equilibrium configuration is at the "top of a hill" and therefore does not define a barrier energy.[15,43]

The effective masses are shown in Fig. 15. The frequencies for the four nonzero modes of oscillation are given in Fig. 16 in liquid-drop units. In Fig. 17 the corresponding quantum energies ($\hbar$ times the frequencies) are given in units of MeV for nuclei along the line of beta stability.[50] For the region in which we will be most interested ($x \approx 0.67$), the stretching and bending quantum energies are each about 1 MeV, the distortion-asymmetry quantum energy is about 1.5 MeV, and the wriggling quantum energy is about 2 MeV.

For values of $x \gtrsim 0.80$, where the saddle point is represented by overlapping spheroids, we again restrict ourselves to the case of symmetrical fragments, specified by the two coordinates ℓ and c. Since there is no cusp in the potential energy at the saddle point, the normal-coordinate transformation for this case is straightforward. There result two normal modes: a stable oscillation (stretching) and an unstable motion in the fission direction. We shall not present here the numerical results obtained for the frequencies of the normal modes.

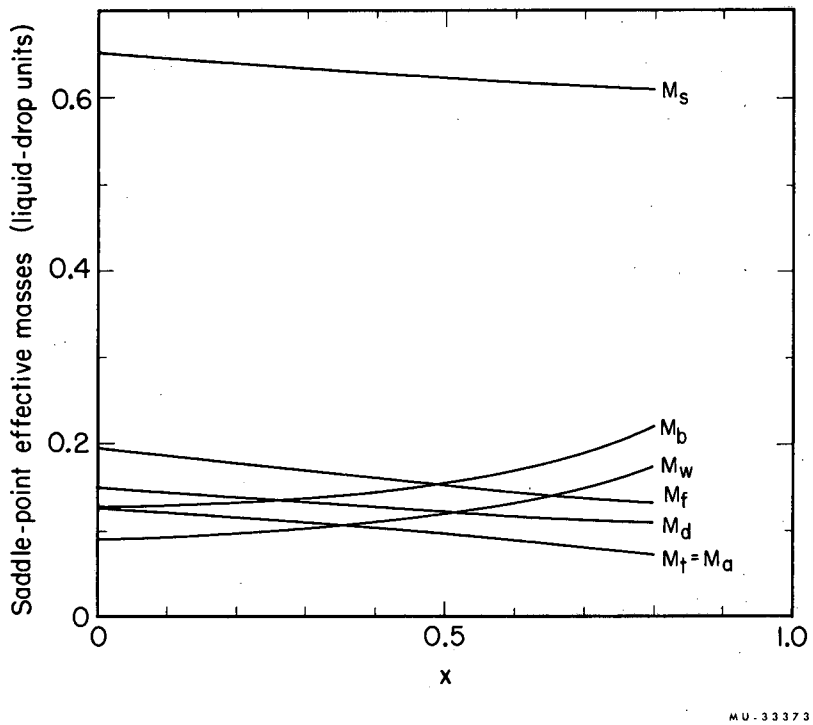

Fig. 15. Saddle-point effective masses, as functions of fissionability parameter x. The masses M_s, M_d, and M_f are in units of M_0; and M_b, M_w, M_t, and M_a are in units of $M_0 R_0^2$.

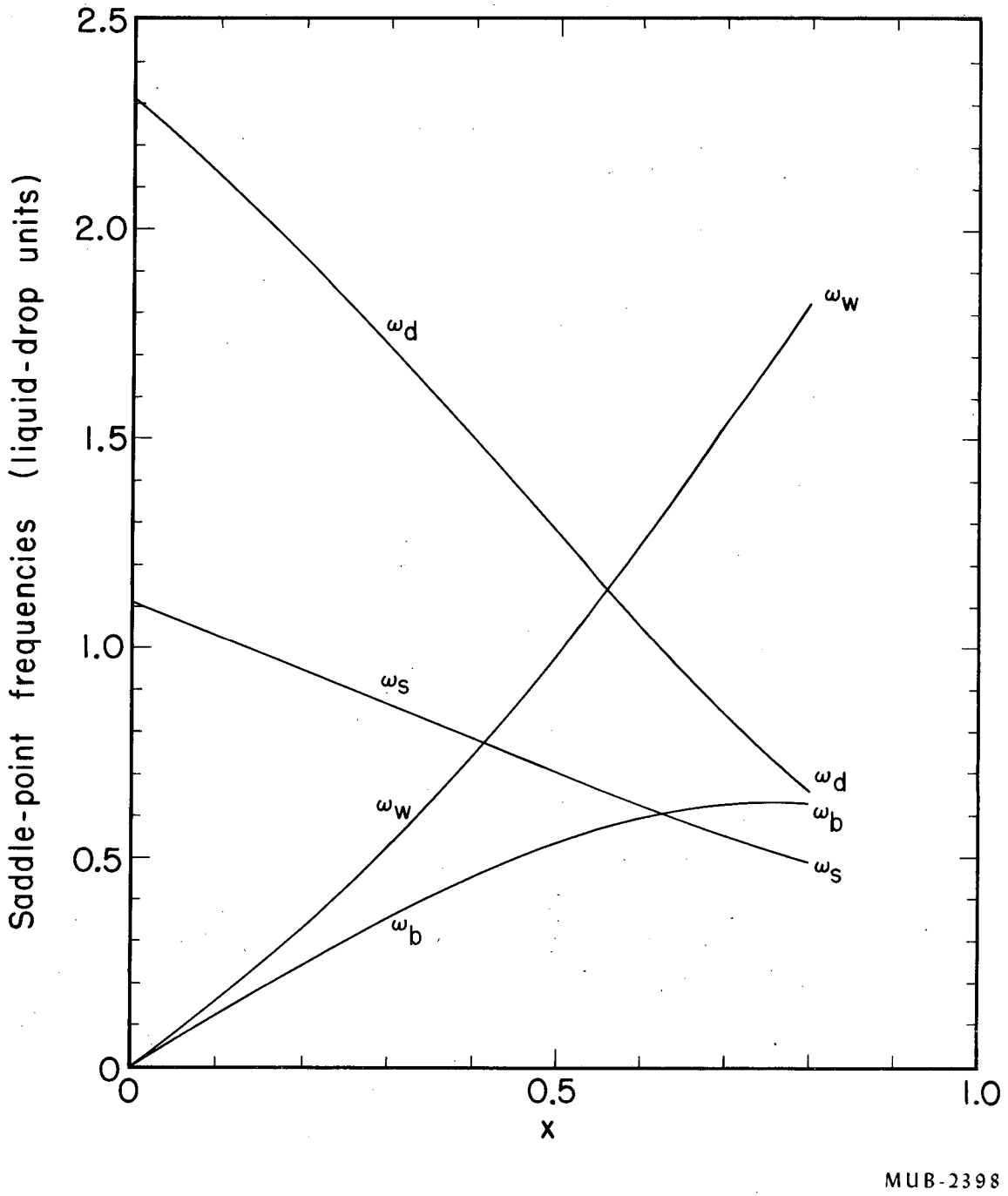

Fig. 16. Saddle-point frequencies of normal-mode oscillations, as functions of fissionability parameter x. The frequencies are in units of Ω_0 [see Eq. (4) for value of Ω_0].

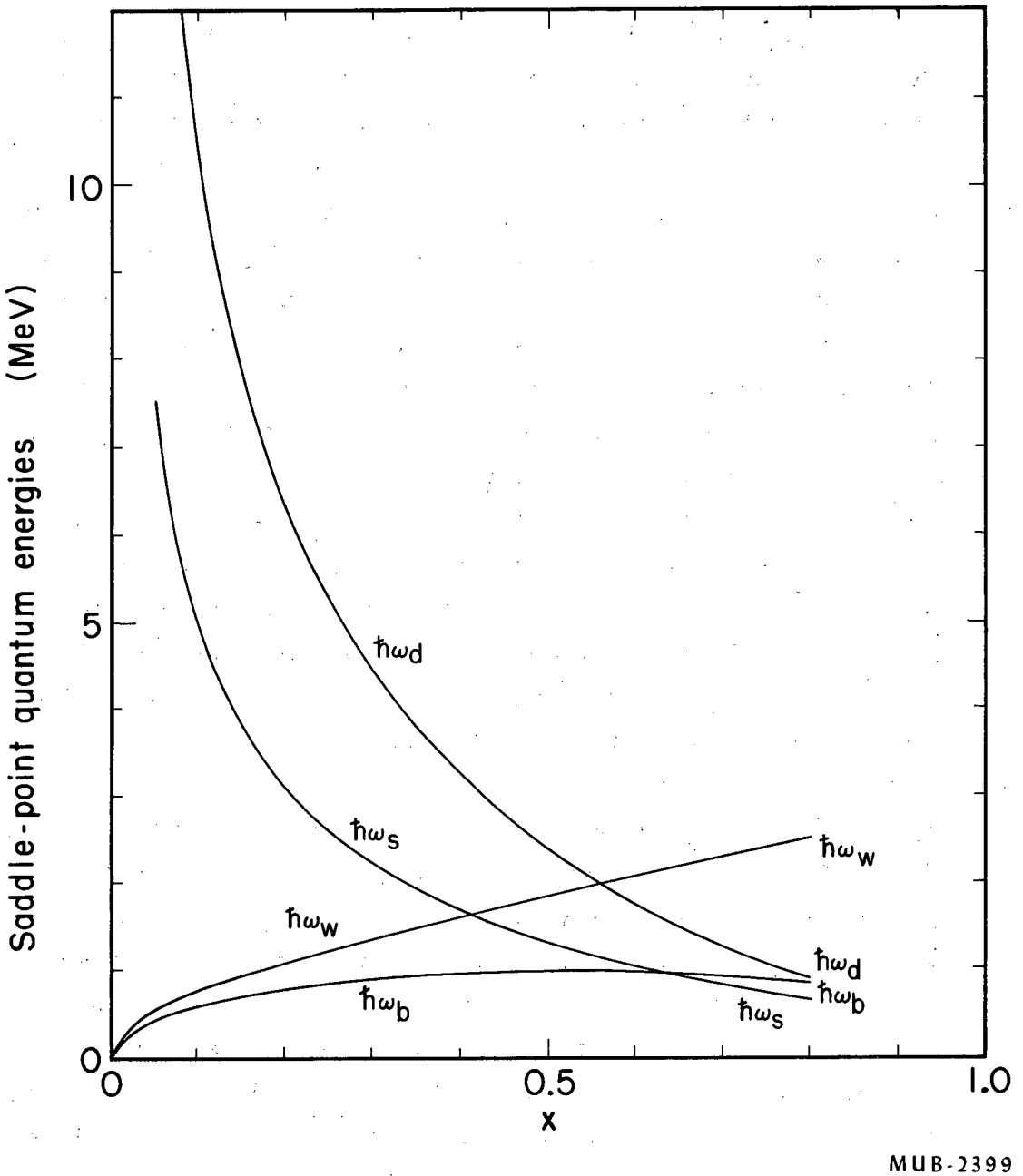

Fig. 17. Saddle-point quantum energies of normal-mode oscillations for nuclei along the line of beta stability,[50] as functions of fissionability parameter x.

C. Solution of Equations of Motion

We consider in this subsection the solution of Hamilton's classical equations of motion for the system. For given initial conditions, solution of these equations gives the subsequent motion of the system. The question naturally arises of what initial conditions to use. One could, for example, study the behavior of the system by always starting with the original sphere, giving it different sets of initial momenta. Because most of this work will be concerned with tracing out the consequences of assuming statistical equilibrium at the saddle point (see Section IV), we examine the solutions for initial conditions that are specified in the neighborhood of the saddle point.

We have seen that for both the potential energy and the kinetic energy different formulae are used, depending upon whether the system consists of spheroids that are separated or spheroids that are overlapping. The equations of motion are also different for these two cases. We will consider first the simpler case in which the saddle point consists of two tangent spheroids ($x \lesssim 0.80$); then, in the region from the saddle point to infinity, the system consists of two separated spheroids.

We saw from the Hamiltonian (12) that four of the normal modes at the saddle point—stretching, distortion-asymmetry, bending, and wriggling—are simply bounded harmonic oscillations with frequencies given by (13). The mass-asymmetry normal mode, on the other hand, was seen to be stable for $x > x_{BG}$ and unstable for $x < x_{BG}$. Since the effective mass for this mode is infinite, the mass-asymmetry coordinate changes with time infinitely slowly at the saddle point. Because the restoring force for the twisting mode is zero, this mode consists of uniform rotations of the fragments.

The remaining normal mode—the fission mode—is always unstable, taking us out of the subspace of touching spheroids. It is motion in the (positive) fission direction that causes the two fragments to start their eventual separation to infinity. Attention is first focused on the system at the critical moment when it is passing over the saddle

point; i.e., the time is chosen to be zero when the fission coordinate f is zero. Our solutions to the equations of motion will then depend upon the values of the remaining normal coordinates and momenta when $f = 0$.

The equations of motion themselves are obtained for the separated case by differentiating the Hamiltonian (10) with respect to the coordinates and momenta. For the case of coplanar symmetry axes with no rotations of the spheroids about them, there result ten first-order linear differential equations, which are listed in Appendix C.1. These equations are given in terms of the original coordinates, since the normal coordinates have meaning as such only in the vicinity of the saddle point. The equations are solved numerically for given initial conditions, determing ℓ, c_1, c_2, θ_1, and θ_2, along with their conjugate momenta, as functions of time. By carrying a solution out to infinity (in practice, to $\ell = 25\ R_0$), one finds the values at infinity of the quantities of interest. A summary of the numerical procedure is given in Appendix G.

Let us now examine the qualitative features of the solutions for various initial conditions. We have indicated in Fig. 3, on each potential-energy map in the range $0.05 \leq x \leq 0.80$, the solution corresponding to starting from rest at the saddle point. For each case the points along the path are equally spaced in time at intervals of $0.1\ T_0$ [see Eq. (2) for the value of T_0]. The motion of the system is a fairly rapid oscillation of the fragments, superimposed on a separation of their centers.

Starting the system from the saddle-point configuration corresponds to the case in which all normal coordinates are initially zero. When a particular normal coordinate is initially nonzero, the solution is altered in a manner characteristic of that normal coordinate:

(a) Mass-asymmetry: The fragment with greater volume and larger semisymmetry axis oscillates with a larger amplitude but with a smaller frequency than the other fragment.

(b) Stretching: When s is positive (corresponding to fragments that are initially more elongated than at the saddle point), both fragments oscillate with greater amplitude and separate more slowly. Conversely, for negative s, the fragments oscillate with less amplitude and separate more rapidly.

(c) Distortion-asymmetry: The fragment with the larger semi-symmetry axis oscillates with greater amplitude than the other fragment.

(d) Bending: In a bent configuration each fragment experiences a torque from the electrostatic field, resulting in an induced angular momentum. One fragment rotates clockwise and the other counterclockwise. The period of rotation is much greater than the period of oscillation.

(e) Wriggling: Both fragments rotate clockwise (or counterclockwise) simultaneously. As with the bent case, the fragments oscillate many times during a single period of rotation.

In general, the initial conditions include several nonzero coordinates, and in addition several nonzero momenta. The corresponding solutions then consist of a superposition of the characteristic features arising from each normal coordinate, modified by the effects of the initial momenta. The motion of the system is thus in general a separation of the two fragments from the saddle point to infinity, with each fragment simultaneously oscillating fairly rapidly and rotating rather slowly.

What is desired is a relationship between the initial conditions and the observable properties of fission fragments at infinity. From this we will be able to obtain, by performing suitable integrations over probability distributions for the initial conditions (see Section IV), probability distributions for the observable quantities of interest. Each quantity of interest depends strongly upon only a few of the initial coordinates and momenta, and very weakly upon the remaining ones. The practicability of our approach lies in being able to neglect the weak dependences of each quantity of interest on most of the initial conditions.

From a consideration of the numerical solutions corresponding to a large number of combinations of initial conditions, we have determined simple approximate equations expressing the quantities of interest in terms of the initial coordinates and momenta. The equations are, in general, valid for small deviations of the coordinates from the saddle point and for small values of the initial momenta. However, we were able to find for the total translational kinetic energy an equation that works well even for moderately large deviations from the saddle point. The accuracy of each equation can be seen from Table E.I (in Appendix E), where we have listed forty-four sets of initial conditions for a particular value of x, along with the values for the quantities of interest determined exactly by solving the equations of motion and determined approximately from the equations listed below. For a given x the constants appearing in each equation are calculated numerically from the solutions to the equations of motion, as described in Appendix G.

The equation established for the translational kinetic energy E of both fragments at infinity is

$$E = \frac{4U(1-U)\,E^0}{1+as} + \frac{p_\ell^2}{2M_\ell}$$

$$= \frac{(1-4m^2)\,E^0}{1+as} + M_\ell \left(\frac{2M_{c_1}^{\;0}\,p_f}{M_0\,M_f} + \frac{p_s}{M_s} \right)^2 , \quad (14)$$

where E^0 and a are constants (for a given x). The second result is simply the first result expressed in terms of the normal coordinates and momenta. This equation can be interpreted physically as giving the final kinetic energy that would result from two effective point charges of relative strengths U and $(1 - U)$ initially separated a certain distance and moving with relative momentum p_ℓ.

The fractional mass U at infinity is equal to the initial fractional mass, since after scission this coordinate does not change with time. Thus, the fractional mass at infinity is related to the mass-asymmetry normal coordinate m by the exact equation

$$U = \frac{1}{2} + m \, . \tag{15}$$

The relationship (14) for E reproduces the exact result extremely well—more accurately than do the equations we established for the individual excitation energies and angular momenta.* For these quantities we retain only terms linear in the more important initial coordinates and momenta:†

$$X_1 = X_1^0 + X_{1,s}\, s + X_{1,d}\, d + X_{1,m}\, m \, , \tag{16a}$$

$$X_2 = X_1^0 + X_{1,s}\, s - X_{1,d}\, d - X_{1,m}\, m \, , \tag{16b}$$

$$L_{1x} = L_{1,p_b}\, p_{b_x} + L_{1,p_w}\, p_{w_x} + L_{1,b}\, b_x + L_{1,w}\, w_x \, , \tag{17a}$$

$$L_{2x} = - L_{1,p_b}\, p_{b_x} + L_{1,p_w}\, p_{w_x} - L_{1,b}\, b_x + L_{1,w}\, w_x \, , \tag{17b}$$

$$L_{1y} = L_{1,p_b}\, p_{b_y} + L_{1,p_w}\, p_{w_y} + L_{1,b}\, b_y + L_{1,w}\, w_y \, , \tag{17c}$$

$$L_{2y} = - L_{1,p_b}\, p_{b_y} + L_{1,p_w}\, p_{w_y} - L_{1,b}\, b_y + L_{1,w}\, w_y \, , \tag{17d}$$

$$L_{1z} = p_{\theta_{1z}} = (1/\sqrt{2})(p_t + p_a) \, , \tag{17e}$$

$$L_{2z} = p_{\theta_{2z}} = (1/\sqrt{2})(-p_t + p_a) \, , \tag{17f}$$

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*There is currently no experimental information on excitation energies and angular momenta for the fission of elements lighter than radium. We content ourselves at this time with treating these quantities to a lower order than the translational kinetic energy.

†It should be recalled that the excitation energy calculated here is the energy associated with the collective vibrations and deformations of the fragments. Any internal excitation energy that a fragment has at the moment of division would be added to the excitation energy we calculate, to obtain the final total excitation energy.

where the quantities X_1^0, $X_{1,s}$, $X_{1,d}$, $X_{1,m}$, L_{1,p_b}, L_{1,p_w}, $L_{1,b}$, and $L_{1,w}$ are constants (for a given x). We are denoting by $X_{1,s}$, for example, the partial derivative of X_1 with respect to s, evaluated at the saddle point. Symmetry arguments have been used to relate the constants in the equation for X_2 to those in the equation for X_1. Similarly, the constants in the equations for L_{1x}, L_{2x}, L_{1y}, and L_{2y} are related by symmetry. The equations for L_{1z} and L_{2z} express the approximate conservation of the z components of angular momentum, since to first order in the angles the z components of the torque are zero. Each of the remaining equations also has a simple physical significance, which we will discuss when we present graphs of the constants appearing in them.

Complete symmetry in the observable quantities of interest could be achieved by a simple transformation from E and U to the individual translational kinetic energies E_1 and E_2 of the two fragments at infinity. The conservation of linear momentum implies that

$$UE_1 = (1 - U)E_2 .$$

From this one finds that

$$E_1 = (1 - U)E,$$

$$E_2 = UE;$$

the inverse transformation is

$$E = E_1 + E_2 , \tag{18a}$$

$$U = E_2/(E_1 + E_2) . \tag{18b}$$

An expression for E to the same order as the equations for excitation energy and angular momentum can be obtained by expanding (14) and retaining only the linear term:

$$E = E^0 - (aE^0)s . \tag{19}$$

The individual rotational energies at infinity are of second order in the initial coordinates and momenta, since they are proportional to the square of the individual angular momenta at infinity, which in turn depend linearly upon the initial coordinates and momenta. Also, the (initial) energy of the system in the neighborhood of the saddle point is of second order in the initial coordinates and momenta. Therefore, to first order, the sum at infinity of the total translational kinetic energy and the individual excitation energies is a constant:

$$E + X_1 + X_2 = E^0 + 2X_1^0 = E^0 + X^0 . \tag{20}$$

The constant $X_{1,s}$ is thus not independent but is instead given by

$$X_{1,s} = \alpha E^0/2. \tag{21}$$

A series of graphs (Figs. 18-27) has been prepared giving each constant as a function of x from 0 to 0.80. The solid line in each case represents the result for the case we have been considering: incompressible, nonviscous fragments with hydrodynamic flow consisting of a superposition of an irrotational flow and a flow corresponding to a uniform rotation. We also indicate (by a dot-dashed curve) the result for the limiting case of infinitely viscous fragments, which would separate to infinity (as rigid bodies) without oscillating. This limiting case would be approached physically if the fragments were sufficiently viscous such that their period of oscillation was large in comparison with the time of separation to a few nuclear diameters.

Figure 18 gives the dependence on x of E^0, the total translational kinetic energy at infinity that would result from fragments initially starting from rest at the saddle point. Note that the translational kinetic energy that would result if the fragments were extremely viscous (top curve) is larger than the kinetic energy corresponding to nonviscous fragments with irrotational flow (middle curve). The difference between these two curves represents the portion of original interaction energy which, for the nonviscous

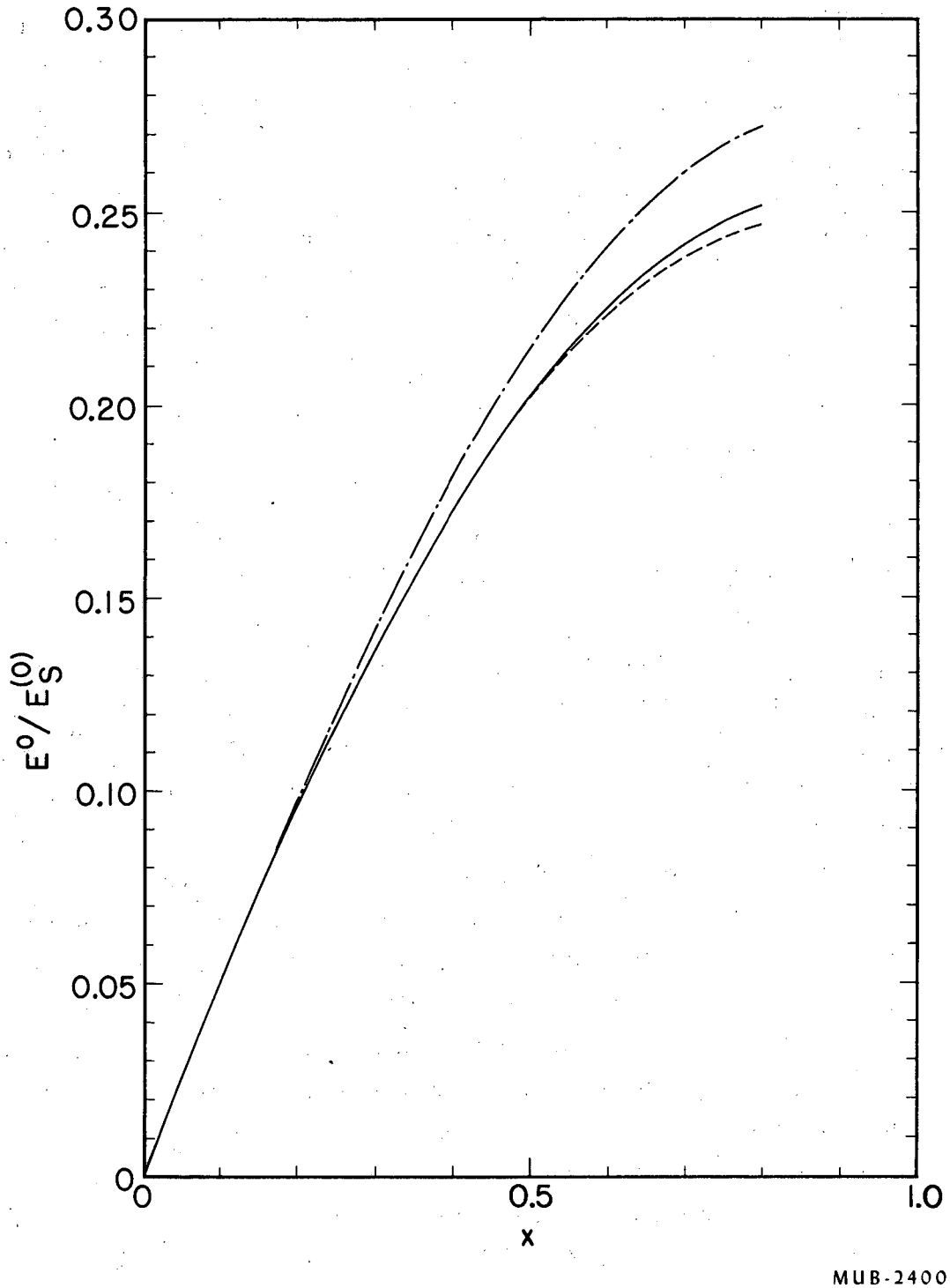

Fig. 18. The total translational kinetic energy E^0 corresponding to the case in which the fragments initially start from rest at the saddle point, as a function of fissionability parameter x. The result calculated for nonviscous fragments is given by the solid line, the result for infinitely viscous fragments by the dot-dashed line, and a simple approximation to the former (see text) by the short-dashed line.

irrotational case, is converted into excitation energy rather than into translational kinetic energy. We also present the result (short-dashed curve) obtained using the very simple approximation that the kinetic energy is equal to the product of the charges of the spheroids divided by the initial distance between their centers. Of course, this is equivalent to replacing the oscillating spheroids by two rigid spheres whose centers initially coincided with the spheroid centers. That this procedure should give a result that is close to the nonviscous irrotational limit is physically very reasonable, since the fairly rapid oscillations of the fragments tend to cancel the opposing effects of the prolate and oblate shapes.

In Fig. 19 is shown the result for the constant α; as with E^0, we also present the result (short-dashed curve) that would be obtained by replacing the spheroids with rigid spheres whose centers initially coincided with the spheroid centers.

The constant X_1^0, the excitation energy of fragment 1 that would result from initially starting the fragments from rest at the saddle point, is given in Fig. 20. We note that the sum of E^0 and $2X_1^0$, which is the energy difference between the two-spheroid saddle point and the configuration of two fragments at infinity, is larger than the corresponding sum would be if calculated from the exact liquid-drop saddle point. This discrepancy is equal to the difference between the energy of the two-spheroid saddle point and the exact liquid-drop saddle point, which, as we noted before, is due principally to the inadequate representation of the neck in the two-spheroid model. One might argue that this discrepancy is therefore more likely to affect the estimates of fragment excitation energies rather than their kinetic energies, but this conclusion cannot be regarded as reliable.

The excitation-energy derivatives $X_{1,s}$, $X_{1,d}$, and $X_{1,m}$ are presented in Figs. 21 - 23, respectively. The physical content of the equations for X_1 and X_2 can be easily seen if we substitute the values of the constants and transform back to the original coordinate system. We find then that, for typical values of the initial

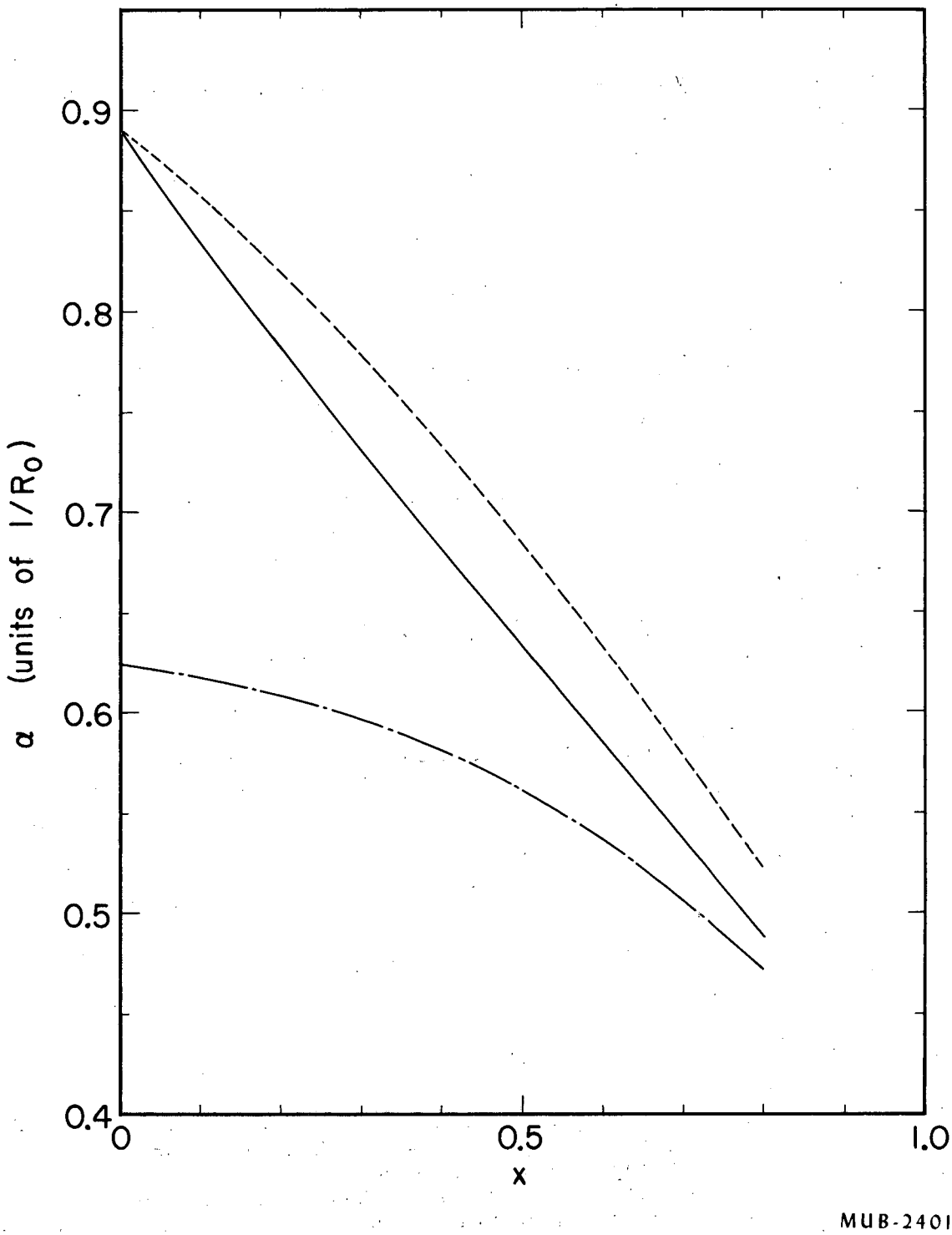

Fig. 19. The constant α [see Eq. (14)], as a function of fissionability parameter x. The result calculated for nonviscous fragments is given by the solid line, the result for infinitely viscous fragments by the dot-dashed line, and a simple approximation to the former (see text) by the short-dashed line.

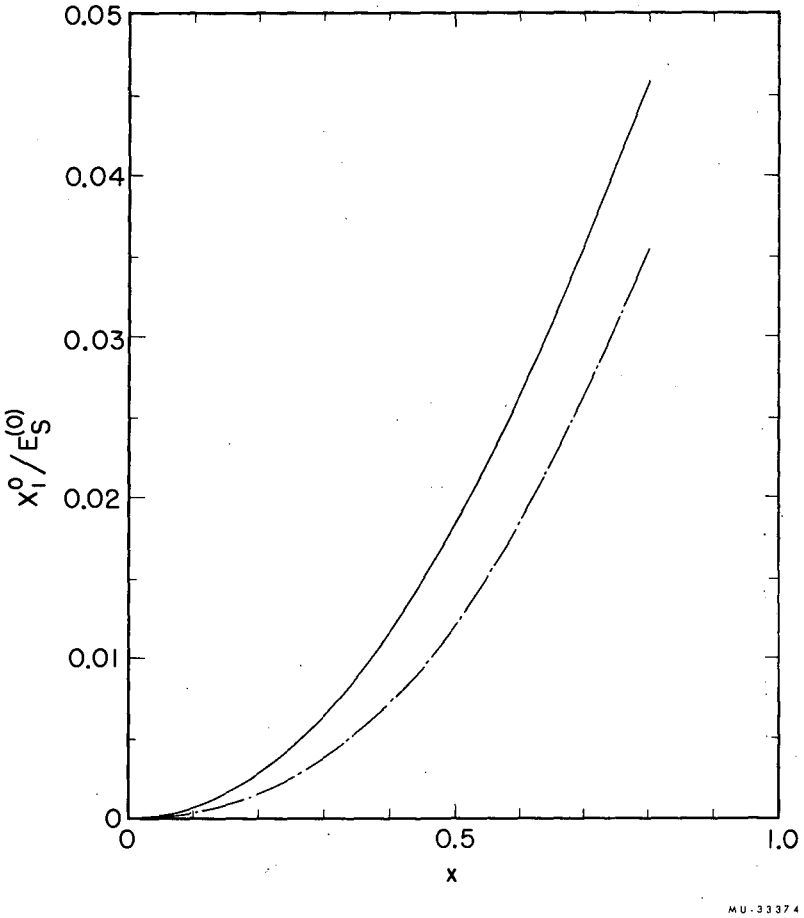

Fig. 20. The excitation energy X_1^0 of a single fission fragment corresponding to the case in which the fragments initially start from rest at the saddle point, as a function of fissionability parameter x. The result calculated for non-viscous fragments is given by the solid line, and the result for infinitely viscous fragments by the dot-dashed line.

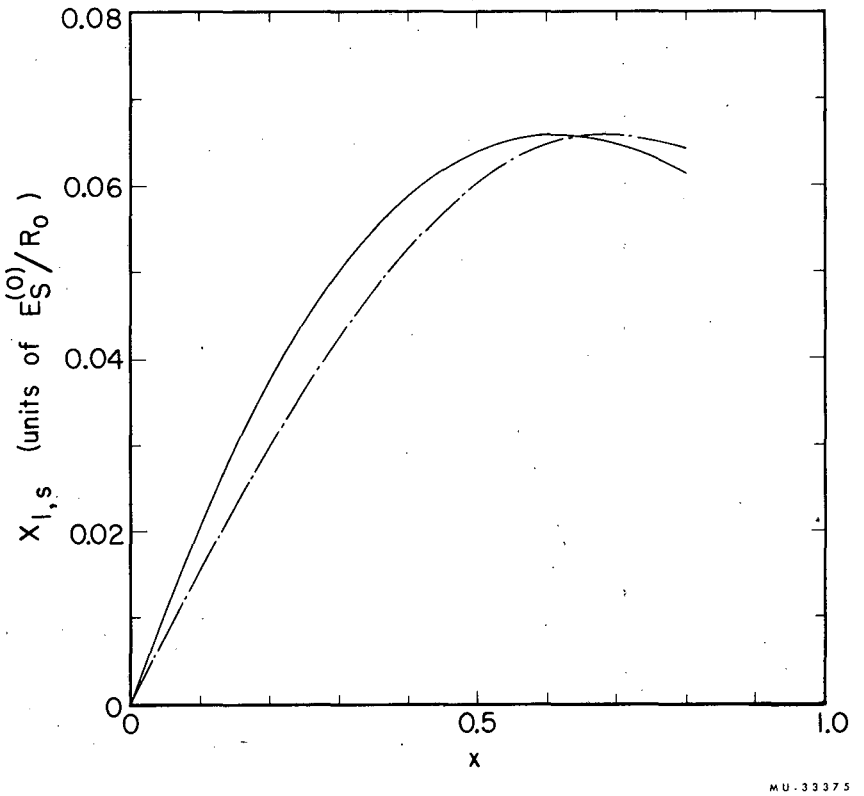

Fig. 21. The excitation-energy derivative $X_{1,s}$ [see Eq. (16)], as a function of fissionability parameter x. The result calculated for nonviscous fragments is given by the solid line, and the result for infinitely viscous fragments by the dot-dashed line.

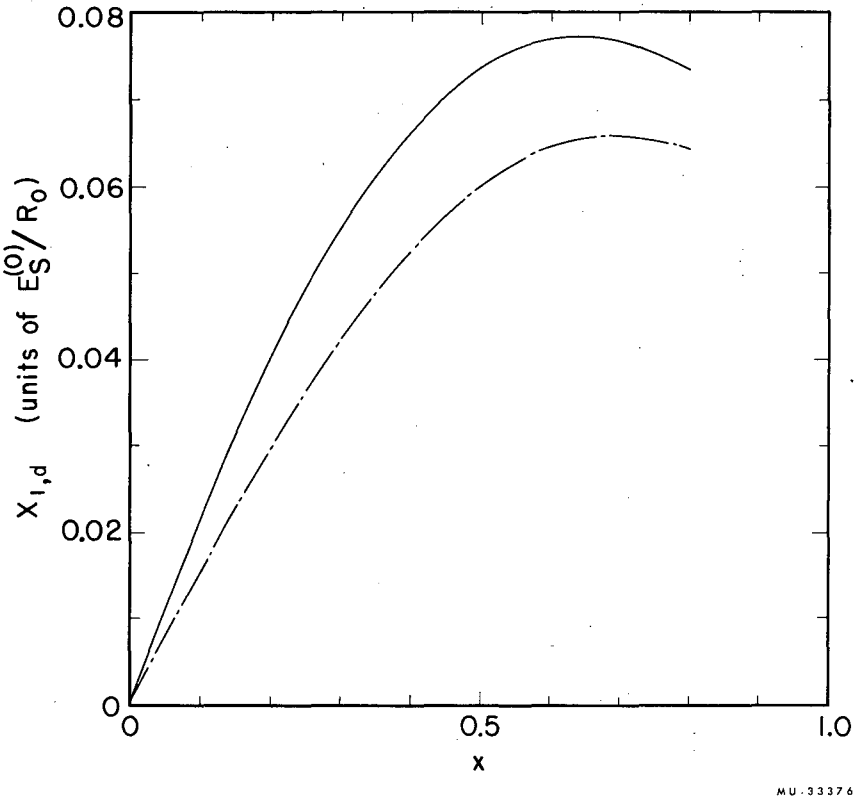

Fig. 22. The excitation-energy derivative $X_{1,d}$ [see Eq. (16)], as a function of fissionability parameter x. The result calculated for nonviscous fragments is given by the solid line, and the result for infinitely viscous fragments by the dot-dashed line.

