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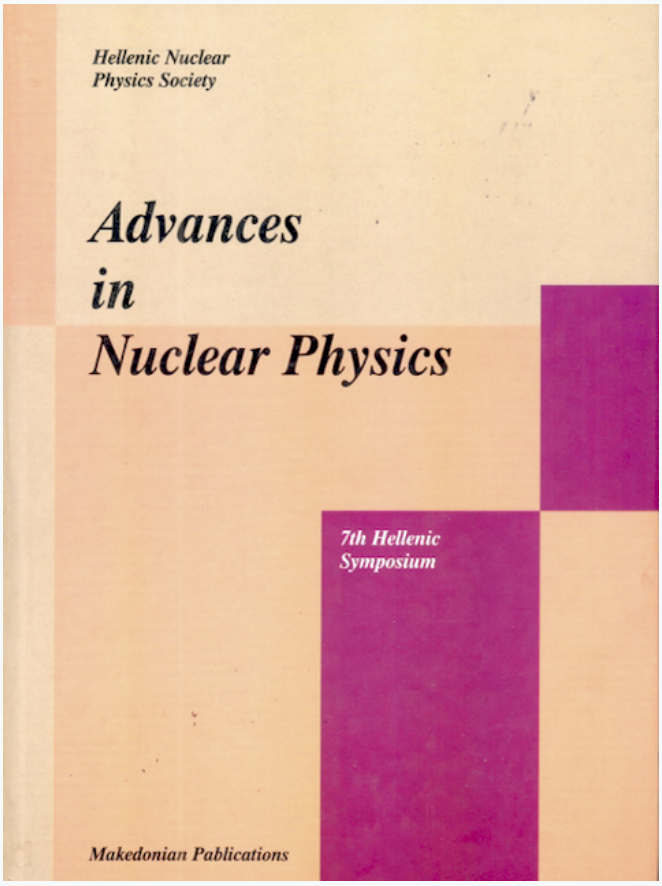

Determination of the Harmonic Oscillator Energy Level Spacing for Atomic Clusters

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Determination of the Harmonic Oscillator Energy Level Spacing for Atomic Clusters

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Abstract

The harmonic oscillator energy level spacing $\hbar\omega$ for atomic clusters as a function of the particle number N is expressed analytically in terms of the parameters of a Woods-Saxon (or Symmetrized Woods-Saxon) potential which approximates the effective spherical self-consistent jellium model potential. The expressions derived depend on the particular scheme adopted to approximate the potential by the harmonic oscillator one and on the assumed dependence of the potential radius R on N. It is also observed, considering the case of sodium clusters,that for large N the expressions of $\hbar\omega$ are in good agreement with the well known expression of $\hbar\omega$ in terms of the Wigner-Seitz radius.

The determination of the harmonic oscillator (HO) energy level spacing $\hbar\omega$ for atomic clusters and its variation with the particle number N is discussed in literature [1-3], by adopting the approach which is traditionally followed in Nuclear Physics [4] and adjusting it to the case of atomic clusters. The essential idea is that the energy scale is directly related to the size of the cluster. It is assumed that the cluster valence electrons have a constant density ϱ_0 equal to the bulk conduction electron density and the size of the (spherical) cluster is normalized so that it contains exactly N electrons,that is $R_0 = r_s N^{1/3}$,where $r_s = (\frac{3}{4\pi\varrho_0})^{1/3}$ is the Seitz-Wigner radius. By equating the mean-square radius of such a density distribution to that which results on the basis of the HO model in the case of large N, there results the following expression for $\hbar\omega$:

$$\hbar\omega = (\frac{375}{8})^{\frac{1}{3}} \frac{\hbar^2}{2m_e} {r_s}^{-2} N^{-\frac{1}{3}} = 49 {r_s}^{-2} N^{-\frac{1}{3}} \qquad (1)$$

where r_s is in atomic units and $\hbar\omega$ in eV. It is of interest to note the similarity of expression (1) with the "order of magnitude" expression for $\hbar\omega$ which results by dividing the bulk Fermi energy ε_F ($\varepsilon_F = (\frac{9\pi}{4})^{\frac{2}{3}} \frac{\hbar^2}{2m_e} {r_s}^{-2}$) by the number of the separate energy levels in a spherical potential which is of the order $N^{\frac{1}{3}}$ [5].

