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THE SURFACE ENERGY OF MULTI-COMPONENT SYSTEMS

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Abstract

Two commonly used non-equivalent ways of defining the surface energy of a multi-component system are discussed, both for a single-phase (isolated) system and for a two-phase system such as nuclei immersed in a neutron gas. For the case of ordinary nuclei, with a relatively small neutron excess, the results are illustrated with the aid of the algebraic Droplet Model. The general case (arbitrary neutron excess, one or two phases) is illustrated by the nuclear Thomas-Fermi model of nucleons interacting by velocity-dependent (Seyler-Blanchard) interactions. The results bring out the subtleties of the surface-energy problem of multi-component systems and stress the need for care in making estimates of the properties of nuclear matter in astrophysical applications.

1. Introduction

For a sufficiently large system, such as a homogenous fluid or a heavy atomic nucleus, the total energy of the system may be approximated as a sum of a volume energy (proportional to the volume V of the system) and a surface-layer energy. (For a recent formulation of this method see ref. [1])). The surface-layer energy is approximately proportional to the area S of the surface bounding the system. The <u>surface-energy coefficient</u> γ (i.e. the surface energy per unit area of a flat surface) may be defined as the following limit:

$$\gamma = \lim_{S/V \to 0} \frac{E - E_{ref}}{S} , \tag{1}$$

where E is the actual energy of the system, E_{ref} is a reference energy taken to represent the volume contribution, and the limit is taken for a large system for which the ratio S/V tends to zero.

It may come as a surprise that, in the case of multi-component systems (such as nuclei consisting of neutrons and protons), there exist two sensible and commonly used definitions of the reference energy E_{ref} which, when inserted in eq.(1), can lead to widely different values of γ. For example, in the case of a model of the nuclear surface of a type used in astrophysical as well as nuclear applications (to be described in Section 6), the two definitions lead to two values of γ whose magnitudes diverge with increasing neutron excess. In the model in question the ratio of the two values of γ reaches 2.7 at the point of "neutron drip" (where the neutrons become

unbound). Beyond this point, when the nucleus becomes immersed in a neutron gas, the ratio of the (interfacial) surface energies increases further, reaching a value of the order of 10 close to the point of protron drip! How can this be and what does it mean Which value of γ should one use in practice, e.g. in astrophysical applications The present paper is concerned with answering these questions.

In section 2 we define the two surface energies, one associated with a reference energy involving the energy per particle in the bulk, the other associated with the chemical potentials of neutrons and protons under bulk conditions (in infinite nuclear matter). We derive a formula relating the difference between the two surface energy coefficients to the neutron excess and the neutron skin thickness. In section 3 we extend the analysis to the interfacial surface energy between the two phases (e.g., a liquid and a vapour). In section 4 we use the algebraic structure of the Droplet Model to illustrate the results in the case of a small neutron excess and a single (liquid) phase. The general result (arbitrary neutron excess and either one or two phases) is illustrated in sections 5 and 6. The former reviews the general formulae needed in a numerical evaluation of the surface energy of a semi-infinite system (and introduces a particularly compact notation). The latter applies this method to a of Thomas-Fermi treatment of a system of nucleons interacting by a velocity-dependent Yukawa interaction (the Seyler-Blanchard interaction). The numerical results are used to verify the formulae of section 2 and to test certain prescriptions for estimating the surface energy of neutron-rich matter in astrophysical applications. Section 7 summarizes the conclusions.

2. The two surface-energy coefficients and their difference

Consider for definiteness a large fluid system consisting of two components. For example, we may think of a large fictitious nucleus consisting of N neutrons and Z uncharged protons, with $N + Z = A$. Assume the nucleus to be sufficiently large, so that a well-defined distinction can be made between the bulk region (where the neutron, proton and total densities are essentially constant and equal to ρ_n, ρ_p and ρ), and a surface region, where the densities fall off to zero. The first definition of γ, which we will refer to as the e-definition, γ_e, is based on subtracting from E in eq.(1) the energy that the total number of particles, A, would have under bulk conditions. Thus, if we denote by e the energy per particle in the bulk, the reference energy is

$$E = eA \quad . \tag{2}$$

In the second definition, γ_μ, one introduces, in place of the energy per particle in the bulk, the chemical potentials μ_n, μ_p of the neutrons and protons and the pressure p associated with the bulk conditions. (These quantities tend to definite limits as the system becomes infinitely large.) The reference energy is now taken as

$$E_\mu = \mu_n N + \mu_p Z - pV \quad , \tag{3}$$

where V is the volume of the system defined by A/ρ. The physical meaning

of E_μ may be appreciated by recalling the thermodynamic relation

$$dE = \mu_n dN + \mu_p dZ - pdV \quad ,$$

which gives the energy increase dE (in the special case of a system at zero temperature) associated with adding dN particles of one kind, dZ of another and increasing the volume by dV. Alternatively, dE may be regarded as the initial energy cost in a process of disassembling a very large system by a removal of dN neutrons, dZ protons and a volume decrease of dV. It follows that E_μ is a reference disassembly energy -- the energy that would be required to disassemble a nucleus completely (by decreasing N, Z, V to zero) <u>if</u> the energy cost continued throughout the process to be the same as at the beginning. The real energy cost to disassemble the nucleus will be different because μ_n, μ_p and p will change as the system becomes smaller and surface effects come into play. Thus, by subtracting from the true energy the reference (disassembly) energy E_μ one again hopes to isolate the surface corrections and, dividing by the area S and going to the limit of an infinite system according to eq.(1), one defines the second kind of surface energy coefficient γ_μ.

Consider now the energy per particle under bulk condition to be a function $e(\rho,\delta)$ of the total density ρ and of the relative neutron excess δ, defined by

$$\delta = \frac{\rho_n - \rho_p}{\rho_n + \rho_p} \quad , \tag{4}$$

so that

$$\rho_n = \frac{1}{2}\rho(1 + \delta) \quad , \tag{5}$$

$$\rho_p = \frac{1}{2}\rho(1 - \delta) \quad . \tag{6}$$

Since the energy density is ρe it follows from the definitions of the chemical potentials that, under bulk conditions,

$$\mu_n \equiv \frac{\partial(\rho e)}{\partial \rho_n} = e + \rho \frac{\partial e}{\partial \rho} + \frac{2\rho_p}{\rho} e'$$

$$= e + \rho \frac{\partial e}{\partial \rho} + (1 - \delta)e' \quad , \tag{7}$$

$$\mu_p \equiv \frac{\partial(\rho e)}{\partial \rho_p} = e + \rho \frac{\partial e}{\partial \rho} - \frac{2\rho_n}{\rho} e'$$

$$= e + \rho \frac{\partial e}{\partial \rho} - (1 + \delta)e' \quad , \tag{8}$$

where e' stands for $\frac{\partial e}{\partial \delta}$. Similarly the definition of the pressure p leads to

$$p \equiv -\frac{\partial e}{\partial(\frac{1}{\rho})} = \rho^2 \frac{\partial e}{\partial \rho} \quad . \tag{9}$$

Introducing the overall neutron excess I, defined by $(N - Z)/(N + Z)$ (which may in general be different, for a finite system, than the neutron excess δ in the bulk), we have

$$N = \frac{1}{2}A(1 + I) \quad , \quad Z = \frac{1}{2}A(1 - I) \quad . \tag{10}$$

Inserting eqs.(7),(8),(9),(10) in eq.(3) we find

$$E_\mu(\delta) = \left[e + \rho \frac{\partial e}{\partial \rho} + (1 - \delta)e'\right]N$$

$$+ \left[e + \rho \frac{\partial e}{\partial \rho} - (1 + \delta)e'\right]Z - \rho \frac{\partial e}{\partial \rho} A$$

$$= \left[e + (1 - \delta)e'\right]N + \left[e - (1 + \delta)e'\right]Z \qquad (11)$$

$$= Ae(\delta) + A(I - \delta)e'(\delta) \qquad (12)$$

or

$$E_\mu(\delta) - E_e(\delta) = A(I - \delta)e'(\delta) \quad . \qquad (13)$$

(We have indicated explicitly the dependences of E_μ, E_e, e, e' on δ, but have suppressed the dependence on ρ, which is immaterial for the present discussion.)