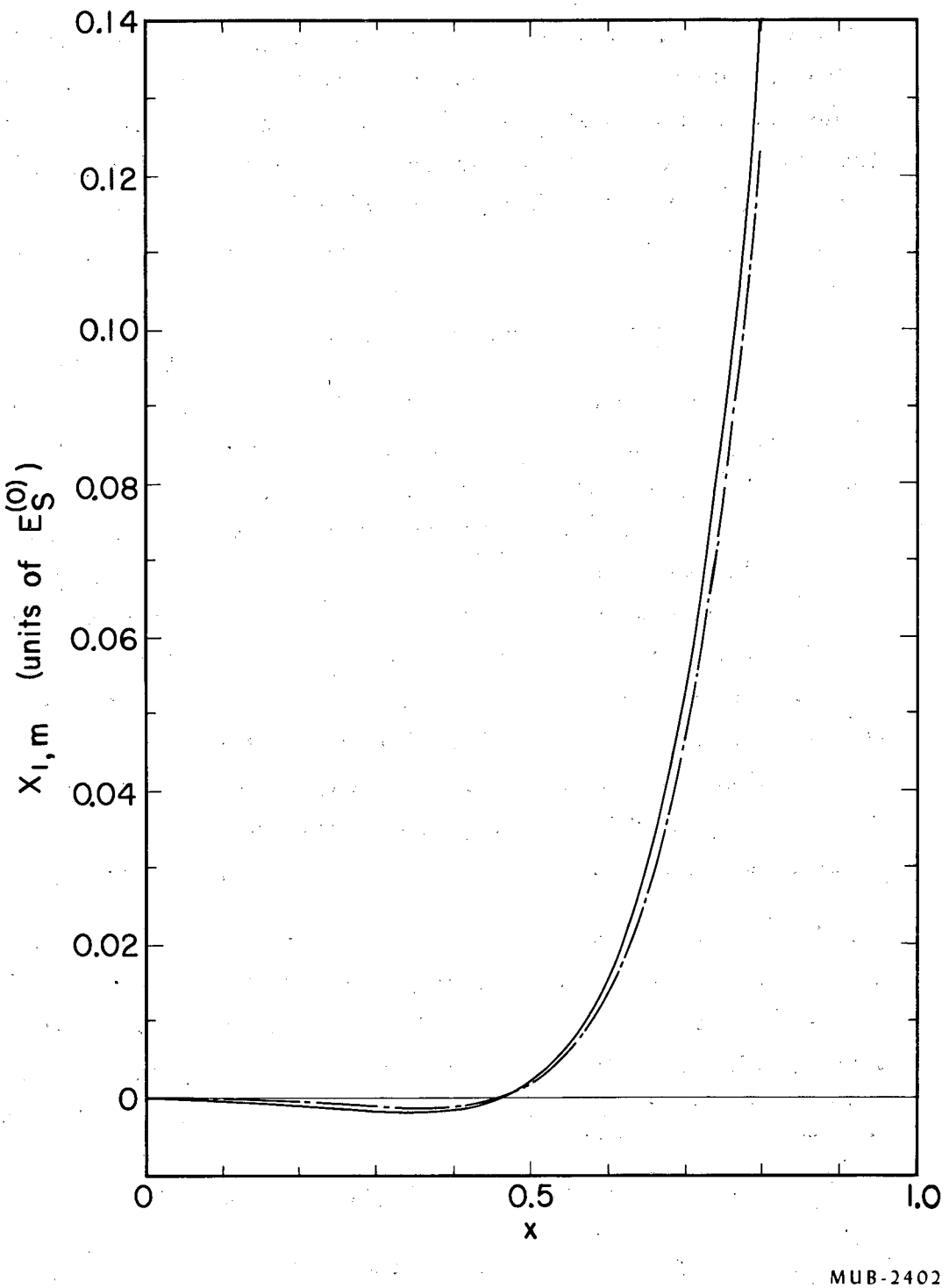

Fig. 23. The excitation-energy derivative $X_{1,m}$ [see Eq. (16)], as a function of fissionability parameter x. The result calculated for nonviscous fragments is given by the solid line, and the result for infinitely viscous fragments by the dot-dashed line.

coordinates, the excitation energy of a fragment at infinity depends primarily upon its initial elongation, is less dependent upon the fractional mass, and is still less dependent upon the initial elongation of the other fragment. Note, as we discussed in connection with the result for E^0, that if the fragments were infinitely viscous they would have less excitation energy than nonviscous fragments oscillating with irrotational flow.

The angular-momentum derivatives L_{1,p_b}, L_{1,p_w}, $L_{1,b}$, and $L_{1,w}$ are shown in Figs. 24 - 27, respectively. By substituting these values for the constants in the equations for the x and y components of angular momentum, and transforming back to the original coordinate system,* we learn the physical content of these equations. For typical initial conditions, the x component of the angular momentum at infinity of fragment 1, say, is found to depend primarily upon its own initial value, is less dependent upon the initial angle θ_{1x}, and is still less dependent upon the initial x component of angular momentum of fragment 2 and the initial angle θ_{2x}. If the fragments did not acquire any additional angular momentum by virtue of the torque exerted by one fragment on the other through the electrostatic interaction, then L_{1,p_b} and L_{1,p_w} would each equal $1/\sqrt{2}$, and $L_{1,b}$ and $L_{1,w}$ would each be zero. Note that the torque mechanism is capable of inducing in infinitely viscous fragments roughly four times as much angular momentum as in nonviscous fragments oscillating with irrotational flow. This is because for an oscillating fragment the torque is reduced as the elongation of the spheroid

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*The relationships $p_{b_x} = (1/\sqrt{2})\,(p_{\theta_{1x}} - p_{\theta_{2x}})$ and $p_{w_x} = (1/\sqrt{2})\,(p_{\theta_{1x}} + p_{\theta_{2x}})$ etc., obtained from the normal-coordinate transformation and the definitions of the momenta, are useful for this purpose. The value at infinity of $p_{\theta_{1x}}$, for example, is L_{1x}.

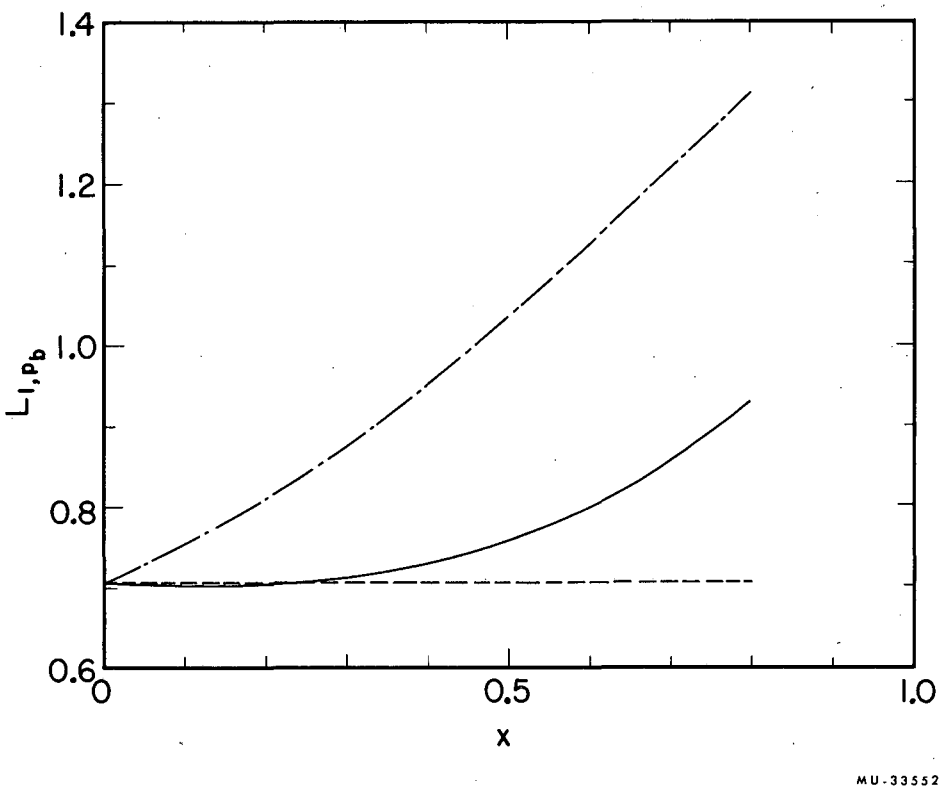

Fig. 24. The angular-momentum derivative L_{1,p_b} [see Eq. (17)], as a function of fissionability parameter x. The result calculated for nonviscous fragments is given by the solid line, and the result for infinitely viscous fragments by the dot-dashed line. The short-dashed line represents the result that would follow if the torque exerted by one fragment on the other through the electrostatic interaction were zero.

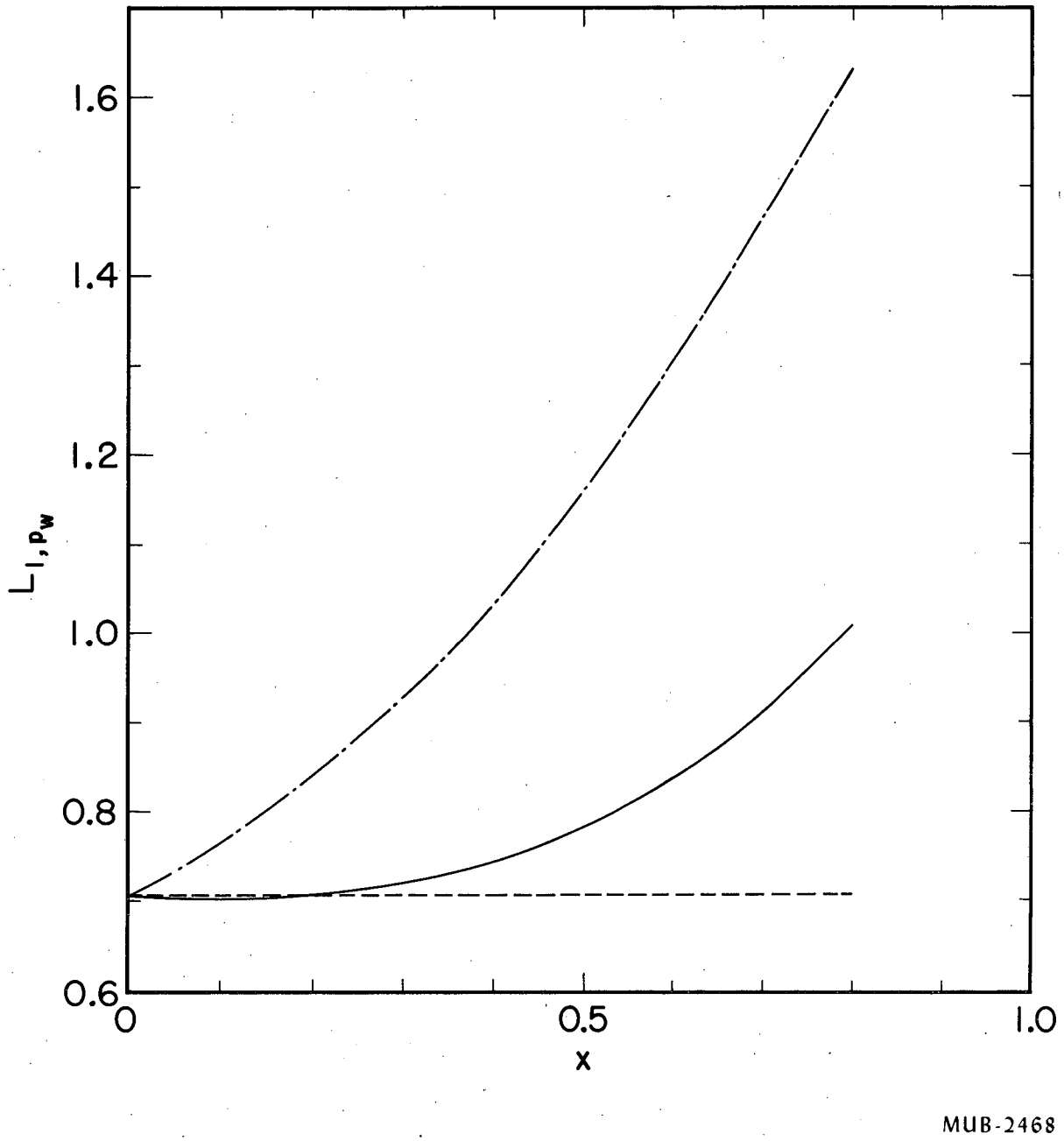

Fig. 25. The angular-momentum derivative L_{1,p_w} [see Eq. (17)], as a function of fissionability parameter x. The result calculated for nonviscous fragments is given by the solid line, and the result for infinitely viscous fragments by the dot-dashed line. The short-dashed line represents the result that would follow if the torque exerted by one fragment on the other through the electrostatic interaction were zero.

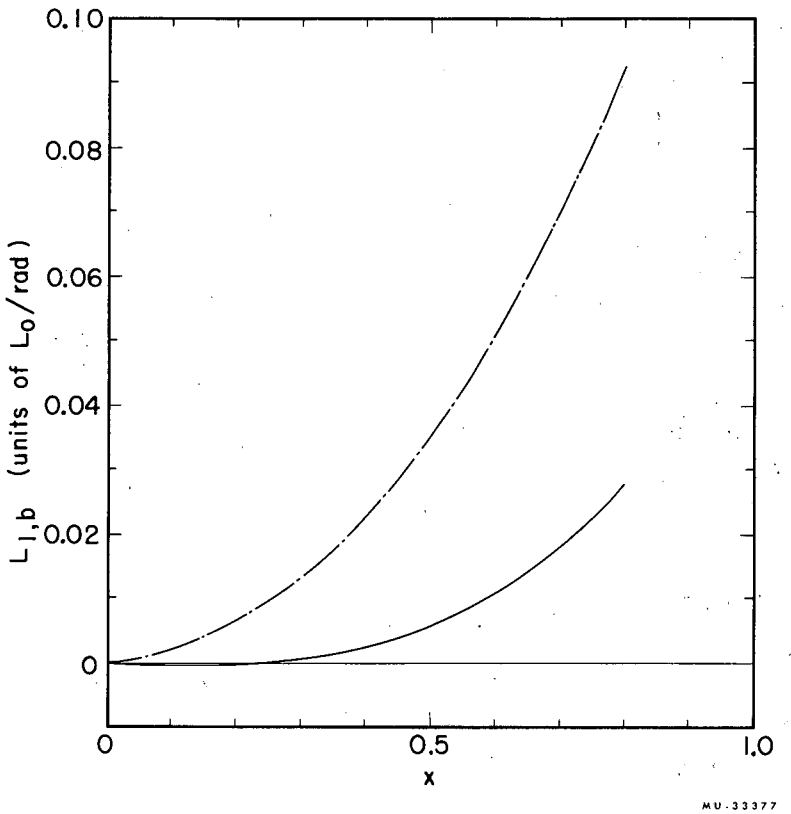

Fig. 26. The angular-momentum derivative $L_{1,b}$ [see Eq. (17)], as a function of fissionability parameter x. The result calculated for nonviscous fragments is given by the solid line, and the result for infinitely viscous fragments by the dot-dashed line. For the value of L_0 see Eq. (3).

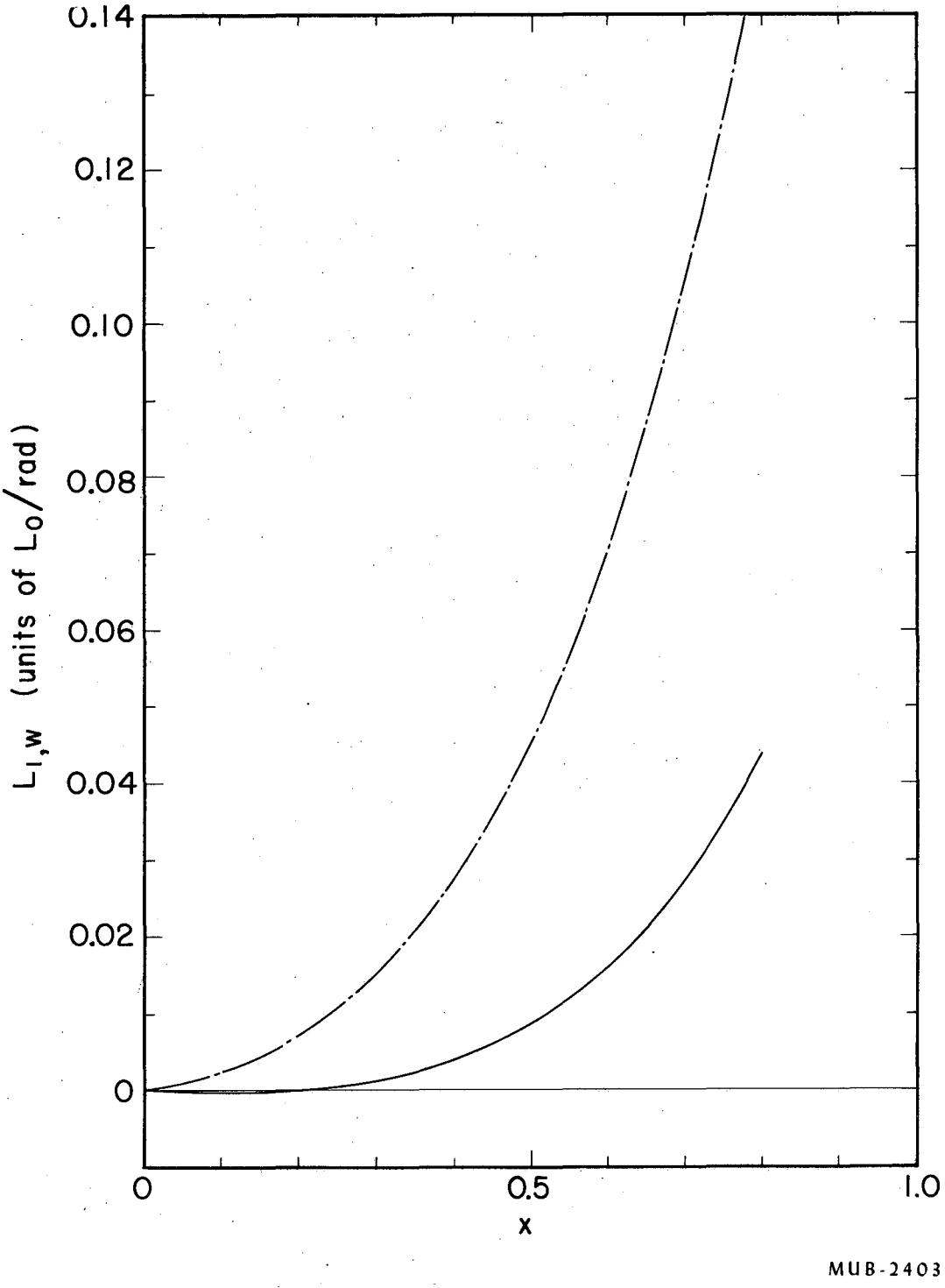

Fig. 27. The angular-momentum derivative $L_{1,w}$ [see Eq. (17)], as a function of fissionability parameter x. The result calculated for nonviscous fragments is given by the solid line, and the result for infinitely viscous fragments by the dot-dashed line. For the value of L_0 see Eq. (3).

is reduced, even changing sign when the spheroid changes from prolate to oblate.

When the saddle point occurs for $\ell < 2c$ ($x \gtrsim 0.80$), it is necessary to consider the equations of motion for overlapping spheroids. As before, we specialize for the overlapping case to symmetrical fragments, specified by the two coordinates ℓ and c. Hamilton's equations of motion for this case are listed in Appendix C.2. In the vicinity of the saddle point, the motion of the system consists of a superposition of the two normal modes: bounded oscillations in one direction and unbounded motion in the fission direction.

The equations of motion for the overlapping case have been solved for only a few isolated values of x and initial conditions. We will here only briefly discuss the solution for $x = 0.90$ corresponding to initially starting the system from rest at the saddle point.* This solution is presented in Fig. 28. The semisymmetry axis c at first increases more rapidly than the distance ℓ between spheroid centers increases; this continues until the system has become fairly elongated. Then, as the distance between centers continues to increase, the semisymmetry axis starts to decrease. When scission occurs, the fragments are already moving apart with a translational kinetic energy of the order of 25 MeV. The scission configuration is less eccentric (and the fragment centers closer together) than the configuration obtained by minimizing the potential energy of symmetric tangent

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*Since the saddle point is a position of (unstable) equilibrium, a system initially at rest at the saddle point would remain there (classically) for an infinite time; we imagine an infinitesimal push in the fission direction to start the system moving. An analytic solution, valid in the neighborhood of the saddle point, is used until the system is a short distance from the saddle point, where the numerical integration begins.

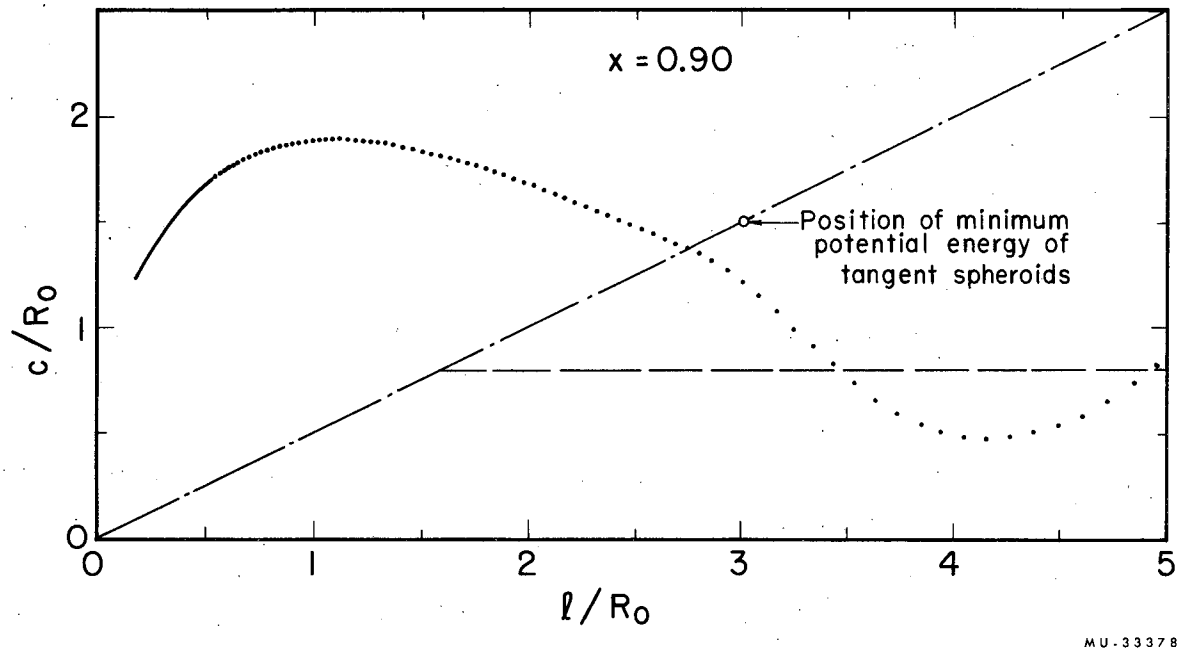

Fig. 28. Solution of equations of motion for $x = 0.90$ corresponding to initially starting the system from rest at the saddle point. The points are equally spaced in time at intervals of $0.107\ T_0$ [see Eq. (2) for value of T_0]. Note that the scission configuration is less eccentric than the configuration of tangent spheroids of minimum potential energy, whose location on the scission line is indicated by the open circle.

spheroids. This results in about a 20-MeV additional gain in translational kinetic energy at infinity over the kinetic energy that would result from initially starting the system at rest from the configuration of tangent spheriods whose eccentricities are obtained by minimizing the potential energy. For this $x = 0.90$ case, the total translational kinetic energy E was found to be $0.315_5\ E_S^{(0)}$, and the total excitation energy X was found to be $0.091_3\ E_S^{(0)}$.

For the higher values of x, where the saddle point is not near the scission configuration, the very subtle question of the dynamics of the motion from saddle to scission becomes important in determining what fraction of the total energy goes into translational kinetic energy and what fraction into excitation energy. Therefore, for the higher values of x, a calculation of the precise division of the total energy into kinetic and excitation energies based on optimum tangent spheroids is likely to be in error.

IV. STATISTICAL MECHANICS

We have obtained approximate formulae [(14), (15), (16), and (17)] relating the observable properties of fission fragments at infinity to the initial values of the coordinates and momenta (when the saddle point is the configuration of two tangent spheroids). We now consider the determination of the probability for the system possessing a given set of initial coordinates and momenta. These two results will be combined in the next section to yield the probability for the two fragments at infinity possessing given total translational kinetic energy, fractional mass, and individual excitation energies and angular momenta.

The conditions at the saddle point would in general depend upon the past history of the system—upon how the system was formed and upon the path it took in reaching the saddle point. However, the system typically undergoes about 10^6 fissionlike oscillations after formation before it reaches the saddle point (if it ever does).[51] This provides ample opportunity for many interchanges of energy, making it unlikely that at the saddle point the system "remembers" the way it was formed or most of its previous motion. It is thus likely that at the saddle point thermal equilibrium is established. This is the central hypothesis of the transition-state method used for discussing the reaction rates of chemical or nuclear systems,[38] and we will base our further considerations on this standard assumption.

The statistical-mechanics discussion is essentially the same for both the case in which the saddle point is represented by tangent spheroids and the case in which it is represented by overlapping spheroids. In the following discussion we will explicitly consider probability distributions for the normal coordinates appropriate to the case in which the saddle point consists of two tangent spheroids; for the other situation (again considering the restricted case of symmetrical fragments) there are simply fewer probability distributions.

The determination of the probability of given initial conditions is very simple in classical statistical mechanics. The probability P

that the system possesses a given set of coordinates and momenta, with the corresponding total energy $\mathcal{H}$ given by (10), is simply

$$P = N \exp(-\mathcal{H}/\Theta) , \tag{22}$$

where Θ is the nuclear temperature at the saddle point (measured in energy units), and N is a normalization constant. (Use of the symbol Θ to denote nuclear temperature should cause no confusion with its earlier use in connection with angles.) When the actual Hamiltonian (10) is used, this expression for P is exact (classically) to all orders in the coordinates and momenta.

If one expands the Hamiltonian about the saddle point and retains only quadratic terms (harmonic approximation), then the probability distribution for each normal coordinate and its conjugate momentum is simply a Gaussian in the normal coordinate or momentum [with the exception of $P(p_a)$ and $P(f)$, which are discussed separately below]. Thus, for example,*

$$P(s) = (2\pi\,\Theta/K_s)^{-1/2} \exp\left(-\tfrac{1}{2} K_s s^2/\Theta\right), \tag{23a}$$

$$P(p_s) = (2\,\pi\,M_s\,\Theta)^{-1/2} \exp\left[-p_s^2/(2M_s\,\Theta)\right] , \tag{23b}$$

$$P(p_f) = 2(2\,\pi\,M_f\,\Theta)^{-1/2} \exp\left[-p_f^2/(2M_f\Theta)\right] . \tag{23c}$$

Equations analogous to these hold for the remaining normal coordinates and momenta. For one of the momenta and two of the coordinates the Gaussian distributions become infinitely broad and hence reduce to constants. This occurs for $P(p_m)$ because the mass-asymmetry effective

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*All probability distributions in this paper are normalized such that unity is obtained when the functions are integrated over the allowed range of variables, which is usually taken to be from $-\infty$ to $+\infty$. The range of integration for p_f is taken to be from 0 to ∞, since for negative values of p_f the system does not fission but instead returns to the pre-saddle-point configuration; this results in the additional factor of 2 in the expression for $P(p_f)$.

mass M_m is infinite,* and for $P(t)$ and $P(a)$ because the twisting and axial-rotation stiffness constants are zero.

Because of the condition that the total angular momentum of the system be zero, the distribution in the axial-rotation momentum is a Dirac delta function:

$$P(p_a) = \delta(p_a) .$$

We recall finally that we do not need a probability distribution in f because our solutions to the equations of motion are in terms of $f = 0$ initially.

As the nuclear temperature approaches zero, the classical distribution for each normal coordinate and momentum approaches a Dirac delta function. However, we know from the Heisenberg quantum-mechanical uncertainty principle that simultaneous localization of the system in a position and conjugate-momentum coordinate can be achieved only within limits. Even at zero temperature each of the distributions should, in general, have a nonzero width, associated with the quantum-mechanical zero-point vibrations. As we shall see, the quantum-mechanical effects may be important at typical nuclear temperatures.

Although the quantum-mechanical determination of the probability distributions would be difficult if one used the complete Hamiltonian (10), the problem can be readily solved in the harmonic approximation. Then, the individual terms in the Hamiltonian (12) are the Hamiltonians for simple harmonic oscillators. (The effect on the distributions of the infinite effective mass M_m and the zero

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*The Gaussian distribution in the mass-asymmetry velocity becomes, on the other hand, infinitely narrow and hence reduces to a Dirac delta function. This means physically that the mass-asymmetry coordinate changes with time infinitely slowly at the saddle point.

stiffness constant K_t will be discussed later.) In our discussion below we will refer only to the stretching mode; results for the remaining modes are completely analogous.

In the coordinate representation the quantum-mechanical solution of the stretching mode yields the harmonic-oscillator wave functions $\psi_n(s)$, with corresponding energy levels $E_n^s = (n + \frac{1}{2})\hbar\omega_s$. When the oscillator is known to be in the quantum-mechanical state n, the probability that its position coordinate has a given value s is

$$P_n(s) = |\psi_n(s)|^2 .$$

In particular, when the oscillator is in the ground state $n = 0$, the probability distribution for the coordinate is[52]

$$P_0(s) = |\psi_0(s)|^2 = (\pi\hbar\omega_s/K_s)^{-1/2} \exp\left[- K_s s^2/(\hbar\omega_s)\right]. \tag{24a}$$

Analogous results for the momentum p_s are obtained just as readily by solving Schrödinger's equation for the stretching mode in the momentum representation.[52] For example, the ground-state probability distribution for the momentum is

$$P_0(p_s) = (\pi M_s \hbar \omega_s)^{-1/2} \exp\left[- p_s^2/(M_s \hbar \omega_s)\right]. \tag{24b}$$

We note that for the stretching oscillator in the ground state, both the distribution in s and the distribution in p_s are Gaussians, with nonzero widths proportional to $(\hbar\omega_s/K_s)^{1/2}$ and $(M_s \hbar \omega_s)^{1/2}$, respectively.

Having thus determined probability distributions for an oscillator in a given quantum-mechanical state, we are now in a position to determine probability distributions for an oscillator in statistical equilibrium with its surroundings. The quantum-mechanical probability for finding the stretching oscillator, in statistical equilibrium with its surroundings at a temperature Θ, at position s is given by

$$P(s) = \sum_{n=0}^{\infty} P_n^s \, |\psi_n(s)|^2 ,$$

where

$$P_n^s = N \exp\left(-E_n^s/\Theta\right)$$

is the statistical probability that the oscillator is in the quantum-mechanical state n. Determination of the normalization constant N yields (the same symbol N is used in this paper to denote each of several normalization constants)

$$N = 2 \sinh\left[\hbar\omega/(2\Theta)\right] .$$

By using properties of the harmonic-oscillator wave functions, this infinite summation can be performed explicitly, yielding the remarkably simple result[53, 54*]

$$P(s) = (\pi C_s)^{-1/2} \exp\left(-s^2/C_s\right) , \tag{25a}$$

where the temperature-dependent constant C_s is given by

$$C_s = \frac{\hbar\omega_s}{K_s} \coth\left(\frac{\hbar\omega_s}{2\Theta}\right) \rightarrow \begin{cases} 2\Theta/K_s , & \Theta >> \hbar\omega_s \\ \hbar\omega_s/K_s , & \Theta << \hbar\omega_s \end{cases} .$$

The temperature dependence of C_s can be seen from the graph of $\coth\left[\hbar\omega/(2\Theta)\right]$ vs $2\Theta/(\hbar\omega)$ in Fig. 29. Note that for high temperatures the quantum-mechanical expression (25a) for $P(s)$ reduces to the classical result (23a), whereas in the low-temperature limit it reduces to

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*An analogous formula has been used by Marshall Blann (University of Rochester) and Wladyslaw J. Swiatecki (University of California Lawrence Radiation Laboratory, Berkeley) in connection with fission-fragment charge distributions (unpublished work).

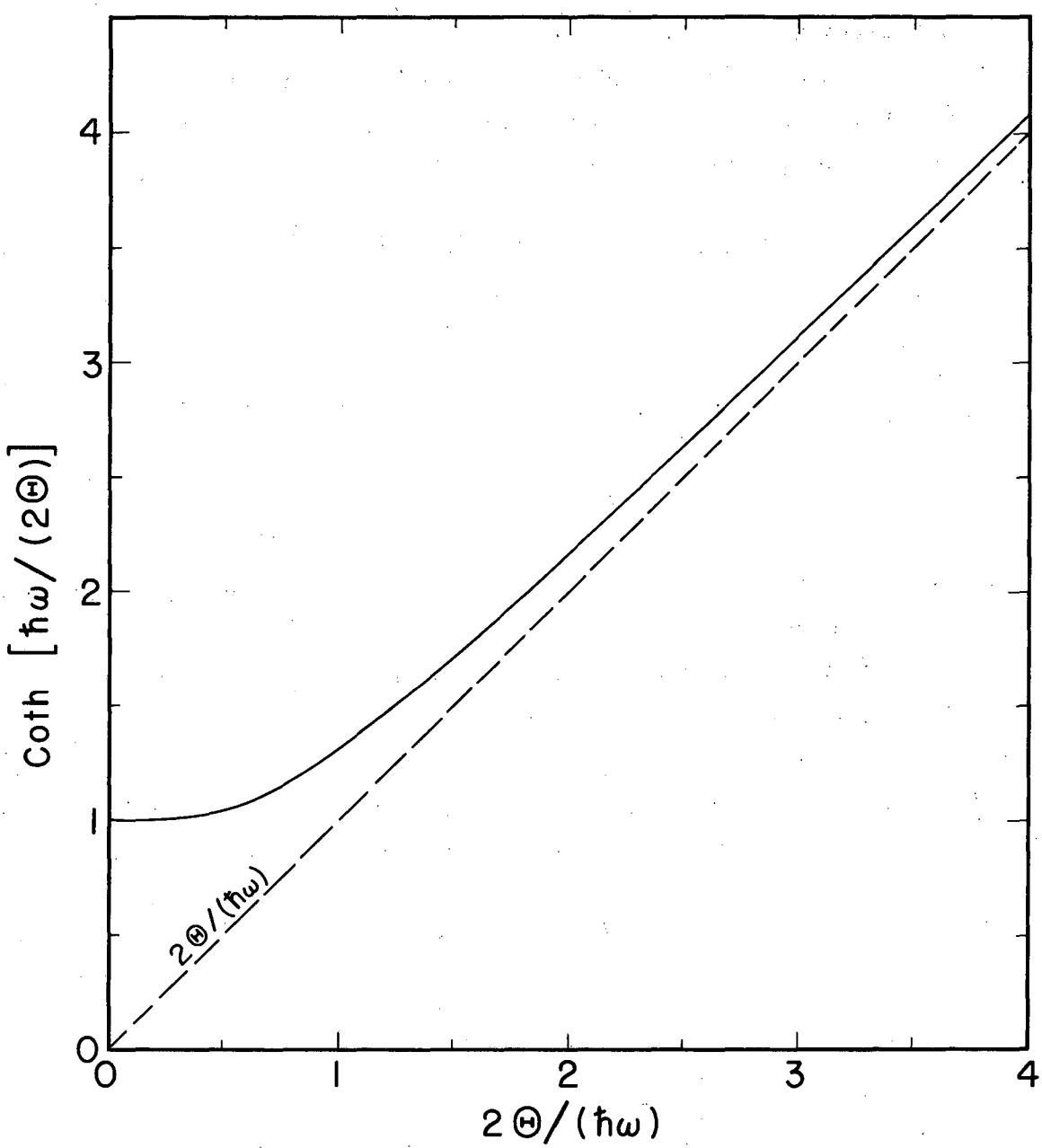

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Fig. 29. Coth [$\hbar\omega/(2\Theta)$], as a function of $2\Theta/(\hbar\omega)$. The temperature dependence of the constants appearing in the probability distributions for the initial conditions is given by this function. Shown also is the asymptote of the function (dashed line).

the distribution (24a) for the quantum-mechanical zero-point motion of a harmonic oscillator.

An analogous probability distribution for p_s is obtained from the momentum-representation solutions to Schrödinger's equation for the stretching mode. The result is[53]

$$P(p_s) = (\pi C_{p_s})^{-1/2} \exp\left(-p_s^2/C_{p_s}\right), \tag{25b}$$

where

$$C_{p_s} = M_s \hbar \omega_s \coth\left(\frac{\hbar \omega_s}{2\Theta}\right) \rightarrow \begin{cases} 2M_s \Theta, & \Theta >> \hbar\omega_s \\ M_s \hbar \omega_s, & \Theta << \hbar \omega_s . \end{cases}$$

For high and low temperatures this expression reduces to (23b) and (24b), respectively.

We saw earlier (Section III.B) that the mass-asymmetry and twisting normal modes have zero frequencies. Thus, $\hbar\omega_m$ and $\hbar\omega_t$ are always small in comparison with Θ, which means that for these modes we are always in the high-temperature (classical) limit. Thus, the constants C_m and C_{p_t} are always given by*

$$C_m = 2\,\Theta/K_m ,$$

$$C_{p_t} = 2\,M_t\,\Theta .$$

---

*Recall that for $x < x_{BG}$ the mass-asymmetry stiffness constant K_m is negative. Thus for $x < x_{BG}$, the probability $P(m)$ increases rather than decreases with increasing absolute value of m.

We have thus derived in the harmonic approximation expressions that are valid quantum-mechanically at any temperature for the distributions in both coordinates and momenta for the normal modes (except the fission mode). Each distribution is a Gaussian with a temperature-dependent width that has simple high- and low-temperature limits.

The probability distribution for the fission momentum p_f is difficult to calculate quantum-mechanically both because this mode is unstable and because the stiffness constant K_f is not defined. For this distribution we use the classical result (23c); rewriting, this is

$$P(p_f) = 2\left(\pi\, C_{p_f}\right)^{-1/2} \exp\left(-p_f^2/C_{p_f}\right),$$

with

$$C_{p_f} = 2M_f\,\Theta\,.$$

Any attempt to improve this result should include an improved description of the shape of the fission barrier. The inaccuracies arising from using the classical expression for $P(p_f)$ are probably not serious for our purposes, since the equations relating the quantities of interest at infinity to the initial conditions are to first order independent of p_f. An error in $P(p_f)$ would thus affect the distributions of the quantities of interest only in second order.

For a given normal mode the probability distribution for the coordinate is independent of the probability distribution for the momentum. Also, each normal mode is completely independent of all the others. Therefore, the probability for observing the system with a given set of initial coordinates and momenta is simply the product of the individual probabilities for each coordinate and momentum.

The probability distributions for the initial conditions are in terms of stiffnesses, masses, and frequencies (all calculated and graphed as functions of x in Section III.B), and the nuclear temperature Θ at the saddle point. The temperature is a function of the internal nuclear excitation energy at the saddle point. The discussion of the determination of Θ in terms of the internal excitation energy will be given in Section VI.

V. PROBABILITY DISTRIBUTIONS FOR OBSERVABLE QUANTITIES OF INTEREST

In this section we derive expressions for the probability of simultaneously observing the two fragments at infinity with given values of the quantities of interest: total translational kinetic energy, fractional mass, individual excitation energies, and individual angular momenta. The derivation utilizes the results obtained in the two preceding sections: the equations relating the observable quantities of interest to the initial conditions, and the probability distributions for the initial conditions. Since the equations relating the quantities of interest to the initial conditions are for the case in which the saddle point is represented by two tangent spheroids ($x \lesssim 0.80$), the probability distributions that we derive for the quantities of interest will be for this case also.

A. The Distribution $P(E, U, X_1, X_2, \vec{L}_1, \vec{L}_2)$ to Lowest Order

We first consider the calculation of the probability $P(E, U, X_1, X_2, \vec{L}_1, \vec{L}_2)$ of simultaneously observing the two fragments at infinity with given total translational kinetic energy E, fractional mass U, individual excitation energies X_1 and X_2, and individual angular momenta $\vec{L}_1$ and $\vec{L}_2$. For this derivation we use the consistent set of lowest-order equations (15), (16), (17), and (19) expressing the quantities of interest in terms of the initial conditions. The resulting probability distributions will then be valid only to lowest order in the quantities of interest. We will later use (14) and (15) to calculate the distribution $P(E, U)$ to a higher order in E and U than that used in the present calculation.

The probability distribution for the observable quantities of interest is obtained by multiplying the probability distribution for the initial coordinates and momenta by the Jacobian for the transformation from the initial coordinates and momenta to the quantities

of interest, and then integrating over the remaining coordinates and momenta.*

Since E, X_1, and X_2 are not independent, but are related to first order by the conservation-of-energy equation (20), the probability distribution $P(E, U, X_1, X_2, \vec{L}_1, \vec{L}_2)$ contains the Dirac delta function $\delta(E + X_1 + X_2 - E^0 - 2X_1^0)$. This probability distribution is then given by

$$P(E, U, X_1, X_2, \vec{L}_1, \vec{L}_2) = P(U, X_1, X_2, \vec{L}_1, \vec{L}_2)\, \delta(E + X_1 + X_2 - E^0 - 2X_1^0),$$

where a particular choice has been made for the remaining variables.