An improved expression for $\hbar\omega$ has also been derived [1-3] by taking into account that the electron density calculated in self-consistent jellium calculations extends beyond the boundary at $R = R_0$. This "spill-out" of the electrons (in the terminology used in this context [3,6,7,8]) was modeled by Clemenger by using an additive correction t to R_0.Thus, the constant density of the N electrons is adjusted to fill a sphere of an effective radius $R_0 + t$. Taking into account this correction, there results the following improved expression for $\hbar\omega$:

$$\hbar\omega = 49 r_s^{-2} N^{-\frac{1}{3}} (1 + \frac{t}{r_s N^{\frac{1}{3}}})^{-2} \tag{2}$$

The value of t may be estimated from polarizability measurements. For sodium ($r_s = 4$) the estimate is $t = 1.44$ a.u.[1,2]

The object of this note is to consider another, quite different, analytic way of determining the HO energy level spacing , which is followed for the first time to our knowledge, pertaining to atomic clusters.This is done by using previous experience in Hypernuclear Physics. Namely, we use the approach described in refs.[9,10] and we adjust it to the cluster case. For this purpose, the effective spherical self-consistent jellium model potential is firstly parametrized by a Woods-Saxon (or symmetrized Woods-Saxon [11]) potential, as has been done by several authors [6,12]. Secondly, this Woods-Saxon potential:

$$V_{W.S.}(r) = -\frac{V_0}{1 + e^{\frac{r-R}{a}}} \tag{3a}$$

or the corresponding symmetrized one:

$$V_{SWS} = -V_0 \left[[1 + exp(\frac{r-R}{a})]^{-1} + [1 + exp(\frac{-r-R}{a})]^{-1} - 1 \right] \tag{3b}$$

is approximated in the interior of the cluster and as far as possible near its surface by an harmonic oscillator potential $V_{HO}(r)$:

$$V_{HO}(r) = -D + \frac{D}{R_0^2} r^2 \tag{4}$$

where $D = V_0$ and R_0 is the distance from the origin where V_{HO} becomes zero. (The R and R_0 used in expressions (3) and (4) should not be confused with the R and R_0 used earlier). More precisely, we make a sort of "best approximation in the mean of the potential $V(r) = -V_0 f(r)$ by the $V_{HO}(r)$ "by choosing R_0 in such a way that the following condition is satisfied:

$$J = \int_0^{R_0} |V(r) - V_{HO}(r)|^2 \, dr = min \tag{5}$$

The optimum value of $R_0 = R_m$ is determined by means of the relation (see

ref 9.).

$$[\frac{8}{15} + f^2(R_m)]{R_m}^3 = 4\int_0^{R_m} f(r)r^2dr \tag{6}$$

which in the case of the Woods-Saxon potential ($V(r) = V_{W.S.}$) or the symmetrized one V_{SWS} leads to the simple expression:

$$R_m = (\frac{5}{2})^{\frac{1}{3}} R[1 + (\frac{\pi a}{R})^2]^{\frac{1}{3}} \tag{7}$$

by omitting terms which are negligible, unless N is too small.

The expression for $\hbar\omega$ follows immediately, since from (4) the spring constant $k = m\omega^2$ equals to $2\frac{V_0}{R_0^2}, R_0 = R_m$ Thus, we obtain the following expression of $\hbar\omega$ in terms of the potential parameters V_0, a and R of the Woods-Saxon (or symmetrized Woods-Saxon) potential:

$$\hbar\omega = [\frac{\hbar^2}{m_e}2V_0]^{\frac{1}{2}} R_m^{-1} = [\frac{\hbar^2}{m_e}2V_0]^{\frac{1}{2}}(\frac{2}{5})^{\frac{1}{3}} R^{-1}[1 + (\frac{\pi a}{R})^2]^{-\frac{1}{3}} \tag{8}$$

Before we proceed, it is instructive to recall that an harmonic oscillator potential, which is very useful because of its valuable analytic advantages, has also its shortcomings. For example, physical quantities sensitive to the surface region are not reproduced well by such a potential. Related to this is the remark made in ref.[13] where a truncated (at R_0) HO potential was used for clusters in the context of the discussion of magic numbers and there was pointed out that it does not lead to the same ordering of levels as the more realistic Woods-Saxon potential. Difficulties of this type are not surprising in view of relevant experience from Nuclear Physics and may be faced by adding to the HO potential a negative term which is proportional to the square of the orbital angular momentum. The variation of $\hbar\omega$ with N, we are discussing here, has, however, a rather different scope. It aims at providing some information on how an "average energy level spacing" of the self-consistent potential varies with the particle number. We also get an idea of how the shape [14] of this self-consistent potential changes "on the average" (i.e. neglecting its fluctuations and its behaviour near the surface of the system) as the particle number varies.

