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To be published in Physical Review

UCRL-9540

UNIVERSITY OF CALIFORNIA
Lawrence Radiation Laboratory
Berkeley, California
Contract No. W-7405-eng-48

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ABSTRACT

Angular distributions and kinetic-energy spectra of fragments, and cross sections for fission of U^{238} with 63- to 124-Mev C^{12} ions, have been measured with the use of a silicon p-n junction detector. The distributions have been analyzed in terms of the formation of a compound nucleus and subsequent decay by evaporation of neutrons in competition with fission. The percent fission from each isotope in the evaporation chain has been calculated and the over-all angular distribution estimated with the use of the theoretical curves of Halpern and Strutinski. At the highest bombarding energies, the observed angular distributions were found to be more nearly isotropic than predicted.

The mean linear momentum of the fissioning nucleus appears to be less than that of the heavy ion. A possible explanation for these discrepancies is that before the fission event there is competition from reactions in which particles are emitted in the forward direction. The contribution from this kind of reaction is estimated to be of the order of 30% at 95 Mev and 124 Mev.

Over the entire range of bombarding energies, the most probable total kinetic energy release is 186 ± 6 Mev. By correspondence this suggests that the fissioning nuclei are californium isotopes.

The fission cross section increases from a value of 40 mb at 63 Mev to 2.35b at 124 Mev. The experimental fission cross sections agree well with the cross sections for compound-nucleus formation calculated by use of a square-well nuclear potential with a radius parameter $r_o = 1.5 \times 10^{-13}$ cm.

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INTRODUCTION

At the first Geneva Conference, Bohr developed some general ideas for understanding the angular distributions of fragments resulting from fission of nuclei having excitation energies slightly higher than their fission barriers.[1] Under these conditions the nucleus goes over the saddle pass "cold"; that is, most of the excitation energy is expended in potential energy of deformation towards fission. Therefore, the spectrum of energy levels of the highly deformed nuclei at the saddle pass should be similar to those of stably deformed nuclei at energies near their ground states. Bohr further assumes that the nuclei retain axial symmetry throughout the deformation, and that the fragments are emitted in the direction of the symmetry axis. The fragment angular distributions are therefore determined by the distributions of the orientations of the symmetry axis with respect to the beam.

Angular distributions based on the Bohr model have been worked out quantitatively and extended to higher energies by a number of authors.[2-5] Among these treatments, that by Halpern and Strutinski is most directly applicable to the systems studied in this work.[2] According to these authors, the fissioning nucleus may be characterized by three quantum numbers: I, the total angular momentum; K, the projection of $\underset{\sim}{I}$ along the direction of the separating fragments (thus, in keeping with the assumptions, along the symmetry axis of the nucleus at the saddle point); and M, the component of $\underset{\sim}{I}$ along the beam direction. For fission induced by high-energy heavy ions, I is approximately equal to that orbital angular momentum of the bombarding particle ℓ which is perpendicular to the beam direction. It is assumed here that M = 0, since the spins of U^{238} and C^{12} are zero, and any component of angular momentum along the beam resulting from particle emission prior to fission is

* This work was supported by the U. S. Atomic Energy Commission.

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expected to be negligible compared to ℓ. With these assumptions, the expression for the angular distribution becomes

$$W(\theta_{c.m.}) = \int dI \int dK\, f(I)g(K) \left[\sin^2\theta_{c.m.} - (K/I)^2\right]^{-1/2}, \tag{1}$$

where f(I) and g(K) are the distributions in I and K of the fissioning nucleus at the saddle point.

In a classical approximation, the possible values of I are distributed uniformly in I^2 from zero to some maximum value I_m^2. In order to evaluate g(K), Halpern and Strutinski have assumed that the division of I between rotation perpendicular and rotation parallel to the symmetry axis is governed by a Boltzmann factor containing the rotational energy.[2] This results in a Gaussian distribution,

$$g(K) \propto C \exp\left[-(K^2/K_0^2)\right], \tag{2}$$

where K_0^2 is the mean value of K^2. Furthermore, K_0^2 is given by

$$K_0^2 = \frac{T\,\mathfrak{J}_{eff}}{\hbar^2}, \tag{3}$$

where T is the nuclear temperature and

$$\frac{1}{\mathfrak{J}_{eff}} = \frac{1}{\mathfrak{J}_{\parallel}} - \frac{1}{\mathfrak{J}_{\perp}} . \tag{4}$$

The $\mathfrak{J}$'s are rigid-body moments of inertia of the prolate saddle-point nucleus, with $\mathfrak{J}_{\parallel}$ being the moment with respect to the symmetry axis and $\mathfrak{J}_{\perp}$ that with respect to an axis perpendicular to the symmetry axis. All of the quantities $\mathfrak{J}_{\parallel}$, $\mathfrak{J}_{\perp}$, and T must be evaluated at the state of the fission process where the K distribution is fixed. Halpern and Strutinski[2] used the level-density formula $\rho(E) = \text{const} \exp\left[2(aE)^{1/2}\right]$, and by assuming $\mathfrak{J}_{eff}$ to be constant, they obtained the relation

$$K_0^2 = \text{const}\,(E-E_f)^{1/2}, \tag{5}$$

where $(E-E_f)$ is the excitation energy in excess of the fission barrier, E_f, for the particular nucleus undergoing fission.

Each of the functions $W_{I(m)}$, K_0, characterized by a single parameter $p = (I_m/2K_0)^2$, follows a $1/\sin\theta_{c.m.}$ curve in the region around 90 deg and falls below $1/\sin\theta_{c.m.}$ near 0 deg and 180 deg.

Griffin[3] assumes a linear distribution of the form $g(K) \propto (K_m - K)$ where K_m is a maximum value of K. His predicted distributions are similar to those of Halpern and Strutinski, but in some cases go above the $1/\sin\theta_{c.m.}$ curve.

Halpern and Strutinski have empirically constructed a curve of K_0^2 as a function of $(E-E_f)$, using experimental angular distributions of fragments from neutron-, proton-, and He^4-induced fission of several heavy nuclides.[2,6,7] It appears that for values of $(E-E_f)$ less than 10 or 15 Mev, relationship (4) is in disagreement with experiments.

The values of parameter p, obtained by fitting the experimental angular distributions with theoretical curves, were used to estimate the mean excitation energy of the fissioning nucleus at the time of fission in the reaction between gold and carbon ions.[8] By obtaining this quantity one was able to determine the mean number of particles emitted prior to fission. The forward motion of the fissioning nuclei was found to be consistent with formation of a compound nucleus over the entire range of bombarding energies studied. At all energies the measured absolute fission cross section in the reaction between Au^{197} and C^{12} was less than the calculated cross section for the formation of a compound nucleus for the square-well model with a radius parameter $r_o = 1.5 \times 10^{-13}$cm. An appreciable amount of the struck nuclei ($\sigma \approx 100$ to 200 mb) result in neutron-evaporation products that survive fission.[9] Also, there may be large numbers of surviving reaction products resulting from emission of charged particles; however, these have not been experimentally measured in this system.

We have chosen U^{238} as the target nucleus in our investigation because we should expect a deviation from this picture. By bombarding with carbon ions, nuclei are formed which have low fission barriers ($\approx$5 Mev) and high level widths for fission. Any non-compound-nucleus processes leading to an excitation energy higher than 5 Mev will therefore, in most cases, lead to fission. For such processes, which include "stripping" and "pickup" reactions, only part of the linear and orbital angular momenta of the heavy ion is deposited in the fissioning nucleus. This will result in a more nearly isotropic fission-fragment distribution if these reactions contribute significantly to the fission cross section. The fission cross section, therefore, should represent the total reaction cross section for processes in which at least 5 Mev excitation energy is deposited. It will be of interest to compare this with any calculated values for cross section for compound-nucleus formation.

II. EXPERIMENTAL PROCEDURES

The experimental arrangement has been described in a previous paper.[8] Carbon-ion beams were obtained from the Berkeley heavy-ion linear accelerator (Hilac), which accelerates heavy ions to 10.4 Mev/nucleon. Occasional lower-energy groups have been observed,[10] and in order to obtain the 125-Mev component of C^{12}, the beam was deflected through 15 deg by a bending magnet before reaching the target chamber. The energy spread of the beam has been shown to have a standard deviation of 0.8%.[11] Lower energies were obtained by inserting weighed aluminum foils into the beam path. The range curve for C^{12} in aluminum, as measured by Walton,[12] was used to estimate the energy. Some of the lower-energy points were also checked by measuring the residual ranges of the ions in nuclear emulsions. They agreed to within 1 Mev of the estimated energies. We have therefore assigned an uncertainty of 1 Mev in the energy.