In order to calculate $P(U, X_1, X_2, \vec{L}_1, \vec{L}_2)$ we need to invert the set of equations (15), (16), and (17) to obtain nine of the initial conditions as functions of the nine quantities of interest and the remaining initial conditions. (Each component of angular momentum of each fragment is regarded here as a separate quantity of interest.) If we choose m, s, d, p_{b_x}, p_{w_x}, p_{b_y}, p_{w_y}, p_t, and p_a as dependent variables, this inversion gives

$$m = U - \frac{1}{2}, \tag{26a}$$

$$s = \frac{X_1 + X_2 - 2X_1^0}{2X_{1,s}}, \tag{26b}$$

$$d = \frac{X_1 - X_2 - 2X_{1,m}\left(U - \frac{1}{2}\right)}{2X_{1,d}}, \tag{26c}$$

$$p_{b_x} = \frac{L_{1x} - L_{2x} - 2L_{1,b}\, b_x}{2L_{1,p_b}}, \tag{26d}$$

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*For a discussion of coordinate transformations, see any advanced-calculus textbook, for example, Brand.[55]

$$p_{w_x} = \frac{L_{1x} + L_{2x} - 2L_{1,w}\, w_x}{2L_{1,p_w}}, \tag{26e}$$

$$p_{b_y} = \frac{L_{1y} - L_{2y} - 2L_{1,b}\, b_y}{2L_{1,p_b}}, \tag{26f}$$

$$p_{w_y} = \frac{L_{1y} + L_{2y} - 2L_{1,w}\, w_y}{2L_{1,p_w}}, \tag{26g}$$

$$p_t = \frac{L_{1z} - L_{2z}}{\sqrt{2}}, \tag{26h}$$

$$p_a = \frac{L_{1z} + L_{2z}}{\sqrt{2}}. \tag{26i}$$

With this choice of dependent variables, the equation transforming the initial probability distribution into the desired probability distribution is

$$P(U, X_1, X_2, \vec{L}_1, \vec{L}_2) = \int_{-\infty}^{+\infty} db_x \int_{-\infty}^{+\infty} dw_x \int_{-\infty}^{+\infty} db_y \int_{-\infty}^{+\infty} dw_y$$

$$\times P(m, s, d, b_x, w_x, b_y, w_y, p_{b_x}, p_{w_x}, p_{b_y}, p_{w_y}, p_t, p_a)$$

$$\times \left| \frac{\partial (m, s, d, p_{b_x}, p_{w_x}, p_{b_y}, p_{w_y}, p_t, p_a)}{\partial (U, X_1, X_2, L_{1x}, L_{2x}, L_{1y}, L_{2y}, L_{1z}, L_{2z})} \right|;$$

we have already integrated over the initial coordinates and momenta not involved in the transformation, obtaining unity in each case. The probability distribution in the integrand is given by the product of the probability distribution for each normal coordinate and momentum appearing in its argument, as determined in Section IV. It is understood that this probability function has the set of equations(26) substituted for the variables $m, s, d, p_{b_x}, p_{w_x}, p_{b_y}, p_{w_y}, p_t,$ and p_a.

The absolute value of the Jacobian for the transformation is given by

$$\left|\frac{\partial(m,s,d,p_{b_x},p_{w_x},p_{b_y},p_{w_y},p_t,p_a)}{\partial(U,X_1,X_2,L_{1x},L_{2x},L_{1y},L_{2y},L_{1z},L_{2z})}\right| = \left(\frac{1}{2X_{1,s}\,X_{1,d}}\right)\left(\frac{1}{2L_{1,p_b}\,L_{1,p_w}}\right)^2 .$$

We note that the transformation equations for m, s, and d are independent of $\vec{L}_1$ and $\vec{L}_2$; and the equations for p_{b_x}, p_{w_x}, p_{b_y}, p_{w_y}, p_t, and p_a are independent of U, X_1, and X_2. Because of this, the probability $P(U, X_1, X_2, \vec{L}_1, \vec{L}_2)$ splits into a product of two marginal probability functions:

$$P(U, X_1, X_2, \vec{L}_1, \vec{L}_2) = P(U, X_1, X_2)\; P(\vec{L}_1, \vec{L}_2),$$

with

$$P(U,X_1,X_2) = \frac{1}{(4\pi^3 C_m X_{1,s}^2 C_s X_{1,d}^2 C_d)^{1/2}} \exp\left\{-\frac{(U-\frac{1}{2})^2}{C_m} - \frac{(X_1+X_2-2X_1^0)^2}{4X_{1,s}^2\,C_s} - \frac{[X_1-X_2-2X_{1,m}(U-\frac{1}{2})]^2}{4X_{1,d}^2\,C_d}\right\}, \quad (27)$$

and

$$P(\vec{L}_1, \vec{L}_2) = \frac{1}{(\pi^9 C_b^2 C_w^2 C_{p_b}^2 C_{p_w}^2 C_{p_t})^{1/2}} \left(\frac{1}{2L_{1,p_b}\,L_{1,p_w}}\right)^2 \int_{-\infty}^{+\infty} db_x \int_{-\infty}^{+\infty} dw_x \int_{-\infty}^{+\infty} db_y \int_{-\infty}^{+\infty} dw_y$$

$$\times \exp\left\{-\frac{(b_x^2+b_y^2)}{C_b} - \frac{(w_x^2+w_y^2)}{C_w} - \frac{[(L_{1x}-L_{2x}-2L_{1,b}b_x)^2+(L_{1y}-L_{2y}-2L_{1,b}b_y)^2]}{4L_{1,p_b}^2\,C_{p_b}} - \frac{[(L_{1x}+L_{2x}-2L_{1,w}w_x)^2+(L_{1y}+L_{2y}-2L_{1,w}w_y)^2]}{4L_{1,p_w}^2\,C_{p_w}} - \frac{(L_{1z}-L_{2z})^2}{2C_{p_t}}\right\} \delta\left(\frac{L_{1z}+L_{2z}}{\sqrt{2}}\right)$$

$$= \frac{\sqrt{2}}{(\pi C_{p_t})^{1/2}\pi^2 C'_{p_b} C'_{p_w}} \exp\left\{ - \frac{[(L_{1x} - L_{2x})^2 + (L_{1y} - L_{2y})^2]}{2C'_{p_b}} \right.$$

$$\left. - \frac{[(L_{1x} + L_{2x})^2 + (L_{1y} + L_{2y})^2]}{2C'_{p_w}} - \frac{(L_{1z} - L_{2z})^2}{2C_{p_t}} \right\} \delta(L_{1z} + L_{2z}). \qquad (28)$$

We have used the abbreviations*

$$C'_{p_b} = 2L_{1,p_b}^2 C_{p_b} + 2L_{1,b}^2 C_b,$$

$$C'_{p_w} = 2L_{1,p_w}^2 C_{p_w} + 2L_{1,w}^2 C_w.$$

The distribution $P(E, U, X_1, X_2, \vec{L}_1, \vec{L}_2)$ is of the form $\exp[-Q(E, U, X_1, X_2, L_{1x}, L_{2x}, L_{1y}, L_{2y}, L_{1z}, L_{2z})]$ times two Dirac delta functions, where $Q(E, U, X_1, X_2, L_{1x}, L_{2x}, L_{1y}, L_{2y}, L_{1z}, L_{2z})$ is a positive-definite quadratic function of its ten variables. In the language of statistics, $P(E, U, X_1, X_2, \vec{L}_1, \vec{L}_2)$ is a 10-variate normal distribution[56] multiplied by two delta functions. Similarly, the marginal probability function $P(U, X_1, X_2)$ is a trivariate normal distribution, and $P(\vec{L}_1, \vec{L}_2)$ is a 6-variate normal distribution multiplied by a delta function.

1. Distributions Obtained from $P(E, U, X_1, X_2)$

We have derived the lowest-order result for the probability of observing the two fragments at infinity with given values of total translational kinetic energy, fractional mass, and individual excitation energies:

$$P(E, U, X_1, X_2) = P(U, X_1, X_2)\, \delta(E + X_1 + X_2 - E^0 - 2X_1^0),$$

where $P(U, X_1, X_2)$ is given by (27). By integrating over the quantities

*If there were no angular momentum induced by the torque arising from the presence of the electrostatic field, then C'_{p_b} would equal C_{p_b}, since for this case $L_{1,p_b} = 1/\sqrt{2}$, and $L_{1,b} = 0$. An analogous statement holds for C'_{p_w}.

not of immediate interest in this probability distribution, we obtain marginal probability distributions for the observation of a smaller number of quantities. Each of the resulting probability functions is also a normal distribution (multiplied possibly by a delta function). For example, we obtain

$$P(X_1, X_2) = \frac{1}{[4\pi^2 X_{1,s}^2 C_s (X_{1,m}^2 C_m + X_{1,d}^2 C_d)]^{1/2}}$$

$$\times \exp\left[- \frac{(X_1 + X_2 - 2X_1^0)^2}{4X_{1,s}^2 C_s} - \frac{(X_1 - X_2)^2}{4(X_{1,m}^2 C_m + X_{1,d}^2 C_d)} \right],$$

$$P(U, X_1) = \frac{1}{[\pi^2 C_m (X_{1,s}^2 C_s + X_{1,d}^2 C_d)]^{1/2}} \exp\left\{ - \frac{(U - \frac{1}{2})^2}{C_m} - \frac{[X_1 - X_1^0 - X_{1,m}(U - \frac{1}{2})]^2}{X_{1,s}^2 C_s + X_{1,d}^2 C_d} \right\},$$

$$P(X_1) = \frac{1}{[\pi(X_{1,m}^2 C_m + X_{1,s}^2 C_s + X_{1,d}^2 C_d)]^{1/2}}$$

$$\times \exp\left[- \frac{(X_1 - X_1^0)^2}{X_{1,m}^2 C_m + X_{1,s}^2 C_s + X_{1,d}^2 C_d} \right],$$

and

$$P(U) = \frac{1}{(\pi C_m)^{1/2}} \exp\left[- \frac{(U - \frac{1}{2})^2}{C_m} \right].$$

We can also obtain from (27) the probability distribution for mass and total excitation energy X by substituting

$$X_1 = X - X_2$$

and integrating over X_2:*

$$P(U, X) = \frac{1}{(4\pi^2 C_m X_{1,s}^2 C_s)^{1/2}} \exp\left[- \frac{(U - \frac{1}{2})^2}{C_m} - \frac{(X - X^0)^2}{4X_{1,s}^2 C_s} \right].$$

An integration over U in this equation then gives

$$P(X) = \frac{1}{(4\pi X_{1,s}^2 C_s)^{1/2}} \exp\left[- \frac{(X - X^0)^2}{4X_{1,s}^2 C_s} \right].$$

Since the total translational kinetic energy and the total excitation energy are related to first order by the conservation-of-energy equation (20), the two preceding equations are completely equivalent to

$$P(E, U) = \frac{1}{[\pi^2 C_m a^2 C_s (E^0)^2]^{1/2}} \exp\left[- \frac{(U - \frac{1}{2})^2}{C_m} - \frac{(E - E^0)^2}{a^2 C_s (E^0)^2} \right] \quad (29)$$

and

$$P(E) = \frac{1}{[\pi a^2 C_s (E^0)^2]^{1/2}} \exp\left[- \frac{(E - E^0)^2}{a^2 C_s (E^0)^2} \right];$$

---

*This equation is also obtainable directly from $P(m, s)$ and the equations $U = \frac{1}{2} + m$ and $X = X^0 + 2X_{1,s}\, s$ [obtained from (16) by adding X_1 and X_2].

we have also used the relationship (21) between $X_{1,s}$ and aE^0. Similarly, Eq. (27) is equivalent to

$$P(E, U, X_1) = \frac{1}{[\pi^3 C_m a^2 C_s (E^0)^2 X_{1,d}^2 C_d]^{1/2}} \exp\left\{ - \frac{(U - \frac{1}{2})^2}{C_m} - \frac{(E - E^0)^2}{a^2 C_s (E^0)^2} - \frac{[X_1 - X_1^0 + \frac{1}{2}(E - E^0) - X_{1,m}(U - \frac{1}{2})]^2}{X_{1,d}^2 \; C_d} \right\}.$$

This result is useful for discussing, for example, the excitation energy of a single fragment, for given values of the total translational kinetic energy and fractional mass.

From the expression (29) for $P(E, U)$ and the transformation (18) relating the total translational kinetic energy E and the fractional mass U to the individual translational kinetic energies E_1 and E_2, we obtain the probability distribution for individual translational kinetic energies:

$$P(E_1, E_2) = P(E, U) \left| \frac{\partial (E, U)}{\partial (E_1, E_2)} \right|$$

$$= \frac{1}{[\pi^2 C_m a^2 C_s (E^0)^4]^{1/2}} \exp\left[- \frac{(E_1 + E_2 - E^0)^2}{a^2 C_s (E^0)^2} - \frac{(E_1 - E_2)^2}{4 C_m (E^0)^2} \right] ;$$

the equation resulting from the transformation has been reduced to lowest order in E_1 and E_2. By integrating over E_2 in this equation we obtain the distribution in translational kinetic energy of a single fragment:

$$P(E_1) = \frac{1}{[\pi (4C_m + a^2 C_s)(E_1^0)^2]^{1/2}} \exp\left[- \frac{(E_1 - E_1^0)^2}{(4C_m + a^2 C_s)(E_1^0)^2} \right] .$$

It should be recalled that for $x < x_{BG}$, C_m is negative, resulting in the mass distribution increasing rather than decreasing with mass asymmetry. The distributions involving U then approach infinity for large values of U and are thus not normalizable. In a contour plot of P(E, U) vs E and U, for example, the lines of constant probability are not ellipses for $x < x_{BG}$, but instead are two families of hyperbolas. One family represents lines of increasing probability for increasing mass asymmetry, whereas the other family represents lines of decreasing probability for deviations in the kinetic energy from its most probable value.

Since the above probability functions are normal distributions (of one or more variables), all the information they contain can be represented in terms of means, variances, and correlation coefficients (or alternatively, covariances).[56] For example, a monovariate normal (Gaussian) distribution is completely specified by two quantities: the mean and the variance (square of the standard deviation).* From the standard form for a Gaussian distribution (whose mean is $\bar{x}$ and whose variance is σ_x^2),

$$P(x) = \frac{1}{(2\pi\,\sigma_x^2)^{1/2}} \exp\left[-\frac{(x-\bar{x})^2}{2\,\sigma_x^2}\right],$$

we can determine by inspection the mean and the variance of each of the Gaussian distributions above.

---

*The mean $\bar{x}$ of the distribution P(x) is defined by

$$\bar{x} = \int x\,P(x)\,dx,$$

where the integration is over the allowed range of x. The variance σ_x^2, which is proportional to the square of the width of the distribution, is defined by

$$\sigma_x^2 = \int (x-\bar{x})^2\,P(x)\,dx\,.$$

To specify a bivariate normal distribution, five quantities are required in general: the mean and variance of each variable, and the correlation coefficient (or alternatively, the covariance). The correlation coefficient ρ_{xy} of the distribution $P(x, y)$ is defined by[57]

$$\rho_{xy} = \frac{\int dx \int dy\, (x-\bar{x})(y-\bar{y})\, P(x, y)}{\sigma_x\, \sigma_y}$$

The value of the correlation coefficient, which has a range of values from -1 to $+1$,[57] indicates the degree of correlation of the variables. A positive value of this coefficient means that the two variables are more likely to be simultaneously large or else simultaneously small; a negative value means that if one variable is large, the other is more likely to be small. The distributions $P(U, X)$ and $P(E, U)$ are seen to have correlation coefficients that are zero. On the other hand, the correlation coefficient for the distribution $P(X_1, X_2)$, for example, is nonzero; we find that it is given by

$$\rho_{X_1 X_2} = \frac{X_{1,s}^2 C_s - X_{1,m}^2 C_m - X_{1,d}^2 C_d}{X_{1,s}^2 C_s + X_{1,m}^2 C_m + X_{1,d}^2 C_d} . \tag{30}$$

Coefficients of correlation could be similarly calculated for the other distributions.

To illustrate the magnitudes of the widths of these distributions, we present sample graphs of some of the more important functions derived above. The results are for nonviscous fragments with the type of hydrodynamic flow we have been considering: a superposition of an irrotational flow and a flow corresponding to a uniform rotation. All graphs refer to the fission of the compound nucleus ${}_{85}At^{213}$, with a nuclear temperature at the saddle point of $\Theta = 1.13$ MeV. (This situation may be obtained experimentally, for example, by bombarding ${}_{83}Bi^{209}$ with 65-MeV alpha particles.) In two of the graphs we

illustrate the effect of temperature on the widths by also presenting the result for $\Theta = 0$. In Fig. 30 is presented a contour map of $P(X_1, X_2)$ vs X_1 and X_2. Figure 31 shows the distribution in excitation energy of a single fragment: $P(X_1)$ vs X_1. The distribution in total excitation energy [$P(X)$ vs X], which is equivalent to the distribution in total translational kinetic energy [$P(E)$ vs E], is shown in Fig. 32. The lowest-order result (29) for the distribution in mass and total translational kinetic energy is presented in Fig. 33 as a contour map of $P(E, U)$ vs E and U.

An interesting feature of the theory is the prediction that the excitation energies should be anticorrelated–if one fragment has a large excitation energy, then the other fragment is more likely to have a small excitation energy, and vice versa. This can be seen either from the contour map of $P(X_1, X_2)$ in Fig. 30, or from evaluating Eq. (30) for the correlation coefficient. For ${}_{85}At^{213}$ and $\Theta = 1.13$ MeV, we find that $\rho_{X_1 X_2} = -0.46$. For this same nucleus in the high-temperature (classical) limit, $\rho_{X_1 X_2} = -0.44$, whereas in the zero-temperature limit, $\rho_{X_1 X_2} = -0.58$. Thus the excitation energies are predicted to be somewhat more strongly anticorrelated at low temperatures than at high temperatures.

The physical reason for the anticorrelation in the excitation energies is very simple in the classical limit. The result can be interpreted in terms of the relative amplitudes of the stretching and distortion-asymmetry modes, since the excitation energy of a fragment at infinity depends primarily upon its initial elongation. Pure stretching-mode oscillations correspond to completely correlated fragment excitation energies, whereas pure distortion-asymmetric oscillations correspond to completely anticorrelated fragment excitation energies. The potential energy in the neighborhood of the saddle point is found to be "stiffer" with respect to stretching than with respect to distortion-asymmetry. The distortion-asymmetric oscillations therefore possess larger amplitudes than the stretching oscillations–hence, anticorrelation.

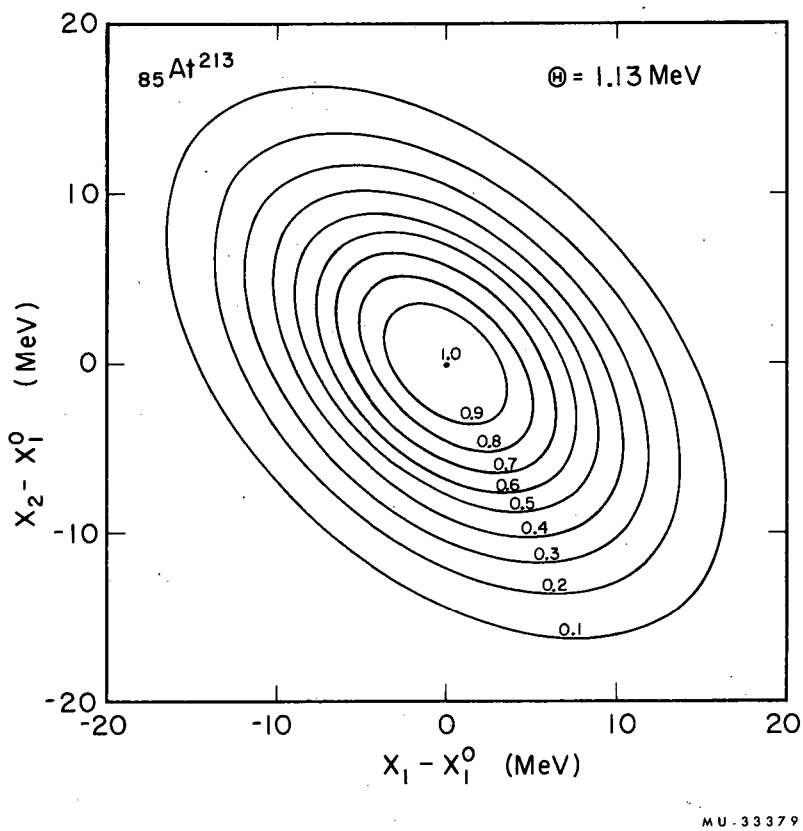

Fig. 30. Contour map of the probability distribution of individual fragment excitation energies, $P(X_1, X_2)$ vs X_1 and X_2. The lines of constant probability (ellipses with axes rotated 45° with respect to E and U axes) are labeled by relative probability. The value of X_1^0 is 21.3 MeV. The calculations are for the case of the compound nucleus $_{85}At^{213}$ (x = 0.677) at a nuclear temperature of 1.13 MeV ($_{83}Bi^{209}$ + 65-MeV α, for example).

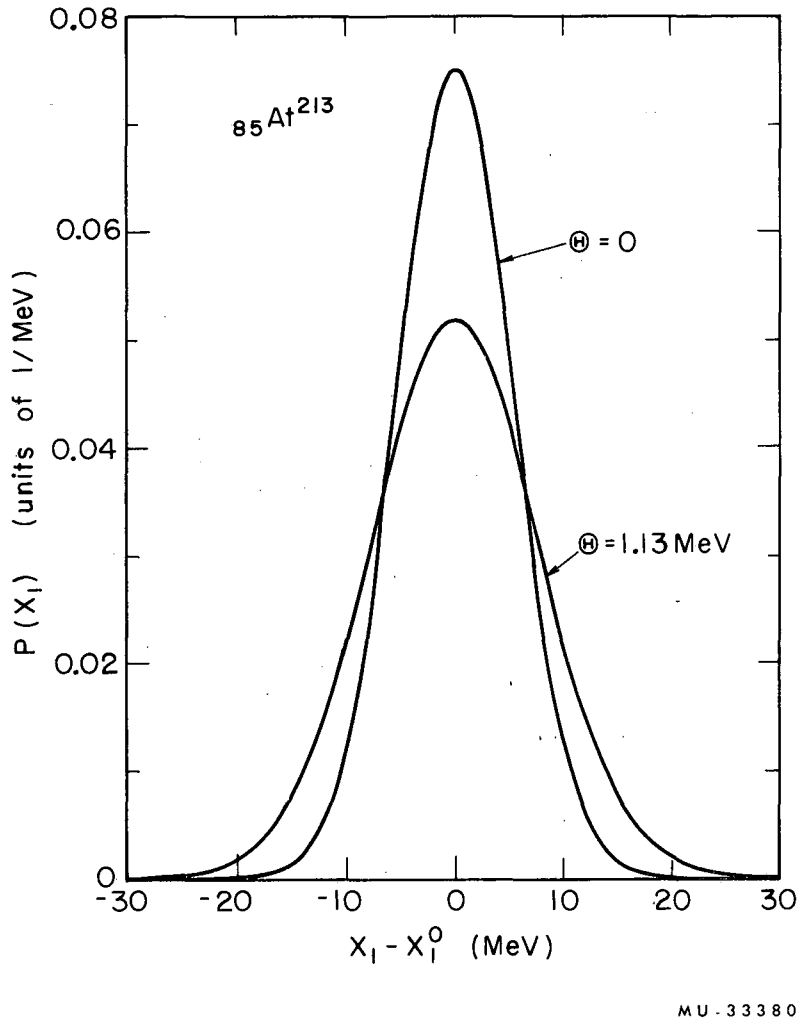

Fig. 31. The probability distribution of excitation energy of a single fragment, $P(X_1)$ vs X_1. The value of X_1^0 is 21.3 MeV. The calculations are for the case of non-viscous fragments and the fission of the compound nucleus $_{85}At^{213}$ (x = 0.677), at two different values of the nuclear temperature Θ.

The mean (or most probable) value of each of the quantities of interest is seen to be independent of nuclear temperature. However, since the constants C_s etc. are temperature-dependent, the widths (or variances) of the distributions are functions of nuclear temperature. As the temperature approaches zero, the widths of the distributions approach finite values determined by the quantum-mechanical zero-point vibrations of the appropriate oscillators. (Exceptions occur for the mass distribution and the distributions in the z-component of angular momentum. Since the mass-asymmetry and twisting frequencies are zero, the widths of these distributions approach zero as the nuclear temperature approaches zero.) For the fission of a typical lighter-than-radium nucleus, the zero-point full width at half maximum of the distribution in total translational kinetic energy is ≈ 10 MeV. The width of the kinetic-energy distribution arises primarily from oscillations in the distance between fragment centers (stretching mode). Because of the near cancellation of the opposing effects of the surface and Coulomb energies near the saddle point, the potential energy in the stretching direction is very flat. This means that a very small quantum-mechanical uncertainty in the stretching-mode potential energy (≈ 0.3 MeV) is "amplified" into a rather large zero-point width in the total translational kinetic-energy distribution. As the nuclear temperature increases, the uncertainty in the stretching-mode potential energy increases, resulting in a corresponding increase in the width of the kinetic-energy distribution.[3]

2. Distributions Obtained from $P(\vec{L}_1, \vec{L}_2)$

From Eq. (28) for $P(\vec{L}_1, \vec{L}_2)$ we can obtain several useful formulae involving the fragments' angular momenta. We first convert from cartesian coordinates to spherical coordinates through the transformation

$$L_{1x} = L_1 \sin\theta_1 \cos\phi_1,$$
$$L_{1y} = L_1 \sin\theta_1 \sin\phi_1,$$
$$L_{1z} = L_1 \cos\theta_1;$$

an analogous set of equations holds for fragment 2. The magnitude of $\vec{L}_1$ is L_1, the angle between the angular-momentum vector of fragment 1 and the z_1 axis (the line connecting the fragment centers) is θ_1, and the azimuthal angle is ϕ_1. We further define

$$\phi = \phi_1 - \phi_2,$$
$$\phi' = (\phi_1 + \phi_2)/2;$$

if one looks along the line connecting the spheroid centers, then ϕ is the angle between the fragments' components of angular momentum perpendicular to this line.

Equation (28) becomes, after an integration over ϕ' (upon which the probability function does not depend) is performed (the normalization is such that the range of ϕ is from 0 to π),

$$P(L_1, L_2, \theta_1, \theta_2, \phi) = \frac{4\sqrt{2}\, L_1^2 L_2^2 \sin\theta_1 \sin\theta_2}{(\pi C_{p_t})^{1/2}\, \pi C'_{p_b} C'_{p_w}}$$
$$\times \exp\left[-\frac{1}{2}\left(\frac{1}{C'_{p_b}} + \frac{1}{C'_{p_w}}\right)(L_1^2 + L_2^2) \right.$$
$$-\frac{1}{2}\left(\frac{1}{C_{p_t}} - \frac{1}{C'_{p_b}} - \frac{1}{C'_{p_w}}\right)(L_1^2 \cos^2\theta_1 + L_2^2 \cos^2\theta_2)$$
$$+\left(\frac{1}{C'_{p_b}} - \frac{1}{C'_{p_w}}\right) L_1 L_2 \sin\theta_1 \sin\theta_2 \cos\phi$$
$$\left. + \frac{1}{C_{p_t}} L_1 L_2 \cos\theta_1 \cos\theta_2 \right] \delta(L_1 \cos\theta_1 + L_2 \cos\theta_2). \tag{31}$$

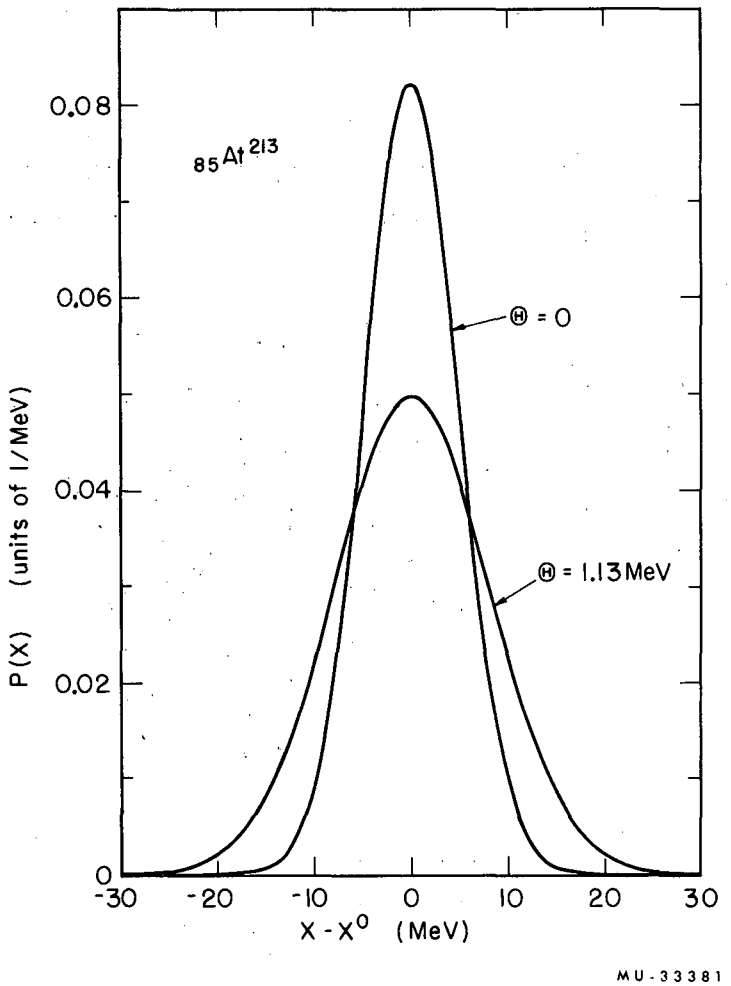

Fig. 32. The probability distribution of total fragment excitation energy, P(X) vs X. The value of X^0 is 42.6 MeV. The calculations are for the case of nonviscous fragments and the fission of the compound nucleus $_{85}At^{213}$ (x = 0.677), at two different values of the nuclear temperature Θ.

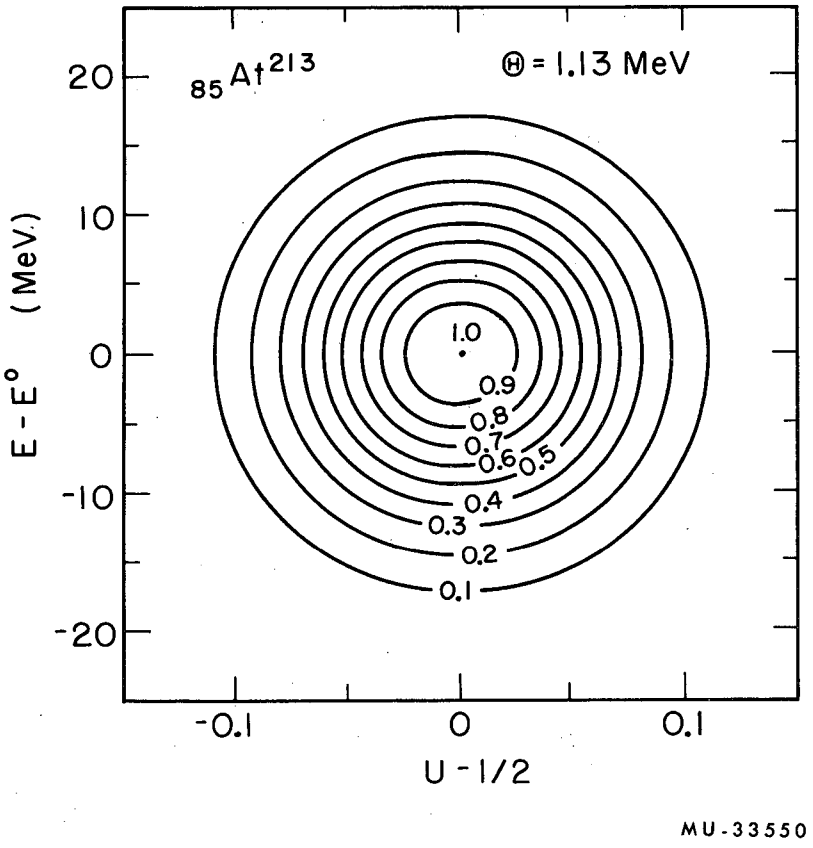

Fig. 33. Contour map of the probability distribution of total translational kinetic energy and fractional mass, P(**E**, U) vs E and U, calculated to lowest order [Eq. (29)]. The lines of constant probability (ellipses with axes parallel to the E and U axes) are labeled by relative probability. The value of E^0 is 151.4 MeV. The calculations are for the case of nonviscous fragments and the fission of the compound nucleus $_{85}At^{213}$ (x = 0.677) at a nuclear temperature of 1.13 MeV ($_{83}Bi^{209}$ + 65-MeV α, for example).

The integration over ϕ in this result can be performed explicitly to yield a probability distribution that is independent of azimuthal angles. Since[58]

$$\int_0^{\pi} \exp(x \cos\phi)\, d\phi = \pi\, I_0(x) = \pi\, J_0(ix)$$

$$= \pi \left[1 + (x/2)^2 + \frac{(x/2)^4}{1^2 \cdot 2^2} + \frac{(x/2)^6}{1^2 \cdot 2^2 \cdot 3^2} + \ldots \right], \quad (32)$$

where J_0 denotes the Bessel function of the first kind of order zero (I_0 is the modified Bessel function of the first kind of order zero); the expression for $P(L_1, L_2, \theta_1, \theta_2)$ can be obtained from (31) by inspection. Also, because of the presence of the delta function, an integration over any one of the four variables L_1, L_2, θ_1, θ_2 can be performed immediately.

In the zero-temperature limit the twisting mode is not excited, and the twisting-momentum constant C_{p_t} approaches zero. This means physically that the z component of angular momentum of each fragment is zero, and the angular-momentum vector of each fragment is perpendicular to its direction of motion. The integrations over both θ_1 and θ_2 in (31) can then be easily performed, yielding the zero-temperature result*

$$P(L_1, L_2, \phi) = \frac{4 L_1 L_2}{\pi C'_{p_b} C'_{p_w}} \exp\left[-\frac{1}{2}\left(\frac{1}{C'_{p_b}} + \frac{1}{C'_{p_w}}\right)(L_1^2 + L_2^2) + \left(\frac{1}{C'_{p_b}} - \frac{1}{C'_{p_w}}\right) L_1 L_2 \cos\phi \right];$$

---

*This expression could also be obtained directly from the set of equations (17) and $P(b_x, w_x, b_y, w_y, p_{b_x}, p_{w_x}, p_{b_y}, p_{w_y})$ by integrating over the bending and wriggling angles, and transforming the result to cylindrical coordinates.

it is understood that the zero-temperature limits of C'_{p_b} and C'_{p_w} are to be used. By employing (32) the zero-temperature result for $P(L_1, L_2)$ can also be written down by inspection.

If, in the original expression (28) for $P(\vec{L}_1, \vec{L}_2)$, we integrate with respect to L_{2x}, L_{2y}, and L_{2z}, we obtain the probability distribution for the angular momentum of a single fragment:

$$P(\vec{L}_1) = \frac{2\sqrt{2}}{(\pi C_{p_t})^{1/2}\pi(C'_{p_b} + C'_{p_w})} \exp\left[-\frac{2(L_{1x}^2 + L_{1y}^2)}{C'_{p_b} + C'_{p_w}} - \frac{2L_{1z}^2}{C_{p_t}}\right].$$

If we transform to spherical coordinates and integrate over the azimuthal angle (upon which the probability function does not depend), we obtain

$$P(L_1, \theta_1) = \frac{4\sqrt{2}\, L_1^2 \sin\theta_1}{(\pi C_{p_t})^{1/2}(C'_{p_b} + C'_{p_w})} \exp\left[-\frac{2L_1^2}{C'_{p_b} + C'_{p_w}} - 2\left(\frac{1}{C_{p_t}} - \frac{1}{C'_{p_b} + C'_{p_w}}\right) L_1^2 \cos^2\theta_1\right].$$

An integration over θ_1 can be carried out to give the probability of observing a single fragment with given magnitude of angular momentum:

$$P(L_1) = \frac{4L_1}{[(C'_{p_b} + C'_{p_w})(C'_{p_b} + C'_{p_w} - C_{p_t})]^{1/2}} \exp\left[-\frac{2L_1^2}{C'_{p_b} + C'_{p_w}}\right] \times H\left\{\left[\frac{2(C'_{p_b} + C'_{p_w} - C_{p_t})}{C_{p_t}(C'_{p_b} + C'_{p_w})}\right]^{1/2} L_1\right\},$$

where $H(x)$ is the error function, defined by[59]

$$H(x) = \frac{2}{\sqrt{\pi}} \int_0^x \exp(-a^2)\,da.$$

In the zero-temperature limit, C_{p_t} approaches zero, and this expression reduces to

$$P(L_1) = \frac{4L_1}{C'_{p_b} + C'_{p_w}} \exp\left[- \frac{2L_1^2}{C'_{p_b} + C'_{p_w}} \right] .$$

The various probability functions obtained from $P(\vec{L}_1, \vec{L}_2)$ are useful as regards specific information on the fragments' angular momenta at infinity. One has available in these formulae predictions regarding the magnitudes of the angular momenta, the angles between the angular momenta and the line connecting fragment centers, and the angle between the components of angular momentum perpendicular to the line connecting fragment centers. These predictions include the correlations between the various quantities, as well as their dependences upon nuclear temperature and fissionability parameter.

As a single example, we present in Fig. 34 the curve $P(L_1)$ vs L_1 for the compound nucleus ${}_{85}At^{213}$ and $\Theta = 1.13$ MeV. The result for nonviscous fragments with hydrodynamic flow of the type we have been considering is given by the solid line, and the result for infinitely viscous fragments by the dot-dashed line. We also indicate the result (short-dashed line) that would follow if the torque exerted by one fragment on the other through the electrostatic interaction were zero—this represents physically the distribution of angular momentum at the scission configuration. At scission, the most probable magnitude of the angular momentum of a fragment is seen to be about $8.5\hbar$. The corresponding most probable value at infinity is about $10\hbar$ for the nonviscous case, and about $15\hbar$ for the viscous case. The relatively large difference in the predicted angular-momentum distribution between the case of viscous fragments and the case of nonviscous fragments will perhaps make it possible to estimate experimentally the degree of nuclear viscosity. For this to be practicable, of course, the

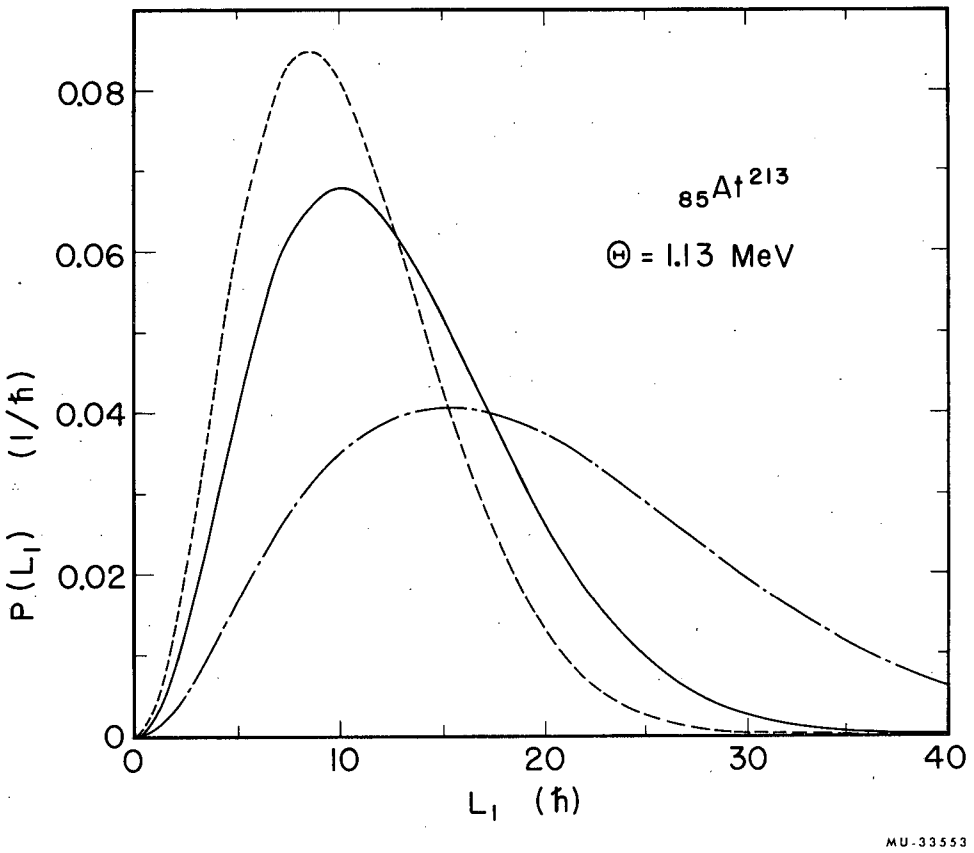

Fig. 34. The probability distribution of the magnitude of angular momentum of a single fragment, $P(L_1)$ vs L_1. The result that is calculated for nonviscous fragments (with hydrodynamic flow consisting of a superposition of an irrotational flow and a flow corresponding to a uniform rotation) is given by the solid line, and the result for infinitely viscous fragments by the dot-dashed line. The short-dashed line represents the result that would follow if the torque exerted by one fragment on the other through the electrostatic interaction were zero. The calculations are for the compound nucleus ${}_{85}At^{213}$ (x = 0.677) at a nuclear temperature of 1.13 MeV (${}_{83}Bi^{209}$ + 65-MeV α, for example).

present theory of fragment angular-momentum distributions would have to be refined to the stage where predictions can be trusted to within considerably better than 50%.