It is clear that in the approach we follow the variation of $\hbar\omega$ with the particle number N depends essentially on the variation of the potential radius R with N. The potential depth V_0 and the parameter a which determines the surface thickness of the potential are roughly independent of N and are given by their best fit values in a fitting procedure [11]. Regarding the expression of $R = R(N)$, the following three possibilities are considered: i)$R = r_0 N^{\frac{1}{3}}$ ii)$R =$

$r_0 N^{\frac{1}{3}} + b$ and iii)

$$R = \frac{1}{2^{\frac{1}{3}}} r_0' N^{\frac{1}{3}} \left[\left[1 + \left[1 + \frac{4}{27} (\frac{\pi a}{r_0' N^{\frac{1}{3}}})^6 \right]^{\frac{1}{2}} \right]^{\frac{1}{3}} + \left[1 - \left[1 + \frac{4}{27} (\frac{\pi a}{r_0' N^{\frac{1}{3}}})^6 \right]^{\frac{1}{2}} \right]^{\frac{1}{3}} \right] \tag{9}$$

where $r_0' = r_0(1 + \frac{\beta}{N})^{\frac{1}{3}}$. The first expression is the mostly used [6, 12], while the ii)[12] and iii) are expected to be better approximations for comparatively smaller clusters and are discussed in ref.[11] where the parameters are determined by least-squares fits. The quality of the fit is almost the same for these two expressions, although with the latter one it is marginally better. The complex expression iii) originates from the observation that the volume integral of the (symmetrized) Woods-Saxon potential for each cluster varies with N as $cN + d$, where c and d are constants. The parameters r_0 and β are given by the expressions $r_0 = (\frac{3c}{4\pi V_0})^{\frac{1}{3}}$ and $\beta = \frac{d}{c}$. Expression (9) is somewhat different from the corresponding "rigid-core" model expression for R, which has been used in Hypernuclear [15-17] and Nuclear [18] Physics, the difference being that in the latter expression $\beta = 0$ and therefore $r_0' = r_0$.

We finally note that the values of the parameters in the above expressions of R depend on the region of N used for the fit. Here we shall use mainly expression (9). The best fit values, if the fitting of the symmetrized Woods-Saxon potential is made to the Ekardt [19] effective potentials for sodium clusters with $8 \leq N \leq 198$, are [11]: $V_0 = 6.03eV, r_0 = 2.295\mathring{A}, a = 0.78\mathring{A}, \beta = 10.49$, while, if the fitting is restricted to the potentials of the above clusters with $N \geq 90$, the corresponding best fit values are : $V_0 = 6.02eV, r_0 = 2.37\mathring{A}, a = 0.88\mathring{A}, \beta = 1.064$.

Expression (8) may be expanded in powers of N as follows:

$$\hbar\omega = \left[\frac{\hbar^2}{m_e} \frac{2V_0}{{r_0}^2} \right]^{\frac{1}{2}} (\frac{2}{5})^{\frac{1}{3}} N^{-\frac{1}{3}} \left[1 - \frac{\beta}{3} N^{-1} + \frac{2\beta^2}{3^2} N^{-2} - \right.$$
$$\left. - \frac{14\beta^3}{3^4} N^{-3} + \frac{35\beta^4}{3^5} N^{-4} + ... \right] \tag{10}$$

We may notice that, at least to order N^{-4}, the above expression for $\hbar\omega$ is independent of the parameter α and only negative integral powers of N appear in the expansion. The coefficients of the terms given in (10), except the first one, become zero if $\beta = 0$.

