

## SHELL AND PAIRING CORRECTIONS FOR ATOMIC CLUSTER PHYSICS

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*Abstract.* The microscopic shell and pairing corrections traditionally used since 1967 in nuclear physics are adapted to atomic clusters. The smoothing effect of pairing corrections is illustrated for a semi-spheroidal sodium cluster with 140 atoms deposited on a planar surface, using as input data the energy levels of a semi-spheroidal harmonic oscillator.

*Key words:* atomic clusters, shell corrections, pairing corrections.

### 1. INTRODUCTION

Single-particle behavior of the nucleon motion is present even in highly collective nuclear phenomena like fission or fusion. Neither phenomenological liquid drop models (LDM) nor microscopic shell models could account for all nuclear properties. Any variant of the LDM fails to explain ground-state deformations and fission fragment mass asymmetry. By subtracting LDM masses from the experimentally determined ones, the extra binding of the nuclei possessing a magic number of nucleons is very clearly seen [1, 2]. Experimentally determined nuclear masses and deformations have been reproduced by Myers and Swiatecki [3] by adding a phenomenological shell correction to the LDM energy. The concept of deformed nuclear shells and the development of Strutinsky [4] microscopic shell correction method allowed to explain the experimentally discovered [5] fission isomers. Shell and pairing correction have been taken into account recently to calculate potential energy surfaces (PES) for cluster emitters [6] and for light and superheavy alpha emitters [7], by using the most advanced asymmetric two center shell model [8].

On the other side, the electronic shell structure in monovalent free-electron metal clusters [9] has shown a strong analogy with the single-particle states of atomic nuclei, despite gross differences in the physical forces binding the two

systems. Moreover, the delocalized electrons of a metallic cluster may be considered to form a Fermi liquid like the atomic nucleus. Consequently several theories and computation techniques from nuclear physics could be adapted to atomic clusters [10]. They have been extensively used particularly to investigate the fission process of ionized clusters as well as the shell and supershell structures, and the shapes of metal clusters [11–13]. Recently we started to apply the macroscopic-microscopic method to semi-spheroidal atomic clusters deposited on a planar surface [14–18].

The purpose of the present work is to outline the method of the nuclear shell and pairing corrections adapted to atomic clusters and to illustrate its capabilities for a semi-spheroidal sodium cluster with 140 atoms. According to the macroscopic-microscopic method, the deformation energy is given by

$$E = E_{LD} + \delta E = E_{LD} + \delta U + \delta P, \quad (1)$$

in which  $E_{LD}$  is the liquid-drop term,  $\delta U$  is the shell correction,  $\delta P$  is the pairing correction and  $\delta E = \delta U + \delta P$ .

## 2. SHELL CORRECTIONS

By summing up the energies  $E_\nu$  of occupied single-particle levels, it is not possible to obtain the true deformation energy. Nevertheless, the variation of  $\sum E_\nu$  with the number of nucleons and with the nuclear deformation are assumed to be correct. The smooth term is replaced by the liquid drop energy  $E_{LD}$ , and the shell correction is obtained by an averaging procedure applied in nuclear physics separately to both neutron and proton level scheme. In atomic cluster physics we have to take into account only one kind of particles, namely the electrons.

$$\delta U = \sum_{\nu=1}^n 2E_\nu - \tilde{U}. \quad (2)$$

We consider infinite-depth potential wells, hence the level spectrum  $\{\epsilon_i\}$  is discrete, and the level density  $g$  may be represented as a sum of delta functions

$$g(\epsilon) = \sum_i \delta(\epsilon - \epsilon_i) = g_0(\epsilon) + \delta g(\epsilon), \quad (3)$$

the smooth component  $g_0(\epsilon)$  describing the average behavior of the level distribution and the oscillating component  $\delta g(\epsilon)$  has a period of about  $\hbar\omega_0$ , which is estimated with the relationship  $\hbar\omega_0 = 41A^{-1/3}$  in nuclear physics. For spheroidally deformed atomic clusters [19] one may use the formula

$$\hbar\omega_0 = \frac{13.72}{r_s R_0} \text{ eV}, \quad (4)$$

where the Wigner-Seitz radius,  $r_s$ , and the radius of a sphere with the same volume,  $R_0$ , are expressed in Å.