Before striking the target, the beam passed through two 1/8-in.-diameter collimators, 10 in. apart. Beam particles were collected in a Faraday cup at the rear of the chamber. Targets were made by vaporizing UF_4 onto 0.03-mil nickel backing foils. Targets most frequently used had 250 $\mu g/cm^2$ of U^{238} and were found to withstand beams of up to 10^{-6} amp/cm^2.

The targets were oriented 45 deg to the beam with the uranium layer facing the detector. The detectors used were made by diffusion of n- or p-type impurities into one face of a silicon wafer containing an excess of the opposite type of impurity.[13,14] A more detailed account of the properties of these detectors is given elsewhere.[15]

The angular position, θ_L, of the detector could be adjusted to within 1 deg and the angular resolution usually was of the order of 3 deg.

The electronic system used with the semiconductor detector has been described in Ref. (15). A pulse generator was used to check the gain and noise level of the system and to make corrections for coincidence losses. A signal from the Hilac electronic system could be used to trigger the pulse generator during the 2-msec bursts of particles.

A Cf^{252} spontaneous-fission source was used to calibrate the detectors. Energies corresponding to the peaks of the Cf^{252} spectrum were taken from the time-of-flight data of Fraser and Milton.[16] An energy deficiency has been observed in the spectrum from the detectors. The assumption has been made that the

ionization defect is negligible, owing to the small amount of energy required for electron-hole pair formation in the semiconductor material.[17] The energy loss has been attributed to a "window" or "dead layer" at the surface of the detector.[15] Correction for energy loss in the detector "window", assumed to be silicon, was made with the help of the fragment range-energy data of Fulmer[18] and Schmitt and Leachman.[19] Recent measurements have shown that a substantial part of the energy defect is due to a different effect, possibly an incomplete collection of ions produced in the detector.[20] We have corrected for this loss, which we have assumed to be proportional to the mass of the fragment. Corrections for energy degradations in the targets were determined empirically by bombardment of targets of various thicknesses. A thickness of 250 $\mu g/cm^2$ of UF_4 was found to degrade the energy by an amount corresponding to 60 $\mu g/cm^2$ of aluminum. The fission fragments were identified by their range-energy relationship in aluminum.

Figure 1 shows a typical fragment kinetic energy spectrum obtained at 90 deg to the beam with a UF_4 target bombarded with 124-Mev C^{12} ions. The large number of counts at the low-energy end of the spectrum resulted from pileup of pulses produced by scattered beam particles and light reaction products. Individual pulses from these light particles were clearly distinguishable from the pulses produced by fission fragments because the sensitive counting region could be made just slightly longer than the range of the densely ionizing fission fragments by varying the voltage applied to the detector. Thus, the lighter particles deposited only small amounts of energy in the counting region. However, pileup of several of the small pulses in the electronic system could result in pulses of the size produced by the fission fragments. This difficulty became serious only at forward angles less than about 40 deg. At those angles a logarithmic subtraction of the pileup background was often necessary.

The same detector was also used to count elastically scattered carbon ions for the determination of the total cross section for fission. It was found that electrons knocked from the target by the beam onto the detector had the effect of worsening the resolution of the carbon-ion peak, but not the resolution of the pulse-generator peak. This effect was overcome by introducing a magnetic field of 1000 gauss, 1 cm in length, in front of the detector. This field removed most of the electrons. Apparently the electrons cause a malfunction of the detector. A typical spectrum for 73-Mev carbon ions

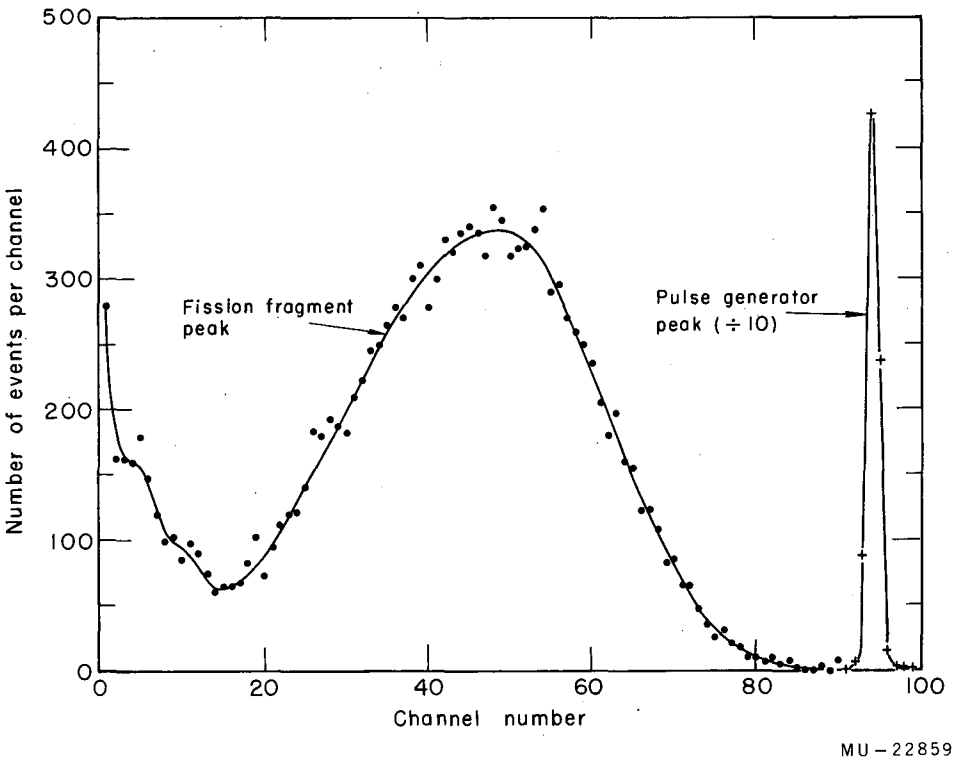

Fig. 1. Spectrum of fragment kinetic energies from fission of U^{238} induced by 124-Mev C^{12} ions. Observed at 90 deg to the beam with a p-n junction detector reverse-biased by 6 v.

elastically scattered at an angle of 25 deg in the laboratory system is shown in Fig. 2. On the low-energy side of the sharp peak is a tail of inelastically scattered ions and reaction products.

III. RESULTS

A. Angular Distribution of Fission Fragments

The fragment kinetic energy spectra, obtained at various angles, were integrated, corrected for coincidence loss, and normalized to the same number of beam particles.

Statistical errors were negligible. Possible systematic errors arose from inhomogeneous thickness of target, errors in angle, and variations in the counting efficiency of the detector. A background of fission activity was introduced onto the surface of the detector by self-transfer of Cf^{252} from the calibration source. The background therefore had to be determined after each calibration.

Another possible error at lower bombarding energy was due to inhomogeneous degrading foils, which, because of the rapid change in fission cross section, could introduce fluctuations in the counting rate. Generally, the errors increased with decreasing counting angle and decreasing energy of the carbon ion. Over the period of the experiments the standard deviation was found to be 3% at 124 Mev and 5% at 73 Mev for the differential cross section at 90 deg. For the differential cross section relative to 90 deg, we have assigned a standard deviation of 5% at 124 Mev, 6% at 95.4 Mev, and 7% at 73 Mev.