B. The Distribution P(E, U) to Higher Order

We have seen [Eq. (29) and Fig. 33] that to lowest order in the variables E and U, the probability function P(E, U) is a bivariate normal distribution; in a contour map of P(E, U) the lines of constant probability are ellipses whose axes are along E and U. This lowest-order result for P(E, U) is completely specified by E^0 (the mean value of E; the mean value of U is one-half from symmetry) and the two variances σ_E^2 and σ_U^2, which measure the widths of the distributions in E and U, respectively. By use of this result we are able to discuss the distribution in mass and total translational kinetic energy in the immediate vicinity of the most probable values.

If one desired to discuss the deviations of the distributions in mass and total translational kinetic energy from a bivariate normal distribution, then it would be necessary to include in the expression for P(E, U) higher-order terms in the variables E and U. Whereas in the bivariate normal distribution the exponent contains only the two quadratic terms $(E-E^0)^2$ and $(U-\frac{1}{2})^2$, the exponent of the distribution to the next higher order contains in addition the two cubic terms $(E-E^0)(U-\frac{1}{2})^2$ and $(E-E^0)^3$, and the quartic term $(U-\frac{1}{2})^4$.* In addition to the mean E^0 and the two variances, three additional quantities, the coefficients of the two cubic terms and the quartic term, are needed to specify P(E, U) to this order. These coefficients have

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*The other two cubic terms, $(E-E^0)^2(U-\frac{1}{2})$ and $(U-\frac{1}{2})^3$, are absent because P(E, U) is an even function of $(U-\frac{1}{2})$. The absence of the $(U-\frac{1}{2})^3$ term means that the $(U-\frac{1}{2})^4$ term is responsible for the first-order deviation of the distribution in mass from a Gaussian, and must then be considered to this order. The distribution P(E, U) would also, in general, have a pre-exponential dependence upon E and U.

simple physical significances: The coefficient of the $(E-E^0)^3$ term strongly affects the skewness[60] of the distribution in E, and determines the lowest-order deviation of the distribution in E from a pure Gaussian—whether the distribution has a high-energy or a low-energy tail, and by how much. Similarly, the coefficient of the $(U-\frac{1}{2})^4$ term strongly affects the kurtosis[60] (peakedness or flatness) of the distribution in U, and determines the lowest-order deviation of the distribution in U from a pure Gaussian—whether the distribution is more peaked than a Gaussian or has a flatter top. We will find that the coefficient of the $(E-E^0)(U-\frac{1}{2})^2$ term determines both how rapidly the most probable (or mean) value of E as a function of U falls off with $(U-\frac{1}{2})^2$, and also the derivative with respect to E (evaluated at $E=E^0$) of the variance of the mass distribution as a function of E. To the next higher order beyond including these three terms, the exponent of the P(E, U) distribution would contain the quartic term $(E-E^0)^4$, the sixth-order term $(U-\frac{1}{2})^6$, along with cross terms in E and U; and so on.

The derivation of the bivariate normal distribution (29) utilized a combination of two lowest-order results: (a) the equation (19) relating E linearly to the initial conditions, and the exact equation (15) for U, and (b) the initial-conditions probability distributions obtained in Section IV by expanding the potential energy about the saddle point and retaining only quadratic terms (harmonic approximation). We have available [Eq. (14)] a much more accurate equation relating E to the initial conditions than the first-order equation (19). In addition to taking into account the dependence of E on the mass-asymmetry coordinate m and the fission and stretching momenta p_f and p_s, Eq. (14) also includes a more precise dependence of E on the stretching coordinate s. However, the harmonic-oscillator probability distributions that we have used for the initial conditions represent the most accurate quantum-mechanical result that can be easily obtained. In order to obtain the best expression for P(E, U) that still incorporates initial conditions determined quantum-mechanically, we will use the same probability distributions for the initial conditions as before, but will

use the more accurate equation (14) for E and the exact equation (15) for U. Although the resulting expression for P(E, U) will be strictly accurate only to lowest order in E and U, it will in addition contain higher-order terms in E and U. The higher-order terms represent the effect of the nonlinear transformation (14) expressing E in terms of the initial conditions. Since there are two separate steps involved in the calculation of P(E, U), the treatment of one step essentially exactly and the other step to lowest order in E and U is not completely inconsistent.

We will later calculate an expression for P(E, U), valid in the classical limit, in which we use probability distributions for the initial conditions obtained by retaining certain anharmonic terms in the expansion of the potential energy about the saddle point.

1. <u>P(E, U) for Initial Conditions Determined in the Harmonic Approximation</u>

In order to calculate P(E, U) we need to invert the two equations (14) and (15) to obtain two of the initial conditions as functions of E and U and the remaining initial conditions. If we choose m and s as dependent variables, this inversion gives

$$m = U - \frac{1}{2}, \tag{33a}$$

$$s = \frac{1}{a}\left[\frac{4U(1-U)\,E^0}{E - M_\ell\left(\frac{2M_{c_1}^0 p_f}{M_0 M_f} + \frac{p_s}{M_s}\right)^2} - 1\right]. \tag{33b}$$

With this choice of dependent variables, the transformation from the initial probability distribution to the desired probability distribution is given by

$$P(E, U) = \int_0^\infty dp_f \int_{-\infty}^{+\infty} dp_s \; P(m, s, p_f, p_s) \left|\frac{\partial(m, s)}{\partial(E, U)}\right| ;$$

we have already integrated over the initial coordinates and momenta not involved in the transformation. It is understood that the set of equations (33) is substituted for m and s in $P(m, s, p_f, p_s)$, which is given by the product of the probability distribution for each normal coordinate and momentum appearing in its argument, as determined in Section IV.

The double integral over p_s and p_f cannot be expressed in a closed form. (By an appropriate change of variables, the double integral can be reduced to a single integral in which the new integrand includes an error function; in practice, this transformation is not very useful.) However, if we expand the expression (33b) for s in powers of the small quantity

$$\frac{M_\ell \left(\frac{2M_{c_1}^0 p_f}{M_0 M_f} + \frac{p_s}{M_s} \right)^2}{E}$$

everywhere it appears in the integrand, we can integrate the resulting expansion term by term, obtaining an asymptotic series expansion for $P(E, U)$. The result is found to be

$$P(E, U) = \frac{4U(1-U)E^0 F(E, U)}{(\pi^2 C_m a^2 C_s)^{1/2} E^2} \exp\left\{ - \frac{(U - \frac{1}{2})^2}{C_m} - \frac{[E - 4U(1-U)E^0]^2}{a^2 C_s E^2} \right\}, \tag{34}$$

where $F(E, U)$ is the asymptotic series

$$F(E, U) = 1 + \left\{ 1 + \frac{4U(1-U) E^0 [E - 4U(1-U) E^0]}{a^2 C_s E^2} \right\} \times \left[\frac{4(M_{c_1}^0 / M_0)^2 C_{p_f}}{M_f^2} + \frac{C_{p_s}}{M_s^2} \right] \frac{M_\ell}{E} + \cdots . \tag{35}$$

The function $F(E, U)$ is close to unity, except where the term in braces becomes very large. This occurs only when E and/or U are far

from their most probable values. (Since this is an asymptotic series rather than a power series, the expansion converges for a definite range of the variables E and U rather than for an infinite number of terms retained in the expansion.) Thus, the approximation $F(E, U) = 1$ is a very good one. [Setting $F(E, U) = 1$ is equivalent to neglecting in (14) the dependence of E on the initial momenta p_f and p_s.]

An alternative way of writing $P(E, U)$ is more useful for some purposes than (34). If we collect terms in (34) according to powers of $(U - \frac{1}{2})$, we obtain

$$P(E, U) = \frac{E^0 F(E, U)}{(\pi^2 C_m a^2 C_s)^{1/2} E^2} \exp\left[- \frac{(E - E^0)^2}{a^2 C_s E^2} \right]$$

$$\times [1 - 4(U - \tfrac{1}{2})^2] \exp\left[- G(U - \tfrac{1}{2})^2 - H^2 (U - \tfrac{1}{2})^4 \right], \tag{36}$$

where the quantities G and H (functions of E) are defined by

$$G = \frac{1}{C_m} + \frac{8 E^0 (E - E^0)}{a^2 C_s E^2}, \tag{37a}$$

$$H = \frac{4 E^0}{(a^2 C_s)^{1/2} E}. \tag{37b}$$

It is instructive to compare the current higher-order result for $P(E, U)$ with the lowest-order expression (29) derived earlier. Whereas (29) is a bivariate normal distribution, the current result is not—in a plot of $P(E, U)$ vs E and U, the lines of constant probability are not ellipses. In Fig. 35 is presented a contour map of the current result for $P(E, U)$; this may be compared directly with the map of the lowest-order result shown in Fig. 33. Near the position of maximum probability the contour lines are close to ellipses, but in the region of smaller probability they tend toward a triangular shape. The reason for this can be seen mathematically from Eq. (36) and the

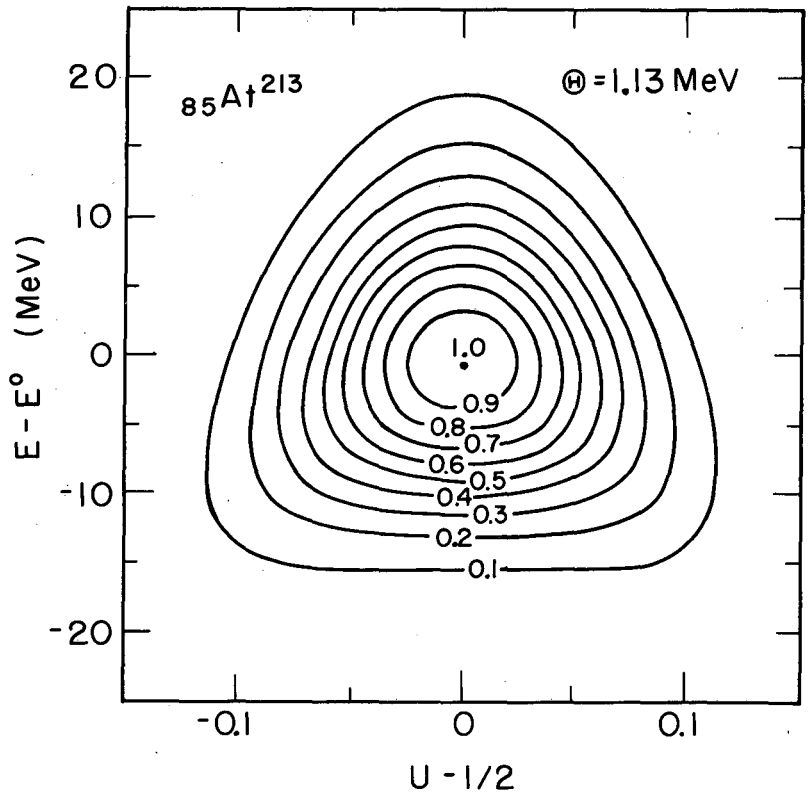

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Fig. 35. Contour map of the probability distribution of total translational kinetic energy and fractional mass, P(E, U) vs E and U, calculated for initial conditions determined in the harmonic approximation [Eq. (36)]. The lines of constant probability are labeled by relative probability. The value of E^0 is 151.4 MeV. The calculations are for the case of nonviscous fragments and the fission of the compound nucleus $_{85}At^{213}$ (x = 0.677) at a nuclear temperature of 1.13 MeV ($_{83}Bi^{209}$ + 65-MeV α, for example).

definition (37a) for G.* Since G is the coefficient of the $(U - \frac{1}{2})^2$ term in the exponent, $1/\sqrt{G}$ is proportional to the width of the mass distribution [as long as the coefficient H^2 of the $(U - \frac{1}{2})^4$ term is small]. Because G increases with increasing E (and vice versa), the mass distribution is narrow for high E and broad for low E. This means that the lines of constant probability acquire the appearance of rounded triangles in a plot of P(E, U) vs E and U.

If one extrapolates Eq. (36) for P(E, U) to very low values of the total translational kinetic energy E, then the lines of constant probability begin to curve in the opposite direction in the region near symmetry. [This occurs because the coefficient G of the $(U - \frac{1}{2})^2$ term becomes negative for sufficiently low values of E. This is just beginning to occur in Fig. 35 for the contour line of one-tenth maximum probability.] This would mean physically that the probability for obtaining a given mass is greater for asymmetric divisions than for symmetric ones. However, this aspect of the theory is changed if, when determining the probability distributions for the initial conditions, one takes into account terms beyond quadratic in the expansion of the potential energy about the saddle point (to be discussed in Section V. B.2). When the lowest-order anharmonic terms are retained, this effect starts occurring at a much lower value of E than that given by (36). [The contour plot of P(E, U) for the case in which these anharmonic terms are included is shown in Fig. 39.] It is not clear what effect the inclusion of still further anharmonic terms would have on P(E, U).

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*The triangular appearance of the contour lines results physically from a combination of two things: (a) the total translational kinetic energy has a linear term in the stretching coordinate s, but only a quadratic term in the mass-asymmetry coordinate m, and (b) the probability distributions in both s and m are Gaussians.

From the distribution $P(E, U)$ one may obtain, by performing suitable integrations or by differentiating, other quantities of physical interest. We now consider the calculation of approximate analytical expressions for such quantities; for these derivations, we use the very good approximation $F(E, U) = 1$.

By integrating over the total translational kinetic energy E, we obtain the distribution $P(U)$ for fractional mass:*

$$P(U) = \int_0^\infty P(E, U)\, dE = \frac{1}{(\pi C_m)^{1/2}} \exp\left[- \frac{(U - \frac{1}{2})^2}{C_m} \right]. \tag{38}$$

The mean total translational kinetic energy, as a function of fractional mass, is defined by

$$\overline{E}(U) = \frac{\int_0^\infty E\, P(E, U)\, dE}{P(U)} . \tag{39}$$

If we use the result (34) for $P(E, U)$, with $F(E, U) = 1$, and the result (38) for $P(U)$, we obtain†

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*This result follows directly from the equation $U = \frac{1}{2} + m$ and the distribution for $P(m)$, without the use of the approximation $F(E, U) = 1$. It may also be obtained by substituting (34) for $P(E, U)$, with $F(E, U) = 1$, and integrating.

†We note that as it stands this integral diverges logarithmically (at the upper limit)! This occurs because both the expression (14) for E as a function of s and the probability distribution (25a) in s are incorrect when s is far from zero–in particular, when the two fragments are so close together that they produce an infinite kinetic energy. In reality, as the distance between the two fragments approaches zero, the actual probability $P(s)$ falls off much more rapidly than a Gaussian distribution, because the potential energy increases much more rapidly than a parabola. Thus there should actually appear in the integrand an additional "damping" factor for large E, which would make the integral converge.

$$\overline{E}(U) = \frac{4U(1-U)E^0}{(\pi a^2 C_s)^{1/2}} \int_0^\infty \frac{dE}{E} \exp\left\{ - \frac{[E - 4U(1-U)E^0]^2}{a^2 C_s E^2} \right\}.$$

If we now make the substitution

$$E = \frac{4U(1-U)E^0}{1 + as},$$

this becomes*

$$\overline{E}(U) = \frac{4U(1-U)E^0}{(\pi C_s)^{1/2}} \int_{-\infty}^{+\infty} \frac{ds}{1+as} \exp\left(- \frac{s^2}{C_s} \right),$$

where we have replaced the lower limit $-1/a$ by $-\infty$. By expanding the denominator of the integrand and integrating term by term, we obtain an asymptotic series expansion in powers of $a^2 C_s$ for $\overline{E}(U)$:

$$\overline{E}(U) = 4U(1-U)E^0(1 + \frac{1}{2} a^2 C_s + \cdots). \tag{40}$$

The variance of the distribution in total translational kinetic energy, as a function of fractional mass, is defined by

$$\sigma_E^2(U) = \frac{\int_0^\infty [E - \overline{E}(U)]^2 P(E, U)\, dE}{P(U)}.$$

We obtain, by methods analogous to those used in calculating $\overline{E}(U)$, the asymptotic series expansion

$$\sigma_E^2(U) = \frac{1}{2} [4U(1-U)E^0]^2 a^2 C_s (1 + 4a^2 C_s + \cdots).$$

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*This result could also have been obtained—more easily—directly from (14), with the neglect of the initial momenta, and the probability $P(s)$.

Higher statistical moments of the distribution in total translational kinetic energy (as functions of fractional mass) could be calculated in a similar manner.

One may also obtain from $P(E, U)$ the most probable total translational kinetic energy, as a function of fractional mass, by solving the equation

$$\frac{\partial P(E, U)}{\partial E} = 0.$$

If we use the expression (34) for $P(E, U)$, with $F(E, U) = 1$, we obtain

$$E_{MP}(U) = \frac{4U(1 - U)E^0[(1 + 4\alpha^2 C_s)^{1/2} - 1]}{2\alpha^2 C_s}$$

$$= 4U(1 - U)E^0(1 - \alpha^2 C_s + \cdots)$$

$$= E^0[1 - 4(U - \tfrac{1}{2})^2](1 - \alpha^2 C_s + \cdots). \tag{41}$$

It is interesting to note that the most probable total translational kinetic energy (for a given U) is slightly less than $4U(1 - U)E^0$, whereas the mean value [Eq. (40)] is slightly greater. This is because the distribution $P(E, U)$ is not a normal distribution but instead has a small high-energy tail. The difference between $E_{MP}(U)$, $\overline{E}(U)$, and $4U(1 - U)E^0$ is small, however, since $\alpha^2 C_s$ is typically ≈ 0.005.

We now consider the calculation of integrals with respect to U over the distribution $P(E, U)$. The distribution of total translational kinetic energy is defined by

$$P(E) = \int_0^1 P(E, U)\,dU. \tag{42}$$

If we substitute (36) for $P(E, U)$, with $F(E, U) = 1$, and make the change of variables $m = U - \frac{1}{2}$, this becomes

$$P(E) = \frac{E^0}{(\pi^2 C_m a^2 C_s)^{1/2} E^2} \exp\left[- \frac{(E - E^0)^2}{a^2 C_s E^2} \right]$$

$$\times \int_{-\infty}^{+\infty} dm(1 - 4m^2) \exp(-H^2 m^4 - Gm^2),$$

where we have replaced the limits $-1/2$ and $+1/2$ by $-\infty$ and $+\infty$, respectively.

The resulting integral is not expressible in terms of elementary functions. We find it convenient for the present purposes to express it in terms of the functions $I_n(\beta)$ that are defined, for all values of n greater than $-1/2$, by the equation

$$I_n(\beta) = \int_{-\infty}^{+\infty} x^{2n} \exp(-x^4 - \beta x^2) dx = \int_0^{\infty} y^{n-(1/2)} \exp(-y^2 - \beta y)\, dy. \qquad (43)$$

Properties of these functions, including the differential equation satisfied by $I_n(\beta)$, recurrence relations, and asymptotic expansion, as well as their relationship with the repeated integrals of the error function,[61-63] are given in Appendix F. We present in Fig. 36 graphs of the functions $I_n(\beta)$ vs β for $n = 0, 1$, and 2.

The total translational kinetic-energy distribution can then be written in terms of these functions as

$$P(E) = \frac{E^0}{(\pi^2 C_m a^2 C_s)^{1/2} E^2} \exp\left[- \frac{(E - E^0)^2}{a^2 C_s E^2} \right]$$

$$\times \frac{1}{H^{1/2}} \left[I_0(G/H) - \frac{4 I_1(G/H)}{H} \right], \qquad (44)$$

where the energy-dependent quantities G and H are defined by (37). The second term in the brackets is much smaller than $I_0(G/H)$ and can usually be neglected.

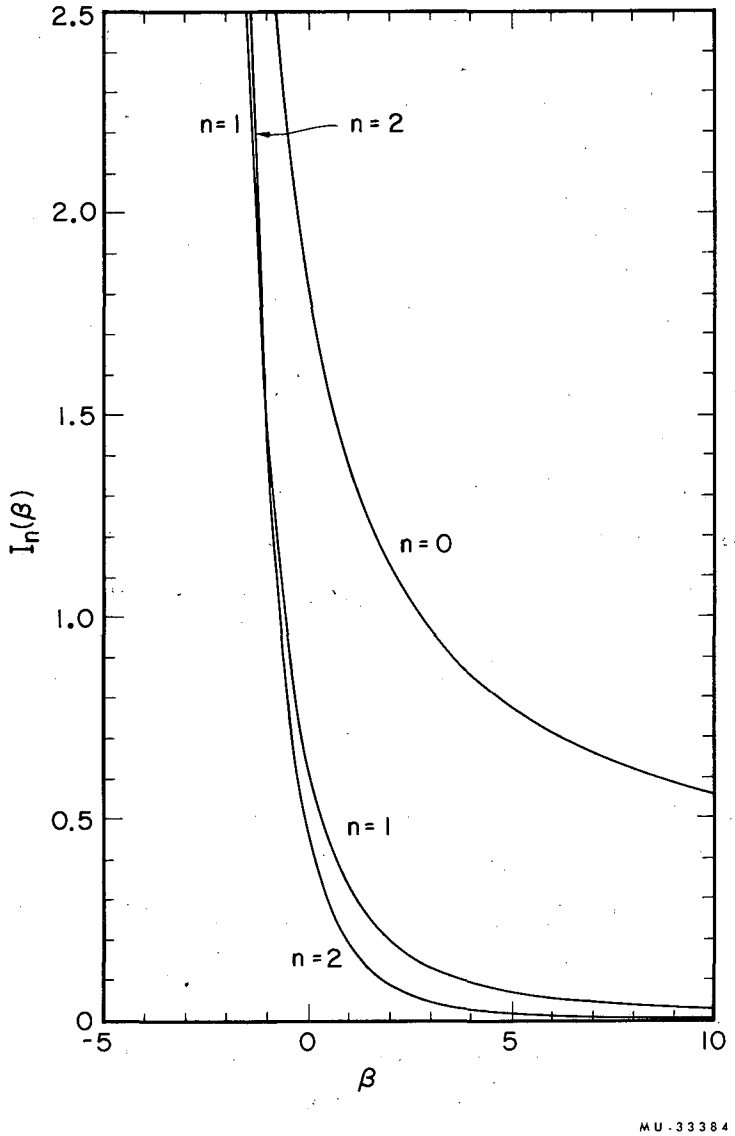

Fig. 36. The functions $I_n(\beta)$ [defined by Eq. (43)] vs β, for $n = 0$, 1, and 2.

The variance of the fractional mass, as a function of total translational kinetic energy, is defined by

$$\sigma_U^2(E) = \frac{\int_0^1 (U - \frac{1}{2})^2 \, P(E, U) \, dU}{P(E)} .$$

We obtain, in a manner analogous to the calculation of $P(E)$, the result

$$\sigma_U^2(E) = \frac{1}{H} \left[\frac{I_1(G/H) - \frac{4 I_2(G/H)}{H}}{I_0(G/H) - \frac{4 I_1(G/H)}{H}} \right] . \tag{45a}$$

If we neglect the second term in the numerator and the second term in the denominator, we obtain the simple result

$$\sigma_U^2 (E) \approx \frac{1}{H} \frac{I_1(G/H)}{I_0(G/H)} . \tag{45b}$$

A graph of the function $I_1(\beta)/I_0(\beta)$ vs β is given in Fig. 37. From the asymptotic expansion of $I_n(\beta)$ listed in Appendix F, we find that for large values of G/H

$$\sigma_U^2 (E) \rightarrow 1/(2G) . \tag{45c}$$

Higher statistical moments of the distribution in fractional mass (as functions of total translational kinetic energy) are obtainable just as readily as $\sigma_U^2(E)$. For example, the fourth central moment is given by

$$\mu_U^{(4)}(E) = \frac{\int_0^1 (U - \frac{1}{2})^4 P(E, U) dU}{P(E)} \approx \frac{1}{H^2} \frac{I_2(G/H)}{I_0(G/H)} ,$$

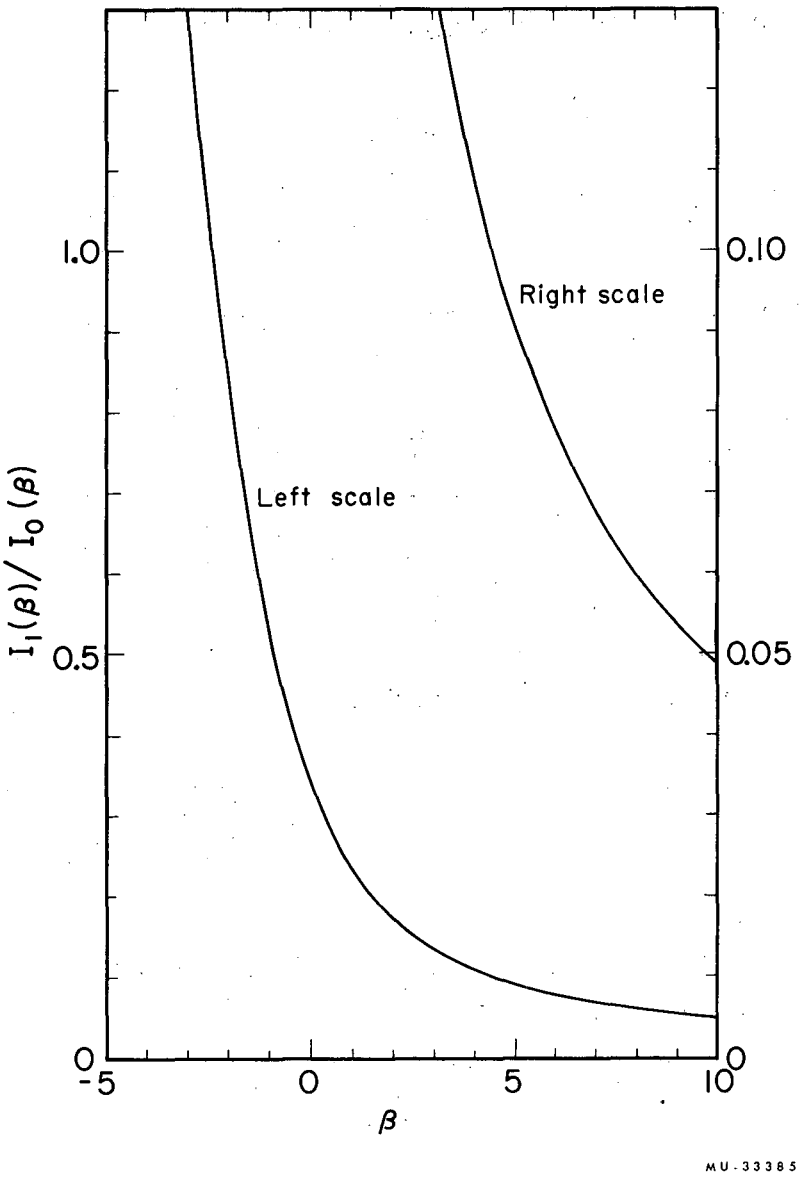

Fig. 37. The function $I_1(\beta)/I_0(\beta)$ vs β. The functions $I_n(\beta)$ are defined by Eq. (43).

where we have neglected all but the leading terms. Then, the kurtosis of the distribution[60] is given by

$$\alpha_U^{(4)}(E) = \frac{\mu_U^{(4)}(E)}{\sigma_U^4(E)} - 3 \approx \frac{I_2(G/H)\, I_0(G/H)}{I_1^2\,(G/H)} - 3. \tag{46}$$

A positive value of this coefficient means the distribution in mass is more peaked than a Gaussian, while a negative value means the distribution has a flatter top and is more rectangular than a Gaussian.

We consider finally the calculation of quantities obtained by integrating over both E and U in $P(E, U)$. The mean total translational kinetic energy (integrated over fractional mass) is defined by

$$\overline{E} = \int_0^\infty E\, dE \int_0^1 dU\, P(E, U) = \int_0^1 dU \int_0^\infty dE\, P(E, U)\, E.$$

If we use the definitions (42) and (39), respectively, we may write this as

$$\overline{E} = \int_0^\infty E\, P(E)\, dE = \int_0^1 \overline{E}(U)\, P(U)\, dU.$$

This is more easily evaluated in the second form; if we substitute (38) for $P(U)$ and (40) for $\overline{E}(U)$, and integrate, we obtain

$$\overline{E} = E^0(1 - 2C_m)(1 + \tfrac{1}{2}a^2 C_s + \cdots) = E^0(1 + \tfrac{1}{2}a^2 C_s - 2C_m + \cdots).$$

The variance of the total translational kinetic energy distribution is defined by

$$\sigma_E^2 = \int_0^\infty (E - \overline{E})^2\, dE \int_0^1 dU\, P(E, U) = \int_0^1 dU \int_0^\infty dE\, P(E, U)(E - \overline{E})^2.$$

The second form may be integrated, by methods analogous to those we have been using, to give the asymptotic series

$$\sigma_E^2 = \tfrac{1}{2}a^2 C_s (E^0)^2 \left(1 + 4a^2 C_s - 4C_m + \frac{16 C_m^{\,2}}{a^2 C_s} + \cdots\right).$$

Since the distribution $P(E, U)$ is symmetric in U about $U = 1/2$, the mean fractional mass (either as a function of E or integrated over E) is $1/2$. The variance of the distribution in fractional mass is obtainable directly from the Gaussian (38) for $P(U)$:

$$\sigma_U^2 = \frac{1}{2} C_m .$$

We shall postpone presenting any graphs of the quantities derived from $P(E, U)$ until we get to Section VI, where we compare the predictions with experimental data.

2. The Effect of Anharmonicity on $P(E, U)$

We are able to easily calculate quantum-mechanical probability distributions for the initial conditions only in the harmonic approximation. However, if the nuclear temperature is sufficiently high that classical statistical mechanics is valid for determining the initial conditions, then the probability distributions may be obtained to any desired order in the initial coordinates and momenta, or even exactly, from Eq. (22).

If we expand the potential energy about the saddle point and retain anharmonic terms in the coordinates, then we are able to discuss classically the effect of the anharmonicity of the potential energy on the distribution $P(E, U)$. By retaining appropriate anharmonic terms, we will be able to obtain classically the coefficients of the $(E - E^0)(U - \frac{1}{2})^2$, $(E - E^0)^3$, and $(U - \frac{1}{2})^4$ terms to a completely consistent order of approximation in both the initial conditions and the equations relating E and U to the initial conditions. We will then be able to discuss classically, to a consistent order of approximation, not only the mean E^0 and the variances σ_E^2 and σ_U^2, but also, for example, the decrease in the most probable (or mean) value of E with $(U - \frac{1}{2})^2$, the skewness of the total translational kinetic-energy distribution, and the kurtosis of the mass distribution.

We consider, then, the retention of anharmonic terms in the expansion of the potential energy about the saddle point. In order to

discuss the distribution $P(E, U)$ to the next higher order beyond a bivariate normal distribution, we need keep only those terms in the expansion of the potential energy that will affect the coefficients of the $(E - E^0)(U - \frac{1}{2})^2$, $(E - E^0)^3$, and $(U - \frac{1}{2})^4$ terms in the exponent of the $P(E, U)$ distribution. Because the equations (14) and (15) for E and U depend only upon the mass-asymmetry coordinate m and the stretching coordinate s (disregarding, for the moment, the initial momenta p_f and p_s), it suffices to consider (in addition to the harmonic terms) only the three anharmonic terms m^2s, s^3, and m^4 in an expansion of the potential energy.* We therefore expand the potential energy about the saddle point with respect to the variables m and s, retaining terms as follows:

$$\mathcal{V} = \mathcal{V}^0 + \frac{1}{2} K_m m^2 + \frac{1}{2} K_s s^2 + \frac{1}{2} K_{mms} m^2 s + \frac{1}{6} K_{sss} s^3 + \frac{1}{24} K_{mmmm} m^4 .$$

The classical probability distribution for the coordinates m and s is therefore given to this order by

$$P(m, s) = N \exp\left(- \frac{\frac{1}{2} K_m m^2 + \frac{1}{2} K_s s^2 + \frac{1}{2} K_{mms} m^2 s + \frac{1}{6} K_{sss} s^3 + \frac{1}{24} K_{mmmm} m^4}{\Theta} \right),$$

where N is a normalization constant.

Figure 38 shows graphs of the third derivatives K_{mms} and K_{sss} (evaluated at the saddle point) as functions of fissionability parameter. In the range of x below 0.78, the third derivative K_{mms}, which couples the mass-asymmetry coordinate and the stretching coordinate, is positive. This means that the most probable value of s decreases with increasing m^2; i.e., the most probable distance between spheroid centers decreases as the mass asymmetry increases. This results in a greater translational kinetic energy at infinity than if

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*The two cubic terms ms^2 and m^3 are absent because $\mathcal{V}$ is an even function of m. Nonzero cubic terms of the form sd^2, for example, introduce only pre-exponential dependences on E and U after the integration over d is performed.

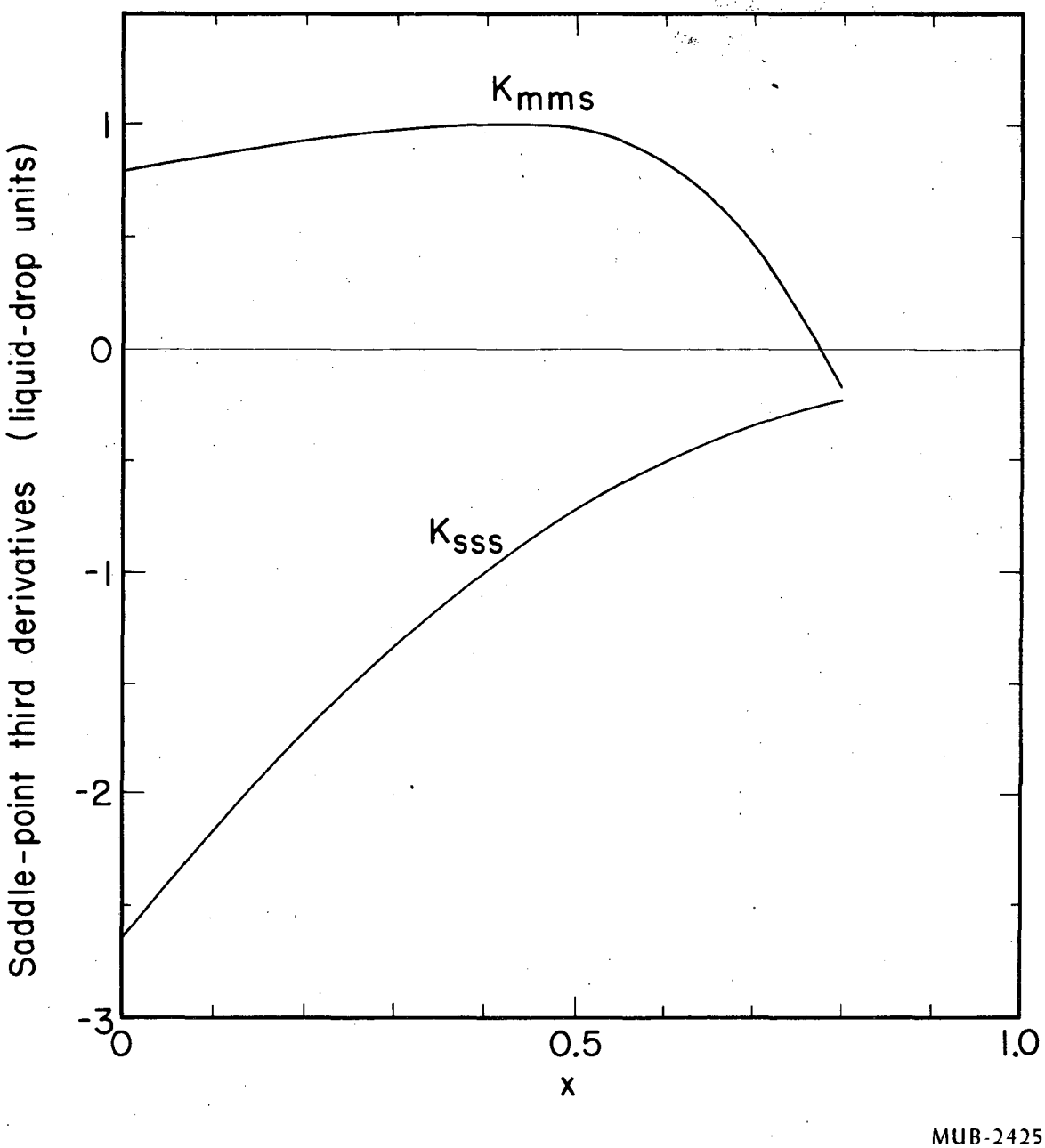

Fig. 38. Third derivatives of the potential energy, evaluated at the saddle point, as functions of fissionability parameter x. The quantity K_{mms} is in units of $E_S^{(0)}/R_0$, and K_{sss} is in units of $E_S^{(0)}/R_0^3$.

the distance remained unchanged. Over the entire range of x (between 0 and 0.80) the stretching third derivative K_{sss} is negative. The effect of this is a contribution toward a low-energy tail on the distribution in E. A rough estimate of the mass-asymmetry fourth derivative K_{mmmm} (evaluated at the saddle point) indicates that it is negative and fairly independent of x. For the $x = 0.677$ the estimate yields $K_{mmmm} \approx -15\, E_S^{(0)}$. The effect of this term is a contribution toward a peaked mass distribution, but the effect is small, since this term accounts for only about 20% of the final value of the $(U - \frac{1}{2})^4$ term in the exponent of $P(E, U)$. (In all numerical results involving anharmonic terms that we present, the quantity K_{mmmm} is taken to be zero. On the scale of the graphs presented here, these results are indistinguishable from those calculated by using the above estimate for K_{mmmm}.)

By using the set of equations (33) for m and s, and by taking into account also the classical probability distributions in the momenta p_f and p_s, we find for the desired distribution

$$P(E, U) = \frac{4U(1-U)NE^0 F(E, U)}{aE^2} \exp\left\{ -\frac{K_m(U-\frac{1}{2})^2}{2\Theta} - \frac{K_s[E - 4U(1-U)E^0]^2}{2\Theta a^2 E^2} + \frac{K_{mms}(U-\frac{1}{2})^2[E - 4U(1-U)E^0]}{2\Theta aE} + \frac{K_{sss}[E - 4U(1-U)E^0]^3}{6\Theta a^3 E^3} - \frac{K_{mmmm}(U-\frac{1}{2})^4}{24\Theta} \right\} . \tag{47}$$

The function $F(E, U)$ is given by (35), where it is understood that the classical limits are to be used for the constants C_s and C_{p_s} appearing in it.

If we collect terms in (47) according to powers of $(U - \frac{1}{2})$, we can alternatively write $P(E, U)$ as

$$P(E, U) = \frac{NE^0 F(E, U)}{a\,E^2} \exp\left[- \frac{K_s (E - E^0)^2}{2\Theta a^2 E^2} + \frac{K_{sss}(E - E^0)^3}{6\Theta\, a^3 E^3} \right]$$

$$\times \left[1 - 4(U - \tfrac{1}{2})^2\right] \exp\left[- G(U - \tfrac{1}{2})^2 - H^2 (U - \tfrac{1}{2})^4 + \frac{32\, K_{sss}(E^0)^3 (U - \frac{1}{2})^6}{3\Theta a^3 E^3} \right], \tag{48}$$

where G and H (functions of E) are now defined by

$$G = \frac{K_m}{2\Theta} + \frac{4K_s E^0 (E - E^0)}{\Theta a^2 E^2} - \frac{K_{mms}(E - E^0)}{2\Theta a E} - \frac{2K_{sss} E^0 (E - E^0)^2}{\Theta a^3 E^3},$$

$$H = \left[\frac{8K_s (E^0)^2}{\Theta a^2 E^2} - \frac{2K_{mms} E^0}{\Theta a E} - \frac{8K_{sss}(E^0)^2 (E - E^0)}{\Theta a^3 E^3} + \frac{K_{mmmm}}{24\,\Theta} \right]^{1/2}.$$

We present in Fig. 39 a contour map of $P(E, U)$ vs E and U for the compound nucleus ${}_{85}At^{213}$ and $\Theta = 1.13$ MeV, calculated by using the completely classical result (48). (The nuclear temperature is sufficiently high that classical statistical mechanics is valid for determining the initial conditions.) By comparing this with Fig. 35 we can see the effect of the anharmonicity of the potential energy on the final distribution (see also Fig. 33). The lines of constant probability still tend toward rounded triangles, but not as markedly as in Fig. 35.

If we neglect the $(U - \frac{1}{2})^6$ term in the exponential of (48), the resulting expression is of the same form as (36). Thus we are able to use the results previously derived for those quantities obtained by integrating over fractional mass: Eq. (44) for $P(E)$, Eq. (45) for $\sigma_U^2(E)$, and Eq. (46) for $a_U^{(4)}(E)$. Closed expressions for the quantities that result from integrating over E are not so readily obtainable because of the presence of the cubic term in E in the exponential of $P(E, U)$.