## 2.1. SMOOTHED-LEVEL DENSITY

In order to calculate  $\tilde{U}$ , a quantity  $\tilde{g}(\epsilon)$ , approximating the function  $g_0(\epsilon)$  has to be subtracted from  $g(\epsilon)$ . The smoothed-level distribution density may be obtained by averaging the actual distribution over a finite energy interval  $\Gamma \simeq \hbar\omega_0$

$$\tilde{g}(\epsilon) = \int_{-\infty}^{+\infty} \zeta\left(\frac{\epsilon - \epsilon'}{\gamma}\right) g(\epsilon') d\epsilon' = \frac{1}{\gamma} \sum_{i=1}^{\infty} \left(\frac{\epsilon - \epsilon'}{\gamma}\right), \quad (5)$$

where  $\gamma = \Gamma/\hbar\omega_0$ . The most utilized smoothing function is

$$\zeta(x) = \frac{1}{\sqrt{\pi}} e^{-x^2} f_m(x), \quad (6)$$

where

$$f_m(x) = L_m^{1/2}(x^2) = \sum_{k=0}^m a_{2k} H_{2k}(x). \quad (7)$$

In this way, not only  $g_0$ , but also its  $2m$  first derivatives are correctly reproduced. This definition of  $g(\epsilon)$ , where  $L_m^{1/2}(x^2)$  is an associate Laguerre polynomial, is equivalent to take into account the first  $m$  terms of the expansion of delta functions sum in a Hermite polynomial series  $H_{2k}(x)$  with a Gaussian weighting function. As lower order Hermite polynomials are oscillating slower than the higher ones, a few first polynomials give the slowly varying contribution to  $g(\epsilon)$ , the other terms – the fluctuating one. The  $f_m$  function with the coefficients  $a_{2k} = (-1)^k / (2^{2k} k!)$ ;  $a_0 = 1$ , connected through the recurrency relationship  $a_{2k} = -a_{2k-2}/4k$  is called sometimes curvature correction to the level density. The correction, of the order  $2m$ , introduced in this way, provides for internal consistency of the smoothing procedure of  $g(\epsilon)$ , expressible as a  $2m$ -order polynomial in  $\epsilon$ .

The Gaussian weighting function is effectively different from zero within a range controlled by the scale factor  $\gamma$ . The quantities  $\gamma$  and  $m$  have no physical meaning; the results have to be independent of their value. This can be checked by testing the “plateau condition”

$$\left. \frac{\partial(\delta U)}{\partial \gamma} \right|_{\gamma=\gamma_0} = 0; \quad \left. \frac{\Delta[\delta U(\gamma_0)]}{\Delta m} \right|_{m=m_0} = 0. \quad (8)$$

As far as the order  $m$  is increased, the range of  $\gamma$  fulfilling this condition is increasing at the same time. The  $m$ -value can not be, however, too large, because in such a case  $\tilde{g}$  could include a contribution from  $\delta g$ . Frequently,  $m = 3$  for the harmonic oscillator. Besides  $f_0(y) = 1$  and  $f_1(y) = 3/2 - y$ , one has

$$f_2(y) = 15/8 - y(5 - y)/2; \quad f_3(y) = 35/16 + y[y(7/4 - y/6) - 35/8], \quad (9)$$

where

$$y = x^2 = \left( \frac{\epsilon - \epsilon_i}{\gamma} \right)^2. \quad (10)$$

The function  $\zeta(x)$  has nonvanishing values in a small range around the origin. The levels  $\epsilon_i$  situated far enough from the energy  $\epsilon$ , for which the level density has to be calculated, are insignificantly contributing to  $\tilde{g}(\epsilon)$ . Thus, for  $|x| \geq 3$ ,  $|\zeta| \leq 6.9 \cdot 10^{-5}$  when  $m = 0$ ;  $5.2 \cdot 10^{-4}$  at  $m = 1$ ;  $1.4 \cdot 10^{-3}$  at  $m = 2$ ; and  $1.2 \cdot 10^{-3}$  at  $m = 3$ . In this way the summation over  $i$  may be restricted to the levels satisfying the above condition.