The angular distributions in the laboratory system for the three bombarding energies are given in Table I in units of the differential cross section at 90 deg. The distributions were transferred to the coordinate system of the fissioning nucleus (hereinafter called the c.m. system) by trial until a distribution symmetric about 90 deg in the new system was obtained. The transformation was performed by using the parameter $\overline{X}$ defined as

$$(\overline{X} = v_{fN}/v_{fF})$$

where v_{fN} is the mean velocity of fissioning nucleus along the beam axis and v_{fF} the mean velocity of the fission fragment in the c.m. system. The relation between the laboratory-system angle, θ_L, and the c.m. angle, $\theta_{c.m.}$ is given by

$$\tan \theta_L = \frac{\sin \theta_{c.m.}}{\overline{X} + \cos \theta_{c.m.}} .$$

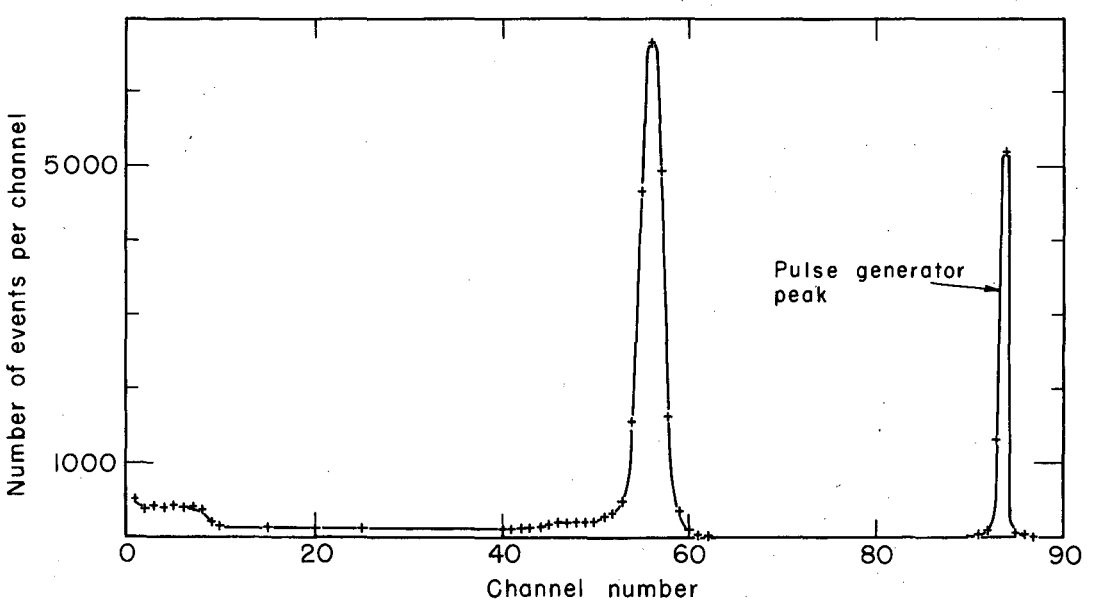

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Fig. 2. A typical spectrum for 73-Mev C^{12} ions scattered from UF_4 at a scattering angle of 25 deg in the laboratory system. Observed with a p-n junction detector reverse-biased by 9 v.

Table I. Differential cross sections (relative to 90 deg) of fission fragments as a function of lab angle, θ_L, in the reaction between U^{238} and C^{12} ions of various energies, E_2.

	$(d\sigma/d\omega)_{\theta_L}/(d\sigma/d\omega)_{90_L}$		
θ_L	E_2 = 73 Mev	95.4 Mev	124 Mev
20		2.15	2.51
30	1.66	1.94	1.92 1.95
35			1.72
40	1.41 1.32	1.55	1.55
50	1.29 1.20	1.39	1.34
60	1.18	1.21	1.26
65			1.15
70	1.03	1.12	1.17
75			1.09
80	1.03		1.09
90	1.00	1.00	1.00
100		1.03	1.00
110	0.98		1.01
120	1.09	1.01	1.01
130	1.14	1.09	1.13
140	1.21	1.21	1.20
145			1.32
150	1.25 1.25	1.30	1.38
160	1.23	1.41	1.46 1.50
162	1.25		
164		1.44	1.55
165			1.57
170	1.36		1.61

The distributions in the c.m. system are shown in Figs. 3, 4, and 5, and the values of $\bar{X}^2$ are listed in Table II.

B. Kinetic Energy Determination

The most probable kinetic energy in the laboratory system, E_L, as a function of lab angle at 73-, 95.4- and 124-Mev C^{12} energies, is shown in Figs. 6, 7, and 8. We have estimated the error in the energy measurements to be of the order of 4%. This involved uncertainty in determination of energy degradation in the target and the detector "window", and fluctuations in the response of the detector. The quoted limit of error attempts to take possible systematic effects into account.

The laboratory energy E_L is related to X_{mp}, the most probable X value, and $E_{c.m.}$, the most probable kinetic energy in the c.m. system, through the equation

$$E_L = E_{c.m.} (1 + X_{mp}^{\ 2} + 2X_{mp} \cos \Theta_{c.m.}), \qquad (6)$$

where $\Theta_{c.m.}$ is defined above.

Equation (6) is valid if $E_{c.m.}$ is a constant independent of $\Theta_{c.m.}$. The values of $E_{c.m.}$ and X_{mp} were adjusted to fit the experimental data. The resulting curves are shown in Figs. 6, 7, and 8. The values of X_{mp} are given in Table II.

The value of $E_{c.m.}$ was found to be 93±3 Mev, independent of bombarding energy. This gives 186±6 Mev for the total kinetic energy release, which is to be compared with 182±5 Mev for the total kinetic energy release for the spontaneous fission of Cf^{252}.[16]

The full width at half maximum (FWHM) of the kinetic energy distribution for single fragments was approximately 40 Mev at all angles and bombarding energies. In the bombardment of gold with carbon ions, the same quantity was found to be 30 Mev. The FWHM for the total kinetic energy release for Cf^{252} has been determined to be 26 Mev.[16]

C. Total Fission Cross Section

The total absolute cross section for fission was determined by a direct comparison of the counting rate of carbon ions in the region of pure Coulomb scattering with the fission counting rate under the assumption of binary fission.

Table II. Various experimental and calculated quantities obtained for fission of U^{238} with C^{12} ions (see text for definition of terms).

E_2 (Mev)	Fissioning nucleus	E (Mev)	$\bar{I}$ ($\hbar$)	f_F (%)	$\bar{\nu}$	$\bar{A}_{fF}$	$\overline{X^2_{CN}}$	$\overline{X^2}$	X^2_{mp}	K^2_0	P_{CN}
73	Cf^{250}	45.7	19.4	26	9.1	120.5	0.018	0.018	0.016	202	1.1
	Cf^{249}	36.0	19.2	20	7.7	120.7				175	1.2
	Cf^{248}	27.4	19.0	30	6.5	120.8		(+.004)[a]	(+.004)[a]	150	1.4
	Cf^{247}	17.4	18.8	12	5.2	120.9				100	2.0
	Cf^{246}	8.5	18.6	12	4.0	121.0				10	5.6
95.4	Cf^{250}	67.0	33.5	28	11.6	119.2	0.024	0.018	0.023	249	2.5
	Cf^{249}	57.3	33.2	25	10.3	119.4				227	2.7
	Cf^{248}	48.7	32.9	22	9.0	119.5		(+.004)[a]	(+.005)[a]	209	2.9
	Cf^{247}	38.7	32.6	16	7.7	119.7				183	3.3
	Cf^{246}	29.8	32.3	8	6.6	119.7				158	3.7
124	Cf^{250}	94.2	45.9	33	14.9	117.6	0.030	0.022	0.023	298	4.0
	Cf^{249}	83.4	45.4	27	13.4	117.8				279	4.2
	Cf^{248}	75.9	45.0	20	12.5	117.8		(+.004)[a]	(+.005)[a]	266	4.3
	Cf^{247}	65.9	44.5	12	11.0	118.0				246	4.5
	Cf^{246}	57.0	44.1	6	9.8	118.1				228	4.8

[a] Upper limit of the error.