In order to show that the right hand side of eq.(13) is proportional to a surface area, we define effective neutron and proton volumes by

$$V_n \equiv \frac{N}{\rho_n} \quad , \quad V_p \equiv \frac{Z}{\rho_p} \qquad (14)$$

and write the relative difference $\Delta V/V$ as follows:

$$\frac{\Delta V}{V} = \frac{V_n - V_p}{V} = \frac{N/\rho_n}{A/\rho} - \frac{Z/\rho_p}{A/\rho}$$

$$= \frac{N/A}{\rho_n/\rho} - \frac{Z/A}{\rho_p/\rho} = \frac{1 + I}{1 + \delta} - \frac{1 - I}{1 - \delta}$$

$$= \frac{2(I - \delta)}{1 - \delta^2} \quad . \tag{15}$$

Using this equation to express $(I - \delta)$ in terms of $\Delta V/V$ we find

$$E_\mu(\delta) - E_e(\delta) = \frac{A}{2}(1 - \delta^2)e'(\delta)\frac{\Delta V}{V} \quad . \tag{16}$$

In the limit of a very large system, when the surface is essentially flat (and the system may be considered as semi-infinite) the separation between the effective neutron and proton surfaces (the thickness of the "neutron skin") must obviously tend to a constant value, t say, independent of position on the surface. (Any deviation from constancy could depend only on the local curvature of the surface, which tends to zero.) In the limit $S/V \to 0$ the difference $V_n - V_p$ may be written as St so that

$$\frac{St}{V} = \frac{2(I - \delta)}{1 - \delta^2} \tag{17}$$

and

$$E_\mu(\delta) - E_e(\delta) = \frac{1}{2}\rho(1 - \delta^2)e'(\delta)St \quad . \tag{18}$$

It follows that

$$\boxed{\gamma_e - \gamma_\mu = \frac{1}{2}\rho(1-\delta^2)e'(\delta)t} \quad . \tag{19}$$

All the symbols on the right refer to quantities evaluated in the limit $S/V \to 0$, i.e., for semi-infinite matter. Thus the right hand side is, like a surface-energy coefficient, some constant characteristic of the (semi-infinite) matter in question.

Equation (18) itself states that, in the limit, the difference between E_e and E_μ is a quantity proportional to the surface area of the system under consideration — a result not at all obvious from the structure of eqs.(2) and (3).

The relation to each other of the reference energies E_μ and E_e as regards their dependences on I and δ, may be clarified by a graphical construction. Thus fig. 1 shows, first, the quantity $E_e(\delta)$, proportional to the bulk energy per particle $e(\delta)$. This reference energy is, by definition, a function of the bulk neutron excess only and does not know about the actual overall neutron excess I of the system. By contrast, E_μ is a function, $E_\mu(I,\delta)$, of both I and δ. According to eq.(13) it may be obtained by constructing the tangent line to $e(\delta)$ at δ and intersecting it with a vertical line at $\delta = I$. The ordinate corresponding to the intersection point gives the value of E_μ at δ for a nucleus with overall neutron excess I. The resulting curve is sketched in fig. 1 as $E_\mu(I,\delta)$. We note that this curve is stationary at $\delta = I$, with value $E_\mu(I,I)$. This quantity, equal also to $E_e(I)$, i.e. to $e(I)A$, is just the volume energy of a nucleus with $\delta = I$, i.e. with no neutron skin. It may be called the Liquid Drop reference energy $E_{LD}(I)$, shown as a horizontal line in fig. 1.

(In the Liquid Drop model — as distinct from the Droplet Model -- the neutron skin is disregarded and neutrons and protons are assumed to fill one and the same volume V.) Fig. 1 thus shows altogether three reference energies: $E_{LD}(I)$, $E_\mu(I,\delta)$ and $E_e(\delta)$. We note that on account of eq.(17) the deviation $(I - \delta)$ is proportional to the small quantity St/V. We thus see that the difference between E_μ/A and E_e/A is first order in St/V but the deviation between E_μ/A and E_{LD}/A is second order in St/V. Thus up to and including the surface energy term, the reference energies E_μ and E_{LD} are equal. This is somewhat unexpected, since E_μ is defined thrugh eq.(3) in terms of the chemical potentials of bulk nuclear matter with neutron excess δ, whereas E_{LD} knows only about the overall neutron excess I. The stationary nature of the difference between E_μ and E_{LD} is readily verified algebraically using eq.(13). Thus, writing $y = E_\mu(I,\delta) - E_{LD}(I)$, we have

$$y = E_e(\delta) + A(I - \delta)e'(\delta) - E_{LD}$$

$$= A[e(\delta) - e(I) + (I - \delta)e'(\delta)] \quad ,$$

$$\frac{dy}{d\delta} = A(I - \delta)e''(\delta) = 0 \quad \text{for} \quad \delta = I \quad ,$$

$$\frac{d^2y}{d\delta^2} = A[(I - \delta)e'''(\delta) - e''(\delta)]$$

$$= -Ae''(I) \quad \text{for} \quad \delta = I \quad .$$

(This is a negative quantity, so long as e'' is positive.)

Let us summarise by collecting together the definitions of the three reference energies E_{LD}, E_e, E_μ for a large nucleus with A nucleons (N neutrons and Z protons) and characterized by a bulk neutron excess δ (which implies that the nucleus has a neutron skin of thickness t, related to δ and S by eq.(17)).

$$E_{LD} = Ae(I) \tag{20}$$

$$E_e = Ae(\delta) \tag{21}$$

$$E_\mu = \mu_n N + \mu_p Z - pV = e_n(\delta)N + e_p(\delta)Z \quad , \tag{22}$$

where

$$\begin{aligned} e_n(\delta) &= e + (1 - \delta)e' \\ e_p(\delta) &= e - (1 + \delta)e' \quad . \end{aligned} \tag{23}$$

We have learnt that, to the order of the surface energy term, the total energy E may be approximated either as

$$E = E_e + \gamma_e(\delta)S + \text{curvature and higher-order terms} \tag{24}$$

or

$$E = E_\mu + \gamma_\mu(\delta)S + \text{curvature and higher-order terms} \quad . \tag{25}$$

Since we have shown that the difference between E_{LD} and E_μ is of second order in S/V we may also approximate the total energy by

$$E = E_{LD} + \gamma_\mu(\delta)S + \text{higher-order terms} \quad . \tag{26}$$

The values of γ_e and γ_μ are, in general, quite different, but the values of E obtained by any one of the equations 24, 25, 26 are equal (to within higher order corrections) because the reference energies E_e and E_μ (or E_{LD}) differ by precisely the difference between $\gamma_\mu S$ and $\gamma_e S$. Any one of the equations 24, 25, 26 may, therefore, be used in practice. What must <u>not</u> be used are the equations

$$E = E_\mu + \gamma_e(\delta)S \tag{27}$$

$$E = E_e + \gamma_\mu(\delta)S \tag{28}$$

$$E = E_{LD} + \gamma_e(\delta)S \quad . \tag{29}$$

These equations are, in general, grossly incorrect at the level of the surface energy term.

