

# Nuclear diffuseness as a degree of freedom

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The response of the nuclear energy to changes in neutron and proton surface diffusenesses is investigated using the Thomas-Fermi model. Algebraic expressions are provided for the energy cost of changing the two diffusenesses away from their equilibrium values. This will make it possible to generalize the macroscopic-microscopic calculations of nuclear masses and deformation energies by the inclusion of the neutron and proton diffusenesses as degrees of freedom (to be varied along with the shape degrees of freedom). One result, which is suggested by the relatively low cost in macroscopic energy of increasing the diffuseness of a heavy nucleus by 10% (about 4 MeV), is that superheavy nuclei near  $Z=126$ ,  $N=184$  may have a fair chance of becoming stabilized by shell effects. An appendix introduces an improved measure of surface diffuseness, with certain advantages over the conventional Süssmann width  $b$ . [S0556-2813(98)03612-7]

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## I. INTRODUCTION

Estimating the dependence of the nuclear surface energy on surface diffuseness may turn out to be important for locating more reliably the magic numbers in the region of superheavy nuclei. The argument goes as follows. In macroscopic-microscopic approaches to extrapolations into the superheavy regime the nuclear mean field is parametrized as a shape-dependent Woods-Saxon or similar potential, in which the Strutinsky shell correction is then evaluated. In order to find the ground-state energy and shape of a nucleus, in particular a superheavy nucleus, the sum of the microscopic shell correction and a macroscopic (e.g., liquid drop) energy is varied as a function of the shape degrees of freedom. In such variations the surface diffuseness is usually kept constant, but one may well ask how the result would change if, when locating the energy minimum, the diffuseness were to be treated as an additional degree of freedom, to be varied simultaneously with the shape degrees of freedom.

This question has, in fact, a long history going back at least to Refs. [1–3]. There have also been indications as long ago as 1966 (See Fig. 4 in [4]), that an increased surface diffuseness would begin to favor the magic proton number  $Z=126$  over 114. This possibility has been examined in the recent comprehensive study in [5], where macroscopic-microscopic extrapolations were confronted with self-consistent Hartree-Fock calculations, in which the mean field is not parametrized, but is allowed to seek out its optimum form, including whatever changes in the surface diffuseness are called for.

The resulting possibility of a reappearance of the magic proton number 126 would affect profoundly forthcoming searches for spherical superheavy nuclei, and it is extremely important to throw further light on this question by performing up-to-date macroscopic-microscopic calculations generalized to include the surface diffuseness degree of freedom. This should be done in parallel with further refinements of the Hartree-Fock calculations, aimed at eliminating ambiguities associated with different choices of the effective interaction, as well as making the calculations more nearly comparable with macroscopic-microscopic treatments as regards the accuracy with which *known* properties of nuclei are re-

produced (see [6]). The Extended Thomas-Fermi-Strutinsky Integral method of [7], comparable to but faster than Hartree-Fock, should have an especially important role to play in such studies.

In order to carry out a macroscopic-microscopic calculation with the diffuseness degree of freedom included, it is necessary to investigate the response to diffuseness of both the macroscopic and microscopic parts of the energy. The machinery for calculating the latter is already in place: simply recalculate the Strutinsky shell correction for a series of diffusenesses. As regards the former, a new question arises: besides the known response of the Coulomb energy to diffuseness, one needs the response of the macroscopic surface-layer energy. To answer this question there is now available a reliable Thomas-Fermi model of nuclei fitted accurately to a wide range of nuclear properties, [8]. The model has already served as the basis for a variety of macroscopic calculations [8,9], and the present paper describes the application of the model to estimating the response of the macroscopic energy to variations of the surface diffuseness (an early attempt to estimate this response was made in [10]).

## II. THE THOMAS-FERMI MODEL

The model is described in [8]. It provides the self-consistent mean-field solution of the problem of nucleons interacting by an effective Yukawa potential, the solution following from a straightforward application of the semiclassical Thomas-Fermi approximation of two fermions per  $h^3$  of phase space. After allowing for shell and pairing effects and for the Congruence/Wigner energy, the model, with a suitably adjusted Yukawa interaction, reproduces accurately the measured nuclear binding energies and fission barriers, gives a good account of nuclear sizes and of the surface diffuseness, and predicts neutron matter properties in substantial agreement with independent theoretical calculations. The model is thus expected to be a fair guide to macroscopic nuclear properties calculated within the limitations of a semiclassical approach.