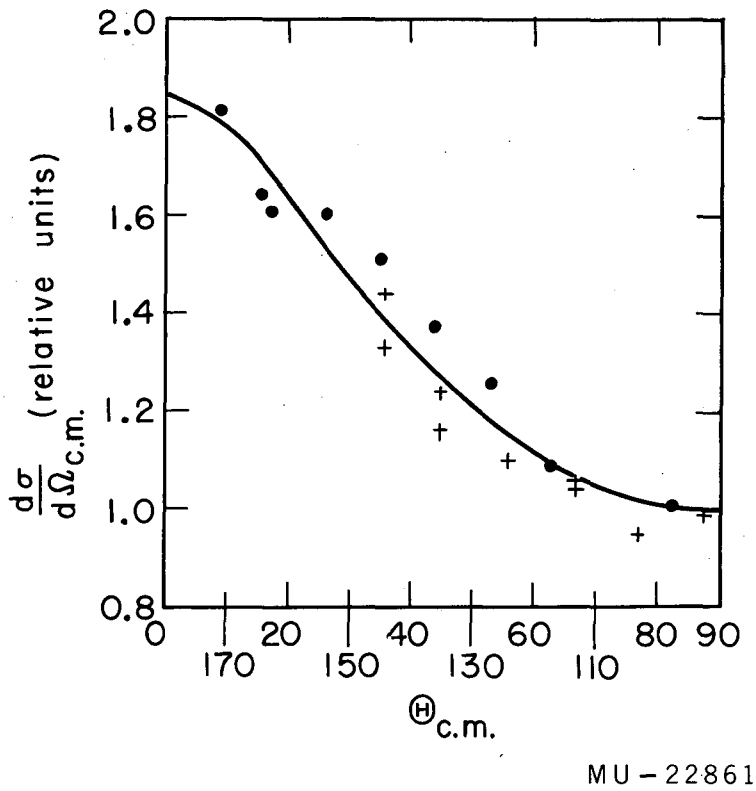

Fig. 3. Differential cross section (in the system of the fissioning nucleus) for fission fragments from the reaction between U^{238} and 73-Mev C^{12}; $X_m^2 = 0.018$. The curve is calculated (see text).

+ - Experimental point observed in forward hemisphere
● - Experimental point observed in backward hemisphere

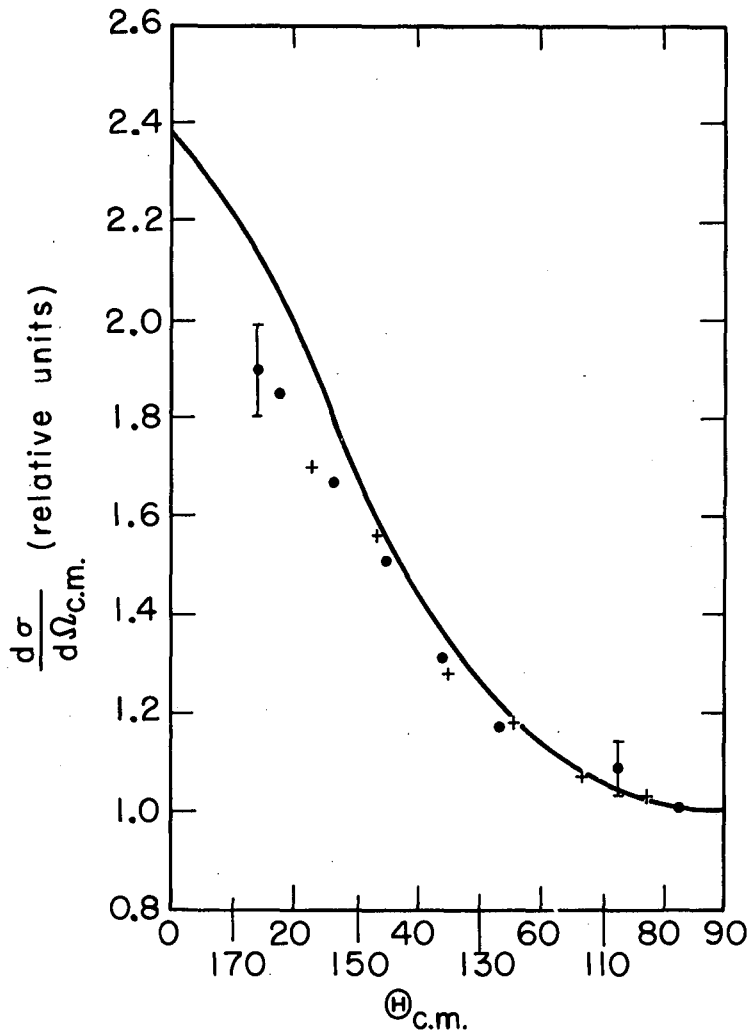

Fig. 4. Differential cross section of fission fragments from the reaction between U^{238} and 95.4-Mev C^{12} ions. The transformation to the system of the fissioning nucleus was performed with $X_m^2 = 0.018$. The curve is theoretical (see text).

+ - Experimental point observed in forward hemisphere
● - Experimental point observed in backward hemisphere

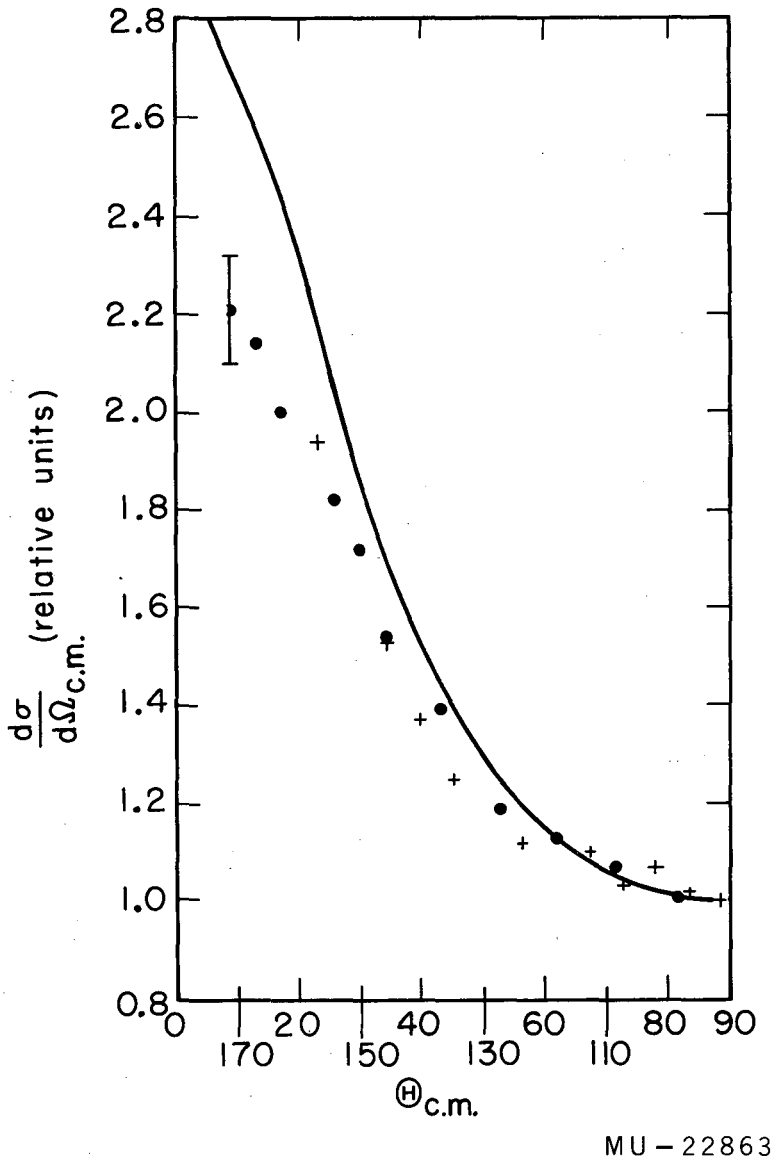

Fig. 5. Differential cross section of fission fragments from the reaction between U^{238} and 124-Mev C^{12} ions. The transformation to the system of the fissioning nucleus was performed with $X_m^2 = 0.022$. The curve is theoretical (see text).