3. Interfacial surface-energy coefficients between two phases

Consider now the case when the two-component system consists of two phases (say a liquid and a vapour). For example, if in our fictitious nucleus the neutron excess became large enough, the neutrons would become unbound and, if confined by external forces, would form a vapour surrounding the liquid phase. In general, let us consider N neutrons and Z protons in a confining volume V and distributed in two phases (conventionally referred to as the "inside" and "outside" phases), with bulk densities ρ_{ni}, ρ_{pi}, $\rho_i = (\rho_{ni} + \rho_{pi})$ and ρ_{no}, ρ_{po}, $\rho_o = (\rho_{no} + \rho_{po})$. The effective inside and outside volumes for the neutrons, protons and nucleons (denoted by V_{ni}, V_{no}, V_{pi}, V_{po}, V_i, V_o) as well as the numbers of particles (denoted by N_i, N_o, Z_i, Z_o, A_i, A_o) that these volumes would contain if filled with matter at the bulk densities are determined by four equations for each kind of particle:

$$V_{ni} \equiv N_i/\rho_{ni} \quad , \quad V_{pi} \equiv Z_i/\rho_{pi} \quad , \quad V_i \equiv A_i/\rho_i \tag{30}$$

$$V_{no} \equiv N_o/\rho_{no} \quad , \quad V_{po} \equiv Z_o/\rho_{po} \quad , \quad V_o \equiv A_o/\rho_o \tag{31}$$

$$V_{ni} + V_{no} = V \quad , \quad V_{pi} + V_{po} = V \quad , \quad V_i + V_o = V \tag{32}$$

$$N_i + N_o = N \quad , \quad Z_i + Z_o = Z \quad , \quad A_i + A_o = A \quad . \tag{33}$$

Solving for the volumes and particle numbers in terms of the parameters of the problem, taken to be N, Z, V, ρ_{ni}, ρ_{no}, ρ_{pi}, ρ_{po}, we find

$$V_{ni} = \frac{-V\rho_{no} + N}{\rho_{ni} - \rho_{no}} \quad , \quad V_{pi} = \frac{-V\rho_{po} + Z}{\rho_{pi} - \rho_{po}} \quad , \quad V_i = \frac{-V\rho_o + A}{\rho_i - \rho_o} \tag{34}$$

$$V_{no} = \frac{V\rho_{ni} - N}{\rho_{ni} - \rho_{no}} \quad , \quad V_{po} = \frac{V\rho_{pi} - Z}{\rho_{pi} - \rho_{po}} \quad , \quad V_o = \frac{V\rho_i - A}{\rho_i - \rho_o} \tag{35}$$

$$N_i = V_{ni}\rho_{ni} \quad , \quad Z_i = V_{pi}\rho_{pi} \quad , \quad A_i = V_i\rho_i \tag{36}$$

$$N_o = V_{no}\rho_{no} \quad , \quad Z_o = V_{po}\rho_{po} \quad , \quad A_o = V_o\rho_o \quad . \tag{37}$$

Note that the "insideness" and "outsideness" is defined with respect to three different effective surfaces for the three kinds of particles (neutrons, protons and nucleons) so that, somewhat unexpectedly, $N_i + Z_i \neq A_i$ and $N_o + Z_o \neq A_o$, in general.

Interfacial surface-energy coefficients between the two phases may again be defined by eq.(1), with reference energies E_e and E_μ taken to be

$$E_e = e_i A_i + e_o A_o \tag{38}$$

and

$$\begin{aligned} E_\mu = {} & \mu_{ni}N_i + \mu_{pi}Z_i - p_i V_i \\ & + \mu_{no}N_o + \mu_{po}Z_o - p_o V_o \quad . \end{aligned} \tag{39}$$

In eq.(38) e_i is the energy per particle, $e(\rho_i,\delta_i)$, in the bulk region of the inside phase and e_o is the energy per particle, $e(\rho_o,\delta_o)$, in the bulk of the outside phase. In eq.(39) μ_{ni}, μ_{pi}, p_i are the chemical potentials and the pressure for an infinite medium characterised by densities ρ_{ni} and ρ_{pi}. Similarly for μ_{no}, μ_{po}, p_o. (For a system in equilibrium we would have $\mu_{ni} = \mu_{no}$, $\mu_{pi} = \mu_{po}$, $p_i = p_o$ but, in what follows, we do not need to specialize to systems where the two phases are in equilibrium.)

Using once again the definitions of the chemical potentials and the pressure we find

$$
\begin{aligned}
E_\mu - E_e = {} & \left[e_i + \rho_i \frac{\partial e_i}{\partial \rho_i} + (1-\delta_i)e_i'\right]N_i + \left[e_o + \rho_o \frac{\partial e_o}{\partial \rho_o} + (1-\delta_o)e_o'\right]N_o \\
& + \left[e_i + \rho_i \frac{\partial e_i}{\partial \rho_i} - (1+\delta_i)e_i'\right]Z_i + \left[e_o + \rho_o \frac{\partial e_o}{\partial \rho_o} - (1+\delta_o)e_o'\right]Z_o \\
& - \rho_i \frac{\partial e_i}{\partial \rho_i} A_i - \rho_o \frac{\partial e_o}{\partial \rho_o} A_o - e_i A_i - e_o A_o \\
= {} & \left(N_i + Z_i - A_i\right)\left(e_i + \rho_i \frac{\partial e_i}{\partial \rho_i}\right) + \left(N_o + Z_o - A_o\right)\left(e_o + \rho_o \frac{\partial e_o}{\partial \rho_o}\right) \\
& + e_i'[(1-\delta_i)N_i - (1+\delta_i)Z_i] + e_o'[(1-\delta_o)N_o - (1+\delta_o)Z_o] \; . \quad (40)
\end{aligned}
$$

In order to demonstrate that, as before, the right hand side of eq.(40) is proportional to the surface area separating the two phases, we have to express the particle number differences in that equation in terms of the bulk densities and the neutron skin volume. With this in mind, we denote by

ΔV_n the difference between the effective volumes occupied by the neutrons and by the nucleons of the inside phase, i.e., $\Delta V_n = V_{ni} - V_i$. Similarly we write $\Delta V_p = V_i - V_{pi}$, so that

$$\Delta V_n + \Delta V_p = St \quad , \tag{41}$$

where S is the area of the system and t is the neutron skin, as before. Using eqs.(34)-(37) we may verify that

$$\frac{\Delta V_n}{\Delta V_p} = \frac{\Delta\rho_p}{\Delta\rho_n} \quad , \tag{42}$$

where $\Delta\rho_n = \rho_{ni} - \rho_{no}$, $\Delta\rho_p = \rho_{pi} - \rho_{po}$ are the neutron and proton density differences between the inside and outside phases. Solving eqs.(41),(42) for ΔV_n, ΔV_p, we find

$$\Delta V_n = \frac{1}{2}\, St\left[\frac{\rho_i}{\rho_i - \rho_o}(1 - \delta_i) - \frac{\rho_o}{\rho_i - \rho_o}(1 - \delta_o)\right] \tag{43}$$

$$\Delta V_p = \frac{1}{2}\, St\left[\frac{\rho_i}{\rho_i - \rho_o}(1 + \delta_i) - \frac{\rho_o}{\rho_i - \rho_o}(1 + \delta_o)\right] \quad . \tag{44}$$