## III. NUCLEAR SURFACE-LAYER ENERGY

The two leading terms in the part of the energy associated with the nuclear surface (the “surface-layer energy”) are a

surface energy proportional to the surface area of the nucleus and a curvature correction proportional to the integrated surface curvature. For a spherical nucleus of mass number  $A$  the surface energy  $E_s$  can be written as

$$E_s = a_2(w_n, w_p)A^{2/3} \quad (1)$$

and the curvature energy as

$$E_\kappa = a_3(w_n, w_p)A^{1/3}, \quad (2)$$

where the surface and curvature energy coefficients  $a_2$  and  $a_3$  are now considered to be functions of the neutron and proton surface diffusenesses, as specified by the widths  $w_n$  and  $w_p$  (defined later, but almost identical with the standard Süssmann widths  $b_n$ ,  $b_p$ , [11]). According to the Thomas-Fermi model of [8], the optimum diffusenesses for standard, semi-infinite nuclear matter are given by  $w_n = w_p = w_o \approx 1.0$  fm, with  $a_2(w_o, w_o) = 18.63$  MeV and  $a_3(w_o, w_o) = 12.11$  MeV.

We have determined the quadratic response of the surface-layer energy to small variations of  $w_n$  and  $w_p$  by performing a series of Thomas-Fermi calculations for finite nuclei in which  $w_n$  and  $w_p$  were constrained to have values that were  $\lambda_n$  or  $\lambda_p$  times their equilibrium values. The scaling factors  $\lambda_n, \lambda_p$  were varied, in steps, between about 0.8 and 1.2, and cubic expressions were fitted to the series of energy changes induced in this way. The details are described in Appendix A. The final result may be summarized by the following expression for the deviation of the energy from its equilibrium value for an uncharged spherical nucleus with mass number  $A$  and  $N=Z$ :

$$\begin{aligned} \Delta E = & 18.63A^{2/3}[\tfrac{1}{2}\phi_1(\lambda_n - 1)^2 - \phi_2(\lambda_n - 1)(\lambda_p - 1) \\ & + \tfrac{1}{2}\phi_1(\lambda_p - 1)^2] \text{ MeV} \\ & + \text{cubic terms in } (\lambda_n - 1), (\lambda_p - 1), \end{aligned} \quad (3)$$

where, for  $A > 40$ , the coefficients  $\phi_1$  and  $\phi_2$  are given approximately by the following functions of  $A$ :

$$\phi_1 = 0.7388 + 1.1787\alpha + 12.5929\alpha^2, \quad (4)$$

$$\phi_2 = 0.4836 + 0.4178\alpha + 5.2180\alpha^2, \quad (5)$$

with  $\alpha = A^{-1/3}$ . [The factor  $18.63A^{2/3}$  was included in defining  $\phi_1$  and  $\phi_2$  in Eq. (3) so that for  $\alpha \rightarrow 0$  the constants  $\phi_1 = 0.7388$ ,  $\phi_2 = 0.4836$  would represent the response of the surface energy coefficient  $a_2$  to diffuseness changes.]

The small anharmonic (cubic) terms in Eq. (3) are discussed in Appendix A.

The diffuseness dependence of the curvature energy itself turned out to be amazingly simple. Thus, in the case  $w_n = w_p$ , the dependence on the mean diffuseness  $w = (w_n + w_p)/2$  was found to be indistinguishable from a straight line through the origin, viz.:

$$a_3(w) = 12.11w/w_o \text{ MeV}. \quad (6)$$

By symmetry, there is no linear dependence of  $a_3$  on  $w_n - w_p$ , so that Eq. (6) is correct to leading (linear) order in  $w_n$  and  $w_p$ .

Due to quantal effects, Eqs. (3), (6) become inappropriate for small values of  $\lambda$ , but should be applicable for  $\lambda$  close to 1, where the Thomas-Fermi approximation is known to be useful.

#### IV. COULOMB ENERGY

For an arbitrarily shaped nucleus the diffuseness correction to the Coulomb energy was derived to leading order in [12]. For a spherical nucleus with atomic number  $Z$  and radius  $R$  the energy may be expanded to higher orders in  $\omega \equiv w_p/R$  as follows:

$$E_c = C \left( 1 - \frac{5}{2}\omega^2 + k_3\omega^3 + k_4\omega^4 + \dots \right), \quad (7)$$

where  $C = 3e^2Z^2/5R$ , with  $e^2 = 1.44$  MeV fm. In the case of a Fermi-function density profile one finds  $k_3 = 3.0216$  and  $k_4 = 3/2$ . (see Appendix B). In what follows we shall neglect the last term in Eq. (7) and use the above value of  $k_3$  in our estimates.

