Temperature dependence of the nuclear free energy based on a finite-range mass formula

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The Yukawa-plus-exponential mass formula is generalized to describe the Gibbs free energy of hot, finite nuclei. The temperature dependence is obtained by fitting the results of former temperature-dependent Thomas-Fermi calculations with a finite-range mass formula. The temperature dependence of the pairing and Wigner terms in the mass formula is also given. [S0556-2813(99)04405-2]

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I. INTRODUCTION

Modeling fusion-fission reactions one has to calculate the conservative part of the generalized forces, which govern the collective motion, from the derivatives of the Gibbs free energy with respect to the shape parameters [1]. Therefore a reliable expression is needed for this quantity as function of the nuclear size, charge, shape, and temperature. The accuracy must be such that fission-barrier heights are better reproduced than within 1 MeV. For zero temperature this is achieved by fitting an appropriate mass formula to a standard set of data consisting of all empirically known ground-state binding energies, corrected for shell and deformation effects [2], fission barrier heights [3], corrected in the same way, equivalent sharp radii [4] and the average charge diffuseness [4], and of some fusion barrier heights [5]. Such extensive data do not exist for hot nuclei. One has therefore to rely on extended Thomas-Fermi calculations at finite temperature [6] to obtain the temperature dependence of the free energy [7]. Unfortunately, the droplet expansion, used in the latter reference, is in general not suitable to infer fusion barrier heights and not sufficiently accurate for fission barriers of lighter nuclei at zero temperature. But we shall assume that the temperature coefficients, obtained in Ref. [7], are accurate enough to infer the temperature dependence of the free energy, $\Delta G = G(T) - G(T=0)$, for compact shapes.

II. TEMPERATURE DEPENDENCE OF THE COEFFICIENTS IN THE YUKAWA-PLUS-EXPONENTIAL MASS-FORMULA

In the following we shall use the Yukawa-plus-exponential mass formula for the macroscopic part of the ground-state binding and the deformation energies at zero temperature in the specific form, fitted in Ref. [2] to a standard set of data

$$E(Z,N,\text{shape}) = M_H Z + M_n N + E_{ld} + E_W + E_{\text{pair}} + f(k_f r_p)\frac{Z^2}{A} - c_a(N-Z) - a_{el} Z^{2.39} \tag{1}$$

with the liquid-drop energy proper

$$E_{ld} = -a_V(1-\kappa_V I^2)A + a_S(1-\kappa_S I^2)B_1 A^{2/3} + c_0 A^0 + c_1 \frac{Z^2}{A^{1/3}} B_3 - c_4 \frac{Z^{4/3}}{A^{1/3}}, \tag{2}$$

the Wigner term

$$E_W = W\left(|I| + \begin{cases} 1/A & Z \text{ and } N \text{ odd and equal} \\ 0 & \text{otherwise} \end{cases}\right), \tag{3}$$

and the average pairing energy [8]

$$E_{pair} = \begin{cases} \overline{\Delta}_p + \overline{\Delta}_n - \delta_{np} & Z \text{ and } N \text{ odd}, \\ \overline{\Delta}_p & Z \text{ odd and } N \text{ even}, \\ \overline{\Delta}_n & Z \text{ even and } N \text{ odd}, \\ 0 & Z \text{ and } N \text{ even}, \end{cases} \tag{4}$$

with

$$\overline{\Delta}_n = \frac{rB_{\text{surf}}}{N^{1/3}} e^{-sI-tI^2}, \quad \overline{\Delta}_p = \frac{rB_{\text{surf}}}{Z^{1/3}} e^{-sI-tI^2},$$

$$\delta_{np} = \frac{h}{B_{\text{surf}} A^{2/3}} \tag{5}$$

and the four constants $r = 5.72$ MeV, $h = 6.82$ MeV, $s = 0.118$, and $t = 8.12$. Expressions (1)–(3) involve the neutron excess parameter $I = (N-Z)/(N+Z)$, the Coulomb parameters

$$c_1 = \frac{3e^2}{5r_0} \text{ and } c_4 = c_1 \frac{5}{4}\left(\frac{3}{2\pi}\right)^{2/3}, \tag{6}$$

the proton form factor

$$f(k_f r_p) = -\frac{r_p^2 e^2}{8 r_0^3}\left[\frac{145}{48} - \frac{327}{2880}(k_f r_p)^2 + \frac{1527}{1209600}(k_f r_p)^4\right]$$