Writing $N_i = \rho_{ni}(V_i + \Delta V_n)$, $Z_i = \rho_{pi}(V_i - \Delta V_p)$, $A_i = \rho_i V_i$, $N_o = \rho_{no}(V_o - \Delta V_n)$, $Z_o = \rho_{po}(V_o + \Delta V_p)$, $A_o = \rho_o V_o$ and substituting in eq.(40) we find

$$E_\mu - E_e = \frac{1}{2}\, St\left\{e_i'\rho_i(1 - \delta_i^2) - e_o'\rho_o(1 - \delta_o^2) + \frac{\rho_i\rho_o(\delta_o - \delta_i)}{\rho_i - \rho_o}\left[e_i + \rho_i\frac{\partial e_i}{\partial\rho_i} - e_o - \rho_o\frac{\partial e_o}{\partial\rho_o}\right]\right\} \quad . \tag{45}$$

It follows that

$$\boxed{\gamma_e - \gamma_\mu = \frac{1}{2}\ t\left\{e_i'\rho_i(1-\delta_i^2) - e_o'\rho_o(1-\delta_o^2) + \frac{\rho_i\rho_o(\delta_o-\delta_i)}{\rho_i-\rho_o}\left[e_i + \rho_i\frac{\partial e_i}{\partial\rho_i} - e_o - \rho_o\frac{\partial e_o}{\partial\rho_o}\right]\right\}} \quad . \tag{46}$$

This is the generalized expression for the difference between the two kinds of interfacial surface-energy coefficients in terms of quantities referring to the bulk properties of the two phases. When the outside phase is absent ($\rho_o = 0$) eqs.(45),(46) reduce to eqs.(18),(19).

4. Case of small neutron excess δ and the Droplet Model

The nuclear Droplet Model[2]) analyzes the behaviour of nuclear sizes and binding energies (including the dependence of the surface energy on neutron excess for small values of δ) and it may be used to illustrate the general discussion given in the previous sections.

For purposes of illustration let us consider an uncharged spherical nucleus with radius R, neutron excess δ in the bulk and a neutron skin t. The part of the Droplet Model energy expressly relevant to our discussion is

$$E(\delta) = (-a_1 + J\delta^2)A + (a_2 + H\tau^2 + 2P\tau\delta - G\delta^2)A^{2/3} + \text{higher-order terms} \quad , \quad (47)$$

where $\tau = t/r_0$ and $r_0 = R/A^{1/3}$ is the nuclear radius constant, a_1 is the volume binding energy per particle, J is the volume symmetry energy coefficient, a_2 is the surface energy coefficient for standard semi-infinite nuclear matter and H,P,G are coefficients describing the response of this surface energy coefficient to small deviations of δ and τ from zero. [In order to isolate the features of the Droplet Model relevant for the present discussion we focus attention on energy terms that are left over after effects of compressibility (including spacial redistribution of densities) have been removed. Thus δ in eq.(47) stands for the average value of the neutron excess in the bulk (denoted by $\bar{\delta}$ in ref. [2])) and R stands for the radius before the (slight) relaxation to its equilibrium value. The additional terms in the energy expression which have to do with

compressibility (and redistribution) are (for small compressibilities) uncoupled from the terms we keep and need not be displayed. The same is true of other additional refinements of the Droplet Model (curvature, diffuseness and exchange corrections).]

For a finite nucleus, δ and τ are not independent, but are related by the "geometrical" condition

$$I - \delta = \frac{3}{2}\tau A^{-1/3} \quad . \tag{48}$$

(This relation follows from eq.(15) by writing $\frac{\Delta V}{V} = \frac{4\pi R^2 t}{\frac{4}{3}\pi R^3} = 3\tau A^{-1/3}$ and neglecting δ^2 compared to unity.)

Minimizing $E(\delta)$ with respect to δ (under the restriction given by eq.(48)) one readily finds the equilibrium value, δ_1, of the bulk neutron excess and the associated binding $E(\delta_1)$:

$$\delta_1 = I(1 - kA^{-1/3} + \text{higher powers of } A^{-1/3}) \quad , \tag{50}$$

$$E(\delta_1) = E_e + (a_2 + kJ\delta_1^2)A^{2/3} + \ldots\ldots \quad , \tag{51}$$

where

$$E_e = (-a_1 + J\delta_1^2)A \quad , \quad k = \frac{3}{4H}(3J - 2P) \quad . \tag{52}$$

[Most of the literature on the Droplet Model[2]) uses in place of k the parameter Q defined by

$$Q = H/(1 - \frac{2}{3}\frac{P}{J}) \quad , \tag{53}$$

so that the relation between k and Q is

$$k = \frac{9}{4}\frac{J}{Q} \quad . \Big] \tag{54}$$

From eq.(50) it follows that the equilibrium skin thickness τ_1 is given by

$$\tau_1 = \frac{2}{3} k\delta_1 \tag{55}$$

and the difference between I and δ_1 is

$$I - \delta_1 = IkA^{-1/3} \quad . \tag{56}$$

Applying eq.(13) to the present case we find

$$E_\mu - E_e \approx A(I - \delta_1) \cdot 2J\delta_1 \quad ,$$

$$= 2kJI\delta_1 A^{2/3} \quad ,$$

or

$$E_\mu - E_e = 2kJ\delta_1^2 A^{2/3} \tag{57}$$

to leading order in δ_1.

Hence, eq.(51) may be rewritten as

$$E(\delta_1) = E_\mu + (a_2 - kJ\delta_1^2)A^{2/3} \quad . \tag{58}$$

Eqs.(51),(58) illustrate explicitly how the two surface energy terms $(a_2 + kJ\delta_1^2)$ and $(a_2 - kJ\delta_1^2)$, diverge linearly with increasing δ_1^2 (for small δ_1). The magnitude of the difference, $2kJ\delta_1^2$, is predicted on general grounds by eq.(57). The fact that the average of the two values is constant (to order δ_1^2) is a more specific prediction of the Droplet Model treatment.

5. Calculating the surface-energy coefficients

In the three preceding sections we have discussed γ_e and γ_μ and the general relations that exist between them, first for an isolated system, then for a two phase system and finally for two component systems with a small neutron excess that can be treated with the Droplet Model. In this section we present the expressions for calculating the actual values of the surface energy coefficients and then, in the next section, we apply these expressions to a specific model.

In ref. [3]) Gibbs' expression for the surface energy coefficient, which corresponds to our γ_e, is given as [4]),

$$\gamma_e = \int_{-\infty}^{+\infty} \varepsilon(x)dx - \left[\varepsilon_1 \int_{-\infty}^{L_\rho} dx + \varepsilon_2 \int_{L_\rho}^{+\infty} dx\right] , \qquad (59)$$

where x is an integration variable along a normal to the surface (assumed plane), $\varepsilon(x)$ is the local energy density, ε_1 and ε_2 are the asymptotic values of the energy density inside and outside the surface, and L_ρ is the surface location of the density distribution.