At a given  $\gamma$ , all states producing a finite contribution to  $\tilde{g}$ , for  $\epsilon \leq \tilde{\lambda}$ , where  $\tilde{\lambda} = E_F / \hbar \omega_0$  represents the Fermi energy, must be included. The level density fluctuations of faraway occupied levels could affect the level density  $\tilde{g}$  and, consequently, the uniform level distribution energy  $\tilde{U}$ . This fact is of great importance for calculation of the shell correction obtained as a small difference of two large quantities.

In case of  $m = 3$ , frequently used in practice, the level density of the continuous level distribution is expressed as

$$\tilde{g}(\epsilon) = \left\{ \sum_{i=1}^{n_m} [2.1875 + y_i(y_i(1.75 - y_i/6) - 4.375)] e^{-y_i} \right\} (1.77245385\gamma)^{-1} \quad (11)$$

The summation is performed up to the level  $n_m$  fulfilling the condition  $|x_i| \geq 3$ .

## 2.2. FERMI ENERGY OF THE UNIFORM DISTRIBUTION OF LEVELS

The Fermi energy  $\tilde{\lambda}$  of a smoothed level distribution may be obtained from

$$N = 2 \int_{-\infty}^{\tilde{\lambda}} \tilde{g}(\epsilon) d\epsilon, \quad (12)$$

where  $N$  is the number of atoms in the cluster. By substituting the above expressions, we obtain

$$N = \frac{2}{\sqrt{\pi}} \sum_{i=1}^{\infty} \int_{-\infty}^{x_{iF}} f_m(x_i^2) e^{x_i^2} dx_i. \quad (13)$$

After substitution and integration we get

$$N = \sum_{k=1}^{\infty} \left[ 1 + \operatorname{erf}(x_{iF}) - \frac{2}{\sqrt{\pi}} e^{-x_{iF}^2} \sum_{k=1}^m a_{2k} H_{2k-1}(x_{iF}) \right] \quad (14)$$

or

$$N = \sum_{i=1}^{\infty} \left[ 1 + \operatorname{erf}(x_{iF}) + \frac{e^{-x_{iF}^2}}{\sqrt{\pi}} P_m(x_{iF}) \right], \quad (15)$$

where  $P_0 = 0$ ;  $P_1 = x_{iF}$ ;  $P_2 = -x_{iF}(0.5y_{iF} - 1.75)$ ;  $P_3 = x_{iF}[y_{iF}(y_{iF} - 8) + 14.25]/6$ ;  $y_{iF} = x_{iF}^2 = (\tilde{\lambda} - \epsilon_i)^2/\gamma^2$ .

This nonlinear equation, with  $\tilde{\lambda}$  as unknown, can be solved numerically by using an iteration scheme (Newton-Raphson), which refines an initial guess  $\tilde{\lambda}_0 = (\epsilon_n + \epsilon_{n+1})/2$  with  $n = N/2$ . For levels far away from the Fermi energy, satisfying the relationship  $|x_{iF}| \geq 3$ , we can approximate  $N = \sum_i 1$ , if  $\epsilon < \tilde{\lambda}$  and  $N = 0$ , if  $\epsilon > \tilde{\lambda}$ .

### 2.3. SMOOTHED-LEVEL ENERGY AND SHELL CORRECTIONS

The total energy of the uniform level distribution,  $\tilde{U}$ , reproduces the average behavior of the total energy but not its local fluctuations. It can be obtained from the relationship

$$\tilde{u} = \tilde{U}/\hbar\omega_0 = 2 \int_{-\infty}^{\tilde{\lambda}} \tilde{g}(\epsilon) \epsilon d\epsilon, \quad (16)$$

where  $\tilde{g}(\epsilon)$  is measured in number of levels per  $\hbar\omega_0$  energy interval. By inserting the above results and performing the integrations we get

$$\tilde{u} = \sum_{i=1} \left\{ \epsilon_i \left[ 1 + \operatorname{erf}(x_{iF}) + \frac{e^{-x_{iF}^2}}{\sqrt{\pi}} P_m(x_{iF}) \right] + \frac{\gamma e^{-x_{iF}^2}}{\sqrt{\pi}} Q_m(x_{iF}) \right\}, \quad (17)$$

where

$$Q_m = 1 + \sum_{k=1}^m a_{2k} (H_{2k} + 4kH_{2k-2}) = -a_{2m} H_{2m} \quad (18)$$

and  $Q_0 = -1$ ;  $Q_1 = y_{iF} - 0.5$ ;  $Q_2 = y_{iF}(1.5 - 0.5y_{iF}) - 0.375$ ;  $Q_3 = \{y_{iF}[11.25 + y_{iF}(y_{iF} - 7.5)] - 1.875\}/6$ .