with the Fermi wave number $k_f = (9\pi Z/4A)^{1/3} r_0^{-1}$, the Wigner-term constant $W = 35$ MeV, and the three shape functions

$$B_1=\frac{1}{8\pi^2a^4r_0^2A^{2/3}}\int_V\int_V\left(2-\frac{|\mathbf{r}-\mathbf{r}'|}{a}\right)\frac{e^{-|\mathbf{r}-\mathbf{r}'|/a}}{|\mathbf{r}-\mathbf{r}'|/a}d\mathbf{r}d\mathbf{r}',$$

$$B_3=\frac{15}{32\pi^2r_0^5A^{5/3}}\int_V\int_V\left[1-\left(1+\frac{|\mathbf{r}-\mathbf{r}'|}{2a_{den}}\right)\times e^{-|\mathbf{r}-\mathbf{r}'|/a_{den}}\right]\frac{d\mathbf{r}d\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|},$$

and

$$B_{\rm surf}=\frac{1}{4\pi r_0^2A^{2/3}}\int_{\rm surf}d\boldsymbol{\sigma}.$$

For spherical nuclei the shape functions can be evaluated in closed form [5,9]. One obtains $B_{\rm surf}=1$,

$$B_1=1-\frac{3}{x_0^2}+(1+x_0)\left(2+\frac{3}{x_0}+\frac{3}{x_0^2}\right)e^{-2x_0}, \tag{7}$$

$$B_3=1-\frac{5}{y_0^2}\left[1-\frac{15}{8y_0}+\frac{21}{8y_0^3}-\frac{3}{4}\left(1+\frac{9}{2y_0}+\frac{7}{y_0^2}+\frac{7}{2y_0^3}\right)e^{-2y_0}\right] \tag{8}$$

with $x_0=r_0A^{1/3}/a$ and $y_0=r_0A^{1/3}/a_{\rm den}$.

If the expression (1) is to be used for the free energy, the constants $M_H=7.28903$ MeV, $M_n=8.07143$ MeV, $e^2=1.439976$ MeV fm, $r_p=0.80$ fm, $c_a=0.145$ MeV, $c_0=5.8$ MeV, and $a_{el}=1.433\times10^{-5}$ MeV [2] are either obviously temperature independent or shall be taken to be temperature independent because of their minor importance in the mass formula. At zero temperature the seven main liquid-drop parameters have the values [2] $r_0=1.16$ fm, $a=0.68$ fm, $a_{den}=0.7$ fm, $a_V=16.00$ MeV, $\kappa_V=1.911$, $a_S=21.13$ MeV, and $\kappa_S=2.3$. They shall be assumed to have a quadratic dependence on the temperature when Eq. (1) is taken to represent the macroscopic part of the Gibbs free energy

$$r_0=1.16(1-x_{r_0}T^2),\quad a=0.68(1-x_aT^2),$$
$$a_{\rm den}=0.7(1-x_{a_{\rm den}}T^2) \tag{9}$$

and similarly for the other four parameters. The justification for this parametrization is the same as in Refs. [6,7]: The leptodermous expansion of the Thomas-Fermi expression for the Gibbs free energy can be represented by the droplet-type formula used in Ref. [7]

$$G=G_{\rm vol}+G_{\rm asy}+G_{\rm surf}B_{\rm surf}+G_{\rm curv}B_{\rm curv}+a_0'+G_{\rm Coul} \tag{10}$$

with

$$G_{\rm vol}=a_V'A,\quad G_{\rm asy}=JAI^2,$$
$$G_{\rm surf}=a_S'A^{2/3}-JAI^2k(1+k)^{-1}, \tag{11}$$

$$G_{\rm curv}=a_c'A^{1/3},\quad G_{\rm Coul}=c_1'B_{\rm Coul}Z^2/A^{1/3}+c_2'Z^2/A, \tag{12}$$

and the abbreviation $k=9J/(4QA^{1/3})$ and the shape functions $B_{\rm surf}$,

$$B_{\rm Coul}=\frac{15}{32\pi^2r_0^5A^{5/3}}\int_V\int_V\frac{d\mathbf{r}d\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|},$$

$$B_{\rm curv}=\frac{1}{8\pi r_0A^{1/3}}\int_{\rm surf}\left(\frac{1}{R_1(\boldsymbol{\sigma})}+\frac{1}{R_2(\boldsymbol{\sigma})}\right)d\boldsymbol{\sigma},$$