#### V. SURFACE DIFFUSENESS OF FINITE NUCLEI

We may write the sum of surface, curvature and Coulomb energies in the following form:

$$\begin{aligned} E = & S[1 + \tfrac{1}{2}\phi_1(\lambda_n - 1)^2 - \phi_2(\lambda_n - 1)(\lambda_p - 1) \\ & + \tfrac{1}{2}\phi_1(\lambda_p - 1)^2] \\ & + K(\lambda_n + \lambda_p)/2 \\ & + C - C_2\lambda_p^2 + C_3\lambda_p^3, \end{aligned} \quad (8)$$

where

$$S = 18.63A^{2/3} \text{ MeV}, \quad K = 12.11A^{1/3} \text{ MeV},$$

$$C = 3e^2Z^2/5R = 0.7579Z^2/A^{1/3} \text{ MeV},$$

$$C_2 = 3e^2Z^2w_o^2/2R^3 = 1.4579Z^2/A \text{ MeV},$$

$$C_3 = 3e^2Z^2w_o^3k_3/5R^4 = 1.5457Z^2/A^{4/3} \text{ MeV},$$

and where we took  $R = 1.14A^{1/3}$  fm,  $w_o = 1$  fm. We have disregarded the cubic terms in Eq. (3) and the last term in Eq. (7). This has a negligible effect on the results that follow.

Equating to zero the partial derivatives of  $E$  with respect to  $\lambda_n$  and  $\lambda_p$  gives

$$S[\phi_1(\lambda_n - 1) - \phi_2(\lambda_p - 1)] + K/2 = 0, \quad (9)$$

$$S[-\phi_2(\lambda_n - 1) + \phi_1(\lambda_p - 1)] + K/2 - 2C_2\lambda_p + 3C_3\lambda_p^2 = 0. \quad (10)$$

Eliminating  $\lambda_n - 1$  gives the quadratic equation  $c_2\lambda_p^2 + c_1\lambda_p - c_0 = 0$ , with the solution

$$\lambda_p = \frac{\sqrt{c_1^2 + 4c_0c_2} - c_1}{2c_2}, \quad (11)$$

where

$$c_0 = 1 - K/2S\chi = 1 - 0.3250/A^{1/3}\chi, \quad (12)$$

$$c_1 = 1 - 2C_2/S\psi = 1 - 0.1565Z^2/A^{5/3}\psi, \quad (13)$$

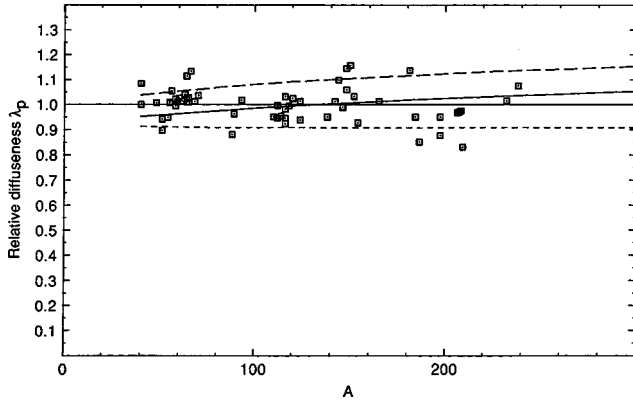


FIG. 1. The relative proton diffuseness  $\lambda_p$ , as predicted by Eq. (11), is compared with the experimental trend of the relative “RMS diffuseness,”  $w_p/1.022$  fm, as deduced from two-parameter Fermi-function fits listed in [13]. The long-dashed curve shows the result of disregarding the curvature energy, the short-dashed curve the result of disregarding the Coulomb energy.

$$c_2 = 3C_3/S\psi = 0.2489Z^2/A^2\psi, \quad (14)$$

where  $\psi$  stands for  $(\phi_1^2 - \phi_2^2)/\phi_1$  and  $\chi$  stands for  $(\phi_1 - \phi_2)$ . With  $\lambda_p$  determined, Eq. (9) may be used to calculate  $\lambda_n$ .