The two equivalent expressions (47) and (48) for $P(E, U)$, although strictly correct only to the next higher order beyond quadratic in E and U, contain further higher-order terms representing the

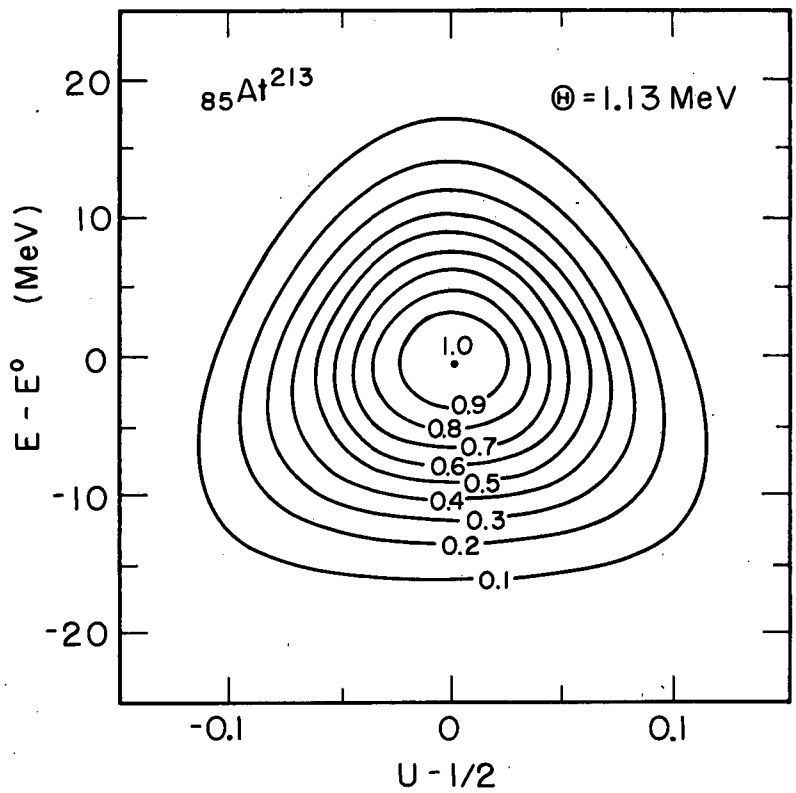

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Fig. 39. Contour map of the probability distribution of total translational kinetic energy and fractional mass, P(E, U) vs E and U, including effects of anharmonicity [Eq. (48)]. The lines of constant probability are labeled by relative probability. The value of E^0 is 151.4 MeV. The calculations are for the case of nonviscous fragments and the fission of the compound nucleus $_{85}At^{213}$ (x = 0.677) at a nuclear temperature of 1.13 MeV ($_{83}Bi^{209}$ + 65-MeV α, for example).

effects of the nonlinear equation (14) for E. If we retain in either of these expressions only terms that are one order higher than quadratic in E and U, we obtain (we regard any pre-exponential factors as of higher order)

$$P(E, U) = \frac{N}{aE^0} \exp\left[-\frac{K_m(U-\frac{1}{2})^2}{2\Theta} - \frac{K_s(E-E^0)^2}{2\Theta a^2 (E^0)^2} - \frac{(8K_s - aK_{mms})(E-E^0)(U-\frac{1}{2})^2}{2\Theta a^2 E^0} + \frac{\left(K_s + \frac{K_{sss}}{6a}\right)(E-E^0)^3}{\Theta a^2 (E^0)^3} - \frac{\left(\frac{8K_s}{a^2} - \frac{2K_{mms}}{a} + \frac{K_{mmmm}}{24}\right)(U-\frac{1}{2})^4}{\Theta} \right] . \tag{49}$$

This result represents a consistent order of approximation in both the initial conditions and the equations for E and U.

By solving

$$\frac{\partial P(E, U)}{\partial E} = 0$$

we obtain the most probable total translational kinetic energy as a function of fractional mass:

$$E_{MP}(U) = E^0 \left[1 - 4\left(1 - \frac{a K_{mms}}{8 K_s}\right)(U - \tfrac{1}{2})^2 + \cdots \right] . \tag{50}$$

It is instructive to compare this classical result with the result (41) obtained by use of the harmonic approximation for the initial conditions. For x in the neighborhood of 0.67, $a K_{mms}/(8K_s) \approx 1/5$; thus, the most probable total translational kinetic energy decreases with increasing $(U - \frac{1}{2})^2$ only about four-fifths as rapidly in (50) as in (41). The physical reason for this difference is that in (41) the decrease in total translational kinetic energy with increasing mass asymmetry results solely from the decrease in the product of the charges of the two fragments, whereas in (50) account is also taken of the decrease in distance between

spheroid centers (which tends to increase the kinetic energy) as the mass asymmetry increases.

In addition to determining the rate of decrease of $E_{MP}(U)$ with $(U-\frac{1}{2})^2$, the coefficient of the $(E-E^0)(U-\frac{1}{2})^2$ term also gives the derivative of the variance of the fractional mass at $E=E^0$. From (49) we find that

$$\sigma_U^2(E^0) = \Theta/K_m$$

and that

$$\frac{d\,\sigma_U^2(E^0)}{dE} = -\frac{\Theta(8K_s - aK_{mms})}{K_m^{\,2}\,a^2\,E^0}\,.$$

Since $8K_s > aK_{mms}$, the derivative is negative.

For values of x of interest in fission, the magnitude of the negative quantity $K_{sss}/(6a)$ is roughly one-half K_s; thus, the coefficient of the $(E-E^0)^3$ term in (49) is positive, indicating that the deviation of $P(E, 1/2)$ (the distribution in E for a symmetric mass division) from a pure Gaussian is toward a high-energy tail (positive skewness). The origin of the K_s term, which contributes toward the high-energy tail, is the nonlinear relationship (14) between $\dot{E}$ and s. The opposing contribution from the $K_{sss}/(6a)$ term toward a low-energy tail arises physically because the potential energy increases more rapidly as the fragments approach one another than as they separate.

An examination of the coefficient (including the over-all minus sign) of the $(U-\frac{1}{2})^4$ term in (49) indicates that it is negative. Thus, the prediction is that $P(E^0, U)$ (the distribution in mass for $E=E^0$) is less peaked and more rectangular than a Gaussian distribution (negative kurtosis).

In this section our concern has been the mathematical derivation and compilation of the formulae relevant to a discussion of the distributions in fragment total translational kinetic energy, mass, individual excitation energies, and individual angular momenta. By using these formulae and the graphs presented earlier for the constants appearing in them, curves expressing the theoretical predictions may be prepared for direct comparison with experiment. This will be done in the next section for distributions in total translational kinetic energy and fragment mass.

VI. COMPARISON OF THEORY WITH EXPERIMENT

In the previous section we derived probability distributions for certain observable characteristics of fission fragments at infinity: their total translational kinetic energy, fractional mass, individual excitation energies, and individual angular momenta. From our earlier discussion regarding the applicability of the two-spheroid model (Section II.B), we know that these distributions are expected to describe only the fission of nuclei that have values of the fissionability parameter $x \lesssim 0.67$.

Experimentally, there is observed a marked transition in the properties of fission at about radium ($x = 0.684$)—mass divisions are predominantly symmetric for elements lighter than radium and predominantly asymmetric for elements heavier than radium. It is not clear whether this experimentally observed transition is associated with the transition in saddle-point properties at $x \approx 0.67$; no explanation of asymmetric fission for the heavier elements is foreseen within the framework of the two-spheroid model. We will nevertheless use the experimentally observed transition between symmetric and asymmetric mass divisions as the dividing point for determining what data the theory should be compared with.* We will therefore compare the theoretical predictions of the model with existing experimental data for the fission of nuclei lighter than radium.

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*A recent experimental determination of the fission barrier of ${}_{81}Tl^{201}$ indicates that $(Z^2/A)_{crit}$ has a value[64] of 48.4 ± 0.5 (rather than Green's value[40] of 50.13 that is used here). When this value of $(Z^2/A)_{crit}$ is used, the value of the fissionability parameter x for each compound nucleus is increased somewhat. For the comparisons between theory and experiment that are made here, the largest value of fissionability parameter that occurs is 0.677 when Green's value of $(Z^2/A)_{crit}$ is used, and 0.701 when the newly reported value is used.

Fission-fragment mass distributions, obtained by use of radiochemical techniques, are available for the fission of several of the lighter elements.[65-70] Single-fragment translational kinetic-energy distributions have been determined for the fission of a number of light nuclei.[71, 72] In addition, two-dimensional distributions in mass and total translational kinetic energy, obtained by use of semiconductor detectors and two-dimensional analyzers,* have recently been reported for various nuclei lighter than radium.[71,73-78] On the other hand, for the fission of the lighter elements, there exists at the present time no experimental information regarding the distributions in individual excitation energies and individual angular momenta. The immediate test of the theory will therefore be the comparison of predictions with experiment for distributions in total translational kinetic energy and mass [P(E, U) and quantities derived from it].

The more detailed comparisons will be made with the data of Burnett[76] and with the data of Plasil,[77] whose experiments were carried out at the Lawrence Radiation Laboratory, Berkeley, simultaneously with the development of the theory. Burnett bombarded ${}_{83}Bi^{209}$ and ${}_{79}Au^{197}$ with alpha particles of energies 65.0 and 70.0 MeV, respectively, to form the compound nuclei ${}_{85}At^{213}$ and ${}_{81}Tl^{201}$. The corresponding values of the fissionability parameter x are 0.677 and 0.651, respectively. Plasil studied the heavy-ion-induced fission of the compound nuclei ${}_{76}Os^{186}$ (x = 0.619) and ${}_{82}Pb^{198}$ (x = 0.677) at several bombarding energies ranging from 102 to 165 MeV. The former was produced in two ways from the reactions ${}_{68}Er^{170} + {}_{8}O^{16}$ and ${}_{70}Yb^{174} + {}_{6}C^{12}$, and the latter from the reaction ${}_{74}W^{182} + {}_{8}O^{16}$.

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*These experiments consist of measuring in coincidence the translational kinetic energies E_1 and E_2 of the two fission fragments at infinity and recording the corresponding number of events. From this experimentally constructed distribution $P(E_1, E_2)$, the experimental distribution P(E, U) is obtained by use of the transformation (18).

We will also refer frequently to the data of Britt, Wegner, and Gursky,[75] and the data of Unik and Huizenga.[78] The former have studied the 25.5-MeV $_2He^3$-induced fission of $_{79}Au^{197}$, $_{81}Tl^{nat}$, $_{82}Pb^{206}$, and $_{83}Bi^{209}$; for the $_{83}Bi^{209}$ case, a bombarding energy of 22.1 MeV was also used. The latter authors have studied the 42-MeV helium-ion-induced fission of $_{83}Bi^{209}$. Comparisons will also be made with other data.

The theoretical distributions depend upon the nuclear temperature Θ at the saddle point, which is a function of the internal excitation energy E_{ex}^{SP} at the saddle point. The determination of Θ in terms of E_{ex}^{SP} is subject to an appreciable error. For the comparisons between theory and experiment made in this work, we use the semiempirical nuclear equation of state[79]

$$E_{ex}^{SP} = (A/8)\,\Theta^2 - \Theta,$$

where both Θ and E_{ex}^{SP} are in MeV, and A is the number of nucleons in the compound nucleus. The excitation energy at the saddle point is in turn given by the total bombarding energy in the center-of-mass system, plus the binding energy of the projectile to the target, minus the fission-barrier energy. The fission-barrier energy is equal to the liquid-drop fission-barrier energy,[15,17] * minus the shell correction to the ground-state mass,[80,81] plus the shell correction to the saddle-point mass. The saddle-point shell correction is not known, but probably does not exceed about 2 MeV—it was neglected here. These considerations are for a compound nucleus that is not rotating. The determination of the fission-barrier energy and the nuclear temperature for a rotating nucleus is discussed by Plasil;[77] his procedure is briefly touched upon below.

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*A plot of the liquid-drop fission-barrier energy vs x is given in Figs. 4 and 8. For the determination of Θ, the true liquid-drop fission-barrier energy is used rather than the two-spheroid approximation to the barrier.

Two complications present in the heavy-ion-induced fission reactions should be mentioned. First of all, because of the high excitation energies involved, it is possible that the compound nucleus will fission following partial de-excitation by the emission of one or more neutrons. If this occurs, there will be a spread in the saddle-point excitation energy (and hence the nuclear temperature Θ) at the time of fission. Secondly, heavy ions are capable of creating compound nuclei possessing large amounts of angular momentum. Because the ions strike the target with varying impact parameters, the angular momentum ranges from zero to some maximum value. Since the fission barrier is a sensitive function of angular momentum,[77,82,83] it will have a range of values, resulting in a spread in the saddle-point excitation energy (and hence Θ). The effect of both these complications on nuclear temperature was taken into account by Plasil.[77] He obtained an average value of Θ by calculating the competition between fission and neutron emission throughout the de-excitation chain,* and by integrating over the distribution of angular momentum.

Although the effect of angular momentum on the nuclear temperature can be taken into account as described above, it should be recalled that the entire theory developed here is restricted to the case of a nonrotating compound nucleus. This must be borne in mind when comparing the theoretical predictions with the data for the heavy-ion-induced reactions. All conclusions drawn on the basis of such comparisons are thus subject to the provision that angular momentum has little effect on the fission process, except in determining the average nuclear temperature at the saddle point.

The theoretical distributions are calculated for fragments observed (at infinity) before they have emitted any neutrons, whereas the experimental kinetic-energy measurements are made after the emission

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*For the cases studied it was found that the average number of neutrons emitted before fission seldom exceeds one; this means that the uncertainty in the nuclear temperature arising from this effect is small.

of neutrons from the fragments. Fragment neutron emission decreases the average translational kinetic energy and introduces a dispersion in the distributions, thereby increasing the variances. When comparing theory and experiment, it is desirable to correct the experimental results for such neutron-emission effects. The problem of neutron-emission corrections has been discussed by Terrell,[84] by Haines,[85] and by Burnett.[76] By making certain standard assumptions regarding the neutron emission, it is possible to derive formulae for correcting the mean total translational kinetic energy $\overline{E}(U)$, the variance $\sigma_E^2(U)$ of the total translational kinetic-energy distribution, and the variance $\sigma_U^2(E)$ of the mass distribution.

Using Burnett's neutron-correction formulae, Burnett and Plasil have corrected all statistical moments obtained from their data. (The formulae of Burnett differ slightly from those of Haines because certain higher-order terms are retained by Burnett.) With the exception of the mean total translational kinetic energies, the data of Britt, Wegner, and Gursky[75] have not been corrected for the effects of fragment neutron emission. The data of Unik and Huizenga[78] have been corrected for neutron-emission effects as regards the mean total translational kinetic energy and the full width at half maximum of the over-all total translational kinetic-energy distribution, but not otherwise.

Although neutron-emission corrections can be made for the statistical moments of the distributions, it is not possible to easily correct the distributions themselves. Thus, when comparing theoretical and experimental distributions, it must be borne in mind that the former refers to pre-neutron-emission and the latter to post-neutron-emission. To distinguish these two cases we denote by a superscript asterisk a quantity measured after neutron emission has occurred; e.g., E^* is the total translational kinetic energy measured following neutron emission.

We begin our comparison of theory with experiment by examining a series of graphs. [With the exception of Figs. 40 and 46, these figures have been reproduced (with additions, and changes in notation and format) from references 76 and 77.]

While making these comparisons it should be kept in mind that no arbitrary parameters have been adjusted—that the theoretical and experimental distributions have not been normalized to one another in any way. Also, it should be remembered that the theory is strictly valid only in the neighborhood of the most probable events; we will, however, extrapolate the theoretical curves to cover the entire region for which there are experimental data.

Figure 40 shows the experimental contour map of $P(E^*, U)$ vs E^* and U for the compound nucleus ${}_{85}At^{213}$, for which the nuclear temperature at the saddle point is $\Theta = 1.13$ MeV.[86] Apart from the effects of neutron emission, as discussed above, this experimental contour map may be compared directly with the theoretical maps calculated for this experimental situation in each of the three successively improved orders of approximation discussed in Section V — see Figs. 33, 35, and 39. (The same relative scales for E and U are used for all four graphs. However, the experimental contour lines are labeled differently from the theoretical ones.) Since the nuclear temperature is sufficiently high that classical statistical mechanics is valid for determining the initial conditions, the completely classical map calculated by including the effects of anharmonic terms on the initial conditions (Fig. 39) represents the best theoretical estimate available, and we will confine our discussion to a comparison with it.

The first thing to look for in comparing the theoretical and experimental distributions is how well the most probable values are reproduced; the experimental and theoretical most probable values of total translational kinetic energy are seen to agree remarkably well. In addition the theory predicts that the most probable value of U should be one-half, and this is verified experimentally—the fission events are predominantly symmetric in mass rather than asymmetric. The second thing to compare is the widths of the distributions in E and U; the widths of both distributions are seen to compare excellently. Finally, we may compare the shapes of the distributions with the theoretical predictions of approximately bivariate normal distributions

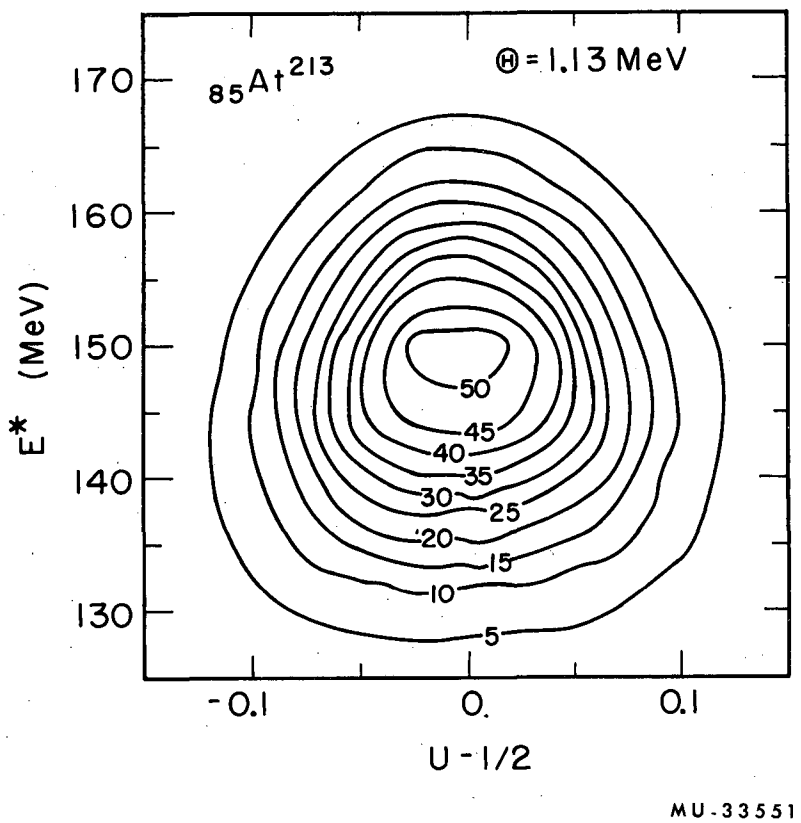

Fig. 40. Experimental contour map of the probability distribution of total translational kinetic energy and fractional mass, $P(E^*, U)$ vs E^* and U. The superscript asterisk denotes that E is measured after neutron emission from the fragments has occurred. The data are those of Burnett[86] for the compound nucleus $_{85}At^{213}$ (x = 0.677), formed from the reaction $_{83}Bi^{209}$ + 65-MeV α, for which Θ = 1.13 MeV. The labels on the lines of constant probability have the following significance: the contour labeled by 10, for example, passes through those regions of the E^* - U plane where an area of 6 MeV by 3 amu contains 1% of the total number of events.

modified by certain characteristic higher-order terms. The over-all agreement between the shapes of the experimental and theoretical contour lines is strikingly good. In the region of maximum probability the experimental contour lines are approximately ellipses, whereas in the region of smaller probability they tend to become rounded triangles, as predicted by the theory: The experimental mass distribution, as predicted, is broader for low values of the total translational kinetic energy than for high values. Other details of the agreement between the theoretical and experimental maps will be compared later in the form of statistical moments.

In reference 77 comparison is made by Plasil between theoretical and experimental contour maps of P(E, U) for the fission of two heavy-ion-induced reactions. The agreement presented there is not as good as for the ${}_{85}At^{213}$ case we have discussed. However, the theoretical maps there are calculated using the intermediate approximation for P(E, U) in which the initial conditions are determined in the harmonic approximation. The agreement is significantly improved when the effects of anharmonicity on the initial conditions are taken into account. Also, the compound nuclei undergoing fission possessed considerable angular momentum, which could possibly affect the experimental distributions.

Figure 41 shows the theoretical and experimental distributions in mass, P(U), and the theoretical and experimental distributions in total translational kinetic energy, P(E), for the compound nucleus ${}_{82}Pb^{198}$, each at two different nuclear temperatures. The theoretical curves are calculated in the intermediate approximation for P(E, U) in which the initial conditions are determined in the harmonic approximation.* As before, we first compare the most probable values of

*Although the approximate formulae derived in Section V for P(U), P(E), and the various statistical moments are sufficiently accurate for calculating the theoretical curves of this section, the curves have actually been calculated by numerical integrations over the full expression (36) [or the full expression (48)] for P(E, U), with F(E, U) retained.

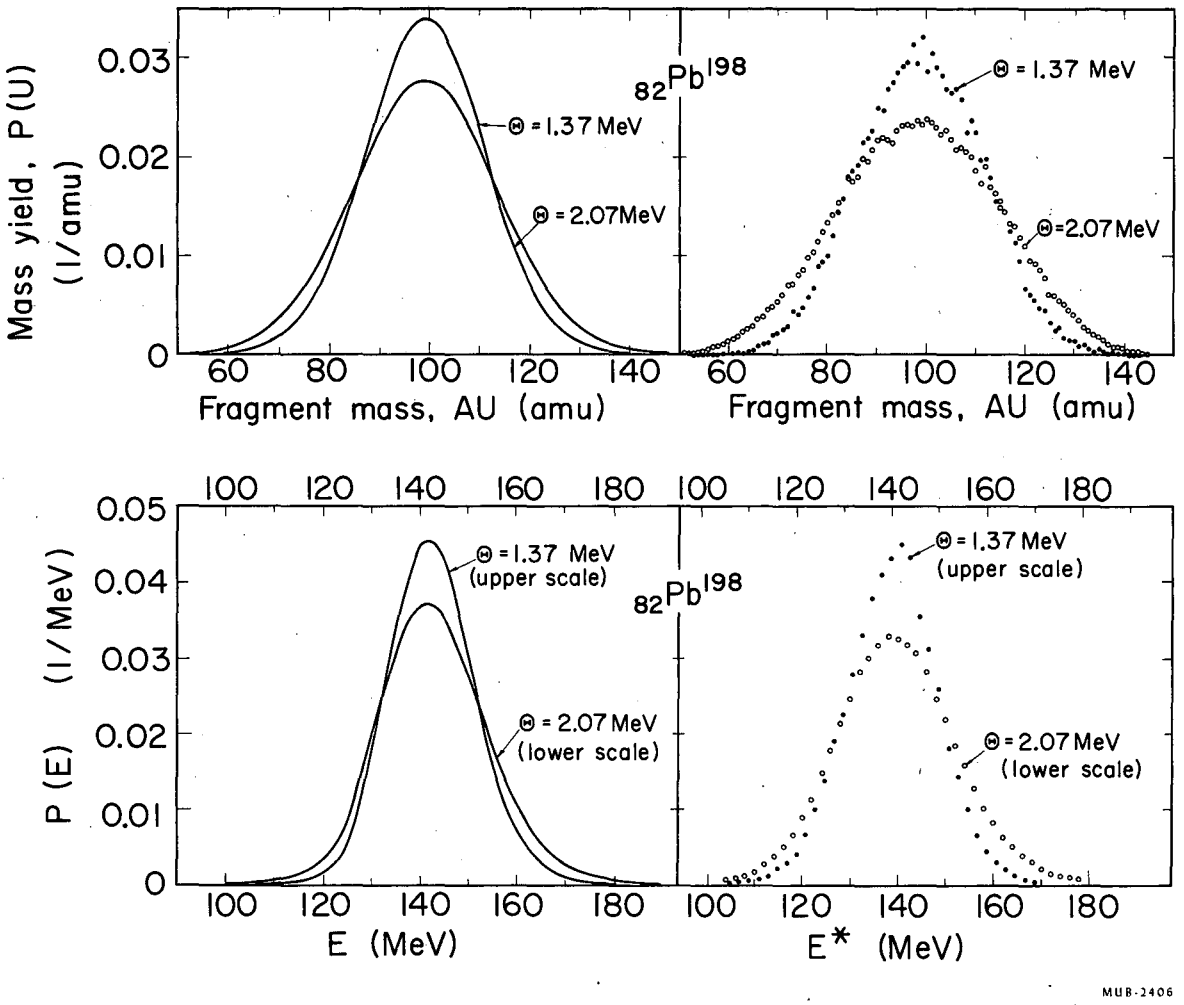

Fig. 41. Probability distributions for fragment mass (top) and for total translational kinetic energy (bottom). The theoretical curves (solid lines) are calculated for initial conditions determined in the harmonic approximation [from Eq. (36)]. The data are those of Plasil [77] for the compound nucleus $_{82}Pb^{198}$ (x = 0.677), formed from the heavy-ion-induced reaction $_{74}W^{182}$ + $_{8}O^{16}$. The results for a bombarding energy of 102 MeV (Θ = 1.37 MeV) are given by the solid points, and for a bombarding energy of 165 MeV (Θ = 2.07 MeV) by the open circles. The superscript asterisk denotes that E is measured after neutron emission from the fragments has occurred.

the total translational kinetic energy, which are seen to agree favorably. Next, the widths of the experimental distributions in both kinetic energy and mass are seen to be reproduced well by the theory. In addition, as the nuclear temperature increases, the theory is seen to correctly predict an increase in width of each distribution. Finally, we note that the over-all shapes of the experimental and theoretical distributions in both E and U are in approximate agreement (each distribution is approximately Gaussian). Because of experimental uncertainties, including the uncertainty of the effects on the distributions of neutron emission from the fragments, we will not compare numerical results for skewness in P(E) and kurtosis in P(U).

We turn now to a comparison of the mean values of the total translational kinetic energy as functions of fragment mass. Shown in Fig. 42 is $\overline{E}(U)$ vs fragment mass AU for the compound nuclei ${}_{85}At^{213}$, ${}_{81}Tl^{201}$, and ${}_{82}Pb^{198}$, the latter for two temperatures. In this figure, as well as in the two succeeding ones, we indicate by a solid line the result obtained by use of the intermediate approximation (36) for P(E, U), in which the initial conditions are determined in the harmonic approximation. The dashed line represents the result calculated from the expression (48) for P(E, U), in which anharmonic terms are considered in determining the initial conditions. Since the nuclear temperature is sufficiently high that classical statistical mechanics is valid for determining the initial conditions, the dashed line in each case represents the better theoretical estimate, and we will confine our discussion to a comparison of the experimental results with it.

We note first of all in Fig. 42 that for three of the four cases the agreement between theory and experiment as regards the mean total translational kinetic energy at symmetry is excellent. For the remaining case (${}_{82}Pb^{198}$ at $\Theta = 2.07$ MeV) the experimental value is slightly higher than the theoretical one. For the ${}_{85}At^{213}$ and ${}_{81}Tl^{201}$ cases the experimental decrease in $\overline{E}(U)$ with increasing mass asymmetry is essentially as predicted by the theory, although the experimental points lie somewhat above the theoretical curves. However,

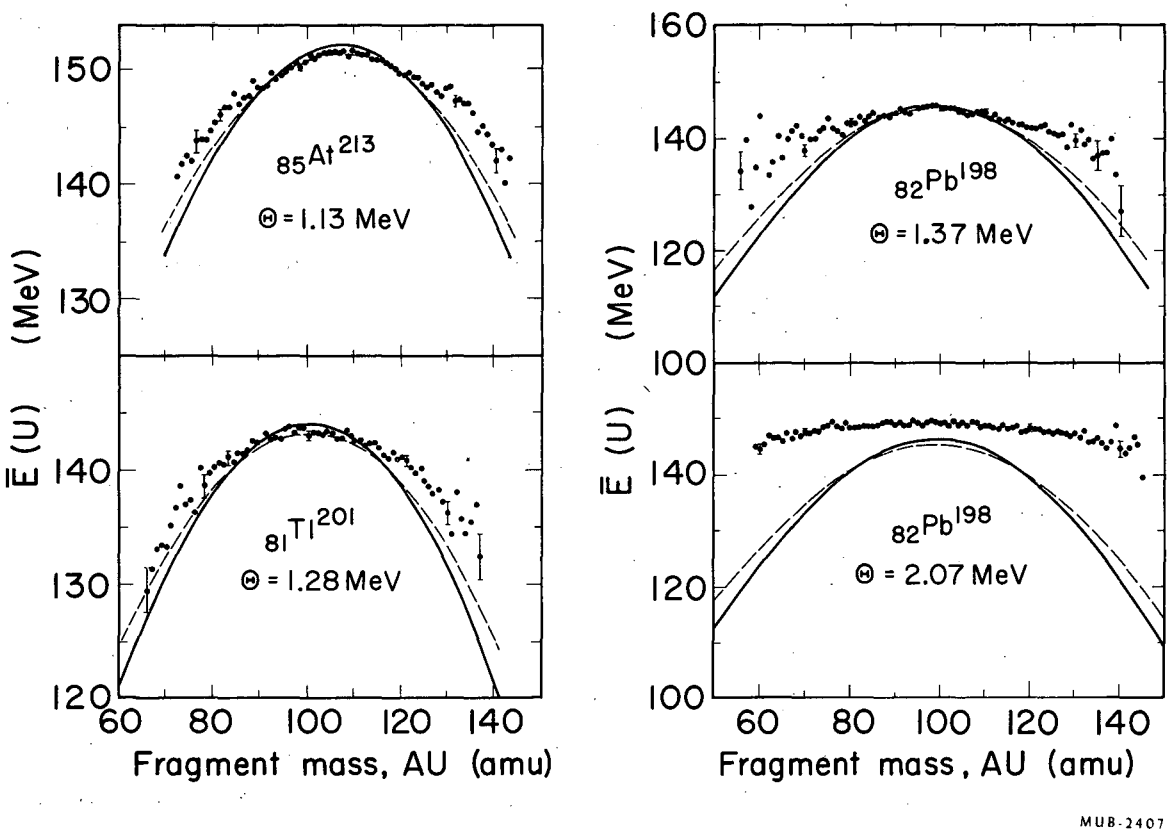

Fig. 42. Mean total translational kinetic energies, as functions of fragment mass AU. The theoretical results that are calculated for initial conditions determined in the harmonic approximation [from Eq. (36)] are given by the solid lines, and the results that are calculated by including effects of anharmonicity [from Eq. (48)] are given by the dashed lines. The data (solid points) are as follows: Burnett:[76] $_{83}Bi^{209}$ + 65-MeV α = $_{85}At^{213}$ (x = 0.677, Θ = 1.13 MeV), and $_{79}Au^{197}$ + 70-MeV α = $_{81}Tl^{201}$ (x = 0.651, Θ = 1.28 MeV); Plasil:[77] $_{74}W^{182}$ + 102-MeV $_{8}O^{16}$ = $_{82}Pb^{198}$ (x = 0.677, Θ = 1.37 MeV), and $_{74}W^{182}$ + 165-MeV $_{8}O^{16}$ = $_{82}Pb^{198}$ (x = 0.677, Θ = 2.07 MeV). Note that the left-hand scale and the right-hand scale are different.

for the two $_{82}Pb^{198}$ cases (formed by heavy-ion bombardments), the experimental points do not drop off with increasing asymmetry nearly as fast as the theoretical curves. Indeed, for the case in which $\Theta = 2.07$ MeV, the experimental values are essentially independent of asymmetry.

The data of Britt, Wegner, and Gursky indicate that $\overline{E}(U)$ decreases with increasing mass asymmetry, although they find in some cases (in particular, $_{83}Bi^{209}$ + 25.5-MeV $_{2}He^{3}$) a tendency for $\overline{E}(U)$ to actually increase at large asymmetry.[75] (Note that these authors plot their data as functions of fragment mass ratio rather than fragment mass or fractional mass.) The experimental $\overline{E}(U)$ curve of Unik and Huizenga also decreases with increasing mass asymmetry.[78]

As Haines has pointed out, the discrepancy between the calculated and experimental $\overline{E}(U)$ curves would be reduced somewhat if account were taken of the change in the mean charge density of fission fragments with a change in mass asymmetry.[85] On the average, the lighter fragment acquires a slightly larger number of protons than its proportionate share, and the heavier fragment acquires a slightly smaller number. The product of the charges of the two fragments, and hence their translational kinetic energy at infinity, is thus increased somewhat over the corresponding value calculated here (in which both fragments have the same charge density). This effect is of the same order of magnitude as the effect of the anharmonicity of the potential energy, and in the case of $_{85}At^{213}$ and $_{81}Tl^{201}$ taking this effect into account would come close to removing the difference between the calculated and experimental $\overline{E}(U)$ curves. On the other hand, the large discrepancy between the calculated and experimental curves for the $_{82}Pb^{198}$ cases would still exist even if the effect arising from the difference in fragment charge densities were taken in account.

The suggestion has been made by Plasil that the angular momentum present in the $_{82}Pb^{198}$ cases is possibly responsible for the marked deviation of the experimental $\overline{E}(U)$ curves from the theoretical ones.[77] This suggestion is consistent with the fact that for the two cases in which very little angular momentum is present ($_{85}At^{213}$

and $_{81}Tl^{201}$, formed by alpha-particle bombardments) the theory and experiment are in substantial agreement, and for the $_{82}Pb^{198}$ case in which there is less angular momentum present (Θ = 1.37 MeV) the agreement is better than for the case of greater angular momentum present (Θ = 2.07 MeV). To see if this actually is the explanation, the theory should be worked out taking into account angular momentum; this presents an interesting problem for the future.

In Fig. 43 we compare the theoretical and experimental variances of the total translational kinetic energy distributions as functions of fragment mass [σ_E^2(U) vs AU] for the same experimental situations as in Fig. 42. The agreement near symmetry is excellent for the $_{85}At^{213}$ and $_{81}Tl^{201}$ cases. For $_{82}Pb^{198}$ (heavy-ion reactions) the agreement near symmetry is good for the Θ = 1.37-MeV case, but for the Θ = 2.07-MeV case the experimental points are somewhat higher than the theoretical values. In all cases the theoretical curves decrease with increasing mass asymmetry. This prediction is not borne out experimentally in the $_{85}At^{213}$ and $_{81}Tl^{201}$ cases—these experimental variances increase with increasing mass asymmetry. The trends of the two $_{82}Pb^{198}$ variances with mass asymmetry are rather uncertain.

Britt, Wegner, and Gursky have found in three of the four cases reported that the variance σ_E^2(U) is essentially independent of fragment mass.[75] In the fourth case ($_{83}Bi^{209}$ + 25.5-MeV $_2He^3$) σ_E^2(U) was observed to remain essentially constant near symmetric mass divisions and to increase for more asymmetric divisions. The data of Unik and Huizenga also indicate that σ_E^2(U) is essentially independent of fragment mass.[78]

The comparisons made for σ_E^2(U) indicate a fairly significant disagreement between theory and experiment. The theoretical curves would be scarcely changed if one were to take into account further anharmonic terms in the potential energy—any pure liquid-drop result that predicts that the mean total translational kinetic energy should decrease with increasing mass asymmetry (see Fig. 42) will also predict that the variance of the total translational kinetic energy should decrease

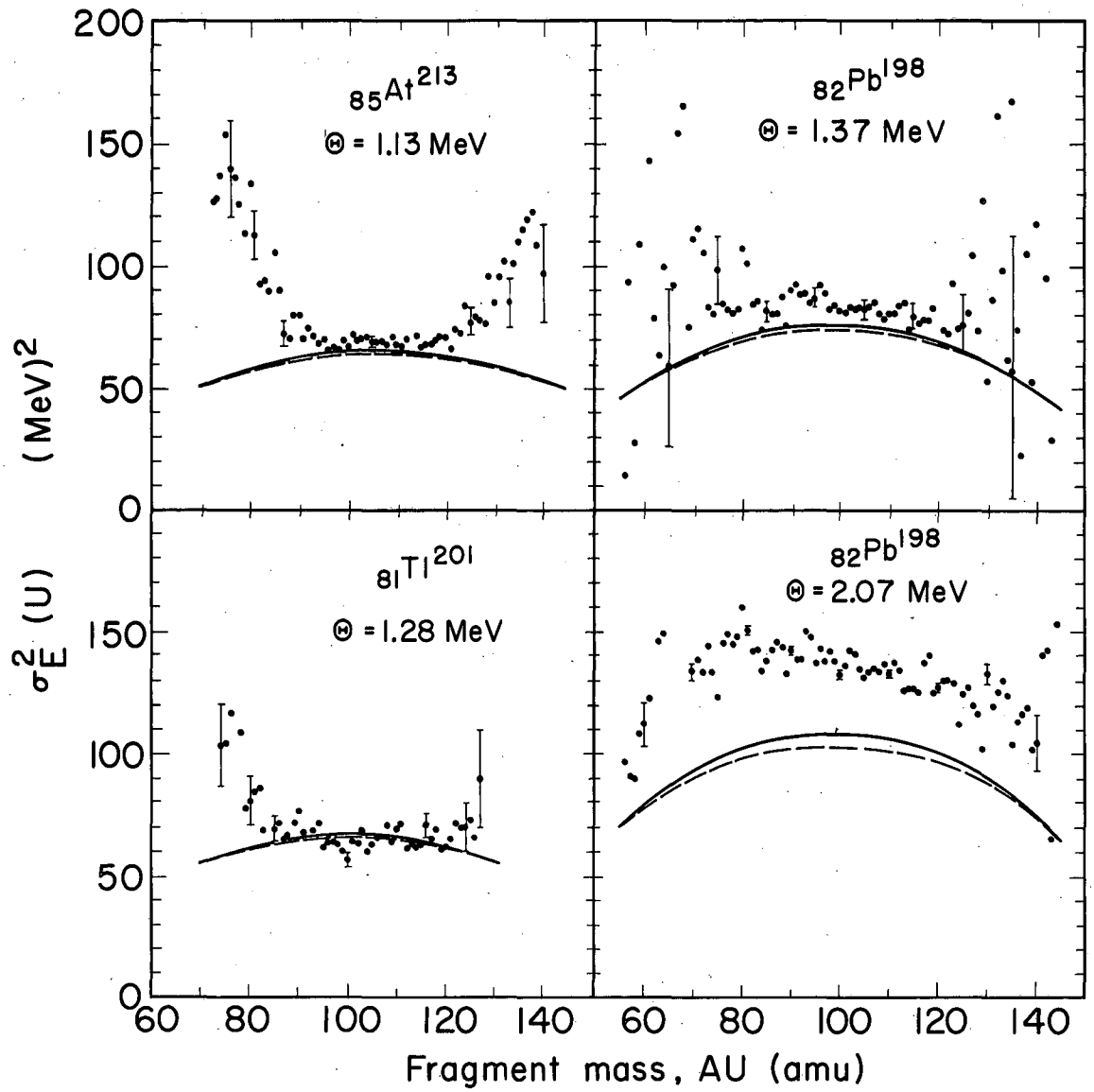

Fig. 43. Variances in the distributions of total translational kinetic energy, as functions of fragment mass AU. The theoretical results that are calculated for initial conditions determined in the harmonic approximation [from Eq. (36)] are given by the solid lines, and the results that are calculated by including effects of anharmonicity [from Eq. (48)] are given by the dashed lines. The data (solid points) are as follows: Burnett:[76] $_{83}Bi^{209}$ + 65-MeV α = $_{85}At^{213}$ (x = 0.677, Θ = 1.13 MeV), and $_{79}Au^{197}$ + 70-MeV α = $_{81}Tl^{201}$ (x = 0.651, Θ = 1.28 MeV); Plasil:[77] $_{74}W^{182}$ + 102-MeV $_{8}O^{16}$ = $_{82}Pb^{198}$ (x = 0.677, Θ = 1.37 MeV), and $_{74}W^{182}$ + 165-MeV $_{8}O^{16}$ = $_{82}Pb^{198}$ (x = 0.677, Θ = 2.07 MeV).

with increasing mass asymmetry. It thus appears that the experimental constancy or increase in $\sigma_E^2(U)$ with increasing mass asymmetry is caused by some effect other than those present in a pure liquid-drop model.