where $R_1(\boldsymbol{\sigma})$ and $R_2(\boldsymbol{\sigma})$ are the main curvature radii at the surface point $\boldsymbol{\sigma}$. It was shown in Ref. [6] that the temperature dependence of the eight coefficients a_V', a_S', a_c', c_1', c_2', a_0', J, and Q of this mass formula can be expanded in a way analogous to Eq. (9) for $T\leqslant4$ MeV if the Thomas-Fermi calculation is based on the SkM* potential. The corresponding temperature coefficients x_i' are given in Table I of [7].

Alternatively, assuming strict incompressibility of nuclear matter, also the expression (2) can be derived from a leptodermous expansion [10]. Below, the temperature dependence of Eq. (10) shall therefore be mapped onto the temperature dependence of Eq. (2) in the sense of a least squares fit of the seven coefficients x_i in Eq. (9) to the similarly parametrized temperature dependence of the expressions (10)–(12).

We first notice that because of Eq. (6) the temperature coefficient x_{c_1}' in the direct Coulomb term of Eq. (12) yields $x_{r_0}=-x_{c_1}'=-7.63\times10^{-4}$ MeV^{-2}. For the sake of simplicity we shall assume that the temperature coefficients of the charge diffuseness $a_{\rm den}$ and of the total diffuseness a are the same, $x_a=x_{a_{\rm den}}$. In order to relate x_a to x_{c_2}' we compare the charge-diffuseness correction term $c_2'Z^2/A$ in Eq. (12) with the corresponding term in Eq. (2), $c_1Z^2A^{-1/3}(B_3-1)$. To obtain the same A dependence in both expressions, we first take the limit of (B_3-1) for large y_0, i.e., $a_{\rm den}\ll R$, which yields $-5y_0^{-2}$. In this limit $c_2'\approx-5c_1(a_{\rm den}/r_0)^2=-3e^2a_{\rm den}^2r_0^{-3}$. Inserting the T dependence of c_2', $a_{\rm den}$, and r_0 according to Eq. (9) we obtain

$$x_a=\frac{3}{2}x_{r_0}-\frac{c_2'r_0^3}{6e^2a_{\rm den}^2}x_{c_2'}=-7.37\times10^{-3}\ \ {\rm MeV}^{-2}$$

by collecting the quadratic terms in T.

There is no one-to-one correspondence between the nuclear terms proper in Eq. (2) and (10). Therefore the four temperature coefficients x_{a_V}, x_{a_S}, x_{κ_V}, and x_{κ_S} shall be determined by requiring the temperature dependence of the expression (2) with coefficients (9) to optimally approximate the temperature dependence of the expression (10) as function of its arguments A, I and the shape variables in their relevant ranges. For that purpose we first define the entropy following from Eq. (2) $S=-\partial_TE_{ld}(T)$ and similarly the entropy following from Eq. (10) $S'(T,A,I,\text{ shape})=-\partial_TG(T)$. Since S and S' are rather smooth, analytic functions of A, I and the deformation parameters, the four temperature coefficients can be determined by minimizing the

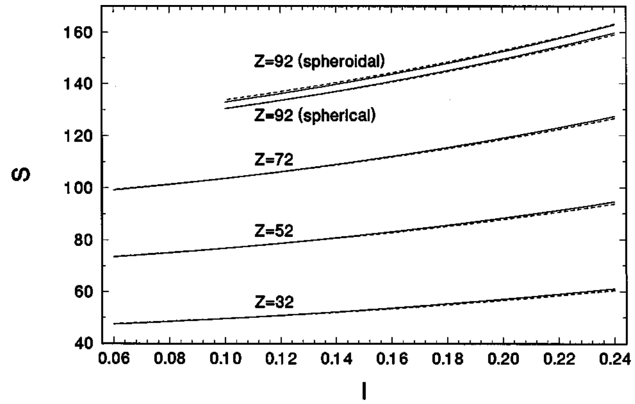

FIG. 1. Nuclear entropy S from Ref. [2] (full lines) and S', the result of our fit (dashed lines), for several sequences of isotopes with spherical shape. The highest pair of curves corresponds to spheroidal uranium with half-axis ratio 1:2. The temperature is 4 MeV.