To leading order in the diffuseness correction to the Coulomb energy (i.e., with  $c_2$  disregarded) we would have

$$\lambda_p \approx \frac{c_0}{c_1} = \frac{1 - 0.3250/A^{1/3}\chi}{1 - 0.1565Z^2/A^{5/3}\psi}, \quad (15)$$

which illustrates the competition between the curvature and Coulomb forces in driving  $w_p$  away from  $w_o$ .

Equation (11) predicts that the relative proton diffuseness  $w_p/w_o$  should be a definite function of  $A$ , resulting from the  $A$ -dependent difference between the curvature and Coulomb forces working against the stiffnesses  $\phi_1$  and  $\phi_2$ , which themselves increase rapidly with decreasing  $A$  according to Eqs. (4) and (5). The net result of these opposing forces turned out to be—somewhat unexpectedly—an almost constant value of  $w_p/w_o$  as illustrated in Fig. 1, where  $\lambda_p$  is plotted vs  $A$  along the valley of  $\beta$  stability. Experimental values of  $w_p$  were deduced from the radius and diffuseness parameters  $c$  and  $z$  of two-parameter Fermi-function fits to data, as reported in [13]. (The more recent reference [14], pointed out to us by a referee, does not change significantly the appearance of Fig. 1.) The average value of  $w_p$  for the points displayed is 1.022 fm and the measured values of  $w_p$  were divided by this average in order to compare the trend of the relative diffuseness with theory.

Figure 1 shows also what the theory would predict for this trend if the curvature energy were disregarded (long-dashed curve) or if the Coulomb energy were disregarded (short-dashed curve).

## VI. ENERGY COST OF DIFFUSENESS CHANGES

The stiffnesses of the macroscopic energy against deviations of  $\lambda_n, \lambda_p$  from 1 are given by the second derivatives of Eq. (8). If  $\lambda_p$  is held fixed, we find

$$\left. \frac{\partial^2 E}{\partial \lambda_n^2} \right|_{\lambda_p} = S\phi_1. \quad (16)$$

If  $\lambda_n$  is held fixed we have

$$\begin{aligned} \left. \frac{\partial^2 E}{\partial \lambda_p^2} \right|_{\lambda_n} &= S\phi_1 - 2C_2 + 6C_3\lambda_p \\ &\approx S\phi_1 - 2C_2 + 6C_3 \quad \text{for } \lambda_p \approx 1. \end{aligned} \quad (17)$$

If  $\lambda_n$  is constrained to be equal to  $\lambda_p$  we have

$$E = S[1 + (\phi_1 - \phi_2)(\lambda_p - 1)^2] + K\lambda_p + C - C_2\lambda_p^2 + C_3\lambda_p^3, \quad (18)$$

and

$$\left. \frac{\partial^2 E}{\partial \lambda_p^2} \right|_{\lambda_n = \lambda_p} = 2S(\phi_1 - \phi_2) - 2C_2 + 6C_3. \quad (19)$$

If for a given  $\lambda_p$  the value of  $\lambda_n$  is chosen so as to minimize the energy we find

$$\left. \frac{\partial^2 E}{\partial \lambda_p^2} \right|_{\text{optimized } \lambda_n} = S\psi - 2C_2 + 6C_3. \quad (20)$$

As an example take  $A = 310$ ,  $Z = 126$  and  $\lambda_n = \lambda_p = \lambda$ . Equation (19) gives the energy cost to change  $\lambda_n, \lambda_p$  from their optimum values as

$$\begin{aligned} \Delta E &= \frac{1}{2} \frac{d^2 E}{d\lambda^2} (\lambda - 1)^2 \\ &= [(\phi_1 - \phi_2)S - C_2 + 3C_3](\lambda - 1)^2 \\ &= (451.1 - 74.7 + 35.1)(\lambda - 1)^2 \text{ MeV} \\ &= 411.5(\lambda - 1)^2 \text{ MeV}. \end{aligned} \quad (21)$$

Thus a 5% increase in the diffuseness would entail a macroscopic energy cost of 1.03 MeV, and a 10% increase would cost 4.12 MeV.