Burnett has suggested that the discrepancy between theory and experiment as regards $\sigma_E^2(U)$ may possibly arise from the effects of single-particle shell structure.[76] If the doubly-closed shell at fragment mass 132 tended to make the heavier fragment nearly spherical a portion of the time, then the separation of the fragment centers would decrease a portion of the time, resulting in an increase in translational kinetic energy. Since only some fraction of the total fission events would be affected by the doubly-closed shell, there would also still be normal fissions with lower translational kinetic energy. This mixture of fission events—some predominantly low in kinetic energy and some predominantly high—would cause the variance to be greater than it otherwise would be. The suggestion that shell effects are responsible is consistent with two pieces of experimental evidence: First of all, as was pointed out by Burnett, the increase in $\sigma_E^2(U)$ begins for both ${}_{85}At^{213}$ and ${}_{81}Tl^{201}$ at fragment masses 120-125 rather than at a constant mass ratio. Secondly, the increase in $\sigma_E^2(U)$ with mass asymmetry becomes less pronounced as the nuclear temperature increases; indeed, for ${}_{82}Pb^{198}$ at $\Theta = 2.07$ MeV, $\sigma_E^2(U)$ is essentially independent of mass asymmetry. This disappearance of the marked disagreement between theory and experiment as the temperature increases would correlate with the disappearance of single-particle shell structure at high excitations.* Work on the cause of the discrepancy represents another interesting problem.

Variances of the mass distributions as functions of total translational kinetic energy [$\sigma_U^2(E)$ vs E] are shown in Fig. 44. The magnitudes of the theoretical and experimental variances of the mass

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*The large amount of angular momentum present in the two ${}_{82}Pb^{198}$ cases could, of course, be responsible for the near-constancy of $\sigma_E^2(U)$ as a function of U (rather than a disappearance of shell structure).

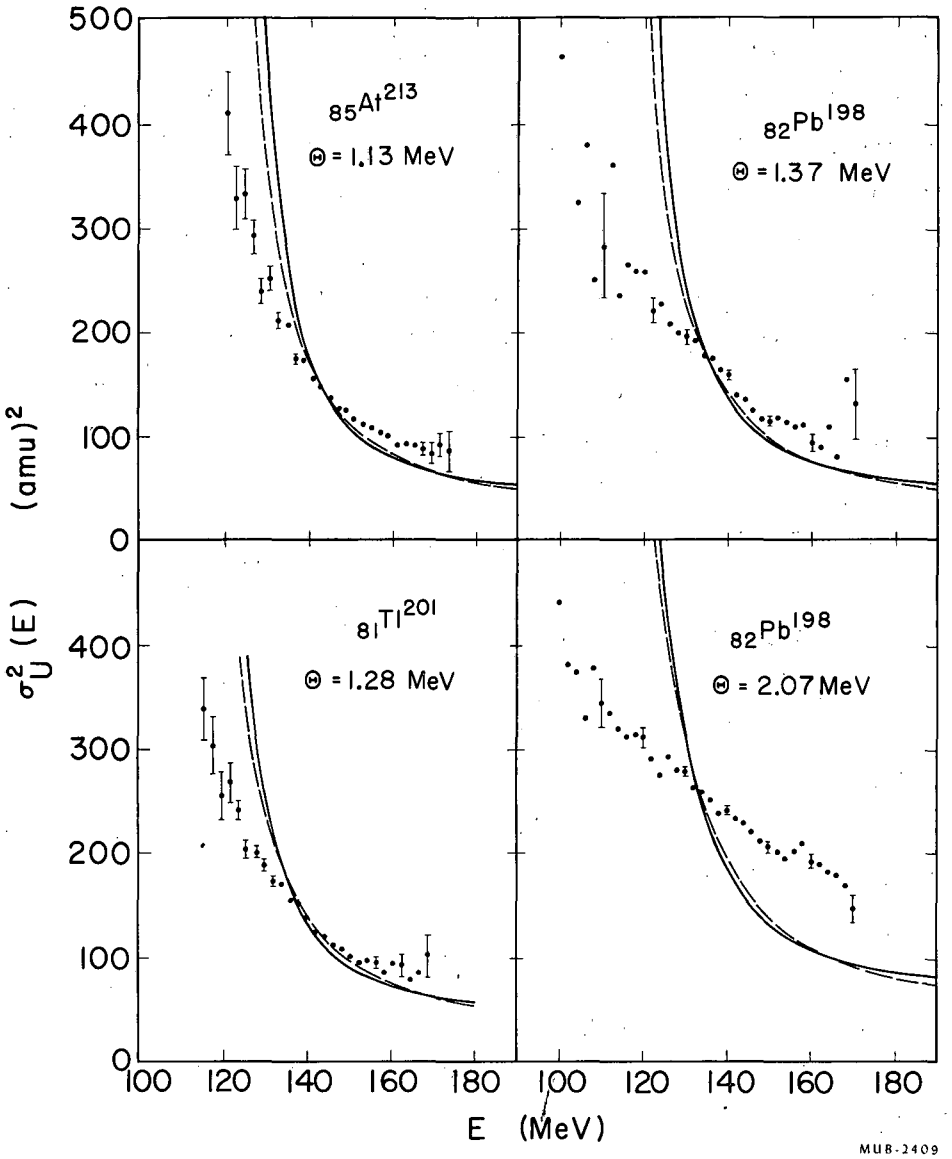

Fig. 44. Variances in the distributions of fragment mass, as functions of total translational kinetic energy E. The theoretical results that are calculated for initial conditions determined in the harmonic approximation [from Eq. (36)] are given by the solid lines, and the results that are calculated by including effects of anharmonicity [from Eq. (48)] are given by the dashed lines. The data (solid points) are as follows: Burnett:[76] $_{83}Bi^{209}$ + 65-MeV α = $_{85}At^{213}$ (x = 0.677, Θ = 1.13 MeV), and $_{79}Au^{197}$ + 70-MeV α = $_{81}Tl^{201}$ (x = 0.651, Θ = 1.28 MeV); Plasil:[77] $_{74}W^{182}$ + 102-MeV $_{8}O^{16}$ = $_{82}Pb^{198}$ (x = 0.677, Θ = 1.37 MeV), and $_{74}W^{182}$ + 165-MeV $_{8}O^{16}$ = $_{82}Pb^{198}$ (x = 0.677, Θ = 2.07 MeV).

distributions at the most probable total translational kinetic energy are in excellent agreement for the $_{85}At^{213}$ and $_{81}Tl^{201}$ cases, and in good agreement for the two $_{82}Pb^{198}$ cases. Furthermore, for the two former cases the theoretical curves correctly predict the over-all shape of the curves, including a flattening at high E and a very rapid rise at low E. At both high and low E, however, the experimental points deviate somewhat from the theoretical curves. The experimental curves for the two $_{82}Pb^{198}$ cases do not possess the characteristic "hyperbolic" shape predicted by the theory, and observed in the former cases. Indeed, for the $\Theta = 2.07$-MeV case the experimental curve is essentially linear. It is again possible that the discrepancies between theory and experiment for the two $_{82}Pb^{198}$ cases arise from the effects of angular momentum; this possibility should be investigated.

We have thus far been mainly concerned with comparing details of the distributions in mass and total translational kinetic energy for a given nucleus and excitation energy (or rather four such combinations). The theory we have developed also predicts the dependence of the distributions on nuclear temperature, and their dependence on fissionability parameter.

We turn now to a comparison of theory and experiment as regards the temperature dependence of the distributions. The mean total translational kinetic energy is prediced by the theory to be (for all practical purposes) independent of nuclear temperature. Plasil finds that for five different nuclear temperatures, ranging from $\Theta = 1.37$ MeV to $\Theta = 2.07$ MeV, the mean total translational kinetic energy $\bar{E}$ for the $_{82}Pb^{198}$ case varied by 3 MeV (with experimental errors on the values of $\bar{E}$ set at ± 5 MeV).[77] For $_{76}Os^{186}$ (formed in two ways) he found that for six different nuclear temperatures covering the range $1.49\ \text{MeV} \leq \Theta \leq 2.06$ MeV the values of $\bar{E}$ varied by 5 MeV (errors on $\bar{E}$ of ± 6 MeV). These data are thus in substantial agreement with the theory.

The variances of the distributions in E and in U are predicted to increase with increasing nuclear temperature. The comparison of theory and experiment as regards this point is made for the

$_{82}Pb^{198}$ and $_{76}Os^{186}$ cases in Fig. 45.* We again note the significant agreement between the magnitudes of the theoretical and experimental variances in both mass and total translational kinetic energy. Furthermore, theory and experiment are in substanital agreement as regards the dependence of the variances upon nuclear temperature.† This is especially true for the $_{82}Pb^{198}$ case; for $_{76}Os^{186}$ the experimental slopes for both σ_U^2 and σ_E^2 are somewhat smaller than the theoretical slopes.

The theoretical dependence of the most probable total translational kinetic energy on fissionability parameter is compared in Fig. 46 with the data of Viola and Sikkeland.[72] (This figure is reproduced from reference 3.) As in Fig. 18 the solid curve represents the result for nonviscous fragments with hydrodynamic flow of the type we have been considering (a superposition of an irrotational flow and a flow corresponding to a uniform rotation). The dot-dashed curve represents the result for infinitely viscous fragments (which would separate to infinity without oscillating), and the short-dashed line represents a simple approximation (see Section III. C) to the former curve. In addition to the data for nuclei with fissionability parameter $x \lesssim 0.67$, we have also included the data for heavier elements, with fissionability parameters up to $x = 0.807$. The experimental most probable kinetic energies for nuclei with $x \lesssim 0.67$ are seen to be reproduced well both

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*For the benefit of those making a comparison between this figure and Fig. 6 of reference 3, the preliminary data presented there were overcorrected for the effects of neutron emission from the fragments; the agreement is better than indicated there.

†It should be recalled that the variance σ_U^2 retains its linear dependence upon Θ at low temperatures in the two-spheroid model because the mass-asymmetry frequency is zero (as a consequence of the zero neck radius of the saddle-point shape). In the actual situation, of course, the neck radius of the saddle-point shape and the mass-asymmetry frequency are not zero, but are small. The variance σ_U^2 of the mass distribution should therefore approach in the real case a small finite value, rather than zero, as the temperature goes to zero.

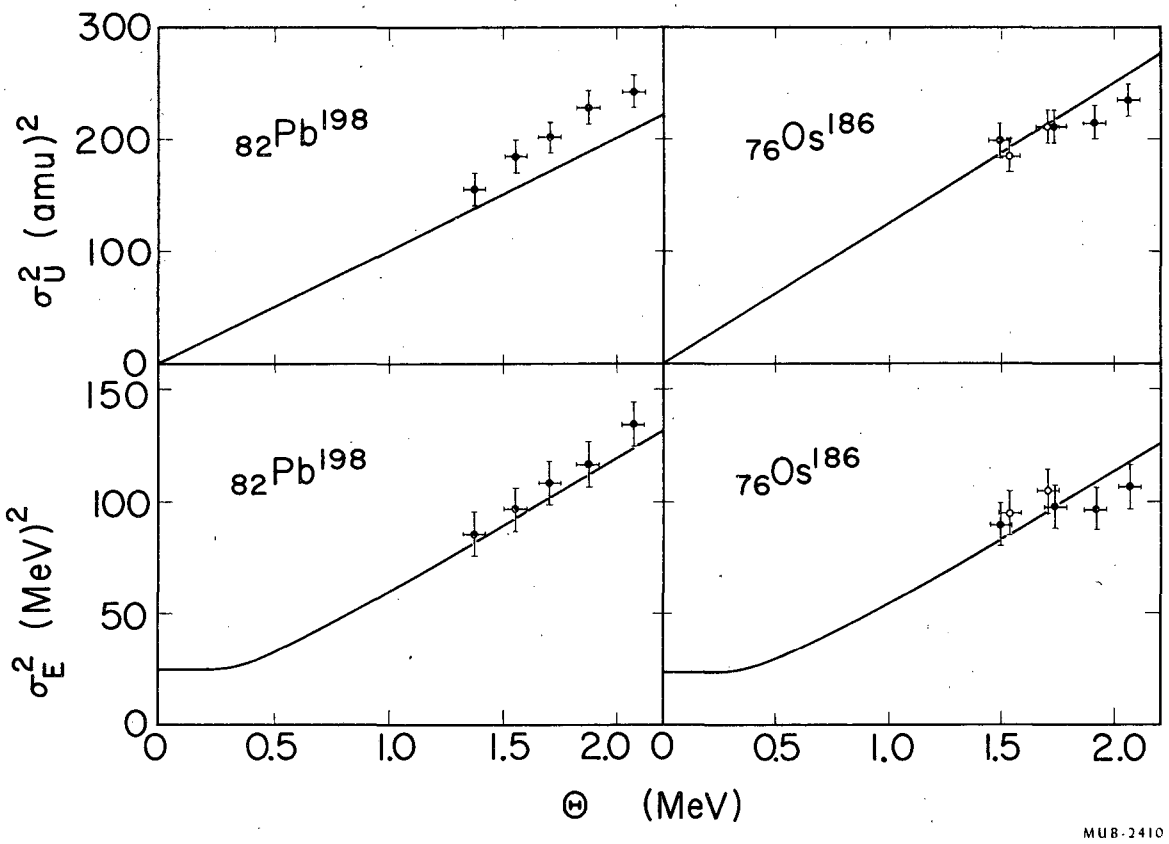

Fig. 45. Variances in the distributions of fragment mass (top), and variances in the distributions of total translational kinetic energy (bottom), as functions of nuclear temperature Θ. The theoretical curves (solid lines) are calculated for initial conditions determined in the harmonic approximation [from Eq. (36)]. The data are those of Plasil.[77] The compound nucleus $_{82}Pb^{198}$ (x = 0.677) was formed from the reaction $_{74}W^{182}$ + $_{8}O^{16}$, and the compound nucleus $_{76}Os^{186}$ (x = 0.619) from each of two reactions: $_{68}Er^{170}$ + $_{8}O^{16}$ (solid points) and $_{70}Yb^{174}$ + $_{6}C^{12}$ (open circles).

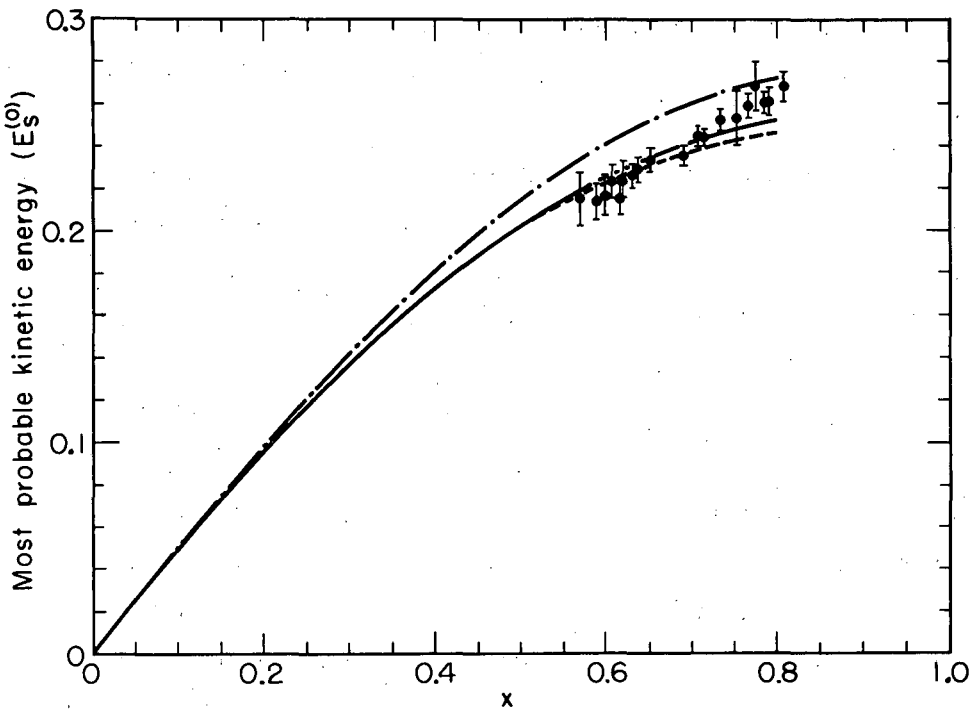

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Fig. 46. Most probable total translational kinetic energy, as a function of fissionability parameter x. The result calculated for nonviscous fragments is given by the solid line, the result for infinitely viscous fragments by the dot-dashed line, and a simple approximation to the former by the short-dashed line. The data are those of Viola and Sikkeland.[72]

in magnitude and in their dependence on x by the solid line (nonviscous fragments). For $x \gtrsim 0.67$ the experimental points are higher than the theoretical estimate represented by the solid line. Recall that for $x \gtrsim 0.67$ the two-spheroid model is inadequate, among other respects, in that its saddle-point configuration is more elongated than the true liquid-drop saddle-point shape. Thus, the translational kinetic energies predicted by the model for $x \gtrsim 0.67$ are expected to be too low.

The comparison between theory and experiment as regards the variation of widths with fissionability parameter has not yet been made. There are insufficient data on either the widths of the mass distributions or the widths of the total translational kinetic-energy distributions to establish their dependence on x. However, the single-fragment translational kinetic-energy distributions of Viola and Sikkeland[72] cover a wide range of x. In the region $0.569 \leq x \leq 0.650$ they have reported the full widths at half maximum of single-fragment kinetic-energy distributions for nine nuclei. Since the reactions studied were formed by heavy-ion bombardments, considerable analysis (similar to that performed by Plasil[77]) is required to determine the average nuclear temperature at the saddle point. If the nuclear temperature for each of these reactions were determined, the experimental widths could then be compared directly with those predicted by the theory.

The remaining comparisons between theory and experiment will be made in a series of three tables. We compare in Table I for several compound nuclei and nuclear temperatures the over-all mean total translational kinetic energy $\overline{E}$, the over-all variance σ_E^2 of the total translational kinetic-energy distribution, and the over-all variance σ_U^2 of the mass distribution. Table II is a similar comparison, but in terms of full widths at half maximum of the distributions rather than variances. (For a Gaussian distribution, the full width at half maximum is equal to the square root of the variance multiplied by 2.3548.) In Table III we compare with theory the full widths at half maximum of mass distributions obtained by use of radiochemical techniques. From the comparisons made in these three tables there is no indication of any significant disagreement between theory and experiment.

Table I. Moments of distributions in total translational kinetic energy and mass. The calculated mean total translational kinetic energy $\overline{E}$, the variance σ_E^2 of the distribution in total translational kinetic energy, and the variance σ_U^2 of the distribution in fragment mass are compared with the data of Britt, Wegner, and Gursky[75] for $_2He^3$ - induced fission reactions. The experimental values of σ_E^2 and σ_U^2 are not corrected for the effects of neutron emission from the fragments.

Target	Compound nucleus	x	Bombarding energy (MeV)	Θ (MeV)	$\overline{E}$ (MeV) Theory	$\overline{E}$ (MeV) Exp.	σ_E^2 $(MeV)^2$ Theory	σ_E^2 $(MeV)^2$ Exp.	σ_U^2 $(amu)^2$ Theory	σ_U^2 $(amu)^2$ Exp.
$_{79}Au^{197}$	$_{81}Tl^{200}$	0.654	25.5	0.65	143	140.3	39	55	73	96.7
$_{81}Tl^{nat}$	$_{83}Bi^{(208)}$	0.661	25.5	0.53	148	141.7	35	50	61	89.0
$_{82}Pb^{206}$	$_{84}Po^{209}$	0.673	25.5	0.57	149	145.4	38	50	62	79.1
$_{83}Bi^{209}$	$_{85}At^{212}$	0.680	25.5	0.54	151	147.3	37	57	58	85.0
$_{83}Bi^{209}$	$_{85}At^{212}$	0.680	22.1	0.40	151	146.5	30	48	43	72.7

Table II. Properties of distributions in total translational kinetic energy and mass. The calculated mean total translational kinetic energy $\overline{E}$, the full width at half maximum $(FWHM)_E$ of the distribution in total translational kinetic energy, and the full width at half maximum $(FWHM)_U$ of the distribution in fragment mass are compared with experimental data.

Target	Projectile	Compound nucleus	x	Bombarding energy (MeV)	Θ (MeV)	$\overline{E}$ Theory (MeV)	$\overline{E}$ Exp. (MeV)	$(FWHM)_E$ Theory (MeV)	$(FWHM)_E$ Exp. (MeV)	$(FWHM)_U$ Theory (amu)	$(FWHM)_U$ Exp. (amu)	Ref.
$_{83}Bi^{209}$	α	$_{85}At^{213}$	0.677	42	0.65[a]	151	150	15[a]	16[b]	20[a]	24[c,d]	78
$_{83}Bi^{209}$	α	$_{85}At^{213}$	0.677	43	0.68	151	148	15	20[d]	20	26[c,d]	71
$_{82}Pb^{nat}$	α	$_{84}Po^{(211)}$	0.667	43	—	149	146	—	—	—	—	71
$_{81}Tl^{nat}$	α	$_{83}Bi^{(209)}$	0.658	43	—	147	143	—	—	—	—	71
$_{79}Au^{197}$	α	$_{81}Tl^{201}$	0.651	43	—	143	138	—	—	—	—	71
$_{83}Bi^{209}$	d	$_{84}Po^{211}$	0.667	21.5	0.35	149	143	12	19[d]	—	—	71

[a] In reference 78 the experimental (rather than calculated) fission barrier is used to obtain Θ = 0.8 MeV; using this value, $(FWHM)_E$ = 16 MeV, and $(FWHM)_U$ = 22 amu.

[b] Value is corrected (in reference 78) for fragment neutron emission and experimental dispersion.

[c] Value is read off experimental curve.

[d] Value is not corrected for fragment neutron emission and experimental dispersion.

Table III. Widths of fragment-mass distributions. The calculated full width at half maximum $(FWHM)_U$ of the distribution in fragment mass is compared with data obtained by radiochemical techniques.

Target	Projectile	Compound nucleus	x	Bombarding energy (MeV)	Θ (MeV)	$(FWHM)_U$ Theory (amu)	$(FWHM)_U$ Exp. (amu)	Ref.
$_{82}Pb^{206}$	α	$_{84}Po^{210}$	0.670	42	0.62	20	21[a]	70
$_{83}Bi^{209}$	p	$_{84}Po^{210}$	0.670	36	0.76	22	18[a]	68
$_{83}Bi^{209}$	p	$_{84}Po^{210}$	0.670	58	1.20	27	24[a]	68
$_{79}Au^{197}$	α	$_{81}Tl^{201}$	0.651	42	0.72	21	34	67
$_{82}Pb^{204}$	α	$_{84}Po^{208}$	0.677	42	0.71	20	27[a]	67
$_{82}Pb^{206}$	α	$_{84}Po^{210}$	0.670	42	0.62	20	22	67
$_{83}Bi^{209}$	d	$_{84}Po^{211}$	0.667	22	0.38	15	17	65, 66

[a]Value is read off experimental curve.

To summarize this section, we have compared the predictions of our model with existing experimental data for the fission of nuclei lighter than radium—distributions in mass and total translational kinetic energy. From these comparisons we have learned that the two-spheroid model is capable of accounting for a large number of the observed properties of the distributions, but that some discrepancies remain. The significance of the comparisons will be discussed in Section VII.

VII. SUMMARY AND CONCLUSION

We have studied in connection with nuclear fission the division of an idealized charged drop, using a simplified version of the liquid-drop model. The coordinates for our model were selected so as to take into account the appropriate degrees of freedom essential to a discussion of the division of a charged drop and the separation of the fragments to infinity. To the Hamiltonian of our idealized system we have applied standard static, dynamical, and statistical methods in order to trace out the essential features of the process.

This has included, first of all, the calculation of the potential energy of the system (a sum of surface and Coulomb energies), and the location and study of the properties of the saddle point. From such a study we learned that our model is expected to be useful for discussing certain aspects of the fission of nuclei with fissionability parameter $x \lesssim 0.67$ (nuclei lighter than about radium), but not, in general, for discussing the fission of heavier elements.

The dynamical study was concerned with calculating the kinetic energy of the system, with setting up the equations of motion, and with solving them in terms of given initial conditions. This made it possible to trace out the division of the nucleus and the separation of the fragments from some given initial configuration to infinity. For the major portion of the study we worked out the theory for completely nonviscous fragments with hydrodynamic flow consisting of a superposition of an irrotational flow and a flow corresponding to a uniform rotation. For certain aspects of the theory we also considered the case of infinitely viscous fragments.

In the application of statistical mechanics we focused attention on the system at the saddle point, making the standard transition-state-method assumption of statistical equilibrium at the saddle point. This made it possible to calculate the probability of observing the system in a given state of motion close to the saddle point.

The results of the dynamical and statistical studies were then appropriately combined to give the probability of observing the two fragments in a given state of motion at infinity. This probability corresponds directly to the probability of certain observable characteristics of fission fragments: their total translational kinetic energy, fractional mass, individual excitation energies, and individual angular momenta.

As stated in the introduction, our purpose has been to study in detail the properties of the division of an idealized liquid drop whose size, surface tension, and charge are those of a nucleus; and to compare the results with what is observed experimentally in the fission of real nuclei. From this point of view there are no adjustable parameters in the problem. The comparison between theory and experiment is thus expected to tell us unambiguously to what extent an idealized liquid-drop model is capable of accounting for the properties of fission.

We have been able to make comparisons of calculations with experiment for several nuclei lighter than radium as regards distributions in mass and total translational kinetic energy. These preliminary comparisons suggest the following conclusions. First and most important, the magnitudes of the experimental most probable kinetic energies and masses are reproduced by the calculations—the experimental and theoretical most probable (or mean) kinetic energies agree to within a few percent, and predominantly symmetric rather than asymmetric mass divisions are observed, as predicted. Secondly, the magnitudes of the experimental widths of the distributions in both kinetic energy and mass are essentially as calculated, usually to within several percent.

As far as the finer details of the distributions are concerned, the calculations are capable of reproducing the correct trend in two out of the three details that we have compared. There is essential agreement as regards the decrease in average total translational kinetic energy with increasing mass asymmetry, and as regards the

rapid broadening of the mass distribution at low values of the total translational kinetic energy. The experimental constancy or increase of the widths of the kinetic-energy distributions with increasing mass asymmetry is not reproduced by the theory, which predicts a slight decrease.

The dependence of the experimental distributions on the nuclear temperature of the system is in basic agreement with the theory. The theory predicts, and experiment seems to confirm, that the most probable kinetic energies should be essentially independent of temperature. The variances (squares of widths) of the distributions in both mass and total translational kinetic energy are predicted to increase in a characteristic way with increasing nuclear temperature. Experimentally, the variances of both the kinetic-energy distributions and the mass distributions were observed to increase with temperature, some with slopes in approximate agreement with theory. There were no experimental points in the interesting region of very low temperature, where the variances of the kinetic-energy distributions are predicted to become independent of temperature.

The experimental trend of the most probable kinetic energies with fissionability parameter x was approximately reproduced by the theory for those nuclei with $x \lesssim 0.67$. No comparisons of theory and experiment have been made as regards the variation of the widths of the distributions with fissionability parameter.

On the whole, the preliminary comparison of theory with experiment suggests that the limitations of the liquid-drop model—in its simplified two-spheroid approximation—are not yet in evidence to a serious degree for the fission of the lighter nuclei. The model has stood the test of comparison with an impressive number of properties of the distributions in mass and total translational kinetic energy, without the introduction of adjustable parameters. The model seems capable of accounting not only for the over-all orders of magnitudes of the most probable values and the widths of the distributions in kinetic energy and mass, but also more detailed properties of the distributions. It appears from preliminary comparisons that for the fission of elements lighter than about radium, single-particle effects are of little

importance in influencing the distributions in kinetic energy and mass.

The conclusions drawn must not be regarded as final, however, since only a fraction of the full predictive power of the two-spheroid model has been subjected to experimental verification. Even as regards distributions in kinetic energy and mass, there are several remaining areas of exploration. The experiments that have been performed have provided us with information regarding the mean values and variances of the distributions, including the dependence of the mean value and variance of one distribution on the other variable, and vice versa. Future experiments of this type should aim at determining not only these quantities but, in addition, the further deviations of the distributions from normal distributions—the skewness of the kinetic-energy distribution, and the kurtosis of the mass distribution, for example. In addition, experimental points at higher and at lower nuclear temperatures are necessary to establish definitely the dependence of the variances of the distributions on nuclear temperature, in particular, whether or not the variance of the kinetic-energy distribution "flattens" to a constant value at low temperatures.

The extension of the measurements of mass and kinetic-energy distributions over a range of fissionability parameter is necessary to establish the trend of the widths with x, and to better confirm the trend of the most probable kinetic energies with x. It would be particularly desirable to perform experiments in the neighborhood of the Businaro-Gallone bifurcation point at $x = x_{BG} \approx 0.39_4$, which would include elements in the neighborhood of silver.[15,43] At $x = x_{BG}$ there should be a transition in the qualitative features of the two-dimensional distributions in total translational kinetic energy and mass. For $x > x_{BG}$ the lines of constant probability should be ellipses (to lowest order), whereas for $x < x_{BG}$ they should consist of two families of hyperbolas. The distribution in mass should become extremely broad for $x \approx x_{BG}$, and for $x < x_{BG}$ the division process should become one of fragmentation, as distinguished from fission,[43] with the probability for obtaining a given mass increasing with increasing mass asymmetry.

A major area of experimental exploration that is untouched for nuclei lighter than radium is the study of the distributions in individual fragment excitation energies. The most direct way of experimentally determining these distributions is to measure the distributions of the number of neutrons emitted from each fragment.[87-89]* A less direct method of obtaining such information is the measurement of the distributions of kinetic energies of the neutrons emitted from each fragment;[88-90] this method would involve the use of the relationship between fragment excitation energy and the resulting kinetic-energy distribution of the evaporated neutrons.[79,91-94]

Experimental information on distributions of individual fragment excitation energies would serve a twofold purpose. First of all, the information is needed for comparison with the theoretical predictions of the model. A particularly important experimental determination is the correlation coefficient of fragment excitation energies, which could be compared directly with the prediction that fragment excitation energies should be rather strongly anticorrelated. Secondly, such information could be used to accurately correct experimental distributions in mass and total translational kinetic energy for fragment neutron-emission effects. This would make the conclusions drawn from comparisons of these experimental distributions with theory more reliable.

The determination of the distributions of individual fragment angular momenta for the lighter elements represents another new area of experimental exploration. Experimental information regarding these distributions is potentially obtainable from at least three different types of experiments. One is a measurement of the distributions

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*Note that a measurement of the distribution of the total number of neutrons emitted (from both fragments) would determine only the distribution in total excitation energy, which would be equivalent (to lowest order) to the distribution in total translational kinetic energy.

of the total prompt gamma-ray energy emitted by each fragment.* The distribution of individual fragment angular momenta could then be estimated from a knowledge of the effect of angular momentum on the competition between neutron emission and gamma-ray emission in the fragment de-excitation process.[100-102]† The second method involves the measurement of the angular distributions of prompt gamma rays emitted from the fragments.[47]‡ The third method, which would yield information only for specific fragment masses, is the study of shielded isomer ratios in the fission products.[103, 104]§

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*For the heavier elements the average total gamma-ray energy per fission (for both fragments) has been found experimentally to be about 8 MeV.[95-98] Fragment de-excitation calculations made for nonrotating fragments indicate that roughly one-half this amount of gamma-ray energy is expected.[88-90,99] It has been suggested that this discrepancy is due to the presence of a total fragment rotational energy of several MeV.

†Recall (Section V.A.2) that for the compound nucleus $_{85}At^{213}$ and a nuclear temperature at the saddle point of 1.13 MeV, the most probable value of the angular momentum of each fragment at infinity is estimated as about $10\hbar$ if the fragments are nonviscous and about $15\hbar$ if the fragments are infinitely viscous. If one uses for the moment of inertia of the fragments at infinity the rigid-body moment of interia of a sphere, for example, this corresponds to total rotational energies for both fragments of about 3 MeV for the nonviscous case and about 7 MeV for the viscous case.

‡For the thermal-neutron-induced fission of heavier elements, this method indicates that the average angular momentum per fragment is about $7\hbar$.[47]

§The angular momentum per fragment in the low- and medium-energy fission of heavy elements deduced by this method is about 6 to $10\hbar$.[104]

The experimental determination of the distributions of individual fragment angular momenta could possibly be of value in helping decide the important question of the degree of nuclear viscosity.

The large number of predicted quantities for which there is little or no experimental information available perhaps calls for a re-examination of the directions in which basic experimental fission research should proceed. For the past quarter of a century experimentalists have concentrated on the more easily accessible region of the heavy elements, and have accumulated a prodigious amount of data. Until a more adequate theory of the fission of the heavy elements is worked out that is capable of explaining more of these data, it appears that a larger fraction of the future experiments might profitably be diverted to the region of the lighter elements.

Further progress in our understanding of fission involves, of course, not only further work on the experimental side but also on the theoretical. Ultimately, any theory of fission will have to take into account single-particle effects, but, even apart from that, there are several important refinements that should be studied within the liquid-drop model.

The present work has been concerned with tracing out the implications of the two-spheroid model on an essentially classical basis. The entire treatment of that stage of the fission process from the saddle point to infinity has involved the solution of classical equations of motion. Only in the neighborhood of the saddle point have we attempted to discuss the effects that quantum mechanics would be expected to have on the process, and it is not clear that quantum mechanics has been introduced in a consistent way. Our quantum-mechanical discussion involved the determination of the probability for initially finding the system in a given state of motion near the saddle point. These quantum-mechanical probability distributions were then combined in the sense of initial conditions with solutions to classical equations of motion.

The classical solution of the equations of motion corresponding to the two-spheroid Hamiltonian represents an essential step in our

understanding. An important next step would involve the completely consistent quantum-mechanical solution of Schrödinger's equation corresponding to the Hamiltonian—both in the vicinity of the saddle point and in the separated region. This would involve, among other things, a study of the restrictions imposed by symmetry upon the wave functions for the system, for which there are no classical analogues. From such a complete quantum-mechanical solution, the classical solutions discussed here could be obtained as a limiting case. (For most of the data compared here, however, the nuclear temperature is sufficiently high that classical statistical mechanics is valid for determining the initial conditions, and the ambiguities associated with this mixture of classical and quantum mechanics are not present. For these cases the entire treatment given here may be regarded as completely classical.)

There is a second important investigation that remains to be carried out for the two-spheroid model. The discussion in this paper was restricted to a system with zero total angular momentum. The theory should also be worked out in which account is taken of the three rotational degrees of freedom of the system as a whole. This would make it possible to draw stronger conclusions when comparing predictions of the theory with data from heavy-ion-induced reactions, in which there is considerable angular momentum present. Explicit consideration of the system's three rotational degrees of freedom would also make it possible to incorporate the existing discussions of angular distributions of fission fragments.[51,105-108]

A third extension of the work of this paper is possible. By relaxing the restriction that the charge density be constant throughout the nucleus, and by taking into account a charge-fluctuation degree of freedom, the discussion of fission-fragment charge distributions could be included.*

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*Historically, the (unpublished) work of Marshall Blann and Wladyslaw J. Swiatecki on fission-fragment charge distributions led to some of the ideas presented in this paper.

To us the most exciting prospect for the future is the extension of the two-spheroid model by the introduction of a conicoid of revolution between the two spheroids (see Section II. B and Appendix B.3). By use of this extension of the model, one should be able to discuss certain aspects of the fission of nuclei over the entire range of fissionability parameter from 0 to 1. Since the saddle-point energies and shapes calculated in this extended model agree so remarkably well with the true liquid-drop saddle-point energies and shapes (see Figs. 8 - 11), the model should make practicable the calculation of the predictions of the liquid-drop model not only as regards the distributions discussed in this paper, but also as regards such questions as the probability for fission-cross sections for induced fission, and half lives for spontaneous fission.

The general version of the extended model has the capability of representing the transformation of the hyperboloidal neck into a spheroidal third body between the two side fragments. This provides the possibility for the division of the nucleus into <u>three</u> fragments. Thus it might be possible to discuss with this three-spheroid approximation the interesting questions of ternary fission and long-range alpha-particle emission, in particular the angular distributions and the kinetic-energy distributions of the long-range alpha particles.

In conclusion, we would like to suggest that the procedure to be adopted in discussing any extensions of the theory should be identical as far as possible with that underlying the present work—the writing down of the Hamiltonian describing the idealized situation, followed by the systematic application of standard static, dynamical, and statistical methods. In this way a degree of unity and continuity could be achieved in the development of fission theory.

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APPENDICES

A. Comments on the Validity of the Liquid-Drop Model for Discussing Fission

A nucleus can be regarded for practical purposes as consisting of protons and neutrons held together by short-range nuclear forces. Solution of the resulting many-body problem would presumably yield an accurate description of all fission phenomena. However, this problem is at present impossible to solve both because of the mathematical difficulties associated with the existence of a large number of particles and because the potential between nucleons is not accurately known.

The short-range character of the nuclear force provides a means for approximate solution. The dimensionless ratio characterizing the nuclear problem–the range of the nuclear force divided by the nuclear radius–is for all but the lightest nuclei a small quantity. The energy of the system (apart from the Coulomb energy) may then be calculated as an expansion in increasing powers of this dimensionless ratio. The coefficients of the various terms, which will in general depend upon nuclear composition (the difference between the number of neutrons and the number of protons), are determined by fitting the resulting expansion to experimental masses. The four leading terms in such an expansion of the energy are of order $(R_0/r_n)^3$, $(R_0/r_n)^2$, $(R_0/r_n)^1$, and $(R_0/r_n)^0$, where R_0 is the radius of the undistorted nucleus, and r_n is the range of the nuclear force. With respect to the number of nucleons A, the expansion has leading terms of order A^1, $A^{2/3}$, $A^{1/3}$, and A^0.

A physical interpretation may be attached to each term in the expansion of the energy. The term of order A^1 represents the approximation in which the size of the nucleus is infinite compared with the range of nuclear forces, i.e., the nuclear-matter approximation. The contribution to the energy associated with the A^1 term is a negative quantity proportional to the volume of the nucleus; for a heavy nucleus its magnitude is a few thousand MeV. This volume energy, which is independent of the shape of the nucleus, represents the energy decrease arising from the binding of each nucleon with its close neighbors. Since it is a constant for a particular nucleus it need not be

considered when discussing fission, where only shape-dependent energies are relevant.

The term of order $A^{2/3}$ represents the approximation in which the range of nuclear forces is no longer neglected in relation to the size of the nucleus. This term represents the loss of binding of nucleons near the surface of the nucleus, but since one is considering this effect only to lowest order, the approximation is equivalent to assuming a semi-infinite distribution of nuclear matter bounded by a plane surface. In this approximation the effect of the $A^{2/3}$ term may be represented as the loss of binding per unit area of the bounding surface, i.e., as a surface energy proportional to the surface area of the nucleus. The size of this term depends on the shape of the nucleus—a typical value for a heavy nucleus is several hundred MeV.

The actual value of the specific surface tension depends in a very complicated way on the details of nuclear forces and nuclear structure. On the other hand, the proportionality of the $A^{2/3}$ term to the nuclear surface area is a consequence only of the smallness of r_n in comparison with R_0, i.e., of the assumption that the causes for the decrease of binding at the surface can be localized to the immediate neighborhood of a given nucleon. The situation is analogous to the case of ordinary liquids: despite the immensely complicated nature of intermolecular forces the proportionality of the surface energy to the area of the drop is extremely accurate except for droplets whose radii become comparable with molecular distances.

The term of order $A^{1/3}$ represents a number of corrections to the volume and surface energies associated with the finite rather than infinite size of nuclei. In particular, the compressibility correction to the volume energy, and the curvature correction to the surface energy appear at this stage.[18] The available information regarding these terms is very inadequate; in order of magnitude, they are some tens of MeV. Since these terms are smooth functions of the neutron and proton numbers, our ignorance regarding them is compensated to a certain extent by a readjustment of the empirical coefficients of the volume-, surface-, and electrostatic-energy terms.

The correction terms of order A^0 are presumably even more subtle and less well understood. Insofar as these terms are smooth functions of the neutron and proton numbers they are also partly absorbed in the leading terms. We note, however, that single-particle effects may be formally regarded to be of order A^0–binding-energy anomalies associated with a single nucleon (a few MeV in practice) are of order A times smaller than the total binding of all nucleons. Because of the characteristic oscillating dependence of the single-particle correction on neutron and proton numbers, this correction is presumably the most important of the A^0 terms. Some information on this correction is directly available from the study of ground-state masses of nuclei in the periodic table and from theories of the observed oscillations (see, for example, references 80 and 81).

We have thus far considered only the energy associated with the nuclear forces. The electrostatic repulsion of the positively charged protons gives rise to a Coulomb energy, which is also a function of the shape of the nucleus. This energy is of order $\alpha A^{5/3}$, where $\alpha \approx 10^{-2}$ is the ratio of the electromagnetic coupling constant to the nuclear coupling constant. For a heavy nucleus the Coulomb energy is of the order of a thousand MeV.

In connection with the Coulomb energy it should be pointed out that the discussion of the Coulomb energy of a deformed drop with a thin diffuse surface is as easy as that of a drop with a sharp surface.[80] Consider the expansion of the Coulomb energy of the drop in increasing powers of the ratio of the "thickness" of the surface to the radius of the drop. The first term in such an expansion is the Coulomb energy of a deformed sharp surface and is a function of the shape of the drop. It can be shown, using Gauss' theorem in electrostatics, that the next term is exactly independent of the shape of the drop.[80] Thus the lowest-order diffuseness correction to the Coulomb energy could be included simply by adding a (negative) constant to the Coulomb energy; this would simply alter somewhat the value of the fissionability parameter x.