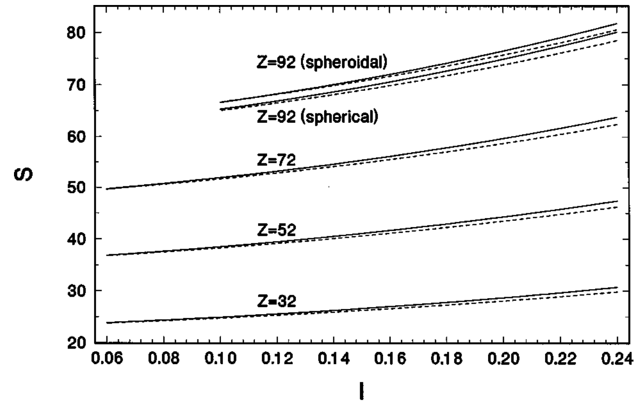

FIG. 2. Same as Fig. 1, but for $T=2$ MeV.

deviation $(S-S')^2$ along ''strategic'' cuts in the A,Z plane: along the line of β stability, i.e., $I(A)=0.4A/(A+200)$ [11], which allows us to fix x_{a_V} and x_{a_S}; along one sequence of isotopes, which determines a combination of x_{κ_V} and x_{κ_S}; and finally the deformation entropy along a sequence of heavy isotopes with a shape deviating sufficiently much from the spherical ground-state shape, which depends only on x_{a_S} and x_{κ_S}. In view of the approximately linear dependence of both, S and S', on T, it is sufficient to fit at one, sufficiently large temperature. We therefore define three sums of squares, all to be evaluated for $T=4$ MeV. First

$$\theta_1=\frac{1}{230}\int_{20}^{250}[S(A)-S('A)]^2 dA.$$

The integral shall be taken along the line of β stability, and for spherical shapes. Second

$$\theta_2=\frac{1}{0.18}\int_{0.06}^{0.24}[S(I)-S'(I)]^2 dI$$

for $Z=70$, i.e., $A(I)=2Z/(1-I)$ and also spherical shapes. And third, θ_3, the same as θ_2, but for $Z=92$ and spheroidal shapes with axis ratio 1:2, corresponding roughly to the fission saddle-point deformation of hot uranium. Minimizing $\theta_1+\theta_2+\theta_3$ with respect to the four remaining temperature coefficients gives

$$x_{a_V}=-3.22\times10^{-3},\quad x_{a_S}=4.81\times10^{-3},$$

$$x_{\kappa_V}=5.61\times10^{-3},\quad x_{\kappa_S}=-14.79\times10^{-3}.$$

The fit achieved with these coefficients is shown in Figs. 1 and 2, in which S and S' are presented for $T=4$ MeV and $T=2$ MeV, respectively, for four sequences of isotopes and for the approximate fission-barrier shapes of uranium isotopes. Considering the intrinsic uncertainties of the extended Thomas-Fermi calculations [6] and the fit [7], the agreement achieved by this fit can be considered as satisfactory. In terms of the entropy the level-density parameter of the Fermi-gas model is $a_F=S/(2T)$ and similarly $a'_F=S'/(2T)$. Figure 3 shows these parameters along the one-dimensional path of the β-stability line, used above in the definition of θ_1. The parameter a_F was calculated for $T=4$ MeV and a'_F for $T=4$, and 0.005 MeV. If S' would be strictly a linear function of T, the level-density parameter would be independent of T. The figure shows to which extent this is actually true. The perfect agreement between a_F and a'_F for $T=4$ MeV shows again the quality of our fit, this time along the line of β stability.

It is useful to compare these level-density parameters, generated by the SkM* potential, with the only independent experimental information on the nuclear entropy, which is derived from the relation between entropy and level density $\rho(E)=\partial S/\partial E e^{S(E)}$. There are several fits of the level-density parameter essentially for nuclei along the line of β stability, using the parametrization $a_F=a_v A+a_s A^{2/3}+a_c A^{1/3}$ and accounting in semiempirical ways for shell and pairing effects and for the influence of low-lying collective states on the level density [12–14]. Depending on the specific way in which these effects are represented, substantially different results are obtained for the Fermi-gas ''background'' contribution to the empirical level density. But all of them yield a larger level density than the SkM* force in the Thomas-