## VII. MAGIC REGION NEAR $Z = 126$ , $N = 184$

According to Eq. (11) the equilibrium value of the relative proton diffuseness is 1.081, and an additional synchronous increase of the proton and neutron diffusenesses by 10% would cost about 4 MeV according to Eq. (21). It was estimated in [2] that for the  $Z = 126$ ,  $N = 184$  magic numbers to show up the diffuseness might have to be increased by something like 10%. The total gain in shell-effect energy, estimated on the basis of Fig. 2 in [2], is of the order of 12 MeV, similar to the value reported in [5], Fig. 5. If the above estimates are taken at face value, the conclusion is suggested that the macroscopic cost in energy might not prevent the nucleus from exploiting the extra binding associated with  $Z = 126$ ,  $N = 184$  in agreement with [5]. It might even turn out that the neighborhoods of both the  $Z = 114$ ,  $N = 184$  and the

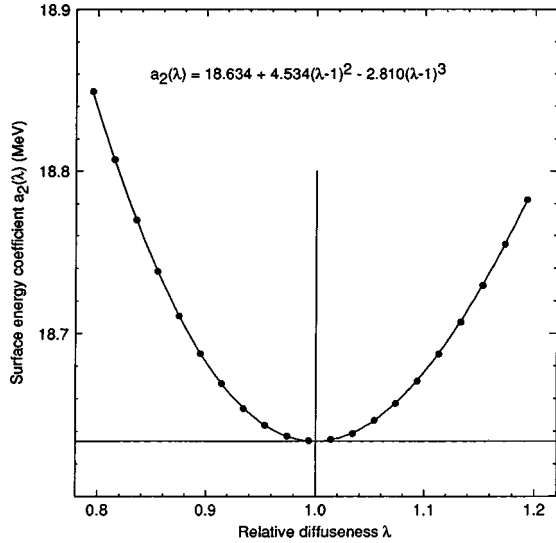


FIG. 2. The surface energy coefficient  $a_2$  for semi-infinite, standard nuclear matter in its dependence on the relative diffuseness  $\lambda$ . The dots refer to the Thomas-Fermi calculations and the curve to a cubic fit, displayed in the figure, with a “stiffness” of 4.534 MeV and an “anharmonicity” of  $-2.810$  MeV.

$Z=126$ ,  $N=184$  doubly magic numbers would exhibit extra stability sufficient to ensure reasonable lifetimes against spontaneous decay. (Such an alternative should not yet be dismissed, even though the 114 proton shell is not favored by the Hartree-Fock calculations with SkP and SLy7 forces in [5].) A straightforward generalization of the current macroscopic-microscopic calculations, made possible by the availability of the Thomas-Fermi stiffnesses as calculated above, should be able to throw further light on these fascinating possibilities.

In the present paper we have focused on the relevance of the diffuseness degrees of freedom to the question of super-heavy elements. But other applications, such as shell effects near the drip lines or surface contributions to giant monopole resonances, readily come to mind.

#### ACKNOWLEDGMENTS

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#### APPENDIX A: DETERMINATION OF THE STIFFNESSES

Figure 2 shows how the surface energy coefficient  $a_2$  for semi-infinite standard nuclear matter changes when the diffuseness of both neutrons and protons is forced to be  $\lambda$  times their equilibrium value. The points represent a series of 21 Thomas-Fermi calculations and the curve is the virtually perfect fit by a cubic in  $(\lambda - 1)$ . The “stiffness” 4.534 MeV is shown as (half) a square at  $A^{-1/3}=0$  and the “anharmonicity”  $-2.810$  MeV is indicated by a (semi)circle in Fig. 3. The other points in Fig. 3 refer to calculations with finite, uncharged,  $N=Z$  nuclei. They were obtained by forcing the diffuseness to be different from its equilibrium value  $w_{eq}$  by the following prescription. Instead of the equilibrium density

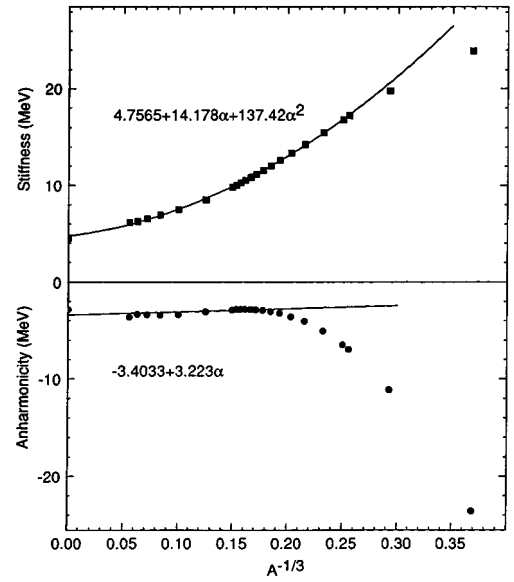


FIG. 3. The stiffness and anharmonicity, deduced from Thomas-Fermi calculations on uncharged, finite nuclei with  $N=Z$  are displayed as functions of  $A^{-1/3}$ . In this case the neutron and proton diffusenesses were made to vary in phase. Fits to the calculated points (stressing the regime of large  $A$ ) are displayed in terms of  $\alpha$ , defined as  $A^{-1/3}$ .