We see that the leading terms of interest in fission–the shape-dependent ones–are of order $\alpha A^{5/3}$, $A^{2/3}$, $A^{1/3}$, and A^0. The liquid-

drop model consists of treating only the first two, the Coulomb and surface energies. Barring accidental cancellations of the changes in Coulomb and surface energies, the remaining terms should in general be less important. In the region of the heavy elements the changes in the Coulomb and surface energies do indeed tend to cancel, and the remaining terms may then be essential for discussing certain aspects of the process. On the other hand, in the region of the lighter elements, the changes in Coulomb and surface energies do not cancel so closely, and the remaining terms should not have the same relative importance.

The point of view that we are trying to bring out is that the liquid-drop model is not to be regarded as a "right" or a "wrong" model of fission, but as a first stage in the development of an approximate theory of nuclear fission that takes into account the principal components of the energy and that may be improved in a systematic way by the inclusion of corrections of higher order.

B. Formulae for Potential Energies

We give here the formulae used for calculating the surface and Coulomb energies of a system consisting of (1) two arbitrarily oriented separated spheroids, (2) two overlapping symmetric spheroids, and (3) two symmetric spheroids connected by a conicoidal neck.

1. Separated Spheroids

The total potential energy of two arbitrarily oriented separated spheroids is written in the form of Eq. (6b): the sum of two individual surface energies, two individual Coulomb self-energies, and the Coulomb interaction energy. We will consider these terms one by one. However, let us first take care of some preliminary definitions.

Recall that c_1 denotes the semisymmetry axis of spheroid 1, and a_1 its transverse semiaxis. When spheroid 1 is prolate its eccentricity e_1 is defined by

$$e_1^2 = 1 - \frac{a_1^2}{c_1^2} = 1 - \frac{U R_0^3}{c_1^3} \quad .$$

When spheroid 1 is oblate we denote by ϵ_1 the quantity

$$\epsilon_1^2 = \frac{a_1^2}{c_1^2} - 1 = \frac{U R_0^3}{c_1^3} - 1 = -e_1^2 .$$

(Note that ϵ_1 is <u>not</u> the eccentricity of an oblate spheroid.) The quantities e_2 and ϵ_2 are defined in an analogous way for spheroid 2.

When spheroid 1 is prolate its surface energy is given by[13]

$$B_S^{(1)} = \frac{1}{2} U^{2/3} (1 - e_1^2)^{1/3} \left[1 + \frac{\sin^{-1} e_1}{e_1 (1 - e_1^2)^{1/2}} \right] .$$

By use of this formula and the relation[109]

$$\sin^{-1} ix = i \ln \left[x + (1 + x^2)^{1/2} \right], \qquad \text{(B.1)}$$

we obtain the result for the surface energy of fragment 1 when it is oblate:

$$B_S^{(1)} = \frac{1}{2} U^{2/3} (1 + \epsilon_1^2)^{1/3} \left\{ 1 + \frac{\ln\left[\epsilon_1 + (1 + \epsilon_1^2)^{1/2}\right]}{\epsilon_1 (1 + \epsilon_1^2)^{1/2}} \right\} .$$

By expanding either of these formulae we obtain a result that is useful for calculating the surface energy when spheroid 1 is nearly spherical (either prolate or oblate):

$$B_S^{(1)} = U^{2/3} \left(1 + \frac{2}{45} e_1^4 + \frac{116}{2835} e_1^6 + \frac{101}{2835} e_1^8 + \frac{1252}{40095} e_1^{10} + \cdots \right) .$$

When spheroid 1 is prolate its Coulomb energy is given by[13]

$$B_C^{(1)} = \frac{1}{2} U^{5/3} (1 - e_1^2)^{1/3} \frac{1}{e_1} \ln \left(\frac{1 + e_1}{1 - e_1} \right) .$$

By use of this formula and the relation[109]

$$\tan^{-1} ix = \frac{1}{2} i \ln \left(\frac{1 + x}{1 - x} \right) ,$$

we obtain the result for the Coulomb energy of fragment 1 when it is oblate:

$$B_C^{(1)} = U^{5/3} \, (1 + \epsilon_1^2)^{1/3} \, \frac{\tan^{-1} \epsilon_1}{\epsilon_1} \, .$$

When spheroid 1 is nearly spherical, the series expansion is useful:*

$$B_C^{(1)} = U^{5/3} \left(1 - \frac{1}{45} e_1^4 - \frac{64}{2835} e_1^6 - \frac{58}{2835} e_1^8 - \frac{1024}{56133} e_1^{10} + \cdots \right) .$$

Formulae for the surface energy and the Coulomb self-energy of spheroid 2 are completely analogous.

We consider now the derivation of the Coulomb interaction energy $E_I = B_I E_C^{(0)}$ between spheroids 1 and 2. For this energy, closed formulae are not available; the final result will be expressed as the sum of two closed formulae and a triple multipole summation. The derivation consists of reducing (for the case of two spheroids) the general formula given by Hirschfelder, Curtiss, and Bird[110] for the interaction energy of two completely separated charge distributions.

The interaction energy of two separated charge distributions 1 and 2 can be written as a fivefold multipole summation:[110]†

$$E_I = \sum_{\substack{n_1 \, n_2 \\ m_1 \, m_2 \\ m}} i^{m_1 - |m_1| - m_2 + |m_2|} (-1)^{n_2 + m} \times \frac{\left[(n_1 - |m_1|)!(n_2 - |m_2|)! \right]^{1/2} (n_1 + n_2)! \, \bar{Q}(1)_{n_1}^{m_1 *} \, \bar{Q}(2)_{n_2}^{m_2}}{\left[(n_1 + |m|)!(n_1 - |m|)! \, (n_1 + |m_1|)! \, (n_2 + |m|)!(n_2 - |m|)!(n_2 + |m_2|)! \right]^{1/2}} \times$$

---

* This expansion is given, through the e_1^8 term, in reference 39.

† In comparing with Eq. (12.1-33) of reference 110, note that a factorial sign is missing there from the last factor in the denominator. A sufficient condition for the convergence of this expression is that one of the charge distributions be completely outside an imaginary sphere in which the second charge distribution is completely enclosed.

$$\times \left[\frac{D^{(n_1)}(\phi_1,\theta_1,\psi_1)^*_{m_1 m}\, D^{(n_2)}(\phi_2,\theta_2,\psi_2,)_{m_2 m}}{\ell^{n_1+n_2+1}} \right] .$$

In this expression $\overline{Q}(1)_{n_1}^{m_1}$ is the multipole moment of charge distribution 1, calculated with respect to a body-fixed coordinate system $\overline{x}_1\ \overline{y}_1\ \overline{z}_1$:

$$\overline{Q}(1)_{n_1}^{m_1} = \int \rho_1(\overline{r}_1, \overline{\theta}_1, \overline{\phi}_1)\, \overline{r}_1^{\,n_1} P_{n_1}^{m_1}(\cos\overline{\theta}_1)\, e^{im_1\overline{\phi}_1}\, d\overline{\tau}_1 ,$$

where $\rho_1(\overline{r}_1, \overline{\theta}_1, \overline{\phi}_1)$ is the charge density of charge distribution 1, $P_{n_1}^{m_1}(\cos\overline{\theta}_1)$ is the associated Legendre polynomial, and $d\overline{\tau}_1$ denotes an element of volume. An asterisk denotes complex conjugation. The orientation of the body-fixed system $\overline{x}_1\ \overline{y}_1\ \overline{z}_1$ with respect to the space-fixed system $x_1\ y_1\ z_1$ is specified by the three Euler angles θ_1, ϕ_1, ψ_1 (see Fig. 2). The coefficients $D^{(n_1)}(\phi_1,\theta_1,\psi_1)_{m_1 m}$ are the representation coefficients of the three-dimensional rotation group.[110] (The representation coefficients being used are identical to those of Margenau and Murphy.[111]) The distance between the origins of coordinate systems $\overline{x}_1\overline{y}_1\overline{z}_1$ and $\overline{x}_2\overline{y}_2\overline{z}_2$ is ℓ.

For uniformly charged spheroidal charge distributions we choose the body-fixed $\overline{z}_1$ axis to lie along the symmetry axis of spheroid 1. Then, because of azimuthal symmetry about the $\overline{z}_1$ axis, $\overline{Q}(1)_{n_1}^{m_1} = 0$ for $m_1 \neq 0$. For $m_1 = 0$, we obtain explicitly

$$\overline{Q}(1)_{n_1}^{0} = \rho_1 \int \overline{r}_1^{\,n_1} P_{n_1}(\cos\overline{\theta}_1)\, d\overline{\tau}_1$$

$$= \frac{2\pi\rho_1}{(n_1+3)} \int_{-1}^{+1} d\mu_1\, P_{n_1}(\mu_1) \left[R_1(\mu_1) \right]^{n_1+3} ,$$

where $P_{n_1}(\mu_1)$ is the (ordinary) Legendre polynomial, $\mu_1 = \cos\overline{\theta}_1$, and

$$R_1(\mu_1) = \frac{a_1}{\left[1-\left(1-\frac{a_1^2}{c_1^2}\right)\mu_1^2\right]^{1/2}} ,$$

is the equation in polar coordinates of the radius vector of spheroid 1.

For n_1 an odd integer, the integrand is an odd function of μ_1, and $\bar{Q}(1)^0_{n_1} = 0$. By use of the result[112]

$$\int_{-1}^{+1} \frac{P_{2n}(\mu)\, d\mu}{(1+k\mu^2)^{n+(3/2)}} = \frac{2(-k)^n}{(2n+1)(1+k)^{n+(1/2)}} , \qquad (-1 < k < +1)$$

we obtain, for n_1 even,

$$\bar{Q}(1)^0_{n_1} = \frac{3\, q_1 (c_1^2 - a_1^2)^{n_1/2}}{(n_1+1)(n_1+3)} ,$$

where q_1 is the total charge of spheroid 1.

Since $\bar{Q}(1)^{m_1}_{n_1} = 0$ for $m_1 \neq 0$, and $\bar{Q}(2)^{m_2}_{n_2} = 0$ for $m_2 \neq 0$, the fivefold summation reduces to a triple summation. Furthermore, since $\bar{Q}(1)^0_{n_1} = 0$ for n_1 odd, and $\bar{Q}(2)^0_{n_2} = 0$ for n_2 odd, the n_1 and n_2 summations need be taken over <u>even</u> integers only. We thus change summation variables by defining

$$n_1 = 2j,$$
$$n_2 = 2k.$$

The formula for the interaction energy becomes, upon simplifying:

$$E_I = \sum_{j=0}^{\infty} \sum_{k=0}^{\infty} \frac{\bar{Q}(1)^0_{2j}\, \bar{Q}(2)^0_{2k}}{\ell^{2j+2k+1}}$$

$$\times \sum_{m=-m_<}^{+m_<} \frac{(-1)^m (2j+2k)!\, D^{(2j)}(\phi_1, \theta_1, \psi_1)^*_{0m}\, D^{(2k)}(\phi_2, \theta_2, \psi_2)_{0m}}{\left[(2j+|m|)!\,(2j-|m|)!\,(2k+|m|)!\,(2k-|m|)!\right]^{1/2}} ,$$

where $m_<$ is the minimum of $2j$ and $2k$.

The representation coefficients with one zero subscript can be written in terms of spherical harmonics:[110]

$$D^{(n)}(\phi, \theta, \psi)_{0m} = \left(\frac{4\pi}{2n+1}\right)^{1/2} Y_n^m(\theta, \phi) \quad ,$$

where θ, ϕ are the usual polar angles specifying the orientation of the body-fixed $\bar{z}$ axis with respect to the space-fixed z axis (see Fig. 2). The spherical harmonics are in turn defined by[110]

$$Y_n^m(\theta, \phi) = i^{|m|-m}\left[\frac{(2n+1)}{4\pi}\,\frac{(n-|m|)!}{(n+|m|)!}\right]^{1/2} P_n^m(\cos\theta)e^{im\phi}.$$

The formula for E_I then becomes

$$E_I = \sum_{j=0}^{\infty}\sum_{k=0}^{\infty}\frac{\bar{Q}(1)_{2j}^{0}\,\bar{Q}(2)_{2k}^{0}}{\ell^{2j+2k+1}}$$

$$\times\sum_{m=-m_<}^{+m_<}\frac{(-1)^m(2j+2k)!\,P_{2j}^m(\cos\theta_1)P_{2k}^m(\cos\theta_2)e^{-im(\phi_1-\phi_2)}}{(2j+|m|)!(2k+|m|)!} \quad .$$

We may eliminate the appearance of imaginary quantities and absolute-value signs to obtain

$$E_I = \sum_{j=0}^{\infty}\sum_{k=0}^{\infty}\frac{\bar{Q}(1)_{2j}^{0}\,\bar{Q}(2)_{2k}^{0}\,(2j+2k)!}{\ell^{2j+2k+1}}\left[\frac{P_{2j}(\cos\theta_1)P_{2k}(\cos\theta_2)}{(2j)!(2k)!}\right.$$

$$\left. + 2\sum_{m=1}^{m_<}\frac{(-1)^m P_{2j}^m(\cos\theta_1)P_{2k}^m(\cos\theta_2)\cos m\phi}{(2j+m)!(2k+m)!}\right] ,$$

where $\phi = \phi_1 - \phi_2$. (When either j or k is zero, the summation over m does not occur.)

We find it convenient at this point to introduce the definitions

$$\lambda_1^2 = \frac{c_1^2 - a_1^2}{\ell^2} = \frac{c_1^2 - \dfrac{UR_0^3}{c_1}}{\ell^2} \quad ,$$

$$\lambda_2^2 = \frac{c_2^2 - a_2^2}{\ell^2} = \frac{c_2^2 - \dfrac{(1-U)R_0^3}{c_2}}{\ell^2} .$$

If we use these definitions and the result previously obtained for the multipole moments, we obtain

$$E_I = \frac{q_1 q_2}{\ell} \sum_{j=0}^{\infty} \sum_{k=0}^{\infty} \frac{3}{(2j+1)(2j+3)} \frac{3}{(2k+1)(2k+3)}$$

$$\times \frac{(2j+2k)!}{(2j)!(2k)!} \lambda_1^{2j} \lambda_2^{2k} \left[P_{2j}(\cos\theta_1) P_{2k}(\cos\theta_2) + 2 \sum_{m=1}^{m<} (-1)^m \frac{(2j)! P_{2j}^m(\cos\theta_1)}{(2j+m)!} \right.$$

$$\left. \times \frac{(2k)! P_{2k}^m(\cos\theta_2)}{(2k+m)!} \cos m\phi \right] .$$

When either j=0 or k=0 in this result, the single remaining summation can be performed explicitly.[113] We thus find it convenient to write E_I in the form

$$E_I = \frac{q_1 q_2}{\ell} \left[s(\lambda_1, \theta_1) + s(\lambda_2, \theta_2) - 1 + S(\lambda_1, \lambda_2, \theta_1, \theta_2, \phi) \right] ,$$

where

$$s(\lambda, \theta) = \sum_{n=0}^{\infty} \frac{3 \lambda^{2n} P_{2n}(\cos\theta)}{(2n+1)(2n+3)} \tag{B.2a}$$

(explicit formulae are given later), and

$$S(\lambda_1, \lambda_2, \theta_1, \theta_2, \phi) = \sum_{j=1}^{\infty} \sum_{k=1}^{\infty} \frac{3}{(2j+1)(2j+3)}$$

$$\times \frac{3}{(2k+1)(2k+3)} \frac{(2j+2k)!}{(2j)!(2k)!} \lambda_1^{2j} \lambda_2^{2k}$$

$$\times \left[P_{2j}(\cos\theta_1) P_{2k}(\cos\theta_2) + 2 \sum_{m=1}^{m<} (-1)^m \frac{(2j)! P_{2j}^m(\cos\theta_1)}{(2j+m)!} \times \right.$$

$$\times \quad \frac{(2k)!\,P_{2k}^{m}(\cos\theta_2)}{(2k+m)!} \cos m\phi \Bigg] .$$

Written in this form, the interaction energy has a simple physical interpretation:[43] If spheroid 2 becomes a sphere, $\lambda_2 = 0$, and the interaction energy is given by $(q_1 q_2/\ell)\, s(\lambda_1, \theta_1)$. If spheroid 1 becomes a sphere, $\lambda_1 = 0$, and the interaction energy is $(q_1 q_2/\ell)\, s(\lambda_2, \theta_2)$. Thus, according to this equation for E_I, the interaction energy of two arbitrarily oriented separated spheroids may be regarded as the sum of what the interaction energy would be if one of the spheroids were a sphere, plus what the interaction energy would be if the other were a sphere, minus the interaction energy of two spheres, plus a correction given by $S(\lambda_1, \lambda_2, \theta_1, \theta_2, \phi)$.

The interaction energy of a sphere and a spheroid (either prolate or oblate) is discussed by Webster.[113] We have transformed the formulae given there into a form more convenient for our purposes. For a prolate spheroid we find that

$$s(\lambda, \theta) = \left[\frac{3}{2\lambda} - \left(\frac{9}{4}\cos^2\theta - \frac{3}{4}\right)\frac{1}{\lambda^3}\right] \ln\left(\frac{g+\lambda}{h}\right)$$

$$+ \frac{3\cos^2\theta}{2\lambda^2 g} - \frac{3\, g \sin^2\theta}{4\lambda^2 h^2} , \qquad \text{(B.2b)}$$

where g and h are defined by

$$g^2 = \frac{1}{2}\left\{1 + \lambda^2 + [1 - 2(2\cos^2\theta - 1)\lambda^2 + \lambda^4]^{1/2}\right\} ,$$

$$h^2 = \frac{1}{2}\left\{1 - \lambda^2 + [1 - 2(2\cos^2\theta - 1)\lambda^2 + \lambda^4]^{1/2}\right\} .$$

For an oblate spheroid,

$$s(\lambda, \theta) = \left[\frac{3}{2\omega} + \left(\frac{9}{4}\cos^2\theta - \frac{3}{4}\right)\frac{1}{\omega^3}\right] \tan^{-1}\left(\frac{\omega}{g}\right)$$

$$- \frac{3\cos^2\theta}{2\omega^2 g} + \frac{3\, g \sin^2\theta}{4\omega^2 h^2} , \qquad \text{(B.2c)}$$

where $\omega^2 = -\lambda^2$.

A graph of the function $s(\lambda, \theta)$ is given in Fig. B.1 as a function of λ^2 for fixed values of θ, and in Fig. B.2 as a function of θ for fixed values of λ^2. Note in Fig. B.2 that $s(\lambda, \theta)$ becomes unity for approximately the same value of θ, independently of λ^2. This value of θ is given by the solution of $P_2(\cos\theta) = 0$; physically this result states simply that the quadrupole interaction term is the most important of the multiple terms (apart from the monopole).

We have thus derived the general result for the Coulomb interaction energy between two arbitrarily oriented separated spheroids. For a number of special cases, the general result simplifies considerably. First of all, if the symmetry axes of the spheroids lie in the same plane, $\phi = 0$, and $\cos m\phi = 1$; this eliminates one factor in S, but the triple summation remains.

If, in addition to $\phi = 0$ the symmetry axes are parallel $(\theta_1 = \theta_2 \equiv \theta)$, then a great simplification occurs. By using properties of the representation coefficients[114] we can show that

$$P_{2j}(\cos\theta)\, P_{2k}(\cos\theta) + 2 \sum_{m=1}^{m<} (-1)^m \frac{(2j)!\, P_{2j}^m(\cos\theta)}{(2j+m)!}$$

$$\times \frac{(2k)!\, P_{2k}^m(\cos\theta)}{(2k+m)!} = P_{2j+2k}(\cos\theta).$$

Then, the formula for S reduces to[43]

$$S(\lambda_1, \lambda_2, \theta, \theta, 0) = \sum_{j=1}^{\infty} \sum_{k=1}^{\infty} \frac{3}{(2j+1)(2j+3)}$$

$$\times \frac{3}{(2k+1)(2k+3)} \frac{(2j+2k)!}{(2j)!(2k)!} \lambda_1^{2j} \lambda_2^{2k} P_{2j+2k}(\cos\theta).$$

If, further, the spheroids are collinear $(\theta = 0)$ then $P_{2j+2k}(\cos\theta) = 1$, and the result for S simplifies slightly. For this case, however, there is considerable simplification in the formulae for s, and we then obtain the formulae given by Cohen and Swiatecki:[43]

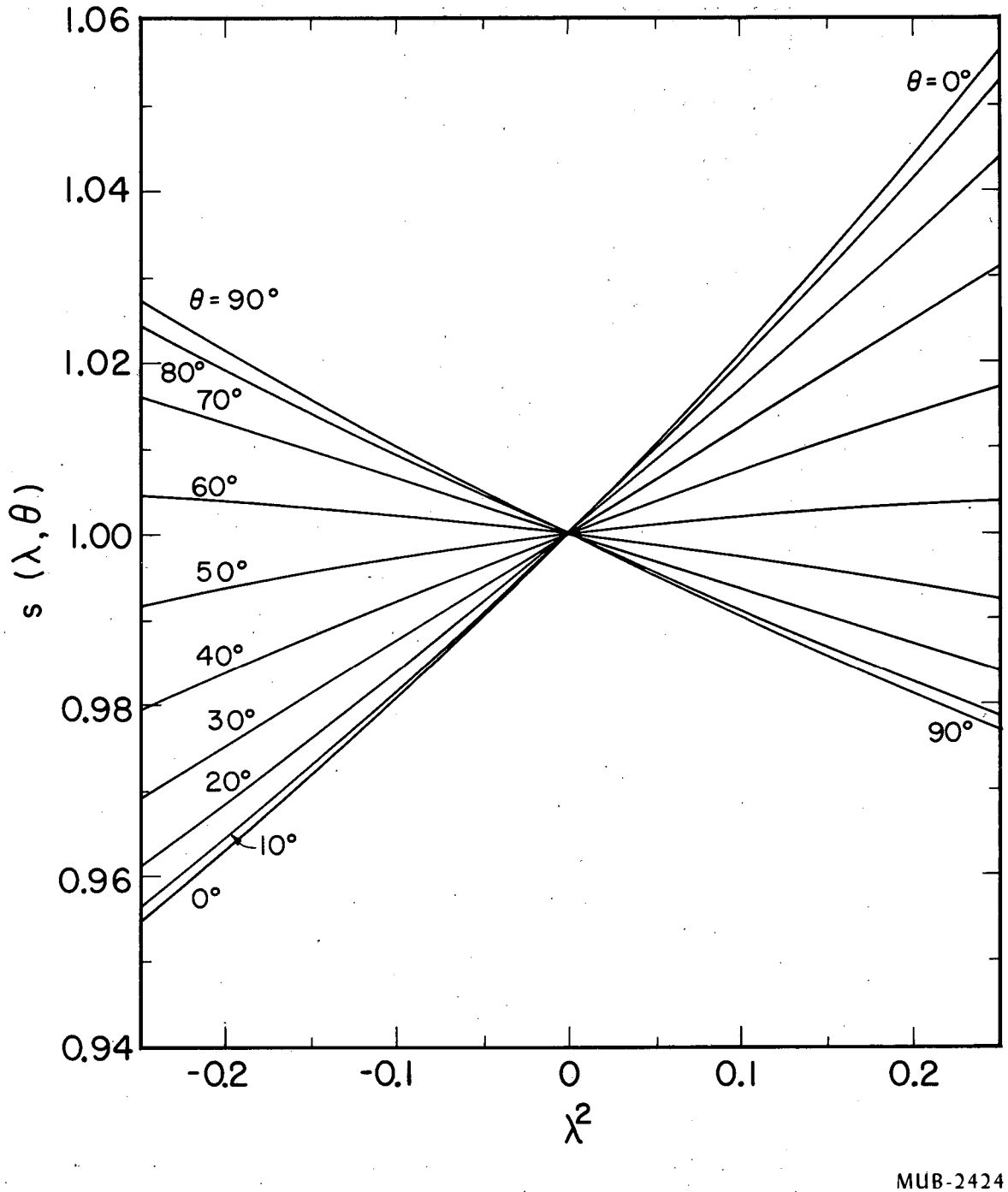

Fig. B.1. The function $s(\lambda,\theta)$ [defined by Eq. (B.2)] vs λ^2, for fixed values of θ.

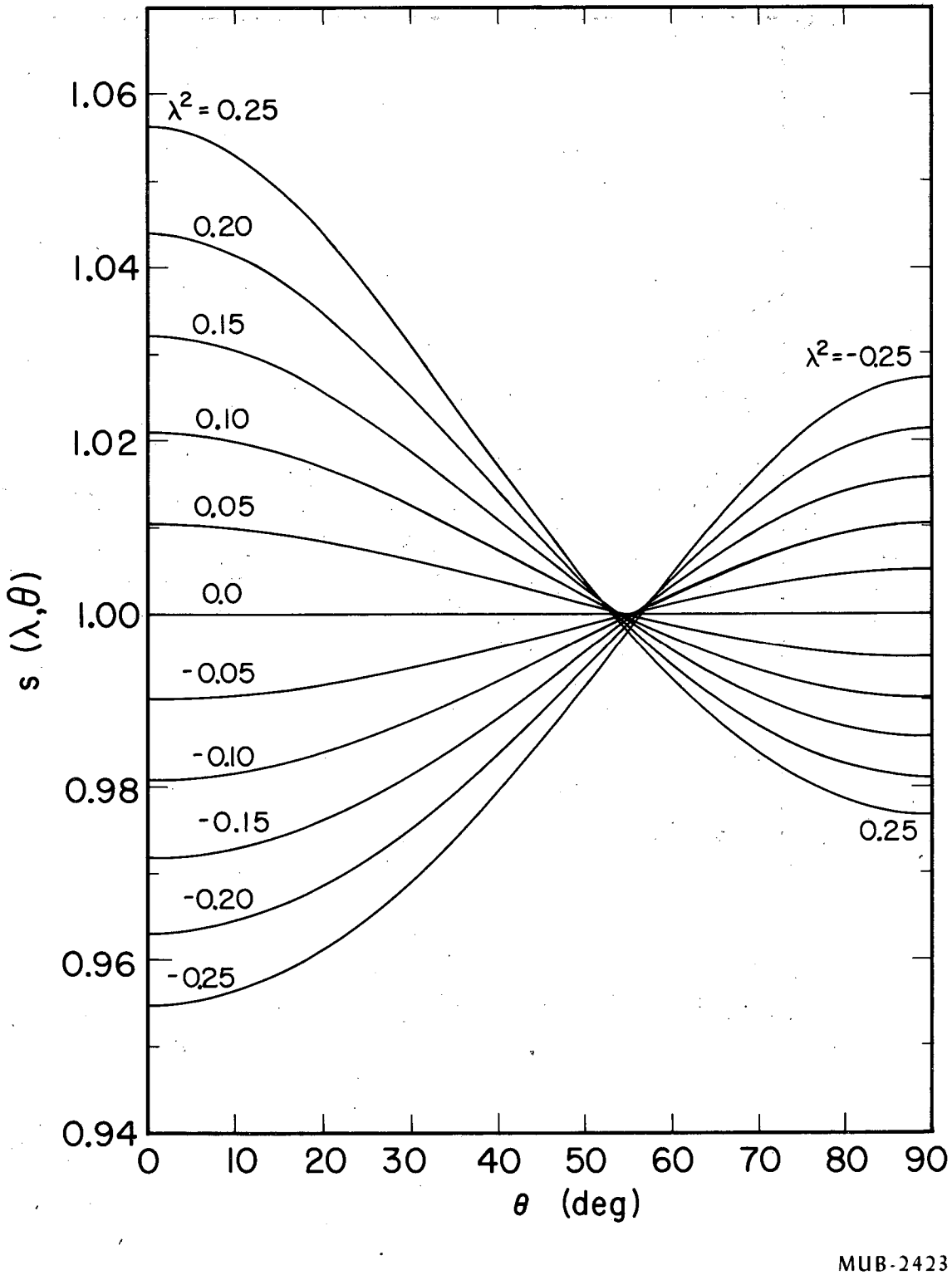

Fig. B.2. The function $s(\lambda, \theta)$ [defined by Eq. (B.2)] vs θ, for fixed values of λ^2.

$$s(\lambda, 0) = \begin{cases} \frac{3}{4}\left(\frac{1}{\lambda} - \frac{1}{\lambda^3}\right)\ln\left(\frac{1+\lambda}{1-\lambda}\right) + \frac{3}{2}\frac{1}{\lambda^2}, & \text{prolate} \\ \frac{3}{2}\left(\frac{1}{\omega} + \frac{1}{\omega^3}\right)\tan^{-1}\omega - \frac{3}{2}\frac{1}{\omega^2}, & \text{oblate.} \quad (\omega^2 = -\lambda^2) \end{cases}$$

2. Overlapping Symmetric Spheroids

The total potential energy of a system consisting of two overlapping symmetric spheroids is written in the form of Eq. (6a): the sum of a surface energy and a Coulomb energy. We will consider these energies in turn. However, let us again first take care of some preliminaries.

Recall that for the symmetric case the semisymmetry axis of either spheroid is denoted by c, and the transverse semiaxis of either spheroid by a; the distance between spheroid centers is ℓ (see the upper part of Fig. 2). It is convenient to define the dimensionless quantity β by

$$\beta = \frac{\ell}{2c} .$$

The semiaxis a is determined in terms of ℓ and c by volume conservation. If we equate the volume of the original drop to the result obtained from a straightforward volume integration of a deformed drop, we find that

$$\frac{a}{R_0} = \left[\frac{2R_0}{c(2 + 3\beta - \beta^3)}\right]^{1/2} .$$

When the spheroids are prolate, the eccentricity is defined by

$$e^2 = 1 - \frac{a^2}{c^2} .$$

For oblate spheroids we denote by ϵ the quantity

$$\epsilon^2 = \frac{a^2}{c^2} - 1 = -e^2 .$$

(Again, note that ϵ is <u>not</u> the ecentricity of an oblate spheroid.) We further define the quantities g and γ by

$$g = \beta e ,$$

$$\gamma = \beta \epsilon .$$

The surface energy of the system is obtained by performing a straightforward surface integration. When the spheroids are prolate, the total surface energy can be written as

$$B_S = \frac{ac}{2R_0^2 e} \left[\sin^{-1} e + \sin^{-1} g + e(1-e^2)^{1/2} + g(1-g^2)^{1/2} \right]. \qquad \text{(B. 3a)}$$

From this equation and the relation (B.1) it follows that when the spheroids are oblate,

$$B_S = \frac{ac}{2R_0^2 \epsilon} \left\{ \ln \left[\epsilon + (1+\epsilon^2)^{1/2} \right] + \ln \left[\gamma + (1+\gamma^2)^{1/2} \right] \right.$$

$$\left. + \epsilon (1+\epsilon^2)^{1/2} + \gamma (1+\gamma^2)^{1/2} \right\}. \qquad \text{(B.3b)}$$

The Coulomb energy of the system is calculated by performing numerically a double integration. We use a cylindrical coordinate system, with the z axis along the system's symmetry axis, and ρ the perpendicular distance from the z axis to the surface of the drop. We denote by ρ_e the (constant) charge density of the drop.

The total Coulomb energy $E_C = B_C E_C^{(0)}$ of the system can be written as[14]

$$E_C = \frac{\rho_e}{5} \int V(z) R^3(z) \, d\Omega , \qquad \text{(B.4a)}$$

where $R(z)$ is the distance from the center of the drop to a point on the surface specified by ρ and z, $V(z)$ is the electrostatic potential on the surface of the drop, and $d\Omega$ denotes an element of solid angle. Because of azimuthal symmetry, a single numerical integration is required to obtain E_C once $V(z)$ is known.

The electrostatic potential on the surface of the drop is in turn given by[19-21]

$$V(z) = 2\rho_e \int \frac{dz' \rho' \left\{ \left[\rho + \rho' + (z-z') \frac{d\rho'}{dz'} \right] K(k) - 2\rho D(k) \right\}}{\left[(\rho+\rho')^2 + (z-z')^2 \right]^{1/2}} , \qquad \text{(B.4b)}$$

where

$$k^2 = \frac{4\rho\rho'}{(\rho+\rho')^2 + (z-z')^2} ,$$

and

$$D(k) = \frac{K(k) - E(k)}{k^2} .$$

The quantities K(k) and E(k) are the complete elliptic integrals in standard notation. Since excellent approximate representations exist for the elliptic integrals,[115] the resulting expression for E_C can be evaluated by performing numerically a double integration.

We give finally the series expansions about a sphere of the surface and Coulomb energies of the overlapping system. It is convenient for this purpose to define the dimensionless quantities δ and ϵ by

$$\delta = \frac{\ell}{R_0} ,$$

$$\epsilon = \frac{c}{R_0} - 1 .$$

The series expansion for the surface energy may be obtained either by expanding the integrand of the surface integral for B_S and integrating, or by expanding the final result for B_S itself. We find, through terms of third order,

$$B_S = 1 + \frac{3}{20}\delta^2 + \frac{9}{20}\delta\epsilon + \frac{2}{5}\epsilon^2 - \frac{5071}{53760}\delta^3 - \frac{657}{1120}\delta^2\epsilon - \frac{99}{112}\delta\epsilon^2 - \frac{52}{105}\epsilon^3 + \cdots .$$

The series expansion for the Coulomb energy is not so readily obtainable because a closed formula for the Coulomb energy is not available. An expansion to second order may be carried out as follows. First, expand to first order the radius vector $R(\theta)$ of the system in spherical harmonics:

$$R(\theta) = R_0 \left[1 + \left(\frac{9}{16}\delta + \epsilon \right) P_2(\cos\theta) + \frac{1}{4}\delta \sum_{\substack{n=4 \\ (\text{even})}}^{\infty} a_n^{TS} P_n(\cos\theta) + \cdots \right] ,$$

where a_n^{TS} denotes the expansion coefficients for a pair of tangent spheres:[43]

$$a_n^{TS} = (-1)^{(n+2)/2}\, 2(2n+1)\, \frac{(n-3)!!}{(n+2)!!}\ , \quad (n \text{ even})\ .$$

Second, insert the coefficients a_n of this expansion of the radius vector into the general result for B_C (to second order):[11, 12]

$$B_C = 1 - 5 \sum_{n=2}^{\infty} \frac{(n-1)}{(2n+1)^2}\, a_n^2 + \cdots\ .$$

We then find that, to second order,

$$B_C = 1 - \kappa\,\delta^2 - \frac{9}{40}\,\delta\epsilon - \frac{1}{5}\,\epsilon^2 + \cdots\ ,$$

where the constant κ is given by

$$\kappa = \frac{81}{1280} + \frac{5}{4} \sum_{\substack{n=4 \\ (\text{even})}}^{\infty} (n-1) \left[\frac{(n-3)!!}{(n+2)!!} \right]^2 = 0.06556 \cdots\ .$$

3. Symmetric Spheroids with Conicoidal Neck

In this generalization of the two-spheroid model the spheroids may be connected by the conicoidal neck either smoothly or in a way that makes the surface discontinuous (as in the original two-spheroid model). In the case of a smooth connection, the requirement of tangency of the conicoid and the spheroids eliminates one of the degrees of freedom. We will consider here only this case. This means, then, that for the symmetric case there are three degrees of freedom, which will be chosen as follows: (1) the distance between spheroid centers, (2) the semisymmetry axis of either of the two spheroids, and (3) the neck radius of the drop (the transverse semiaxis of the conicoid).

The total potential energy of the system is written in the form of Eq. (6a): the sum of a surface energy and a Coulomb energy. We again use a cylindrical coordinate system, with the z axis along the system's symmetry axis, and ρ the perpendicular distance from the z axis to the surface of the drop.

We denote the semisymmetry axis of either of the two symmetric spheroids by c_1, and the other semiaxis of either spheroid by a_1. The quantities a and c are defined in terms of the equation of a hyperboloid of revolution:

$$\frac{\rho^2}{a^2} - \frac{z^2}{c^2} = 1 .$$

When a^2 is positive, a is the neck radius of the drop. When c^2 is negative and a^2 is positive, the hyperboloid of revolution becomes a third spheroid. When a^2 and c^2 are both negative, the conicoid is a hyperboloid of revolution of two sheets.

The requirements that the volume of the drop be constant and that the conicoid be tangent to the spheroids determine a_1 and c in terms of the other variables. As before, ℓ is the distance between spheroid centers. Let z_t be the value of z at which the third conicoid is tangent to the two end spheroids. We obtain

$$z_t = \frac{\ell}{2}\left[1 - \frac{4c_1^2}{\ell^2}\left(1 - \frac{a^2}{a_1^2}\right)\right]. \tag{B.5a}$$

In terms of z_t and the remaining coordinates, c^2 is found to be given by

$$c^2 = \frac{z_t\, a^2\, c_1^2}{[(\ell/2) - z_t]\, a_1^2} . \tag{B.5b}$$

By performing a straightforward volume integration, the total volume $4\pi R_0^3/3$ of the drop is found to be given by

$$\frac{4\pi}{3} R_0^3 = \frac{4\pi}{3} a_1^3 \left[\frac{3}{2} \frac{a_1^2 z_t}{a_1^3} + \frac{3}{4} \frac{\ell}{a_1} + \frac{c_1}{a_1} \right.$$

$$\left. - \frac{\ell^3}{16 c_1^2 a_1} + \frac{3}{2} \frac{z_t}{a_1} \left(\frac{\ell^2}{4 c_1^2} - 1 \right) - \frac{\ell z_t^2}{2 c_1^2 a_1} \right]. \qquad \text{(B.5c)}$$

By simultaneously solving (B.5a) and (B.5c) we could determine a_1 and z_t; then, c would be given by (B.5b). Since no easy solution of (B.5a) and (B.5c) exists it is more convenient in practice to follow an alternative procedure: If we use as our unit of distance a_1 (rather than R_0) then z_t, c, and R_0 are readily obtained from the set of three equations (B.5).

The eccentricity of the spheroids, when they are prolate, is defined by

$$e_1^2 = 1 - \frac{a_1^2}{c_1^2} .$$

For oblate spheroids we denote by ϵ_1 the quantity (<u>not</u> the eccentricity)

$$\epsilon_1^2 = \frac{a_1^2}{c_1^2} - 1 = - e_1^2 .$$

The dimensionless quantity β_1 is defined by

$$\beta_1 = \frac{(\ell/2) - z_t}{c_1} ,$$

and the quantities g_1 and γ_1 by

$$g_1 = \beta_1 e_1 ,$$

$$\gamma_1 = \beta_1 \epsilon_1 .$$

When the third conicoid is either a hyperboloid of revolution of one sheet or a prolate spheroid we define its eccentricity by

$$e^2 = 1 + \frac{a^2}{c^2} .$$

When it is an oblate spheroid we denote by ϵ the quantity

$$\epsilon^2 = - \frac{a^2}{c^2} - 1 = - e^2 .$$

The dimensionless quantity β is defined for all cases by

$$\beta = \frac{z_t}{|c|} ;$$

the quantities g and γ are defined by

$$g = \beta e ,$$

$$\gamma = \beta\epsilon .$$

The surface energy of the system is determined by performing a straightforward surface integration. The result may be written as a sum of two terms:

$$B_S = B_S^{\text{spheroids}} + B_S^{\text{conicoid}} .$$

The expressions for $B_S^{\text{spheroids}}$ (for prolate and oblate spheroids) are identical in form to (B.3); simply use the current definitions of a_1, c_1, e_1, g_1, ϵ_1, and γ_1 for the analogous quantities a, c, e, g, ϵ, and γ, respectively, appearing there.

When the third conicoid is a hyperboloid of revolution of one sheet we find that

$$B_S^{\text{conicoid}} = \frac{ac}{2R_0^2 e} \left\{ \ln \left[g + (1 + g^2)^{1/2} \right] + g(1 + g^2)^{1/2} \right\}.$$

When it is a prolate spheroid we find that

$$B_S^{\text{conicoid}} = \frac{a|c|}{2R_0^2 e} \left[\sin^{-1} g + g(1 - g^2)^{1/2} \right] ,$$

and when it is an oblate spheroid,

$$B_S^{\text{conicoid}} = \frac{a|c|}{2R_0^2\,\epsilon}\left\{\ln\left[\gamma + (1+\gamma^2)^{1/2}\right] + \gamma(1+\gamma^2)^{1/2}\right\}.$$

The Coulomb energy of the system is calculated by using the set of equations (B.4), in a manner analogous to that described in connection with two overlapping symmetric spheroids.

C. Formulae for Kinetic Energies and Equations of Motion

1. Coplanar Separated Spheroids

We consider first the determination of the kinetic energy of a single incompressible fluid spheroid for the type of hydrodynamic flow we are considering: a superposition of an irrotational flow and a flow corresponding to a uniform rotation of the spheroid as a whole.