The notion of the surface location L is made quantitative in this expression, and in others to follow, by the definition

$$\lim_{\substack{x_1 \to -\infty \\ x_2 \to +\infty}} \left[\int_{x_1}^{x_2} f(x)dx = f_1 \int_{x_1}^{L} dx + f_2 \int_{L}^{x_2} dx\right] , \qquad (60)$$

which simply states that "For any function $f(x)$ describing a surface region, consisting of a smooth, localized transition from one constant asymptotic value f_1 to another f_2, the value of the surface location L is determined by the location of a step function having the same integral (the same number of particles if a density distribution is being considered) and the same asymptotic values." Another more compact definition (whose equivalence to eq.(6) may be demonstrated by partial integration) is due to Süssman[6]):

$$L = \int_{-\infty}^{+\infty} x\, g(x)dx \quad , \tag{61}$$

where

$$g(x) = \frac{df(x)}{dx} \Big/ \int_{-\infty}^{\infty} \left(\frac{df(x)}{dx}\right) dx \quad , \tag{62}$$

Surfaces can be further characterized by the distance over which the transition takes place (their width) by the quantity b,[6]) where,

$$b^2 = \int_{-\infty}^{\infty} g(x)(x - L)^2 dx \quad . \tag{63}$$

For a two component system like nuclear matter the relationships between the location and width of the total density and the locations and widths of the separate components is given by:

$$L_\rho = (L_n \Delta\rho_n + L_p \Delta\rho_p)/\Delta\rho \quad , \tag{64}$$

$$b^2 = b_n^2 \left(\frac{\Delta\rho_n}{\Delta\rho}\right) + b_p^2 \left(\frac{\Delta\rho_p}{\Delta\rho}\right) + \Delta L^2 \frac{\Delta\rho_n \Delta\rho_p}{(\Delta\rho)^2} \quad , \tag{65}$$

where $\Delta\rho = (\rho_1 - \rho_2)$, $\Delta\rho_n = (\rho_{n1} - \rho_{n2})$ and $\Delta\rho_p = (\rho_{p1} - \rho_{p2})$.

Of course, the definitions (61) and (62) are completely general, and need not be limited to the characterization of only matter surfaces. Indeed, any quantity that undergoes a smooth, localized transition between two constant asymptotic values can be considered in this way. For example, we can use this procedure to define L_ε, the location of the surface of the local energy density function. If the integrals inside the brackets in eq.(59) are then rewritten so the quantity L_ε appears in the limits of integration in the following way:

$$\gamma_e = \int_{-\infty}^{+\infty} \varepsilon(x)dx - \left[\varepsilon_1 \int_{-\infty}^{L_\varepsilon} dx + \varepsilon_1 \int_{L_\varepsilon}^{L_\rho} dx + \varepsilon_2 \int_{L_\rho}^{L_\varepsilon} dx + \varepsilon_2 \int_{L_\varepsilon}^{+\infty} dx\right], \tag{66}$$

then eq.(60) can be used to rewrite this expression as,

$$\gamma_e = (\varepsilon_1 - \varepsilon_2)(L_\varepsilon - L_\rho) \quad . \tag{67}$$

Alternatively eq.(66) can be transformed by an integration by parts to yield

$$\gamma_e = (\varepsilon_1 - \varepsilon_2) \int_{-\infty}^{\infty} \left[\frac{(\varepsilon - \varepsilon_2)}{(\varepsilon_1 - \varepsilon_2)} - \frac{(\rho - \rho_2)}{(\rho_1 - \rho_2)}\right] dx \quad , \tag{68}$$

which becomes the familiar expression [5]),

$$\gamma_e = \varepsilon_1 \int_{-\infty}^{\infty} \left[\frac{\varepsilon}{\varepsilon_1} - \frac{\rho}{\rho_1}\right] dx \quad , \tag{69}$$

for isolated surfaces where ρ_2 and ε_2 are zero.

The alternative form for the surface energy γ_μ, which results from using a reference energy based on chemical potentials (eq.(3)) can be written

$$\gamma_\mu = \int_{-\infty}^{\infty} \varepsilon(x)dx - \left[\sum_i \mu_i \left(\rho_{i1} \int_{-\infty}^{L_i} dx + \rho_{i2} \int_{L_i}^{\infty} dx\right) - p \int_{-\infty}^{\infty} dx\right], \tag{70}$$

where the summation over i represents a sum over the separate individual components of the system (neutrons and protons in the case of nuclei), and L_i stands for the location of the surface of the i-th component. If use is made of the identity

$$\varepsilon = \sum_i \mu_i \rho_i - p \tag{71}$$

and the same procedure is followed that links eqs.(59) and (67), then (70) becomes

$$\gamma_\mu = \sum_i \mu_i (\rho_{i1} - \rho_{i2})(L_\varepsilon - L_i) \quad . \tag{72}$$

Equations (67) and (72) are the most compact general definitions of the two different surface energies.

6. Thomas-Fermi calculations

The phenomenological two-body interaction of Seyler and Blanchard[7]) has been employed in ref. [2]) for investigating the general properties of saturating two component systems. Later, von Groote[8]) adjusted the parameters of the two-body interaction to give a good fit to nuclear masses and sizes. He also continued the investigation of neutron excess systems to cover the case we are interested in here, where neutrons become unbound and the nuclei are immersed in a neutron gas. In ref. [9]) Kolehmainen also undertook a major study of such systems that compares the results for a wide variety of Skyrme forces. A similar study for nuclei at finite temperatures and in equilibrium with their associated vapor was performed by Küpper[10]).

We have repeated von Groote's calculations here (and corrected some small errors in the calculation of the surface location and surface energy) and we have not only calculated γ_e, as he did, but we also calculated γ_μ for comparison with the work of other authors[11]).

Refs. [2,8]) can be consulted for the details of these calculations but, for the sake of completeness, we list below the values of the parameters of the Seyler-Blanchard interaction. The interaction between two nucleons with separation r and relative momentum of magnitude p is written as

$$V(r,p) = C_{\ell,u} e^{-r/a}[1 - (p/b)^2] \tag{73}$$

and we have used

$$\begin{aligned} C &= 354.069 \text{ MeV} \\ C_u &= 516.638 \text{ MeV} \\ a &= 0.557 \text{ fm} \\ b &= 409.456 \text{ MeV/c} \quad . \end{aligned} \tag{74}$$

These values lead to the following nuclear properties:

$$\begin{aligned} &\text{nuclear radius constant} && r_0 = 1.159 \text{ fm} && , \\ &\text{volume energy} && a_1 = 16.1 \text{ MeV} && , \\ &\text{symmetry energy} && J = 34.0 \text{ MeV} && , \\ &\text{surface energy} && a_2 = 18.011 \text{ MeV} && , \\ &\text{surface symmetry energy} && Q = 28.66 && . \end{aligned} \tag{75}$$

From our earlier definition of k by eq.(54) in section 4 we see that it has the value

$$k = 2.67 \quad . \tag{76}$$

As the asymptotic neutron excess δ increases in the semi-infinite half space to the left of a plane boundary surface, the neutron and proton density distributions were found to evolve in the way shown in fig. 2. At the top of the figure the calculated density distributions are shown for the case of $\delta = 0$. The surface profiles are the same for neutrons and protons and the surfaces are both located at the same point. The second part of the figure is for the case $\delta = 0.2$, where the surface profiles have begun to differ and

the locations of the surfaces (shown by the superimposed vertical bars) have moved apart by about 0.46 fm. Neutron drip occurs at $\delta = 0.30475$, and there is already a substantial outside neutron density shown in the next part of the figure, which is for $\delta = 0.4$. At the bottom of fig. 2 the density distributions for $\delta = 0.6$ are shown.

In fig. 3 the neutron skin thickness for these model calculations (defined here as $t = L_n - L_p$) is plotted against the bulk asymmetry δ. We note the striking appearance of a relatively sharp maximum in this quantity at $\delta \approx 0.4$ (Compare ref. [8].) Similar, but less pronounced maxima are found in ref. [9]. For small δ values the calculated result agrees exactly with the straight line which represents eq.(55), the Droplet Model expression relating τ and δ.