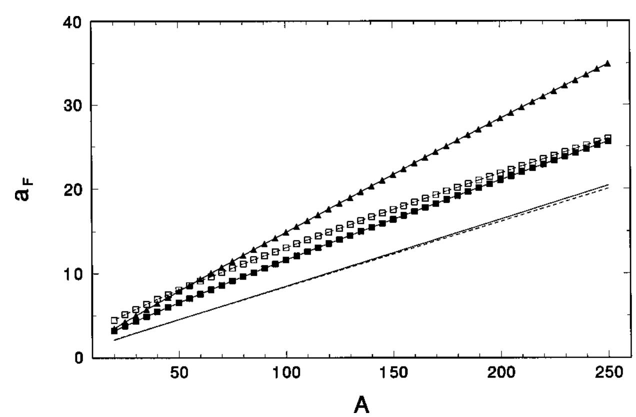

FIG. 3. Level-density parameters a_F, obtained from Ref. [7], and a'_F, both calculated at a temperature of $T=4$ MeV, as a function of the mass number A along the line of β stability (full line)—the two functions are indistinguishable on the scale of this figure—and a'_F, calculated at $T=0.005$ MeV (dashed line); filled squares: empirical fit from Ref. [12], open squares: from Ref. [13], triangles: from an analysis of neutron resonances, Ref. [14].

Fermi mean-field theory and, in fact, than any level density derived from a shell model with a realistic potential. As pointed out already by Bohr and Mottelson [15] it is not clear whether the reasons are deficiencies in accounting for shell and collective-state effects in the empirical fits or whether the effective force in the Thomas-Fermi calculations must be modified to yield a larger effective mass.

III. TEMPERATURE-DEPENDENCE OF THE PAIRING AND WIGNER TERMS IN THE MASS FORMULA

The Wigner and pairing energies in Eq. (1) represent average correlation energies, not contained in the Thomas-Fermi approximation. Their temperature dependence has therefore to be obtained differently. The temperature-dependent version of the one-parameter BCS model used in Ref. [8] can be found in standard text books [16]. We therefore give the results only. There is no pairing above the critical temperature $T_c = 0.568\Delta_0$, where Δ_0 is the zero-temperature energy gap $\overline{\Delta_p}$ or $\overline{\Delta_n}$ of Eq. (5) for protons or neutrons, respectively. The function $\Delta(T)$ is given for all $T \leqslant T_c$ only numerically in terms of T_0 [17]. For temperatures just below T_c one obtains $\Delta(T) = 3.1 T_c (1 - T/T_c)^{1/2}$. For small T a reasonable representation of $\Delta(T)$ is obtained from $\Delta(T)/\Delta_0 = \tanh\{T_c\Delta(T)/(T\Delta_0)\}$ [16]. The parameter δ_{np} in Eq. (5) is not derived from a systematic, microscopic n-body theory, but rather based on qualitative arguments [8]. For simplicity we shall therefore take it to be proportional to $\Delta_p(T)/\Delta_p(0)$ or $\Delta_n(T)/\Delta_n(0)$, whichever is smaller, in order to ensure that above T_c there remains no extra neutron-proton pairing beyond contributions already accounted for in the Thomas-Fermi expression.

In addition to the temperature dependence of the odd-even terms one has to add $\Delta G(T) = T(S_s - S_n)$ to the expression (10), representing the effect on the Gibbs free energy of the change in the entropy when one goes from the superconducting to the normal state. Note that the Thomas-Fermi calculation [6] contains only TS_n. In terms of the critical magnetic field $\mathcal{H}_c(T)$

$$S_s - S_n = \frac{\mathcal{H}_c}{4\pi}\frac{\partial \mathcal{H}_c}{\partial T}.$$

The temperature dependence of $\mathcal{H}_c(T)$ is again given numerically in Ref. [17]. For temperatures just below T_c one has $\mathcal{H}_c(T) = 1.74\mathcal{H}_c(0)(1 - T/T_c)$ in terms of $\mathcal{H}_c(0) = \sqrt{4\pi\rho}\Delta_0$ and $\mathcal{H}_c(T)/\mathcal{H}_c(0) = 1 - (T/T_c)^2$ well below T_c [16]. The density of pairs ρ is given by $\rho_p = \vartheta A^{2/3} Z^{1/3}$ and $\rho_n = \vartheta A^{2/3} N^{1/3}$ for protons and neutrons, respectively, with $\vartheta = (2/3\pi^2)^{1/3} m r_0^2 \hbar^{-2} = 0.019\ \mathrm{MeV}^{-1}$ [8].