In units of  $\hbar\omega_0$  the shell corrections are calculated as

$$\delta u(n, \varepsilon) = \sum_{i=1}^n 2\varepsilon_i(\varepsilon) - \tilde{u}(n, \varepsilon), \quad (19)$$

where the summation is performed over  $n = N/2$  particles.

The dependence of  $\delta u$  on  $N$  at a given deformation, say  $\varepsilon = 3(c-a)/(2c+a) = 0$  are showing minima at the magic numbers (in this case spherical magic numbers). The most pronounced shell structure, except  $\varepsilon = 0$ , takes place at  $\varepsilon = 0.6$  (spheroid with ratio of semiaxes  $c/a = 2$ ).

As far as the maximum shell order taken into consideration, there is no reason to calculate all levels corresponding to a given  $N_m$ , because some are out of interest, exceeding the Fermi energy by at least  $3\gamma$ . It is desirable to establish the energy limit  $\varepsilon_{lim}$  and to calculate the maximum shell of interest at a given deformation. Thus, for  $\varepsilon_{lim} = 13$ , at a deformation as large as  $\varepsilon = 1$ , there are (coming down) levels from many shells up to  $N_m = 20$  in case of an harmonic oscillator without spin orbit coupling.

### 3. PAIRING CORRECTIONS

Among the short-range residual interactions to be considered in nuclear theory, the pairing interaction is the most important [1, 20, 21]. There is much experimental evidence of pairing correlations in nuclei: the energy gap found in the excitation energy spectra of even-even deformed nuclei; the even-odd effect in nuclear masses; low-lying vibrational  $2^+$  states of even-even nuclei in the vicinity of closed shells; the spin zero of even-even nuclei and the spin determined by the last unpaired nucleon of odd-A nuclei; the existence of other spherical nuclei around the magic ones, etc. In a very fast rotating nucleus, pairing correlations disappear (Mottelson-Valatin effect).

Bohr *et al.* [22] suggested the analogy with electron correlations in the theory of superconductivity, developed by Bardeen, Cooper and Schrieffer (BCS) [21]. Second quantization is usually used. The Bogoliubov-Valatin transformation allows to work with independent quasiparticles instead of interacting particles. Pairing corrections have been introduced by Strutinsky in the same time with shell corrections.

We consider, like in the preceding section, a set of doubly degenerate levels  $\{\varepsilon_i\}$  expressed in units of  $\hbar\omega_0$ . In the absence of pairing field, the first  $N/2$  levels

are occupied, from a total number of  $n_t$  levels available. Only few levels below ( $n$ ) and above ( $n'$ ) the Fermi energy are contributing to the pairing correlations. Usually  $n' = n$ . If  $\tilde{g}_s$  is the density of states at Fermi energy obtained from the shell correction calculation  $\tilde{g}_s = dN/d\epsilon$ , expressed in number of levels per  $\hbar\omega_0$  spacing, the level density is half of this quantity:  $\tilde{g}_n = \tilde{g}_s/2$ .

We can choose as computing parameter, the cut-off energy (in units of  $\hbar\omega_0$ ),  $\Omega \approx 1 \gg \tilde{\Delta}$ . Let us take the integer part of the following expression

$$\Omega\tilde{g}_s/2 = n = n'. \quad (20)$$

When from calculation we get  $n > N/2$ , we shall take  $n = N/2$  and similarly if  $n' > n_t - N/2$  we consider  $n' = n_t - N/2$ .

The gap parameter  $\Delta = |G| \sum_k u_k v_k$  and the Fermi energy with pairing correlations  $\lambda$  (both in units of  $\hbar\omega_0$ ) are obtained as solutions of a nonlinear system of two BCS equations

$$n' - n = \sum_{k=k_i}^{k_f} \frac{\epsilon_k - \lambda}{\sqrt{(\epsilon_k - \lambda)^2 + \Delta^2}}, \quad (21)$$

$$\frac{2}{G} = \sum_{k=k_i}^{k_f} \frac{1}{\sqrt{(\epsilon_k - \lambda)^2 + \Delta^2}}, \quad (22)$$

where  $k_i = N/2 - n + 1$ ;  $k_f = N/2 + n'$ .