In fig. 4 the separate surface widths of the neutron and proton density distributions are plotted against the bulk asymmetry δ. Once again we note the occurrence of a relatively sharp maximum in the diffuseness of the neutron distribution. The widths of both the neutron and proton distributions are increasing steeply as the point of proton drip $\delta = 0.67793$ is approached, but the trend of the numerical values (the last one at $\delta = 0.677$) does not seem to indicate an actual singularity.

In fig. 5 the surface energy coefficients γ_e and γ_μ are plotted as the upper and lower solid lines. They are plotted against δ^2 instead of δ because the Droplet Model predicts a δ^2 dependence for small δ values, and the two thin straight lines show the Droplet Model predictions corresponding to eqs.(51) and (58) which are

$$\gamma_e = (a_2 + kJ\delta^2)/(4\pi r_0^2) \quad , \tag{78}$$

$$\gamma_\mu = (a_2 - kJ\delta^2)/4\pi r_0^2) \quad . \tag{79}$$

Here, a maximum in the quantity γ_e was to be expected since the initial behavior is an increase proportional to δ^2, but the value of γ_e is expected eventually to decrease for larger δ because the distinction between the matter inside the surface and the matter outside is then disappearing.

The difference between γ_e and γ_μ in fig. 5 was verified numerically to be in perfect agreement (to several significant figures) with eqs.(46) and (19). The great disparity between γ_e and γ_μ for larger values of δ underlines the absolute necessity of properly matching the surface energy with the associated volume (reference) energy according to eqs.(24)-(26).)

Another aspect of the subtleties of the surface-energy problem is illustrated by the dashed and dot-dashed curves. They test, for the present Thomas-Fermi system with Seyler-Blanchard interactions, two approximations to the surface energy coefficient γ that have appeared in the literature. (Since in those treatments the presence of a neutron skin is disregarded from the beginning, it is not possible to decide unambiguously whether the approximation is meant to represent γ_e or γ_μ.) The dot-dashed line corresponds to applying the prescription suggested by Mackie and Baym[3])

$$\gamma_{MB} = \gamma_0(\Delta\rho/\rho_0)^2(-\Delta e/e_0)^{1/2} \quad , \tag{80}$$

where γ_0 is the value of the surface tension for $\delta_1 = 0$, and ρ_0 and e_0 are the density and energy per particle of infinite (symmetric) nuclear matter. The quantities $\Delta\rho$ and Δe are the differences between the inside and outside values of the density and energy per particle when $\delta_1 \neq 0$. A similar expression was derived earlier by Baym, Bethe and Pethick[12])

$$\gamma_{BBP} = \gamma_0(\Delta\rho/\rho_0)^{3/2}(-\Delta e/e_0)^{1/2} \quad , \tag{81}$$

but was rejected by its authors on physical grounds. The dashed line corresponds to the ad hoc expression,

$$\gamma_{BBP} = \gamma_0(\Delta\rho/\rho_0)^{2/3}(-\Delta e/e_0) \quad , \tag{82}$$

that they chose to use instead of (81). From the content of refs. [3]) and [12]) it is not possible to say whether the above authors intended to approximate γ_e or γ_μ, since these two quantities coalesce if the neutron skin thickness is restricted to be zero (as was done in the above references). The relation of eqs.(80) and (82) to the actual surface energy coefficients is shown in fig. 5. We may note in passing that all the prescriptions, eqs.(80)-(82), fail to reproduce the small-δ behaviour required by the Droplet Model and, in addition, they violate an established theorem[2]) that the surface energy coefficient for $\delta = 0$ is stationary with respect to deviations of the asymptotic (bulk) density from its equilibrium value. (This failing probably explains why the nuclear matter radius constant determined in ref. [3]) is slightly smaller than that found by other authors.)

7. Conclusions

We have discussed the problem of calculating the surface-energy coefficient of multi-component systems, in particular the need to match properly the definition of the surface energy to the definition of the volume energy. The subtleties of the problem have been illustrated by the Drolet Model and by the nuclear Thomas-Fermi model and suggest that there may be important questions left open in current astrophysical applications, in particular regarding supernova collapse and explosion. We intend to pursue this in a future publication.

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Figure captions

Fig. 1. The reference energies (E_e, E_{LD} and E_μ), divided by A, for a hypothetical nucleus having $I = 0.25$ are plotted against the bulk value of the nuclear asymmetry δ. The dashed lines show how E_μ can be constructed from E_e.

Fig. 2. The Thomas-Fermi density distributions for semi-infinite nuclear matter are plotted against the mean location of the surface for four different values of the asymptotic asymmetry δ. For δ values larger than zero the upper curve represents the neutrons and the lower one the protons. The small vertical bars serve to identify the separate locations of the neutron and proton surfaces.

Fig. 3. The neutron skin thickness (the difference between the surface locations of the neutrons and protons) is plotted against the asymptotic asymmetry δ for δ values from zero through the neutron drip point, all the way up to the proton drip point. The thin straight line from the origin is the Droplet Model prediction for this quantity.

Fig. 4. The widths (diffuseness) of the surfaces of the neutron and proton distributions are plotted against the asymptotic asymmetry δ for δ values from zero through the neutron drip point, all the way up to the proton drip point.

Fig. 5. The surface energy coefficients γ_e and γ_μ are plotted against δ^2, the square of the asymptotic nuclear asymmetry. The thin straight lines are the Droplet Model prediction for the behavior of these quantities for small values of δ. The dashed and dot dashed lines represent two schemes that were previously proposed for estimating the dependence of γ_e on δ.