+ - Experimental point observed in forward hemisphere
● - Experimental point observed in backward hemisphere

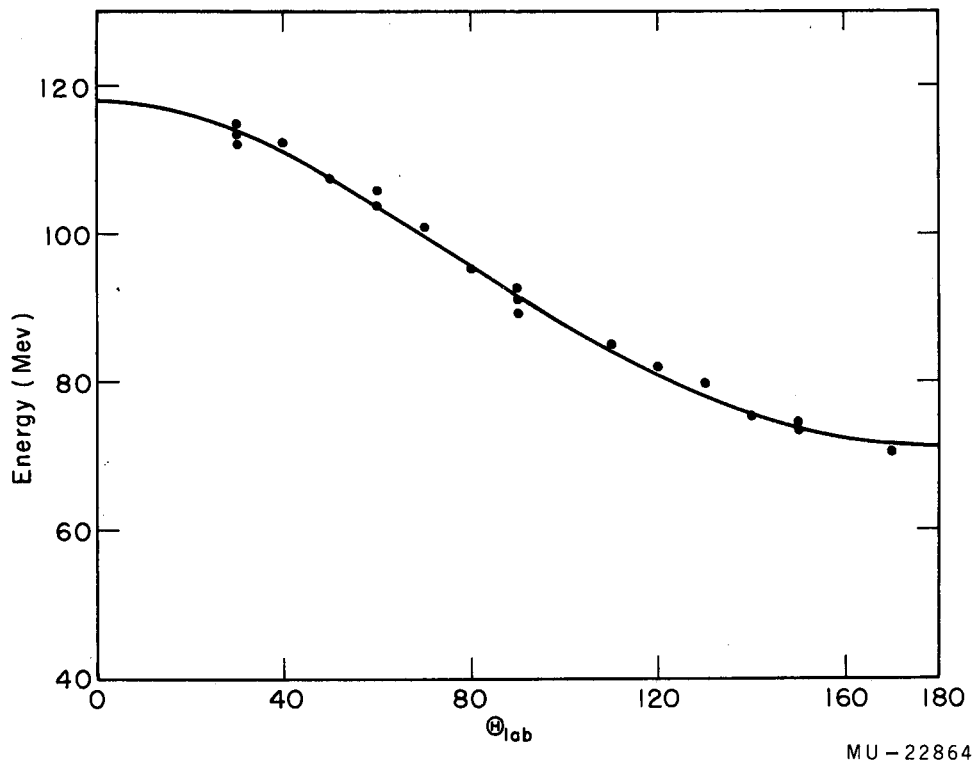

Fig. 6. Most probable kinetic energy of the fission fragments from the reaction between U^{238} and 73-Mev C^{12} as a function of lab angle. The curve is calculated for E_C = 93 Mev and X^2_{mp} = 0.016. The points are experimental.

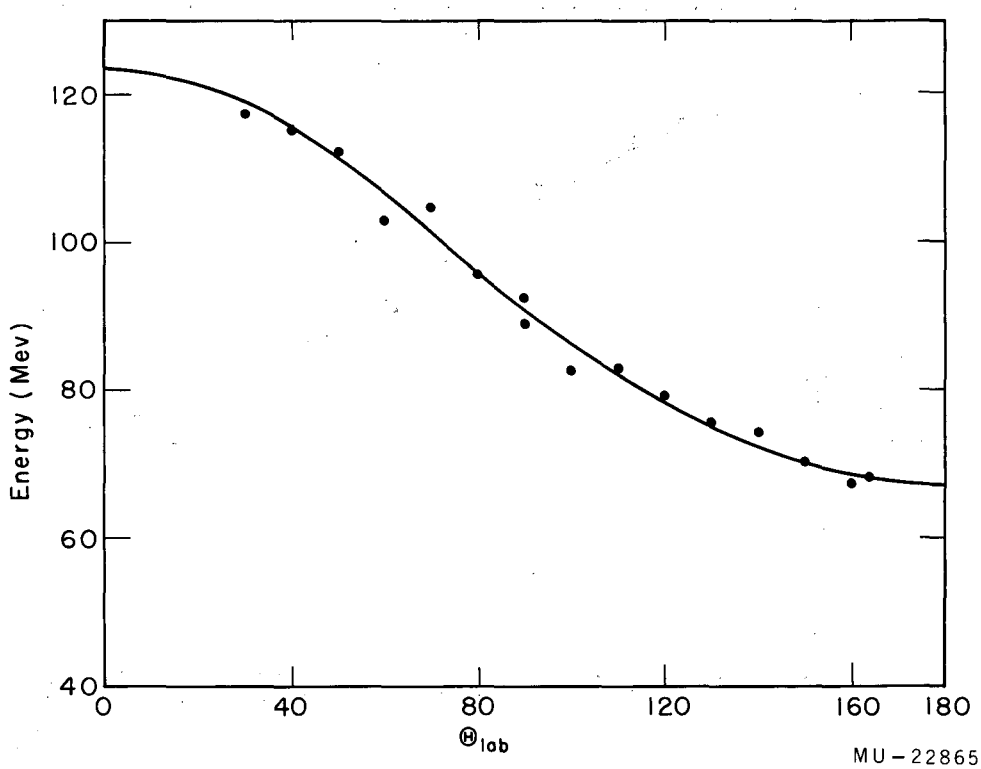

Fig. 7. Most probable kinetic energy of the fission fragments from the reaction between U^{238} and 95.4-Mev C^{12} as a function of lab angle. The curve is calculated for E_C = 93 Mev and X^2_{mp} = 0.023. The points are experimental.

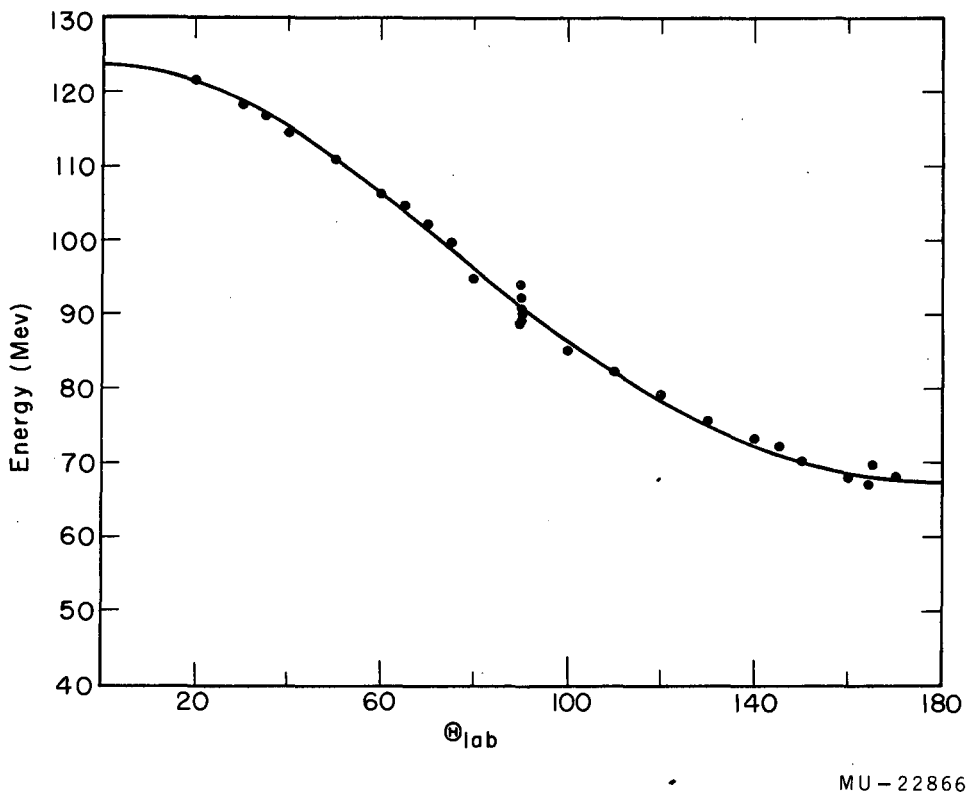

Fig. 8. Most probable kinetic energy of the fission fragments from the reaction between U^{238} and 124-Mev C^{12} as a function of lab angle. The curve is calculated for E_C = 93 Mev and X^2_{mp} = 0.023. The points are experimental.

The differential cross section for elastically scattered carbon ions at 73, 95.4, and 124 Mev was then measured as a function of lab angle. The contribution to the scattering by the nickel backing foil was determined experimentally. The influence on the scattering of the fluorine in the target should be negligible and was ignored. The results were plotted in the c.m. system (relative to the elastic scattering system, not the same as for the fissioning system discussed above), in arbitrary units of the Coulomb scattering cross section. The curves were characterized by a flat portion at low angles followed by a 20% to 30% rise before the sharp drop-off at larger angles. Similar curves have been observed in the scattering of C^{12} by Au, with the nuclear-emulsion technique.[11]

The flat portion of the curve was assumed to represent the region of pure Coulomb scattering from which we evaluated the counting efficiency and target thickness. The values obtained at the two lowest bombarding energies agreed to within 7%. The value at 124 Mev was 20% off.