The pairing interaction strength,  $G$ , is calculated from a continuous distribution of levels

$$\frac{2}{G} = \int_{\tilde{\lambda}-\Omega}^{\tilde{\lambda}+\Omega} \frac{\tilde{g}(\epsilon)d\epsilon}{\sqrt{(\epsilon - \tilde{\lambda})^2 + \tilde{\Delta}^2}}, \quad (23)$$

where  $\tilde{\lambda}$  is the Fermi energy deduced from the shell correction calculations and  $\tilde{\Delta}$  is the gap parameter, obtained from a fit to experimental data. In nuclear physics it is usually taken as  $\tilde{\Delta} = 12/\sqrt{A}\hbar\omega_0$ . From the above integral we get

$$\frac{2}{G} \approx 2\tilde{g}(\tilde{\lambda}) \ln\left(\frac{2\Omega}{\tilde{\Delta}}\right). \quad (24)$$

In order to obtain similar results with those plotted in Fig. 1, we propose  $\tilde{\Delta} = 0.8/[\sqrt{N}\hbar\omega_0]$  for spheroidal atomic clusters, and

$$\tilde{\Delta} = \frac{0.6}{\sqrt{N}\hbar\omega_0} \quad (25)$$

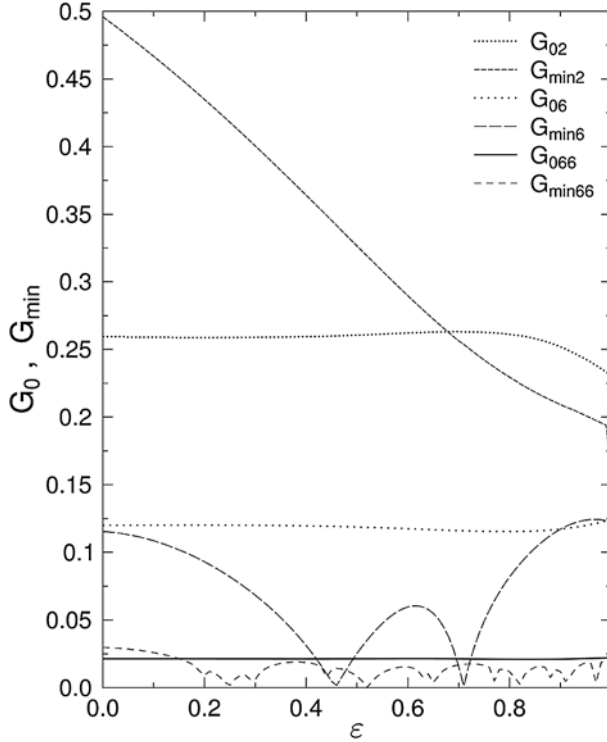


Fig. 1 – The smooth value of the pairing strength,  $G_0 = 1/[\tilde{g} \ln(2/\tilde{\Delta})]$ , with  $\tilde{\Delta} = 12/(\sqrt{A} \hbar \omega_0)$  and the quantity  $G_{min} = 2 \sum |\epsilon_k - \lambda|^{-1}$  for different values of nucleon numbers  $A/2 = 2, 6,$  and  $66$  versus the deformation  $\epsilon = 3(c-a)/(2c+a)$ , where  $c$  and  $a$  are the semiaxes of the spheroid. The three-dimensional spheroidal harmonic oscillator level scheme is used.