We choose a body-fixed xyz coordinate system whose z axis is along the spheroid's symmetry axis. The semisymmetry axis of the spheroid is denoted by c, and its transverse semiaxis by a. The velocity potential ϕ_{ir} for the irrotational flow is given by Lamb:[44]

$$\phi_{ir} = -\frac{1}{2}\left(\frac{\dot{a}}{a}x^2 + \frac{\dot{a}}{a}y^2 + \frac{\dot{c}}{c}z^2\right).$$

The velocity $\vec{v}_{ir}$ of the irrotational motion is then

$$\vec{v}_{ir} = -\operatorname{grad}\phi_{ir} = \frac{\dot{a}}{a}x\,\vec{e}_x + \frac{\dot{a}}{a}y\,\vec{e}_y + \frac{\dot{c}}{c}z\,\vec{e}_z,$$

where $\vec{e}_x$ is a unit vector in the x direction, etc. The condition that the total volume of the drop be constant implies that

$$\frac{\dot{a}}{a} = -\frac{1}{2}\frac{\dot{c}}{c}.$$

Consider now the uniform rotation of the spheroid as a whole about the y axis with angular velocity of magnitude ω. The velocity

$\vec{v}_{rot}$ of this rotational motion (relative to space-fixed axes that are instantaneously coincident with the body-fixed axes) is

$$\vec{v}_{rot} = \omega z \vec{e}_x - \omega x \vec{e}_z ,$$

(We note that $\text{curl}\, \vec{v}_{rot} = 2 \omega \vec{e}_y$.)

The total velocity $\vec{v}$ of the motion consisting of a superposition of these two types of flow (relative to space-fixed axes) is then

$$\vec{v} = \vec{v}_{ir} + \vec{v}_{rot} = (- \frac{1}{2} \frac{\dot{c}}{c} x + \omega z) \vec{e}_x - \frac{1}{2} \frac{\dot{c}}{c} y \vec{e}_y + (\frac{\dot{c}}{c} z - \omega x) \vec{e}_z .$$

From this result we find that the square of the velocity (relative to space-fixed axes) is given by

$$v^2 = [\frac{1}{4} (x^2 + y^2) + z^2] (\frac{\dot{c}}{c})^2 - 3 xz \omega \frac{\dot{c}}{c} + (x^2 + z^2) \omega^2 .$$

The total kinetic energy $\mathcal{T}$ of the drop is

$$\mathcal{T} = \frac{1}{2} \rho_m \int v^2 \, d\tau ,$$

where ρ_m is the (constant) mass density, and $d\tau$ denotes an element of volume. If we substitute for v^2 and perform the straightforward volume integrations we obtain*

$$\mathcal{T} = \frac{1}{2} M_c \dot{c}^2 + \frac{1}{2} M_\theta \omega^2 ,$$

where

$$M_c = \frac{1}{5} (1 + \frac{1}{2} \frac{a^2}{c^2}) M,$$

$$M_\theta = \frac{1}{5} (c^2 + a^2) M ;$$

---

*The result for M_c can also be deduced directly from Eq. (12) of reference 44.

the total mass of the spheroid is M. We note that the cross term involving $\omega \dot{c}$ is zero because the integration of xz over the volume of the symmetric spheroid gives zero.

We now list Hamilton's equations of motion for a system consisting of two coplanar separated spheroids. They are obtained by differentiating the Hamiltonian (10) with respect to the five momenta and the five coordinates that are changing with time.

$$\dot{\ell} = \frac{p_\ell}{M_\ell} ,$$

$$\dot{c}_1 = \frac{p_{c_1}}{M_{c_1}} ,$$

$$\dot{c}_2 = \frac{p_{c_2}}{M_{c_2}} ,$$

$$\dot{\theta}_1 = \frac{p_{\theta_1}}{M_{\theta_1}} + \frac{p_{\theta_1} + p_{\theta_2}}{M_\ell \, \ell^2} ,$$

$$\dot{\theta}_2 = \frac{p_{\theta_2}}{M_{\theta_2}} + \frac{p_{\theta_1} + p_{\theta_2}}{M_\ell \, \ell^2} ,$$

$$\dot{p}_\ell = - \frac{\partial \mathcal{V}}{\partial \ell} + \frac{(p_{\theta_1} + p_{\theta_2})^2}{M_\ell \, \ell^3} ,$$

$$\dot{p}_{c_1} = - \frac{\partial \mathcal{V}}{\partial c_1} + \frac{p_{c_1}^2}{2M_{c_1}^2} \frac{dM_{c_1}}{dc_1} + \frac{p_{\theta_1}^2}{2M_{\theta_1}^2} \frac{dM_{\theta_1}}{dc_1} ,$$

$$\dot{p}_{c_2} = - \frac{\partial \mathcal{V}}{\partial c_2} + \frac{p_{c_2}^2}{2M_{c_2}^2} \frac{dM_{c_2}}{dc_2} + \frac{p_{\theta_2}^2}{2M_{\theta_2}^2} \frac{dM_{\theta_2}}{dc_2} ,$$

$$\dot{p}_{\theta_1} = - \frac{\partial \mathcal{V}}{\partial \theta_1} ,$$

$$\dot{p}_{\theta_2} = - \frac{\partial \mathcal{V}}{\partial \theta_2} .$$

The derivatives of the effective masses are in turn given by

$$\frac{dM_{c_1}}{dc_1} = - \frac{3\, U^2\, R_0^3\, M_0}{10\, c_1^4} ,$$

$$\frac{dM_{\theta_1}}{dc_1} = \frac{U}{5} \left(2c_1 - \frac{U\, R_0^3}{c_1^2} \right) M_0 ;$$

analogous equations hold for dM_{c_2}/dc_2 and dM_{θ_2}/dc_2. The derivatives of the potential energy $\mathcal{V}$ with respect to the coordinates are obtained by straightforward—but lengthy and tedious—differentiations of the results given in Appendix B.1.

2. Overlapping Symmetric Spheroids

We now derive the formula for the kinetic energy of two symmetric overlapping spheroids by use of the approximate method of Werner and Wheeler.[48] For this purpose we use a cylindrical coordinate system, with the z axis parallel to the symmetry axis, and ρ the perpendicular distance to the surface of the drop.

The method of Werner and Wheeler may be stated as follows: First of all, imagine the drop sliced into an infinite number of slices, with the slices' bounding surfaces perpendicular to the symmetry axis. Then, demand that the internal flow be such that when the drop undergoes a displacement all points in one slice remain in that slice—of new position, thickness, and radius—after the displacement. The slice of material undergoes a uniform contraction in the ρ direction and a volume-compensating expansion in the z direction, or vice versa. Werner and Wheeler call this motion a "ρ-independent transport and shear." The motion is consistent with the displacements of the bounding surface, but may deviate from the unique irrotational motion. However, from Kelvin's minimum-energy theorem regarding irrotational motion, the error in the kinetic energy calculated by this method will be of second order in the deviation of the motion from irrotational motion.[116]

The approximation that we are making may be stated mathematically in terms of a single equation:

$$\frac{\dot{\rho}}{\rho} = \frac{\dot{\rho}_0}{\rho_0} = \frac{\dot{a}}{a} \quad , \tag{C.1}$$

where ρ_0 denotes the value of ρ on the surface of the drop, and a is the transverse semiaxis of a spheroid. The equation states simply that the transverse displacement of a point in a slice is proportional to its ρ coordinate.

The kinetic energy $\mathcal{T}$ of the system is given, without approximation, by

$$\mathcal{T} = \frac{1}{2}\rho_m \int v^2 \, d\tau = \pi\rho_m \int dz \int \rho \, d\rho \, (\dot{\rho}^2 + \dot{z}^2) \, ,$$

where ρ_m is the (constant) mass density. If we substitute for $\dot{\rho}$ the result $\rho\,\dot{\rho}_0/\rho_0$ obtained from (C.1), the integration with respect to ρ can be performed to give

$$\mathcal{T} = \frac{1}{2}\pi\rho_m \int dz \, \rho_0^2 \left(\frac{1}{2}\dot{\rho}_0^2 + \dot{z}^2\right).$$

The quantity ρ_0 is given explicitly in terms of z through the equation for the surface of the drop.

The time rates of change of ρ_0 and z are determined in terms of the time rates of change of the two coordinates ℓ and c that specify the system:

$$\dot{\rho}_0 = \left(\frac{\partial \rho_0}{\partial \ell}\right)\dot{\ell} + \left(\frac{\partial \rho_0}{\partial c}\right)\dot{c},$$

$$\dot{z} = \left(\frac{\partial z}{\partial \ell}\right)\dot{\ell} + \left(\frac{\partial z}{\partial c}\right)\dot{c} \quad .$$

The derivation from this point on is fairly straightforward, but lengthy and tedious. It involves taking partial derivatives of the

equation for the surface of the drop and the equation for the volume of the drop, using the second part of (C.1), and performing several integrations. The final result is found to be

$$\mathcal{T} = \frac{1}{2} M_{\ell\ell} \dot{\ell}^2 + M_{\ell c} \dot{\ell}\dot{c} + \frac{1}{2} M_{cc} \dot{c}^2 ,$$

where the effective masses (functions of position) are given by

$$M_{\ell\ell} = \left[\frac{1}{4} + \frac{9(1-\beta)^2(8-9\beta + 3\beta^2)}{160(1+\beta)(2-\beta)^3} \frac{a^2}{c^2} \right] M_0 ,$$

$$M_{\ell c} = \left[\frac{3(1-\beta)^2}{8(2-\beta)} + \frac{3(1+\beta^3)(1-\beta)(8-9\beta + 3\beta^2)}{40(1+\beta)^2(2-\beta)^3} \frac{a^2}{c^2} \right] M_0 ,$$

$$M_{cc} = \left[\frac{(2-2\beta+2\beta^2+3\beta^3-3\beta^4)}{5(1+\beta)(2-\beta)} + \frac{(1+\beta^3)^2(8-9\beta+3\beta^2)}{10(1+\beta)^3(2-\beta)^3} \frac{a^2}{c^2} \right] M_0 .$$

We have defined by β the dimensionless quantity

$$\beta = \frac{\ell}{2c} .$$

The ratio a^2/c^2 is given by

$$\frac{a^2}{c^2} = \frac{2 R_0^3}{c^3(2+3\beta-\beta^3)} .$$

The formula for $\mathcal{T}$ obtained by this method simplifies to the known exact result for two special cases: (1) For a system consisting of two tangent or separated symmetric spheroids we set $\beta = 1$; then

$$M_{\ell\ell} = \frac{1}{4} M_0 ,$$

$$M_{\ell c} = 0 ,$$

$$M_{cc} = \frac{1}{5} \left(1 + \frac{1}{2} \frac{a^2}{c^2}\right) M_0 .$$

(2) For spheroidal distortions ($\dot{\ell} = 0$) of a system consisting of a single spheroid, only a single term contributes to $\mathcal{T}$. Then, for $\beta = 0$,

$$M_{cc} = \frac{1}{5}\left(1 + \frac{1}{2}\frac{a^2}{c^2}\right) M_0 .$$

From the Lagrangian for the system we find that the momenta conjugate to ℓ and c are

$$p_\ell = M_{\ell\ell}\,\dot{\ell} + M_{\ell c}\,\dot{c} ,$$

$$p_c = M_{\ell c}\,\dot{\ell} + M_{cc}\,\dot{c} .$$

Then the Hamiltonian is given by

$$\mathcal{H} = \frac{\frac{1}{2} M_{cc}\, p_\ell^2 - M_{\ell c}\, p_\ell\, p_c + \frac{1}{2} M_{\ell\ell}\, p_c^2}{M_{\ell\ell}\, M_{cc} - M_{\ell c}^2} + \mathcal{V} .$$

Hamilton's equations of motion can then be written as

$$\dot{\ell} = \frac{M_{cc}\, p_\ell - M_{\ell c}\, p_c}{M_{\ell\ell}\, M_{cc} - M_{\ell c}^2} ,$$

$$\dot{c} = \frac{-M_{\ell c}\, p_\ell + M_{\ell\ell}\, p_c}{M_{\ell\ell}\, M_{cc} - M_{\ell c}^2} ,$$

$$\dot{p}_\ell = -\frac{\partial \mathcal{H}}{\partial \ell} ,$$

$$\dot{p}_c = -\frac{\partial \mathcal{H}}{\partial c} .$$

The partial derivatives of $\mathcal{H}$ are obtained by straightforward—but lengthy and tedious—differentiations; formulae for $\mathcal{V}$ as functions of ℓ and c are given in Appendix B.2.

D. Normal-Coordinate Transformation

Our purpose here is to discuss, for the case in which the saddle point consists of two tangent spheroids, two unrelated aspects of the transformation to normal coordinates: the distance between two touching spheroids, and the mass-asymmetry effective mass.

1. Distance Between Two Touching Spheroids

We consider first the determination of the distance ℓ between two touching spheroids as a function of the remaining coordinates. We need the final result only to second order in the angles $\theta_{1x}, \theta_{2x}, \theta_{1y}$, and θ_{2y} (see their definition in Section III.B); because of symmetry at the saddle point the distance ℓ is independent of terms involving $\theta_{1x}\theta_{1y}$, $\theta_{1x}\theta_{2y}$, $\theta_{2x}\theta_{1y}$, and $\theta_{2x}\theta_{2y}$. We therefore consider from the beginning the case in which the symmetry axes of the spheroids are coplanar. (Thus we use the angles θ_1 and θ_2 illustrated in Fig. 2, with $\phi = \phi_1 - \phi_2 = 0$.)

We define a coordinate system whose origin is at the point of tangency of the two spheroids, whose y axis is along the line of tangency, and whose x axis is perpendicular to the y axis. (The x axis is positive in the direction of fragment 2.) Let (x_1, y_1) denote the coordinates of the center of fragment 1, and (x_2, y_2) the coordinates of the center of fragment 2. Then the distance ℓ between fragment centers is

$$\ell = [(x_2 - x_1)^2 + (y_2 - y_1)^2]^{1/2} .$$

The coordinates (x_1, y_1) and (x_2, y_2) are determined in terms of the variables U, c_1, c_2, θ_1, and θ_2 by a geometrical method. The method involves uniformly "squeezing" each spheroid parallel to its symmetry axis into a sphere, leaving all dimensions perpendicular to the symmetry axis unchanged. Then, from the geometry of the sphere and the use of several trigonometric relations, we find that

$$x_1 = -c_1(1 - e_1^2 \cos^2 \gamma_1)^{1/2} ,$$

$$y_1 = \frac{-c_1 e_1^2 \sin \gamma_1 \cos \gamma_1}{(1 - e_1^2 \cos^2 \gamma_1)^{1/2}} ,$$

$$x_2 = c_2(1 - e_2^2 \cos^2 \gamma_2)^{1/2} ,$$

$$y_2 = \frac{-c_2 e_2^2 \sin \gamma_2 \cos \gamma_2}{(1 - e_2^2 \cos^2 \gamma_2)^{1/2}} ,$$

where

$$\gamma_1 = (\pi/2) - \theta_1 - \alpha ,$$

$$\gamma_2 = (\pi/2) + \theta_2 + \alpha ,$$

and

$$\alpha = \tan^{-1}\left(\frac{y_2 - y_1}{x_2 - x_1}\right) .$$

The eccentricities e_1 and e_2 are defined for prolate spheroids by*

$$e_1^2 = 1 - \frac{a_1^2}{c_1^2} = 1 - \frac{UR_0^3}{c_1^3} ,$$

$$e_2^2 = 1 - \frac{a_2^2}{c_2^2} = 1 - \frac{(1 - U)R_0^3}{c_2^3} .$$

---

*Since the saddle point consists of two prolate spheroids, we need not consider oblate spheroids here.

Physically, γ_1 and γ_2 are the angles between the y axis and the symmetry axes of fragments 1 and 2, respectively; α is the angle between the x axis and the line connecting spheroid centers.

For the general case in which θ_1 and θ_2 are arbitrary, no easy solution exists for the above set of equations. However, it is possible to obtain an explicit expression for ℓ to second order in θ_1 and θ_2. We give here the result to second order for the special case in which $U = 1/2$, $c_1 = c_2 \equiv c$ ($e_1 = e_2 \equiv e$), and the angles θ_1 and θ_2 are equal in magnitude. Then, when the fragments rotate in opposite directions ($\theta_2 = -\theta_1 \equiv \theta$),

$$\ell = 2c\left(1 - \frac{e^2}{2}\theta^2 + \cdots\right).$$

When the fragments rotate in the same direction ($\theta_2 = \theta_1 \equiv \theta$),

$$\ell = 2c\left[1 - \frac{e^2\theta^2}{2(1-e^2)} + \cdots\right].$$

We note that for the same angle of rotation the fragment centers approach more closely for rotations in the same direction.

2. Mass-Asymmetry Effective Mass

For the case of actual saddle-point shapes that have small neck radii (low values of x), the mass-asymmetry effective mass can be estimated by neglecting the kinetic energy of motion everywhere except in the neighborhood of the neck. Consider then, for the moment, that the flow of matter from one fragment to the other proceeds through a small neck of effective radius r and effective length d.* Then the mass of the matter flowing through the neck is $M_0\,\pi r^2 d/V$, where M_0 is the total mass of the drop, and V is the volume of the drop. The kinetic energy associated with this flow is

$$\mathcal{T} = \frac{1}{2}\left(\frac{M_0\,\pi r^2 d}{V}\right)v^2,$$

*An analogous method has been used by Marshall Blann and Wladyslaw J. Swiatecki for estimating the effective mass associated with charge fluctuations (unpublished work).

where v is the velocity of flow. The velocity of flow is in turn determined in terms of $\dot{U}$, the time rate of change of the fractional mass U of the left-hand fragment:

$$\pi r^2 v = V\dot{U} .$$

If we substitute for v in the equation for $\mathcal{T}$, we obtain

$$\mathcal{T} = \frac{1}{2} \left(\frac{M_0 Vd}{\pi r^2} \right) \dot{U}^2 .$$

The effective mass for the flow, $M_0 Vd/(\pi r^2)$, is seen to approach infinity as the effective neck radius r approaches zero, provided the effective neck length d does not approach zero as fast as r^2. For actual saddle-point shapes, d is comparable to r, and the effective mass for the flow is then proportional to $1/r$.

For the case of a saddle point consisting of two tangent spheroids, the situation is somewhat different from the case of a saddle point with a well-defined neck. The flow of matter from one spheroid to the other is then analogous to the flow of an incompressible fluid through a circular aperture in a plane wall of infinite extent and infinitesimal thickness, a problem that is discussed by Rayleigh.[117] From his results we find that the corresponding effective mass for flow through an ideal aperture of radius r is $M_0 V/(2r)$, which is approximately the same result that would be obtaining by simply setting $d = r$ in our earlier result. As before, the mass-asymmetry effective mass approaches infinity as r approaches zero.

E. Accuracy of Approximate Formulae Relating Observable Properties of Fragments to the Initial Conditions at the Saddle Point

Our purpose here is to give some indication of the accuracy of Eqs. (14), (16), and (17), which relate the observable properties of fragments at infinity to the initial conditions at the saddle point. For a large number of combinations of initial conditions and several values

of x, we have numerically integrated the equations of motion and determined the corresponding values of the observable quantities of interest at infinity. These exact results may then be compared with the results calculated by use of (14), (16), and (17).*

We present in Table E.I a comparison of the exact and approximate results for cases in which the fissionability parameter x is equal to 0.677.† Forty-four sets of initial conditions were selected in such a way as to illustrate, for the most part, the accuracy of the formulae as regards a particular coordinate or momentum or group of coordinates and momenta. Consequently, each set of initial conditions includes, in general, several coordinates that are equal to their saddle-point values and several zero momenta. For the last four sets of initial conditions, however, the initial conditions were determined in a completely random manner—whether a particular momentum or the deviation of a coordinate from its saddle-point value was taken to be positive, zero, or negative was determined by a roll of a die.

The magnitude of the various momenta and deviations of the coordinates from the saddle point were selected to correspond very roughly to one-tenth the maximum probability of occurrence (for a system such as ${}_{83}Bi^{209}$ + 65-MeV α, for example). This means that the actual deviations from the saddle point are for the most part much smaller than those chosen here. Since the approximate formulae are designed specifically for small deviations, the agreement between the approximate and exact results is considerably better for typical deviations than for the cases presented here.

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*The equations of motion, recall, are numerically integrated for configurations in which the symmetry axes are coplanar, and in which the spheroids are not spinning about their symmetry axes. For this purpose we use the two angular coordinates θ_1 and θ_2 (see Fig. 2), with $\phi = \phi_1 - \phi_2 = 0$.

†This is the value of the fissionability parameter for such compound nuclei as ${}_{85}At^{213}$, ${}_{84}Po^{208}$, and ${}_{82}Pb^{198}$.

Table E.I. Observable properties of fragments at infinity corresponding to 44 sets of initial conditions, for x = 0.677. For a given set of initial conditions, the values of the total translational kinetic energy E, the individual excitation energies X_1 and X_2, and the individual angular momenta L_1 and L_2 that are determined exactly by solving the equations of motion are compared with the values that are determined from the approximate formulae (14), (16), and (17). The energies E, X_1, and X_2 are in units of $E_S^{(0)}$; the angular momenta L_1 and L_2 are in units of L_0. The quantities δc_1 and δc_2 are in units of R_0; $\delta\theta_1$ and $\delta\theta_2$ are in radians; p_ℓ, p_{c_1}, and p_{c_2} are in units of P_0; p_{θ_1} and p_{θ_2} are in units of L_0. (See discussion of units in Section II.A.) In each case the value taken for $\delta\ell$ is $\delta c_1 + \delta c_2$.

δU / p_ℓ	δc_1 / p_{c_1}	δc_2 / p_{c_2}	$\delta\theta_1$ / p_{θ_1}	$\delta\theta_2$ / p_{θ_2}	E (exact) / E (approx.)	X_1(exact) / X_1(approx.)	X_2(exact) / X_2(approx.)	L_1 (exact) / L_1(approx.)	L_2 (exact) / L_2(approx.)
0.0	0.2	0.0	0.0	0.0	0.2214	0.0544	0.0321	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2213	0.0536	0.0318	0.0	0.0
0.0	-0.2	0.0	0.0	0.0	0.2586	0.0148	0.0355	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2584	0.0133	0.0352	0.0	0.0
0.0	0.2	0.2	0.0	0.0	0.2065	0.0528	0.0528	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2065	0.0520	0.0520	0.0	0.0
0.0	0.2	-0.2	0.0	0.0	0.2388	0.0567	0.0137	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2384	0.0553	0.0117	0.0	0.0
0.0	-0.2	-0.2	0.0	0.0	0.2822	0.0164	0.0164	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2821	0.0150	0.0150	0.0	0.0
0.0	0.0	0.0	0.2	0.0	0.2382	0.0328	0.0334	0.0053	0.0010
0.0	0.0	0.0	0.0	0.0	0.2384	0.0335	0.0335	0.0058	0.0011
0.0	0.0	0.0	0.2	0.2	0.2378	0.0326	0.0326	0.0059	0.0059
0.0	0.0	0.0	0.0	0.0	0.2384	0.0335	0.0335	0.0069	0.0069
0.0	0.0	0.0	0.2	-0.2	0.2381	0.0328	0.0328	0.0043	-0.0043
0.0	0.0	0.0	0.0	0.0	0.2384	0.0335	0.0335	0.0046	-0.0046
0.0	0.0	0.0	0.0	0.0	0.2430	0.0328	0.0328	0.0	0.0
0.04	0.0	0.0	0.0	0.0	0.2416	0.0335	0.0335	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2383	0.0361	0.0328	0.0	0.0
0.0	-0.02	0.0	0.0	0.0	0.2384	0.0335	0.0335	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2387	0.0334	0.0369	0.0	0.0
0.0	0.02	-0.02	0.0	0.0	0.2384	0.0335	0.0335	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2382	0.0354	0.0354	0.0	0.0
0.0	-0.02	-0.02	0.0	0.0	0.2384	0.0335	0.0335	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2421	0.0350	0.0350	0.0	0.0
0.04	-0.02	-0.02	0.0	0.0	0.2416	0.0335	0.0335	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2433	0.0327	0.0362	0.0	0.0
0.04	0.02	-0.02	0.0	0.0	0.2416	0.0335	0.0335	0.0	0.0

Table E.I (cont.)

δU / p_ℓ	δc_1 / p_{c_1}	δc_2 / p_{c_2}	$\delta\theta_1$ / p_{θ_1}	$\delta\theta_2$ / p_{θ_2}	E (exact) / E(approx.)	X_1 (exact) / X_1(approx.)	X_2 (exact) / X_2(approx.)	L_1(exact) / L_1(approx.)	L_2 (exact) / L_2(approx.)
0.0	0.0	0.0	0.0	0.0	0.2409	0.0356	0.0356	0.0	0.0
-0.04	-0.02	-0.02	0.0	0.0	0.2416	0.0335	0.0335	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2361	0.0319	0.0319	0.0417	-0.0417
0.0	0.0	0.0	0.04	-0.04	0.2384	0.0335	0.0335	0.0476	-0.0476
0.1	0.0	0.0	0.0	0.0	0.2288	0.0257	0.0420	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2289	0.0251	0.0418	0.0	0.0
0.1	0.2	0.0	0.0	0.0	0.2123	0.0446	0.0407	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2124	0.0453	0.0402	0.0	0.0
0.1	-0.2	0.0	0.0	0.0	0.2484	0.0096	0.0437	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2481	0.0050	0.0435	0.0	0.0
0.1	0.0	0.2	0.0	0.0	0.2125	0.0242	0.0646	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2124	0.0234	0.0620	0.0	0.0
0.1	0.0	-0.2	0.0	0.0	0.2481	0.0278	0.0211	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2481	0.0268	0.0217	0.0	0.0
0.1	0.2	0.2	0.0	0.0	0.1980	0.0429	0.0632	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.1982	0.0436	0.0603	0.0	0.0
0.1	0.2	-0.2	0.0	0.0	0.2290	0.0470	0.0200	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2289	0.0470	0.0200	0.0	0.0
0.1	-0.2	0.2	0.0	0.0	0.2295	0.0085	0.0664	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2289	0.0033	0.0637	0.0	0.0
0.1	-0.2	-0.2	0.0	0.0	0.2710	0.0111	0.0226	0.0	0.0
0.0	0.0	0.0	0.0	0.0	0.2708	0.0067	0.0234	0.0	0.0
0.1	0.0	0.0	0.2	0.0	0.2287	0.0251	0.0418	0.0042	0.0009
0.0	0.0	0.0	0.0	0.0	0.2289	0.0251	0.0418	0.0058	0.0011
0.1	0.0	0.0	0.0	0.2	0.2285	0.0257	0.0412	0.0008	0.0058
0.0	0.0	0.0	0.0	0.0	0.2289	0.0251	0.0418	0.0011	0.0058
0.1	0.0	0.0	0.2	0.2	0.2282	0.0251	0.0408	0.0047	0.0063
0.0	0.0	0.0	0.0	0.0	0.2289	0.0251	0.0418	0.0069	0.0069
0.1	0.0	0.0	0.2	-0.2	0.2284	0.0252	0.0413	0.0034	-0.0050
0.0	0.0	0.0	0.0	0.0	0.2289	0.0251	0.0418	0.0046	-0.0046

Table E.I (cont.)

δU / P_ℓ	δc_1 / P_{c_1}	δc_2 / P_{c_2}	$\delta\theta_1$ / P_{θ_1}	$\delta\theta_2$ / P_{θ_2}	E (exact) / E (approx.)	X_1(exact) / X_1(approx.)	X_2(exact) / X_2(approx.)	L_1(exact) / L_1(approx.)	L_2(exact) / L_2(approx.)
0.0	0.0	0.0	0.2	0.0	0.2359	0.0304	0.0319	0.0443	-0.0408
0.0	0.0	0.0	0.04	-0.04	0.2384	0.0335	0.0335	0.0534	-0.0465
0.0	0.0	0.0	0.2	0.0	0.2363	0.0316	0.0317	-0.0384	0.0425
0.0	0.0	0.0	-0.04	0.04	0.2384	0.0335	0.0335	-0.0419	0.0488
0.0	0.0	0.0	0.2	0.2	0.2361	0.0302	0.0315	0.0448	-0.0378
0.0	0.0	0.0	0.04	-0.04	0.2384	0.0335	0.0335	0.0545	-0.0407
0.0	0.0	0.0	0.2	-0.2	0.2358	0.0306	0.0306	0.0435	-0.0435
0.0	0.0	0.0	0.04	-0.04	0.2384	0.0335	0.0335	0.0523	-0.0523
0.0	0.0	0.0	0.2	-0.02	0.2366	0.0317	0.0317	-0.0393	0.0393
0.0	0.0	0.0	-0.04	0.04	0.2384	0.0335	0.0335	-0.0430	0.0430
0.0	0.2	0.2	0.2	0.0	0.2060	0.0521	0.0527	0.0085	0.0018
0.0	0.0	0.0	0.0	0.0	0.2065	0.0520	0.0520	0.0058	0.0011
0.0	0.2	-0.2	0.2	0.0	0.2382	0.0557	0.0136	0.0101	0.0007
0.0	0.0	0.0	0.0	0.0	0.2384	0.0553	0.0116	0.0058	0.0011
0.0	-0.2	0.2	0.2	0.0	0.2388	0.0133	0.0567	0.0017	0.0008
0.0	0.0	0.0	0.0	0.0	0.2384	0.0116	0.0553	0.0058	0.0011
0.0	-0.2	-0.2	0.2	0.0	0.2822	0.0158	0.0164	0.0018	0.0004
0.0	0.0	0.0	0.0	0.0	0.2821	0.0150	0.0150	0.0058	0.0011
0.0	0.2	0.0	0.2	0.0	0.2208	0.0535	0.0319	0.0092	0.0013
0.0	0.0	0.0	0.0	0.0	0.2213	0.0536	0.0318	0.0058	0.0011
0.0	-0.2	0.0	0.2	0.0	0.2586	0.0143	0.0355	0.0017	0.0006
0.0	0.0	0.0	0.0	0.0	0.2584	0.0133	0.0352	0.0058	0.0011
0.0	-0.2	0.2	-0.2	0.0	0.2378	0.0164	0.0544	0.0398	0.0433
0.0	-0.02	0.0	0.04	0.04	0.2384	0.0116	0.0553	0.0447	0.0493
0.1	0.2	-0.2	0.0	0.2	0.2262	0.0477	0.0197	-0.0428	0.0403
0.0	-0.02	-0.02	-0.04	0.04	0.2289	0.0470	0.0200	-0.0465	0.0432
0.0	0.0	-0.2	0.2	0.0	0.2581	0.0372	0.0134	0.0051	0.0403
0.0	-0.02	0.0	0.0	0.04	0.2584	0.0352	0.0133	0.0072	0.0502
-0.1	0.2	0.2	0.0	0.2	0.1960	0.0616	0.0446	0.0454	-0.0401
-0.04	-0.02	-0.02	0.04	-0.04	0.2015	0.0603	0.0436	0.0488	-0.0419

It is seen that, for the most part, the approximate equation for E reproduces the exact results more accurately than do the equations for X_1, X_2, L_1, and L_2. The approximate equations for L_1 and L_2 are seen, for example, to give overestimates of the angular momenta at infinity; this is because for the large values of the initial momenta considered here we are outside the region of linearity. Also, since the equations for L_1 and L_2 contain no terms representing the dependence of the angular momentum upon fragment elongation, their accuracy is reduced when δc_1 and/or δc_2 are not close to zero.

F. Properties of Certain Integrals

We give here various properties of the integrals $I_n(\beta)$ defined by Eq. (43). We first show the relationship between these integrals and a generalization of the repeated integrals of the error function.[61-63*]

The complement of the error function is defined by

$$\operatorname{erfc} \beta = \frac{2}{\sqrt{\pi}} \int_{\beta}^{\infty} \exp(-x^2)\, dx .$$

The nth repeated integral of the (complement of) the error function is then defined, for $n \geq 1$, by [61, 62]

$$i^n \operatorname{erfc} \beta = \int_{\beta}^{\infty} i^{n-1} \operatorname{erfc} x \, dx, \tag{F.1a}$$

with

$$i^0 \operatorname{erfc} \beta = \operatorname{erfc} \beta. \tag{F.1b}$$

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*The functions discussed in reference 63 are closely related to the repeated integrals of the error function; they are defined in such a way as to be more useful for statistical applications. If reference 63 is consulted, note that the same symbol I_n is used to denote the functions considered there.

Explicit evaluation of this expression gives

$$i^n \operatorname{erfc} \beta = \frac{2}{\sqrt{\pi}\,\Gamma(n+1)} \int_0^\infty x^n \exp[-(x+\beta)^2]\,dx, \qquad (F.2)$$

where Γ denotes the gamma function $[\Gamma(n+1) = n!]$. Although the definition (F.1) defines $i^n \operatorname{erfc} \beta$ only for integral values of n greater than or equal to zero, the result (F.2) defines the function for all values of n greater than -1.[63]

From a comparison of (F.2) and (43) we obtain the desired relationship between the functions:

$$I_n(\beta) = \frac{1}{2}\sqrt{\pi}\,\Gamma(n+\tfrac{1}{2}) \exp\left[\left(\frac{\beta}{2}\right)^2\right] i^{n-(1/2)} \operatorname{erfc}\left(\frac{\beta}{2}\right), \qquad (F.3a)$$

or, alternatively,

$$i^n \operatorname{erfc} \beta = \frac{2 \exp(-\beta^2)}{\sqrt{\pi}\,\Gamma(n+1)}\, I_{n+(1/2)}(2\beta). \qquad (F.3b)$$

For our purposes we are interested in $I_n(\beta)$ for integral values of n, which means repeated integrals of the error function of half-integral order. Tables of the functions $i^n \operatorname{erfc} \beta$ (or related functions) are available for integral values of n,[61-63] but not for fractional values of n.

The following properties of the integrals $I_n(\beta)$ can be determined either directly from the definition (43), or else from the analogous properties[61] of $i^n \operatorname{erfc} \beta$ and the relationships (F. 3):

(a) Differential equation satisfied by $I_n(\beta)$:

$$\left[4\frac{d^2}{d\beta^2} - 2\beta\frac{d}{d\beta} - (2n+1)\right] I_n(\beta) = 0.$$

(b) Recurrence formulae:

$$I_n(\beta) = \frac{1}{2}\left[(n - \frac{3}{2})\, I_{n-2}(\beta) - \beta\, I_{n-1}(\beta)\right],$$

$$\frac{d}{d\beta}\, I_n(\beta) = \frac{1}{2}\left[\beta\, I_n(\beta) - (n - \frac{1}{2})\, I_{n-1}(\beta)\right].$$

(c) Asymptotic expansion:

$$I_n(\beta) \xrightarrow[\beta \to \infty]{} \frac{\Gamma(n + \frac{1}{2})}{\beta^{n + (1/2)}} .$$

(d) Integral relation:

$$I_n(\beta) = \frac{1}{2}(n - \frac{1}{2}) \exp\left[\left(\frac{\beta}{2}\right)^2\right] \int_\beta^\infty \exp\left[-\left(\frac{x}{2}\right)^2\right] I_{n-1}(x)\, dx .$$

G. Numerical Procedures

The numerical calculations of this work were performed, for the most part, on an IBM 7094 computer, by use of codes written in FORTRAN. We will summarize here the numerical methods that were used in performing some of the more important of these calculations.

When calculating the interaction energy and derivatives of the interaction energy, the triple multipole summations were performed by explicitly summing over each of the three summation indices until terms were reached that were less in magnitude than some specified value. The actual value that was specified ranged from 10^{-6} (when integrating the equations of motion) to 10^{-14} (when calculating the stiffness constants). In performing these summations, the associated Legendre polynomials and their derivatives were evaluated by use of standard recurrence formulae.[118]

The double integrations required for the calculation of the Coulomb energy for the case of two overlapping spheroids and for

the case of two spheroids connected by a conicoidal neck were performed by repeated applications of a ten-point Gaussian quadrature routine.[119] The complete elliptic integrals appearing in the integrand were evaluated by use of approximate representations that are accurate to within 1.5×10^{-8}.[115] The same number of integration points was used in the evaluation of the first integral (for the electrostatic potential on the surface of the drop) as in the evaluation of the second integral (for the Coulomb energy itself). For one of the two halves of a drop, the actual number of points used in the evaluation of one of these two integrals ranged from 10 (when integrating the equations of motion) to 30 (when calculating the Coulomb energies presented in Fig. 3a). For the case of two spheroids connected by a conicoidal neck, the corresponding number of points used was 20.* When 30 such points were used the calculated Coulomb energies B_C were typically accurate to within a few units in the sixth decimal; for 20 points and 10 points they were typically accurate to within a few units in the fifth and fourth decimals, respectively.

For the case in which the saddle point consists of two tangent spheroids, the precise location of the saddle point was determined by finding the minimum of the potential energy along the scission line $\ell = 2c$. This was done by an iterative scheme involving the single variable $c = \ell/2$. An approximate location of the saddle point was used to define a starting value of c. Three evaluations of the potential energy were then made (at this value of c and to either side of it), and a quadratic expression in c was fitted to these values. The value of c corresponding to the minimum of the energy for this quadratic

*Since at the point of connection of the third conicoid with the two spheroids there is a discontinuity in the second derivative of the coordinate ρ with respect to z (cylindrical coordinates), it is crucial that the interval of a single ten-point quadrature does not extend across this point of connection.

expression was used as the starting point for a repetition of the calculation. When the iterations led to a change in c less than a specified amount (taken to be 10^{-10} R_0), the calculation was terminated, and the value of c was taken to be the value corresponding to the saddle point.

After the saddle point was located (for the case of tangent spheroids) the stiffness constants were determined by making further evaluations of the potential energy in the immediate neighborhood of the saddle point and by fitting a quadratic expression in several variables to these values. In order to maintain sufficient accuracy in the final result, the location of the saddle point and the calculation of the stiffness constants were performed by use of double-precision arithmetic; the stiffness constants determined in this way are accurate to within a few units in the fifth decimal (when expressed in liquid-drop units).*

For the case in which the saddle point consists of overlapping spheroids, and for the case of two spheroids connected by a conicoidal neck, the saddle point was determined by an iterative scheme similar to that described above. A quadratic expression in two or three variables, respectively, was fitted to values of the potential energy in the neighborhood of an approximate saddle point, and the extremum of the energy for this expression was used as a better approximation to the saddle point. For these cases, the (single-precision) saddle-point-searching code of Cohen and Swiatecki was used.[15]

The equations of motion for the system were numerically integrated by use of a routine employing Adam's method.[120] This routine numerically integrates a system of simultaneous first-order differential equations, maintaining at each step of the integration a prescribed accuracy (by changing the integration step if necessary). For this work

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*The saddle-point third derivatives K_{mms} and K_{sss} (Fig. 38) were calculated on a desk calculater by use of tables of the potential energy, and may consequently be slightly in error.

the maximum allowed error at each integration step in each of the coordinates and momenta was specified to be 10^{-5}; in practice, the actual error was much less. With this specification of accuracy, about 400 to 500 integration steps were required, in general, to integrate the equations of motion from the neighborhood of a tangent-spheroid saddle point to a separation distance of $\ell = 25\ R_0$. The values of the quantities of interest at infinity are determined sufficiently well at $\ell = 25\ R_0$ that the integrations were stopped there. The value of the total translational kinetic energy at infinity was then taken to be equal to its value at $\ell = 25\ R_0$ plus the current interaction energy of either the two spheroids or of two spheres with coincident centers (there is negligible difference at this distance). The values of the remaining quantities of interest at infinity were taken to be equal to their values at $\ell = 25\ R_0$.

The accuracy of the solutions of the equations of motion for the case of overlapping spheroids was not as great as for the case of separated spheroids. This is because of the relatively large amount of computing time required for the calculation of the Coulomb energy and its derivatives. These quantities were calculated for the purposes of the integrations from a quadratic expression in ℓ and c that was fitted to values of the Coulomb energy calculated in the immediate neighborhood of the current location of the system. The solution for $x = 0.90$ presented in Fig. 28 may consequently be slightly in error.

The constants appearing in the approximate formulae (14), (16), and (17) were determined (for a given value of x) from the solutions of the equations of motion corresponding to a sufficient number of sets of initial conditions. Consider, for example, the determination of the constants X_1^0 and $X_{1,s}$ of Eq. (16). The constant X_1^0 is given by the solution of the equations of motion corresponding to starting the system from rest at the saddle point. Let X_1 denote the excitation energy corresponding to the solution in which all initial conditions are zero except the stretching coordinate s, which is taken to be small in magnitude. Then $X_{1,s}$ is determined from

$$X_{1,s} = \frac{X_1 - X_1^0}{s} .$$

In the calculation of the constants for Eqs. (14), (16), and (17), as well as in the calculation of the stiffness constants, care must of course be taken that the calculated values are independent of the choice of increments in the coordinates.

The integrals $I_n(\beta)$ defined by Eq. (43) were evaluated numerically by use of a ten-point Gaussian quadrature routine.[119]

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