The Coulomb scattering cross section is proportional to $1/\sin^4(\theta_{c.m.}/2)$ and our largest error, therefore, arises from the uncertainty of 1 deg in the determination of the angle. At 124 Mev the flat portion extends to approximately 30 deg, and a 1-deg error at this angle will give an error of 11% in the efficiency determination. At 73 Mev the drop-off is at 90 deg, yielding a corresponding error of only 4%. For this reason the value obtained at 124 Mev was discarded. The absolute fission cross sections determined were: 0.464 barns at 73 Mev, 1.50 barns at 95.4 Mev, and 2.35 barns at 124 Mev. For all three values we assign a standard deviation of 10%.

At other bombarding energies, the differential fission cross section at 90 deg in the laboratory system was measured, and the absolute fission cross section was estimated from the known efficiencies and by assuming a smooth variation of the integration factor from the angular distribution with bombarding energy. The errors introduced by this method should be negligible because of the small variation of the angular distribution from 73 Mev to 124 Mev. The results are given in Fig. 9. In the same figure is given the theoretical curve for the cross section for compound-nucleus formation as calculated by Thomas[21] from the square-well model, using as radius parameter $r_o = 1.5 \times 10^{-13}$ cm. The agreement is satisfactory.

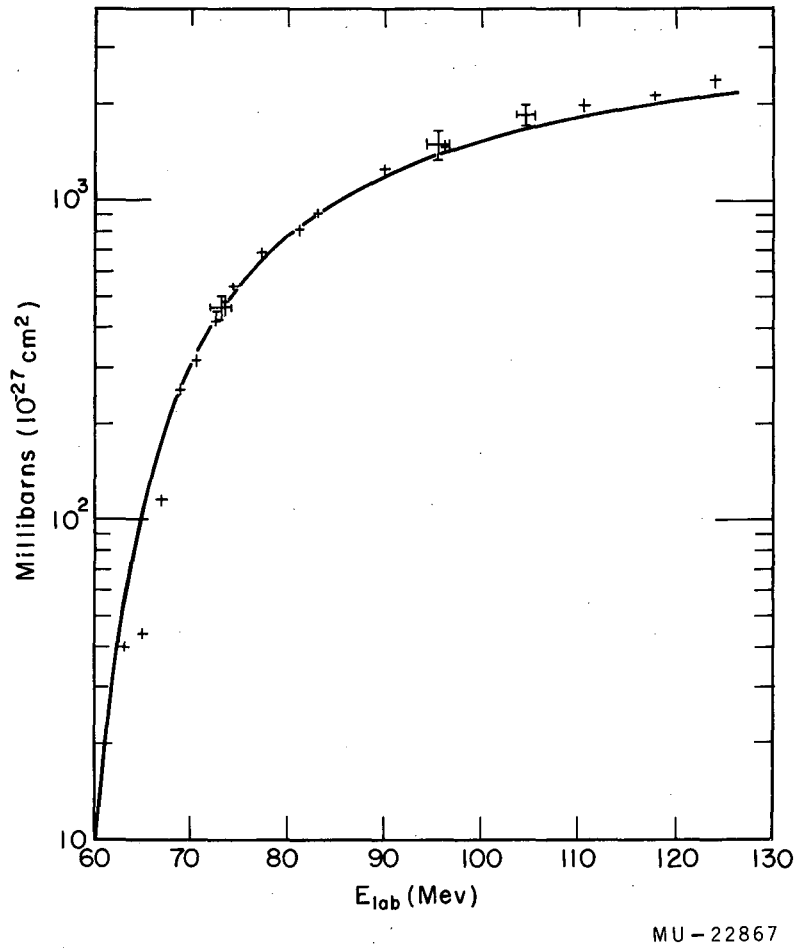

Fig. 9. Cross section for fission of U^{238} with C^{12} ions as a function of C^{12} energy. The curve shows the cross section for formation of the compound nucleus for a square-well model with a radius parameter of 1.5×10^{-13} cm.

IV. DISCUSSION

A. Introduction

In Part B of this section we calculate the over-all fission-fragment angular distributions and values of the quantity X to be expected in the reaction between U^{238} and C^{12} ions if the reactions proceed entirely via the compound-nucleus mechanism. In Part C we compare the results of these calculations with the experimental observations and propose possible explanations for the differences.

B. Compound-Nucleus Calculations

Bohr's original formulation of the concept of a compound-nucleus (CN) reaction implies that the bombarding particle amalgamates with the target nucleus to form a compound nucleus.[22] In so doing, the particle deposits its total linear and angular momenta in the compound nucleus which, after thermal equilibrium is established, decays by evaporation of particles. In the heavy-element region, fission competes with the evaporation of particles.

Monte Carlo calculations performed by Dostrovsky indicate that charged-particle evaporation is negligible compared with neutron evaporation in the reaction between U^{238} and C^{12} at the excitation energies with which we are dealing.[23] We therefore first consider the case of neutron evaporation in competition with fission. The over-all fission-fragment angular distribution will be a combination of the individual distributions from the various fissioning nuclei in the evaporation chain. To the fissioning nuclei at each step in the cascade we have to assign an angular distribution parameter p_{CN}, as defined in Sec. I, and a c.m.-transformation parameter $\overline{X}^2_{CN}$.[24] In order to obtain the combined angular distribution, we must also estimate the fraction f_F of the originally formed compound nuclei that is fissioning at each step in the chain.

1. Estimation of the Angular Distribution

a. Calculation of p_{CN}

In order to evaluate p_{CN} at a given stage in the evaporation chain, one must estimate the excitation energy, E, and the maximum value of the spin, I_m, for the particular fissioning nucleus. The excitation energy of the compound nucleus was computed by using the bombarding energy and the Q of compound-nucleus formation obtained from the mass data given by Glass et al.[25]

Because of barrier-penetration phenomena in compound-nucleus formation, Halpern and Strutinski note that the term I_m^2 is not clearly defined, but may be approximated by $2\langle I^2\rangle$, where $\langle I^2\rangle$ is the average value of the square of the spin of the compound nucleus.[2] In accord with this approximation and the assumptions made in Sec. I, I_m^2 is given by

$$I_m^2 = 2\langle I^2\rangle = 2\langle \ell^2\rangle \quad . \tag{7}$$

Furthermore, for the large ℓ values involved in this work, it may be shown that, to a very good approximation,[26]

$$\langle \ell^2\rangle = \frac{9}{8}(\bar{\ell})^2, \tag{8a}$$

thus

$$I_m^2 = \frac{9}{4}(\bar{\ell})^2 \quad . \tag{8b}$$

Mean values of ℓ, and thus I, were taken from the compound-nucleus-formation calculations by Thomas[21] based on the square-well model with $r_o = 1.5 \times 10^{-13}$ cm. For later isotopes in the evaporation chain, mean values of E were evaluated under the assumption that each evaporated neutron reduces the average excitation energy by the amount $(B_n + 2T)$, where B_n is the neutron binding energy and T is the nuclear temperature. Values for B_n were taken from Ref. 25. The shapes of the excitation functions for neutron-evaporation reactions in this system are consistent with a nuclear temperature equal to 1.5 Mev, independent of excitation energy.[28] With the high excitation energies involved here, the choice of T is not very important and we have used a constant value of 1.5 Mev.

For the change in mean spin along the evaporation chain we assumed, using classical arguments, that a neutron on the average carries off 1% of the spin of the nucleus having a mass of about 250.

Values for E_f, the fission barrier, have been taken from Vandenbosch and Seaborg.[29] K_o^2 was then evaluated from the curve by Halpern and Strutinski.[2] Estimated values for E, ℓ, K_o^2, and the resulting values of p for the various members of the chain involving emission of up to four neutrons at the three

bombarding energies are summarized in Table II. Note that at the two highest bombarding energies the p value has only a small variation along the chain. At 73 Mev, however, the p value, after the evaporation of four neutrons, has increased by a factor of five. This shows that the assignment of a mean fissioning nucleus as a representation for the distribution is not justified in this case.

b. Calculation of f_F

There are several formulas for the calculation of Γ_n/Γ_f, the ratio of the level width for neutron evaporation Γ_n to that for fission Γ_f. Huizenga and Vandenbosch have developed a formula which reproduces quite well the more tedious Monte Carlo calculations and which takes into account the influence of rotational and pairing energies.[30] If the level-density expression $\rho(E) = \text{const} \exp\,[2(aE)^{1/2}]$ is used, Γ_n/Γ_f is given by

$$\frac{\Gamma_n}{\Gamma_f} = \frac{4A^{2/3}(E-B_n' - E_R)}{C_0[2a^{1/2}(E-E_f'-E_R^f)^{1/2}-1]} \exp\{2a^{1/2}[(E-B_n'-E_R)^{1/2}-(E-E_f'-E_R^f)^{1/2}]\}, \qquad (9)$$

where E_R = rotational energy of the undistorted (i.e., spherical) nucleus,

E_F^f = effective rotational energy of the nucleus at the saddle point,

$B_n' = B_n + \Delta$, B_n is neutron binding, Δ is the pairing term for the residual nucleus,

$E_f' = E_f + \Delta_f$, Δ_f is the pairing term for the fissioning nucleus at the saddle point,

E = excitation energy of the nucleus,

$C_0 = \hbar^2/(gmr_0^2)$, r_0 is the nuclear radius parameter, g is the statistical weight for spin (= 2 for neutrons), m is the mass of the neutron.