distribution for the nucleus in question, an equilibrium density distribution of a heavier (or lighter) “substitute” nucleus was calculated and then scaled (along the ordinate and the abscissa) in order to have the same central density and the same number of particles as the original one. If the substitute nucleus was heavier/lighter, the scaled density would be less/more diffuse. The scaled density was then inserted in the Thomas-Fermi computer program to calculate the energy increase  $\Delta E$  of the nucleus in question, Eq. (6). The associated relative diffuseness  $w/w_{eq}$  was calculated using Eq. (B1) in Appendix B. By varying the mass of the substitute nucleus in the appropriate range, a series of values for  $\Delta E$  was obtained and, after division by  $18.63A^{2/3}$ , a fit of a cubic in  $w/w_{eq}$  gave the stiffness and anharmonicity displayed in Fig. 3. Figure 4 shows the corresponding quantities in the case when only one of the diffusenesses was changed, the other remaining fixed. As is readily verified from Eq. (3), the two sets of calculations determine  $\phi_1 - \phi_2$  and  $\phi_1$ , respectively; the values of  $\phi_1$  and  $\phi_2$  follow [Eqs. (4), (5)].

#### APPENDIX B: THE RMS WIDTH $w$

A measure of the surface diffuseness, Süssman’s width  $b$ , is defined in terms of the width of the bump function  $g(r)$  resulting from differentiating the density profile function  $f(r)$ , where  $f(r)$  is assumed to be effectively constant except for a thin surface region around  $r=R$  [11]. It turns out that a slightly different definition of the diffuseness, the “RMS width  $w$ ,” has certain advantages over Süssman’s  $b$ . We define this width in terms of the increase in the root mean square (rms) radius of a spherical distribution when the density profile of the distribution is changed from a sharp cutoff at  $R$  to the diffuse profile in question. Specifically we shall take

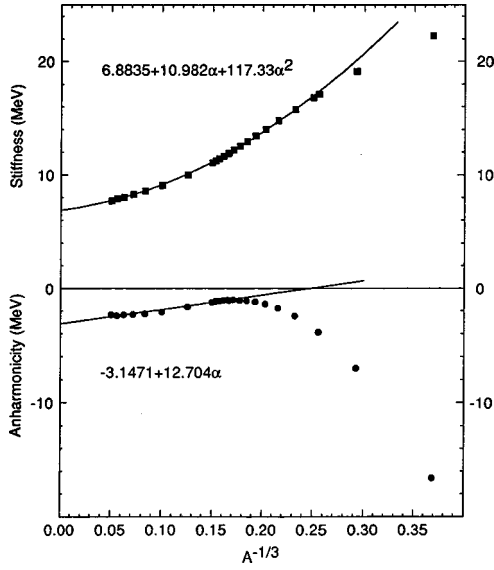


FIG. 4. This is like Fig. 3, but here only one of the two diffusenesses was varied.

$$w^2 \equiv \frac{1}{3} [\langle r^2 \rangle_{\text{diffuse}} - \langle r^2 \rangle_{\text{sharp}}], \quad (\text{B1})$$

where the factor  $1/3$  was chosen to make  $w$  tend to  $b$  in the limit of large  $R$ . Note that the original sharp distribution does not have to be uniform. It could, in fact, be almost any function of  $r$  up to  $r=R$ . Then, as the profile gets diffused according to some given profile prescription,  $\langle r^2 \rangle$  increases and  $w$  provides a measure of the profile's diffuseness. Thus, unlike Süssmann's width  $b$ , the RMS width  $w$  can be defined without difficulty also in the case when there is a central depression in the nuclear density (caused by the Coulomb repulsion).