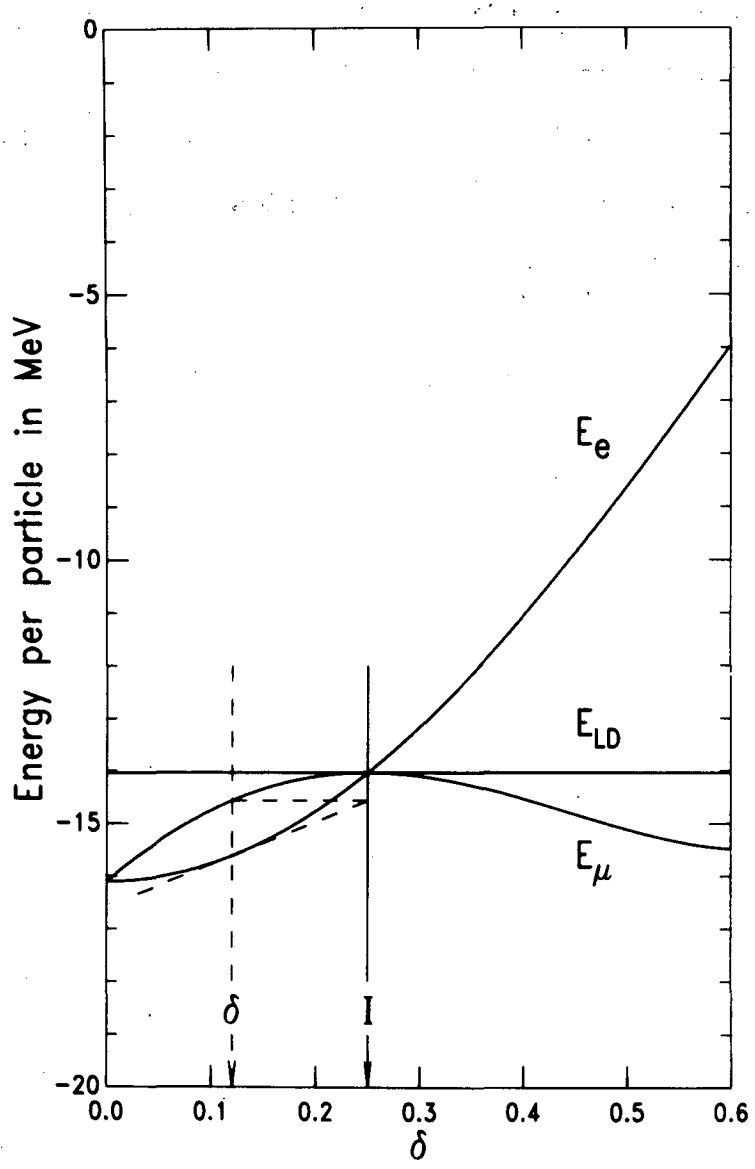

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Fig. 1

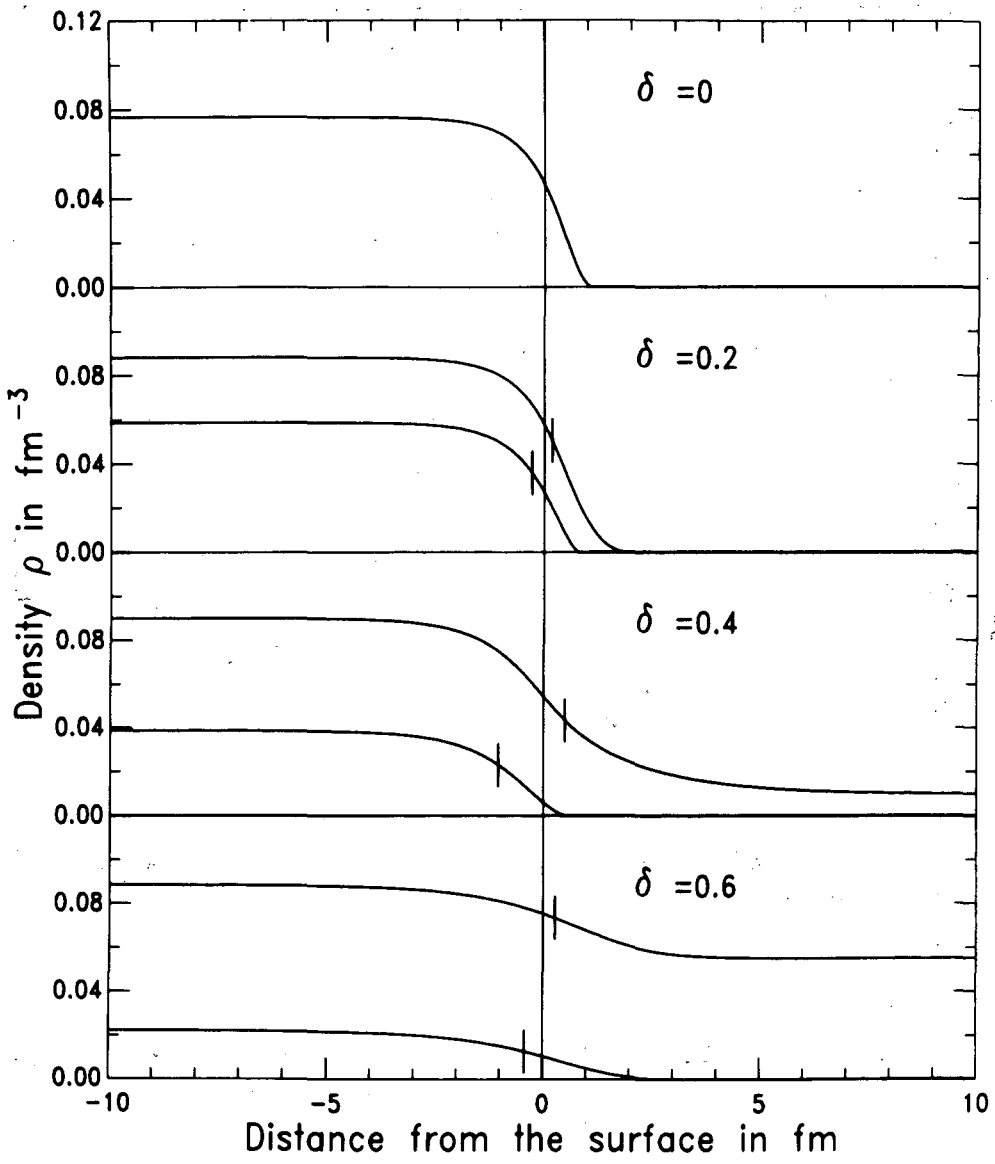

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Fig. 2

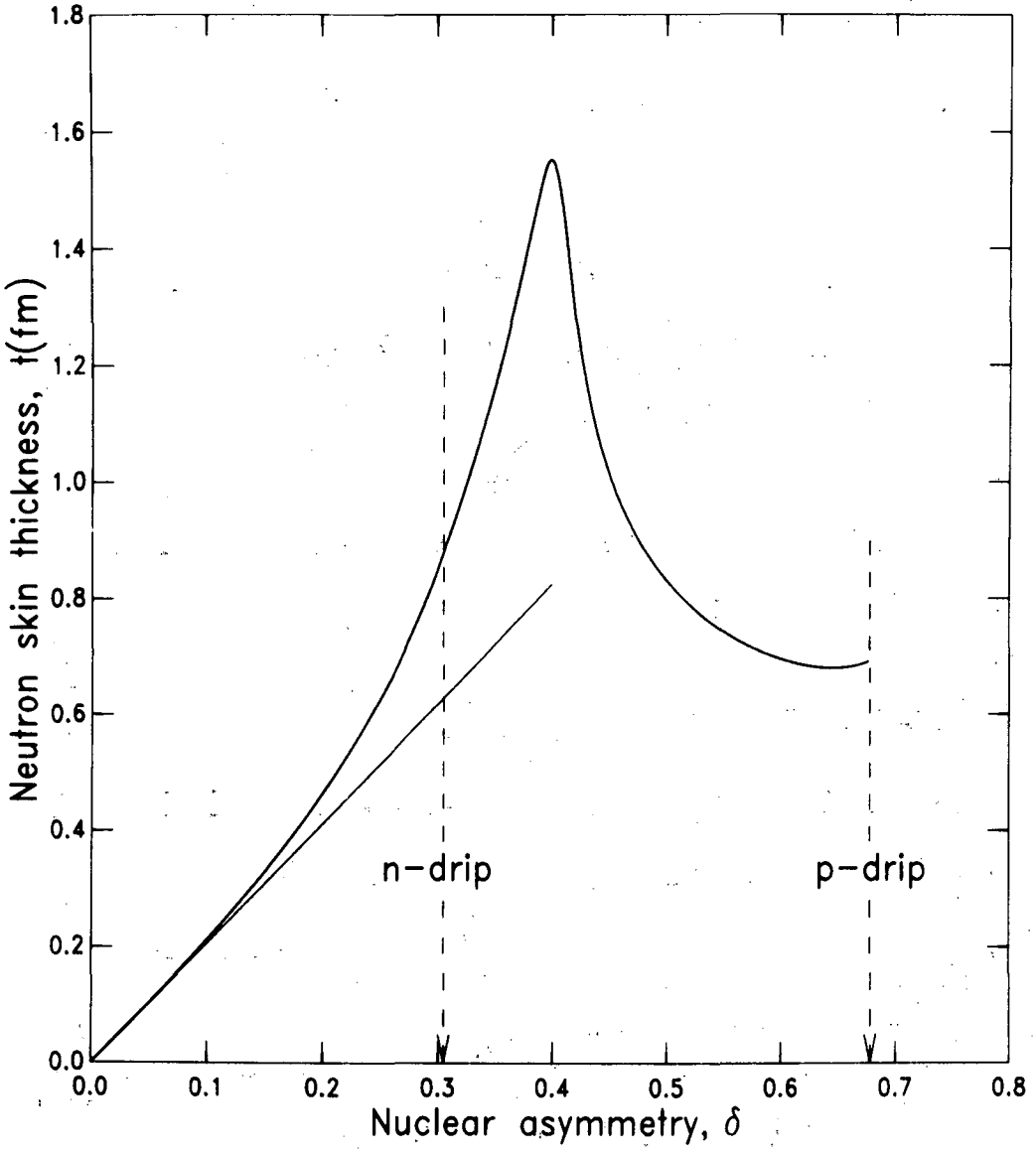

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Fig. 3