A justification of the Wigner term was given in Ref. [18] in terms of the number H of nucleon pairs with the same orbital wave functions. We shall show that the temperature dependence of this quantity can easily be obtained in the constant-level-density model used in the preceding paragraph to calculate pairing correlations. With $\rho = \rho_p + \rho_n$ the average occupation probability of the lth nucleon state with given spin and isospin is $\overline{n_l}(M) = [1 + e^{(4\beta/\rho)(l-M)}]^{-1}$, where M is the number of nucleons with that spin and isospin. In terms of $\overline{n_l}$ we have

$$H(\beta) = \sum_{l=1}^{\infty} \sum_{\{i,j\}} \overline{n_l}(M_i)\overline{n_l}(M_j), \tag{13}$$

where M_i is either the number of protons with spin up or spin down, $M_1 = Z\uparrow, M_2 = Z\downarrow$ or the number of neutrons with spin up or down $M_3 = N\uparrow, M_4 = N\downarrow$. The inner sum in Eq. (13) runs over the six different pairs $\{Z\uparrow Z\downarrow\}$, $\{N\uparrow N\downarrow\}$, $\{Z\uparrow N\uparrow\}$, $\{Z\uparrow N\downarrow\}$, $\{Z\downarrow N\uparrow\}$, and $\{Z\downarrow N\downarrow\}$ between states in the four columns of Fig. 2 in Ref. [18]. We have $|N-Z| = |M_1 + M_2 - M_3 - M_4|$ and for even-even nuclei $M_1 = M_2$, $M_3 = M_4$, for odd-even nuclei $M_1 = M_2 + 1$, $M_3 = M_4$ or $M_1 = M_2$, $M_3 = M_4 + 1$, and for odd-odd systems $M_1 = M_2 + 1$, $M_3 = M_4 + 1$. Using the relations $\Sigma_l \overline{n_l}(M_i) = M_i$ and

$$\overline{n_l}(M_i)\overline{n_l}(M_j) = \frac{e^{-(4\beta/\rho)M_i}}{e^{-(4\beta/\rho)M_i} - e^{-(4\beta/\rho)M_j}}\overline{n_l}(M_i) + \frac{e^{-(4\beta/\rho)M_j}}{e^{-(4\beta/\rho)M_j} - e^{-(4\beta/\rho)M_i}}\overline{n_l}(M_j)$$

we find for the number of pairs (13) in terms of the two temperature-dependent functions

$$f(\beta) = \begin{cases} \coth(\beta/\rho)(|N-Z|) & \text{for even-even nuclei,} \\ \frac{1}{2}[\coth(\beta/\rho)(|N-Z|+1) + \coth(\beta/\rho)(|N-Z|-1)] & \text{for even-odd nuclei} \\ \frac{1}{4}\coth(\beta/\rho)(|N-Z|+2) + 2\coth(\beta/\rho)(|N-Z|) + \coth(\beta/\rho)(|N-Z|-2) & \text{for odd-odd nuclei} \end{cases}$$

and

$$\delta(\beta) = \begin{cases} 0 & \text{for even-even nuclei,} \\ \frac{1}{2}\coth(2\beta/\rho) + [e^{(2\beta/\rho)(|N-Z|+1)} - 1]^{-1} - [e^{(2\beta/\rho)(|N-Z|-1)} - 1]^{-1} & \text{for even-odd nuclei,} \\ \coth(2\beta/\rho) + \frac{1}{2}\coth(\beta/\rho)(|N-Z|+2) - \coth(\beta/\rho)(|N-Z|-2) & \text{for odd-odd nuclei} \end{cases}$$

the expression

$$H(\beta)=\frac{3}{2}A-(|N-Z|)f(\beta)-\delta(\beta).$$

Following the arguments presented in Ref. [18] this yields a Wigner energy of the form $E_W=W[|I|f(\beta)+\delta'(\beta)]$ with

$$\delta'(\beta)=\begin{cases}[\delta(\beta)-1]/A & \text{for } N=Z, \text{ odd-odd nuclei,}\\ 0 & \text{else.}\end{cases}$$

In the limit $T\rightarrow 0$ this expression can easily be shown to give the standard zero-temperature expression for the Wigner energy as given in Ref. [18].

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