for semi-spheroidal atomic clusters. Real positive solutions of BCS equations are allowed if

$$\frac{G}{2} \sum_k \frac{1}{|\epsilon_k - \lambda|} > 1 \quad (26)$$

*i.e.* for a pairing force ( $G$ -parameter) large enough at a given distribution of levels. The system can be solved numerically by Newton-Raphson method refining an initial guess

$$\begin{aligned} \lambda_0 &= (n_s \epsilon_d + n_d \epsilon_s) / (n_s + n_d) + G(n_s - n_d) / 2, \\ \Delta_0^2 &= n_s n_d G^2 - (\epsilon_d - \epsilon_s) / 4, \end{aligned} \quad (27)$$

where  $\epsilon_s$ ,  $n_s$  are the energy and degeneracy of the last occupied level and  $\epsilon_d$ ,  $n_d$  are the same quantities for the next level. Solutions around magic numbers, when  $\Delta \rightarrow 0$ , have been derived by Kumar *et al.* [23]. In Fig. 1 we have plotted the smooth value of the pairing strength,  $G_0 = 1/[\tilde{g} \ln(2/\tilde{\Delta})]$ , with  $\tilde{\Delta} = 12/(\sqrt{A} \hbar \omega_0)$  and the quantity  $G_{min} = 2 \sum |\epsilon_k - \lambda|^{-1}$  for different values of nucleon numbers  $A/2 = 2, 6,$  and  $66$  versus the deformation  $\epsilon = 3(c-a)/(2c+a)$ , where  $c$  and  $a$  are the semiaxes of the spheroid. The three-dimensional spheroidal harmonic oscillator level scheme is used.

As a consequence of the pairing correlation, the levels situated below the Fermi energy are only partially filled, while those above the Fermi energy are partially empty; there is a given probability for each level to be occupied by a quasiparticle

$$v_k^2 = \frac{1}{2} \left[ 1 - \frac{\epsilon_k - \lambda}{\sqrt{(\epsilon_k - \lambda)^2 + \Delta^2}} \right] \quad (28)$$

or a hole

$$u_k^2 = 1 - v_k^2. \quad (29)$$

Only the levels in the near vicinity of the Fermi energy (in a range of the order of  $\Delta$  around it) are influenced by the pairing correlations. For this reason, it is sufficient for the value of the cut-off parameter to exceed a given limit  $\Omega \gg \tilde{\Delta}$ , the value in itself having no significance.

At a given deformation, the pairing correction to the deformation energy is calculated by using the relationship  $\delta p = p - \tilde{p}$ , representing the difference between the pairing correlation energies for the discrete level distribution

$$p = \sum_{k=k_i}^{k_f} 2v_k^2 \epsilon_k - 2 \sum_{k=k_i}^{N/2} \epsilon_k - \frac{\Delta^2}{G} \quad (30)$$

and for the continuous level distribution

$$\tilde{p} = -(\tilde{g}\tilde{\Delta}^2)/2 = -(\tilde{g}_s\tilde{\Delta}^2)/4. \quad (31)$$

A term  $G \left( \sum_{k_i}^{k_f} v_k^4 - \sum_{k_i}^{N/2} 1 \right)$  is supposed to be negligibly small. Compared to shell correction, the pairing correction is out of phase and smaller, as may be seen in Fig. 2 on the example of the semi-spheroidal  $\text{Na}_{140}$  cluster. As a result of

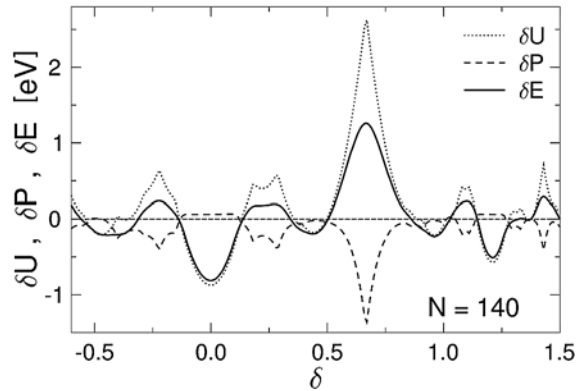


Fig. 2 – Shell corrections,  $\delta U$ , pairing corrections,  $\delta P$ , and their sum,  $\delta E = \delta U + \delta P$ , for semi-spheroidal harmonic oscillator energy levels using the deformation parameter  $\delta = 2(c - a)/(c + a)$ , where  $c$  and  $a$  are the semi-axes of the semi-spheroid. A cluster of Na with 140 atoms is considered.

taking into account the pairing corrections,  $\delta P$ , the total shell and pairing corrections,  $\delta E = \delta U + \delta P$ , are smoother compared to  $\delta U$ .

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