The mean value of Γ_n/Γ_f at each step in the cascade may be evaluated by proper choice of the parameters. For the constant a in the level density formula we have chosen the value 10 Mev^{-1}. The values of E, E_f, and B_n have been determined above, and Δ and Δ_f were taken from Huizenga and Vandenbosch.[30] The rotational energies are estimated from the formulas $E_R = \hbar^2 I^2/2\mathfrak{J}$ and $E_R^f = \hbar^2 I^2/2\mathfrak{J}_\perp$. As given above, I is the spin and $\mathfrak{J}$ the effective moment of inertia of the spherical nucleus. The quantity $\mathfrak{J}_\perp$

is the moment of inertia of the saddle-point nucleus, taken about an axis perpendicular to the fission axis.

For the moment of inertia, only an order-of-magnitude estimate can be made. For a nucleus of mass number 250, $\hbar^2/2\mathfrak{J}_{rig}$ is about 3 kev, where $\mathfrak{J}_{rig}$ is the rigid-body moment of inertia.[29] We have rather arbitrarily chosen a value of 5 kev for $\hbar^2/2\mathfrak{J}$, as a compromise between the value obtained using $\mathfrak{J}_{rig}$ and those obtained from the spacings of rotational energy levels in the ground-state bands of nuclei in this region.[31] As stated by Huizenga and Vandenbosch,[30] the ratio E_R/E_R^f probably lies between 2.5 and 1.25 in fission induced by heavy ions. In our case, the choice of this ratio is not very critical and we have used the value of 2 in our calculations. The values of f_F obtained from this calculation are listed in Table II.

The over-all angular distribution of fission fragments in the c.m. system predicted by the compound-nucleus-reaction assumptions are shown as curves in Figs. 3, 4, and 5 together with the experimental angular distributions.

2. Estimation of $\overline{X}_{CN}$

The mean value, $\overline{X}_{CN}$, at a certain step in the evaporation chain is given by

$$\overline{X}^2_{CN} = \frac{A_2 E_2 A_{fF}}{A_{CN}^{\ 2}\ E_{c.m.}} , \qquad (10)$$

where A_2 = mass of the C^{12} bombarding particle,

E_2 = kinetic energy of the C^{12} particle in the laboratory system,

A_{CN} = mass of the compound nucleus,

A_{fF} = mean mass of the fission fragments,

and $E_{c.m.}$ = the mean kinetic energy of the single-fragment spectrum.

Because of the high excitation energies at which most of the fission events occur, one would expect the mass and kinetic energy distributions to be symmetric. Under this condition, the mean and most probable values should be identical.

Our observation that $E_{c.m.}$ is independent of bombarding energy has also been observed in the bombardment of gold with carbon ions.[8] The value of 93 Mev seems reasonable for the fission of Cf isotopes. Terrell's correlation predicts a linear dependence of kinetic energy release on

$Z^2/A^{1/3}$ of the fissioning nucleus[32]; however, this effect gives a negligible variation of $E_{c.m.}$ along the evaporation chain. We have therefore accepted the value 93 Mev for $E_{c.m.}$ for all the fissioning nuclei involved in the calculations.

The mean mass of the fission fragments, A_{fF}, is given by $A_{fF} = \frac{1}{2}(A_{fN} - \bar{\nu})$, where $\bar{\nu}$ is the mean number of prompt neutrons emitted in the fission process. These neutrons, evaporated as a result of the large fragment excitation energies, are emitted in the frame of reference of the moving fragments. Leachman[33] and Bondarenko et al.[34] have plotted $\bar{\nu}$ as a function of the excitation energy E of the fissioning nucleus for several systems. They obtain the relationship

$$\bar{\nu} = \bar{\nu}_o + 0.12\ E \quad , \tag{11}$$

where $\bar{\nu}_o$ is presumably the mean number of neutrons that would be emitted with spontaneous fission of the particular species. From a compilation by Huizenga and Vandenbosch[30] we have obtained the following values for $\bar{\nu}_o$: 3.6 for Cf^{250}, 3.4 for Cf^{249}, 3.2 for Cf^{248}, 3.1 for Cf^{247}, and 3.0 for Cf^{246}.

Extension of this relationship into the high spin values and excitation energies we are dealing with is, of course, questionable. There is the possibility that the fission fragments will increase their rotational energies with increasing spin of the fissioning nucleus, resulting in a reduction of the number of emitted neutrons. Charged particles may also be emitted. We will, lacking other information, assume Eq. (11) still valid.

Because of the small variation in A_{fF} along an evaporation chain, a constant value was used at each bombarding energy. The final values of $\bar{X}^2_{CN}$ therefore become equal for all the fissioning nuclei in an evaporation chain. The results of the calculations are presented in Table II

c. Comparison of Compound-Nucleus-Model Predictions With Experimental Data

The agreement between the calculated and experimental values of p (over-all) and X^2 is quite satisfactory at the 73-Mev bombarding energy. At the higher bombarding energies, however, we observe considerable differences in these quantities. At these energies the experimental angular distributions are less anisotropic than predicted and, in general, we find for the X values: $\bar{X}^2_{CN} > \bar{X}^2_{mp} > \bar{X}^2$.

It is interesting to note that the discrepancies cannot be explained on the basis of charged-particle evaporation prior to fission, since predictions based on such an assumption are largely the same as those obtained on the assumption of only neutron evaporation in competition. The $\overline{X}^2_{CN}$ values for charged-particle evaporation are calculated by using Terrell's[32] curve to estimate $E_{c.m.}$ and by choosing reasonable values for the mean energy of the evaporated particle. The mean linear velocity of the fissioning nucleus will not change appreciably in the evaporation, and the reduction of $E_{c.m.}$ will be approximately compensated by the reduction in A_{fF}. Similarly the p values, in charged-particle evaporation, will not differ very much from those obtained for neutron evaporation since the charged particle will carry off more spin, but will reduce the excitation energy more than a neutron. For example, emission of an alpha particle will classically reduce the mean spin of the nucleus by about 4%, but this will be compensated by a 20-Mev reduction in the excitation energy.

There are several possible sources of error in the calculations. For the angular distribution, any uncertainty in the estimation of the number of fissions occurring at each step in the evaporation chain will have little effect (except at 73 Mev), since the p value has a negligible variation along the chain at the higher bombarding energies. Changing the X^2 values for the transformation of the experimental angular distribution will also not appreciably alter the distribution in the c.m. system. The most susceptible points for introduction of errors into the calculation of the p values are in the estimations of K_o^2 and the appropriate values of I_m^2 for the various fissioning nuclei.

1. Uncertainty in K_o^2

In the region where the relation $K_o^2 = \text{const.}\ (E-E_f)^{1/2}$ is assumed to be valid, Halpern and Strutinski used the value of 31.5-Mev$^{-1/2}$ for the constant.[2] We have seen that with this value the angular distribution at 73 Mev is reproduced. If we assume that the reactions leading to fission at higher bombarding energies also are CN reactions, then the constant has to be of the order of 50 Mev $^{-1/2}$ at 95 Mev and 68 Mev$^{-1/2}$ at 124 Mev to give a fit to the experimental data. This implies that K_o^2 also is a function of the spin I of the fissioning nucleus. Going back to the formula $K_o^2 = T\mathfrak{J}_{eff}/\hbar^2$ (Eq. 3) and applying the formula $T = [\frac{1}{a}\ (E-E_f)]^{1/2}$ for the nuclear temperature, we obtain that $\hbar^2/2\mathfrak{J}_{eff}$ is decreasing with increasing I.