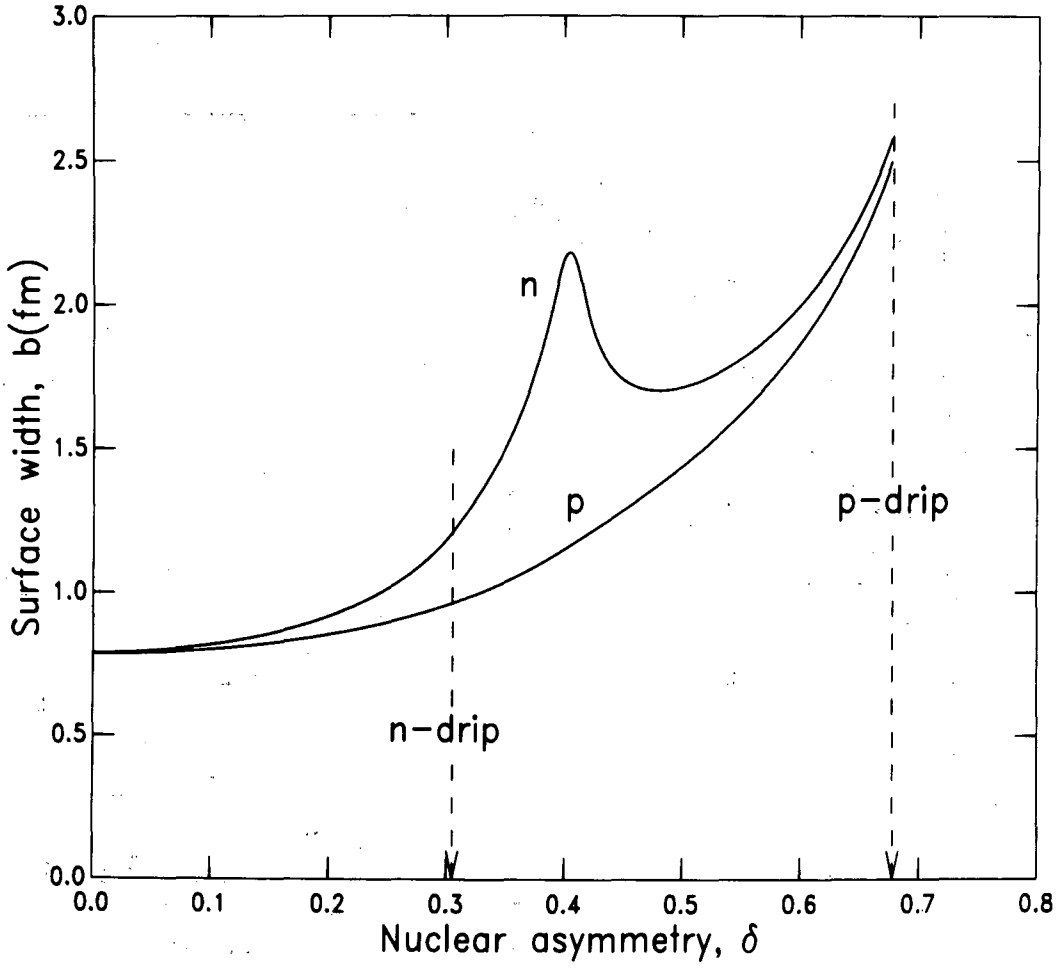

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Fig. 4

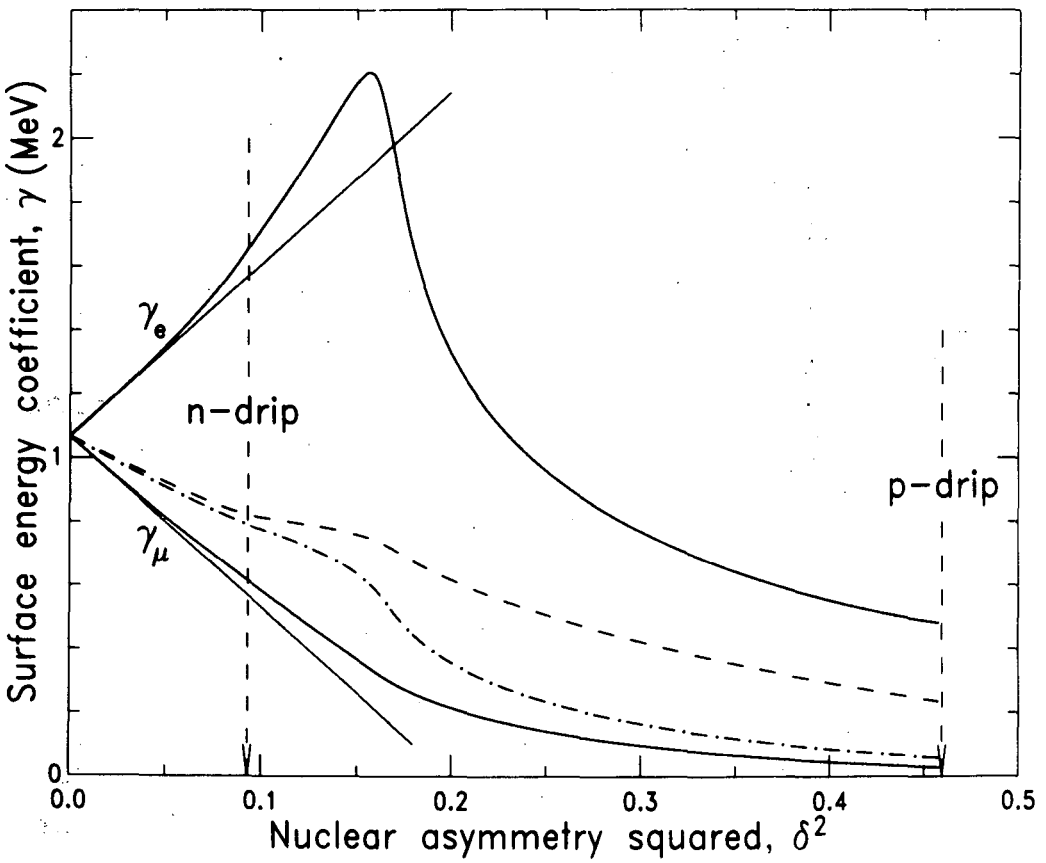

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Fig. 5

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