It is seen from expression (10) that the leading term which determines the HO energy level spacing for large particle number is:

$$\hbar\omega_{as} = \left[\frac{\hbar^2}{m_e}\frac{2V_0}{{r_0}^2}\right]^{\frac{1}{2}} (\frac{2}{5})^{\frac{1}{3}} N^{-\frac{1}{3}} \tag{11}$$

For sodium clusters, we obtain, using the first set of the parameters, $\hbar\omega_{as} = 3.08N^{-\frac{1}{3}}$. If we use the second set of parameters which is more appropriate for large N, we obtain $\hbar\omega_{as} = 2.978N^{-\frac{1}{3}}$. These expressions are in very good agreement with the expression $\hbar\omega = 3.06N^{-\frac{1}{3}}$, which follows from expression (1) applied to the present case ($r_s = 4$). For smaller values of N the $\hbar\omega$ given by expression (8) deviates from its asymptotic behaviour and leads to smaller values, in comparison with those resulting from the asymptotic expression. This is clear from fig 1, where $\hbar\omega$ and $\hbar\omega_{as}$ are plotted. The same is the case if expression (1) and (2) are compared. In this case, however, the difference in the values of the HO energy level spacing is larger.

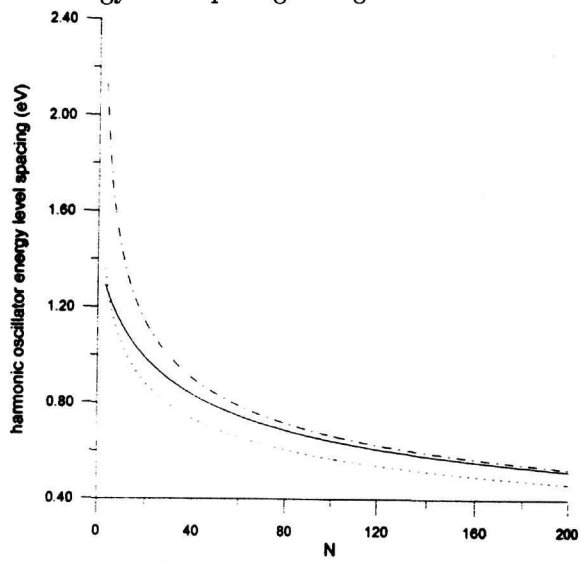

Fig. 1 The HO energy level spacing $\hbar\omega$ versus the particle number N, using the potential parameters $V_0 = 6.03eV, r_0 = 2.295\mathring{A}, a = 0.78\mathring{A}, \beta = 10.49$. The solid line is obtained by means of formula (8), while the dot-dashed line is its asymptotic expression $\hbar\omega_{as}$. The dotted line is obtained by means of formula (2) of Clemenger.

It is a useful to point out, that the way in which one "adjusts" the HO potential is not unique [9]. For example, if the harmonic oscillator is determined by the conditions: $D = V_0$ and $V_{HO}(R) = V_{W.S.}(R)$ that is $R_0 = \sqrt{2}R$ the "reduction factor" would be $\frac{1}{\sqrt{2}} \simeq 0.707$, which does not differ very much from $(\frac{2}{5})^{\frac{1}{3}} \simeq 0.737$, deduced from (8) and (10).

Other possible integral conditions, which differ from (5) may be considered [9]. Another possibility, which in fact has not been used in connection to hy-

pernuclei so far and might be worthy to be investigated would be to determine D and R_0 in (4) by means of the conditions:

$$\int_0^{R_0} V_{HO}(r)dr = \int_0^{\infty} V(r)dr$$

and

$$\int_0^{R_0} V_{HO}(r)r^2dr = \int_0^{\infty} V(r)r^2dr \tag{12}$$

These conditions lead to the following expression of R_0 in terms of the parameters of $V_{W.S.}$:

$$R_0 = (\frac{5}{3})^{\frac{1}{2}}R[1 + (\frac{\pi a}{R})^2]^{\frac{1}{2}} \tag{13}$$

Furthermore, the parameter D of the oscillator potential is given now by the expression:

$$D = V_0\frac{3}{2}(\frac{3}{5})^{\frac{1}{2}}\left[1 + (\frac{\pi a}{R})^2\right]^{-\frac{1}{2}} \tag{14}$$