With a=10, $\hbar^2/2\mathcal{J}_{eff}$ will be 2.3 kev at 124 Mev ($\bar{I} \approx 45\hbar$), 3.2 kev at 95 Mev ($\bar{I} \approx 33\hbar$), and 5 kev at 73 Mev ($\bar{I} \approx 19\hbar$). In comparison $\hbar^2/2\mathcal{J}_{sph}$ is about 2.4 kev for a heavy spherical nucleus. A decrease in $\hbar^2/2\mathcal{J}_{eff}$ with $\bar{I}$ is in accordance with the predictions that the saddle-point shape will be less distorted with higher spin.[35,36] No quantitative calculations have been made of this effect, however. We will make the rather questionable assumption that the change in $\hbar^2/2\mathcal{J}_{eff}$ with I will be less than what the estimations given above, have indicated. The discrepancies between the calculated angular distributions for CN reactions and the experimental distributions are attributed to an admixture of non-compound-nucleus (NCN) reactions, as will be shown in the following paragraph.

2. Uncertainty in I_m^2

There are two possible sources for uncertainty in the assignments of the average spin (or of I_m^2) of the various fissioning nuclei. On one hand, it has been noted above that one expects Γ_f/Γ_n to increase with the spin value of the nucleus at a given excitation energy. Thus, in the early stages of the evaporation chain, fission will occur with greater probability among the high spin states, leaving a lower spin distribution than we have assumed in the later stages of the chain. This difficulty was pointed out in the interpretation of the fission-fragment angular distribution resulting from bombardment of Au^{197} with C^{12} ions.[8] In that system, the effect may be rather serious; however, with U^{238} and C^{12}, one would not expect a strong effect because (a) essentially all the compound nuclei eventually fission, and (b) the p value obtained is not very sensitive to the stage at which fission occurs. On the other hand, uncertainty may arise in estimating the average spin change that occurs when particles are evaporated. We do not feel that large errors have been made in our treatment of this problem, although the situation is not totally clear.[37]

Apparently the estimated angular distributions for a compound-nucleus mechanism are reasonable. Similarly no large uncertainty in $\bar{X}^2_{CN}$ is to be expected.

That some compound-nucleus reactions must occur in the reaction between U^{238} and C^{12} has been shown by studies of spallation reactions of the type (C^{12},xn).[27,28] We are led to the conclusion that fission in this system results both from compound-nucleus reactions and from

other reactions that, at least at energies above 73 Mev, yield less anisotropic angular distributions and smaller forward velocities than expected for compound-nucleus reactions. If, in the latter type of reactions, particles are "stripped" from the carbon ion and emitted in the forward direction, high orbital angular and linear momenta can be carried off. For example, an alpha particle emitted in a stripping reaction could reduce the spin of the struck nucleus by as much as 20ħ if its kinetic energy were 20 Mev. On the other hand, evaporation of an alpha particle would reduce the spin by only about 2ħ on the average at the maximum bombarding energy. In both cases the reduction of the excitation energy of the nucleus is approximately the same. Indications for such reactions have also been observed by other experimenters. Britt and Quinton have measured the energy spectra and angular distributions of alpha particles and protons emitted in the reactions of Au^{197} and Bi^{209} bombarded with various heavy ions.[37] For both alpha particles and protons, the angular distributions are sharply peaked in the forward direction and relatively flat in the backward hemisphere. Also, the energy spectra of the alpha particles are rather broad and peaked at about 35 Mev at far forward angles, suggesting that these particles are emitted with nearly the full velocity of the heavy-ion beam (10.5 Mev-nucleon). At backward angles, the energy spectra are consistent with evaporation of the alpha particles from the compound nucleus. These results suggest that most of the alpha particles observed in the forward hemisphere result from direct interactions, whereas those found at large angles in the backward hemisphere come almost entirely from evaporation reactions. The various observations by Britt and Quinton have led them to the conclusion that the dominant direct process is the breakup of the projectile. It is also suggested that the breakup is caused by a nuclear interaction rather than a Coulomb breakup process. It is to be expected that a similar type of alpha emission occurs in the reaction between U^{238} and C^{12}.

Ghiorso and Sikkeland showed that products from (HI, xnyp) reactions between a heavy element such as Cm and heavy ions (HI) have a much shorter range than expected if the product nuclei had the total linear momentum of the heavy ion.[27] They found that even with C^{12} energies as low as 80 Mev the contribution from such NCN-type reactions is at least 10% of the total compound-nucleus-reaction cross section. The dominant group of products result from reactions that can be written (C^{12}, 2αxn).

In these reactions, which can be characterized by the emission of two alpha particles, the residual nuclei are left with excitation energies sufficiently high to cause fission. It is reasonable to expect that these reactions occur at the nuclear surface and involve C^{12} ions having the highest possible impact parameters. If we further assume that the alpha particles leave the nucleus with the same impact parameter and velocity as the incoming ion, the spin and excitation energy, and thus the X^2 and p values of the fissioning nucleus, can be evaluated. By adjusting the contributions to the fission cross section from the two types of reactions CN and NCN to fit the experimental values, we find the fraction of the NCN-reaction cross section to be about 30% at 95 Mev and 124 Mev.

By comparison, Britt and Quinton[38] have found that the ratio of the cross section for direct production of alpha particles to the calculated cross section for compound-nucleus formation[21] for Bi^{209} and C^{12} rises from a value of approximately 0.15 at 85 Mev to 0.39 at 126 Mev. A direct comparison between these values and our estimate of 30% NCN reactions is not possible because one does not know the cross sections for direct emission of all other particles (although direct emission of protons in this system is known to be small compared with that for alpha particles[37]), or the frequency with which two alpha particles are emitted in a given event.

Some fission doubtless results from nucleon-transfer reactions. These reactions will result in small linear and orbital angular momentum transfer to the fissioning nucleus. The total cross sections observed for these types of reactions are, however, only of the order of millibarns[39] and should therefore have small effect on the angular distribution of the fission fragments.

V. SUMMARY AND CONCLUSION

The most probable total kinetic energy release in fission of U^{238} with C^{12} is 186 ± 6 Mev, independent of bombarding energy. This is consistent with the fissioning nuclei as predominantly californium isotopes. The broad kinetic energy distributions probably are the result of a broad mass-yield curve for the fission fragments. This latter observation, suggested by radiochemical studies of the fission of U^{238} with 115-Mev N^{14} ions[40] and preliminary work in the U^{238} and C^{12} system[41], is likely the result of the high excitation energies at which fission occurs and the wide

range in forward velocity of the fissioning nuclei.

Values for X^2 and the anisotropies of the angular distributions of the fission fragments have been calculated on the assumption of purely compound-nucleus-reaction mechanisms and compared with the experimental results. The comparison reveals that above 73 Mev we have contributions to the fission cross section from reactions in which particles are emitted preferentially in the forward direction prior to the fission process.[42]

If one assumes the main group of these stripping reactions to be of the type (C^{12}, 2α), an estimate based on rather crude assumptions yields an approximate 30% contribution to the fission cross section from this kind of reaction at 95 Mev and 124 Mev. This value has to be regarded as an upper limit due to the possible increase of K_o^2 with I.

The fission cross sections observed agree well with calculated cross sections for compound-nucleus formation based on the square-well nuclear potential with radius parameter $r_o = 1.5 \times 10^{-13}$ cm. From our observations, it would appear that the calculated cross sections would be more aptly termed the "interaction cross section for reactions leading to deposition of excitation energies of more than 5 Mev."

It is evident that only charged-particle-fission-fragment-coincidence experiments can give a clearer picture of the reactions occurring pior to the fission process.

ACKNOWLEDGMENTS

We acknowledge the contributions from Bruce D. Wilkins of this Laboratory and Dr. John R. Walton, Lawrence Radiation Laboratory (Livermore), in the early stages of this work. Discussions with Dr. John R. Huizenga of Argonne National Laboratory and Dr. Albert Ghiorso and Dr. Stanley G. Thompson of this Laboratory are appreciated. S. Harges and B. Garrett were most helpful in the plotting of numerous curves. We wish to thank Charles A. Corum for the mechanical design of our experimental equipment. The Hilac crew was most helpful between and during bombardments.

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