In the case of an originally uniform sharp distribution with radius  $R$  we have

$$w^2 = \frac{1}{3} \left( \frac{3}{5} Q^2 - \frac{3}{5} R^2 \right) = \frac{1}{5} (Q^2 - R^2), \quad (\text{B2})$$

where  $Q$  is the "equivalent RMS radius" of the diffuse distribution. According to Eq. (2.45) in [11], p. 23,  $Q$  is related to  $b$  by

$$Q = R \left( 1 + \frac{5}{2} \frac{b^2}{R^2} + \text{higher powers of } b/R \right). \quad (\text{B3})$$

Thus

$$Q^2 = R^2 + 5b^2 + \dots$$

It follows that  $w$  tends to  $b$  for large  $R$ .

Equation (2.45) in the above reference, taken to higher orders in  $\beta \equiv b/R$ , reads

$$Q = R \left[ 1 + \frac{5}{2} \beta^2 + \frac{25}{6} \gamma_3 \beta^3 + \frac{5}{2} \left( \gamma_4 - \frac{21}{4} \right) \beta^4 + \frac{1}{2} \left( \gamma_5 - \frac{275}{6} \gamma_3 \right) \beta^5 + \dots \right], \quad (\text{B4})$$

where  $\gamma_3, \gamma_4, \gamma_5$  are numbers characterizing the skewness, kurtosis, etc. of the density profile in question, as given on p. 22 of [11]. Substituting in Eq. (B2) we find

$$w^2 = b^2 \left[ 1 + \frac{5}{3} \gamma_3 (b/R) + (\gamma_4 - 4) (b/R)^2 + \left( \frac{1}{5} \gamma_5 - 5 \gamma_3 \right) (b/R)^3 + \dots \right] \quad (\text{B5})$$

as the relation between the RMS width  $w$  and Süssmann's width  $b$ . Taking as an example the Fermi-function profile, we have  $\gamma_3 = \gamma_5 = 0$ ,  $\gamma_4 = 21/5$  (p. 31, [11]), which leads to

$$w = b \left[ 1 + \frac{1}{10} (b/R)^2 + \text{terms in } (b/R)^4 \text{ and higher} \right]. \quad (\text{B6})$$

The Coulomb energy of a Fermi-function density distribution is given on p. 32 of [11] as

$$E_C = E_C^{\text{sharp}} \left( 1 - \frac{5}{2} \beta^2 + 3.0216 \beta^3 + \beta^4 + \dots \right). \quad (\text{B7})$$

Using Eq. (B5) this becomes the following expansion in powers of  $\omega \equiv w/R$ :

$$E_C = E_C^{\text{sharp}} \left( 1 - \frac{5}{2} \omega^2 + 3.0216 \omega^3 + \frac{3}{2} \omega^4 + \dots \right). \quad (\text{B8})$$

The two formulas differ only in the coefficient of the small fourth-order correction. Equation (B6) shows similarly that  $w$  and  $b$  differ by only 0.66% for  $A=40$  and by 0.20% for  $A=240$  in the case of a Fermi-function profile. In the case of "skew" surface profiles, when  $\gamma_3$  is not zero, the differences would be greater.

There are three reasons for preferring  $w$  to  $b$ : (1) The above-mentioned applicability of  $w$  to nonuniform radial distributions. (2) The fact that  $w$  is defined in terms of an *integral* property of the given distribution  $f(r)$ . Süssmann's  $b$  requires differentiating  $f(r)$ , which can lead to serious loss of accuracy when  $f(r)$  is available only as a numerically evaluated quantity, as is the case in Thomas-Fermi (or Hartree-Fock) computer calculations. (3) The quantity  $w$  is a three-dimensional property of the density distribution, whereas  $f(r)$  and  $g(r)$  that are used in defining  $b$  are one-dimensional objects. Why use a one-dimensional property when the problem is intrinsically three dimensional?

Note also that for a diffuse distribution obtained from a sharp distribution by convoluting it with a folding function, it is identically true that (see Ref. [11], p. 23):

$$\langle r^2 \rangle_{\text{diffuse}} = \langle r^2 \rangle_{\text{sharp}} + \langle r^2 \rangle_f, \quad (\text{B9})$$

where  $\langle r^2 \rangle_f$  is the mean-square radius of the folding function. It follows from Eq. (B1) that in the case of a diffuseness induced by convolution, the surface width  $w$  is simply the rms radius of the folding function divided by the square root of 3. This relation is exact for any value of  $R$ , large or small, and for any radial function  $f(r)$  of the sharp distribution.

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