In contrast to the previous cases, the parameter D depends now on N and in addition D is (usually) larger than V_0. The expression of $\hbar\omega$ is in this case:

$$\hbar\omega = [\frac{\hbar^2}{m_e}2V_0]^{\frac{1}{2}}(\frac{3}{2})^{\frac{1}{2}}(\frac{3}{5})^{\frac{3}{4}}R^{-1}[1 + (\frac{\pi a}{R})^2]^{-\frac{3}{4}} \tag{15}$$

This is expanded as follows, using expression (9) for R:

$$\hbar\omega = (\frac{3}{2})^{\frac{1}{2}}(\frac{3}{5})^{\frac{3}{4}}\left[\frac{\hbar^2}{m_e}\frac{2V_0}{r_0^2}\right]^{\frac{1}{2}}N^{-\frac{1}{3}}\left[1 - \frac{5}{12}(\frac{\pi a}{r_0})^2N^{-\frac{2}{3}} - \frac{\beta}{3}N^{-1} + \right.$$

$$\left. + \frac{5}{288}(\frac{\pi a}{r_0})^4N^{-\frac{4}{3}} + \frac{5\beta}{12}(\frac{\pi a}{r_0})^2N^{-\frac{5}{3}} + ...\right] \tag{16}$$

One should notice the additional terms ($\sim N^{-\frac{2}{3}}, \sim N^{-\frac{4}{3}} etc.$) appearing in (16) in comparison with (10). It is further seen that because of the different "reduction factor": $(\frac{3}{2})^{\frac{1}{2}}(\frac{3}{5})^{\frac{3}{4}} \simeq 0.835$ (instead of $(\frac{2}{5})^{\frac{1}{3}} \simeq 0.737$ in the previous case) the values of $\hbar\omega_{as}$ will be about 11% larger in this case. In fig.2 the values of $\hbar\omega$ from (15) and the corresponding values of the leading term $\hbar\omega_{as}$ have been plotted versus N using the set of parameters $V_0 = 6.03eV, r_0 = 2.295\mathring{A}, a = 0.78\mathring{A}, \beta = 10.49$. It is seen again that the values of $\hbar\omega$ lie between the values of $\hbar\omega_{as}$ and those obtained by means of Clemenger's expression (2). For very small N the values obtained from the two expressions of $\hbar\omega$ approach each other considerably.

In the previous analysis we have considered, as usually, electronic states. Analogous treatment can be made for the atoms in the cluster if the corresponding potential is given. Approximation of the latter by harmonic oscillator potentials has been useful [20].

In conclusion, the knowledge of the effective self-consistent potential in jellium model cluster calculations offers the possibility of determining the HO energy level spacing, in quite a different analytic way from the one, which has been followed so far in Cluster Physics.

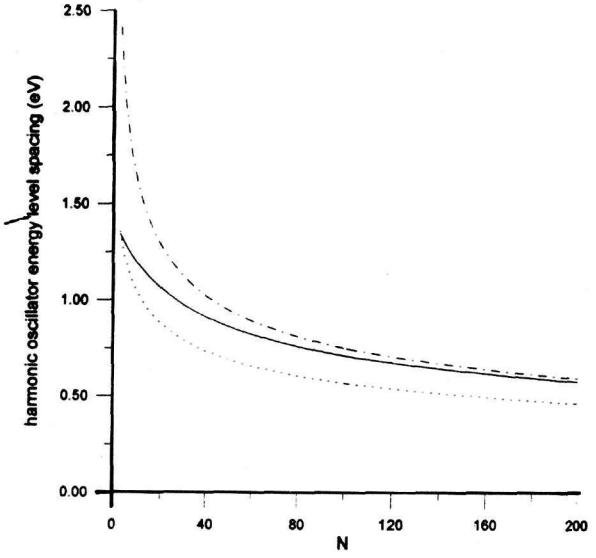

Fig. 2 The HO energy level spacing $\hbar\omega$ versus the particle number N, using the potential parameters $V_0 = 6.03eV, r_0 = 2.295\mathring{A}, a = 0.78\mathring{A}, \beta = 10.49$. The solid line is obtained by means of formula (15), while the dot-dashed line is its asymptotic expression $\hbar\omega_{as}$. The dotted line is obtained by means of formula (2) of Clemenger.

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