

## Static electric polarizabilities and collective resonance frequencies of small metal clusters

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The static electric dipole polarizabilities and surface plasma resonance frequencies of small metal clusters are calculated analytically. The resonance frequencies are evaluated self-consistently and are shown to depend sensitively on the density distribution of the valence electrons. The latter is described by the recently developed statistical approach to the electronic structure of metal clusters. Electric polarizabilities are derived from the results for the resonance frequencies and general sum rules. Very good agreement with experimental data is found.

### I. INTRODUCTION

This paper is devoted to a calculation of static electric polarizabilities and collective resonance frequencies of small metal clusters. Both have long been subjects of active interest (see, e.g., the reviews<sup>1-3</sup>).

The problem of static electric polarizability of size-quantized systems attracted attention when it was predicted<sup>4</sup> that very small metal particles would exhibit an orders-of-magnitude increase in polarizability compared to the classical value for a conducting sphere ( $\alpha = R^3$ , where  $R$  is the sphere radius). Subsequent experiments<sup>5</sup> showed that no such effect was present. Later, this was explained<sup>6,7</sup> by the fact that the strong screening of the applied electric field by the conduction electrons had not been accounted for. Subsequently, a number of calculations were carried out which predicted that the electron screening will lead to the polarizability of metal particles being smaller than the classical value.<sup>6-9</sup> However, recent experiments on small sodium and potassium clusters containing up to 40 atoms per cluster<sup>10</sup> demonstrated that their static electric polarizability is in fact 60–80% higher than  $R^3$ , and only slowly converges to the bulk value. (Here  $R$  is the radius of the positive background in the cluster.)

In the past few years, numerical calculations utilizing the density-functional method have been carried out.<sup>11-14</sup> The predicted cluster polarizabilities are enhanced compared to the classical value, but are still noticeably lower than experiment (by about 20%). This discrepancy becomes more pronounced if one is interested in the deviation of the polarizability from its classical value. (Recently there appeared a density-functional calculation<sup>15</sup> incorporating certain corrections, which is in much better agreement with experiment; however, the authors point out that the approximations they use are not consistent with each other.)

The magnitude of the static electric polarizability is directly related to another response property of metal clusters, namely, the dipole resonance in their optical spectra. This connection follows from well-known sum rules (see Sec. III).

The visible spectrum of small metal particles is dominated by the well-known surface-plasma resonance of the

valence electrons.<sup>2,16</sup> It has been predicted<sup>13</sup> and experimentally indicated<sup>17-19</sup> that the same is true for small spherical clusters. For particles of radius much less than the wavelength of incident light, the classical theory<sup>20</sup> predicts a resonance at the frequency  $\omega_s = \omega_p/\sqrt{3}$ , where  $\omega_p$  is the bulk-plasma frequency of the metal. Much work has been devoted to the prediction of its location and width in microscopic systems; both blue and red shifts with respect to the classical position have been predicted (see the review<sup>2</sup> and the recent numerical calculations<sup>13,21</sup>). Experimentally,<sup>17-19</sup> it is observed that the resonance in isolated small spherical sodium clusters is red-shifted. As mentioned above, there is a connection between this shift and the fact that the electric polarizability of clusters exceeds its classical value.

The response of the valence electrons to an external electromagnetic field depends on the interplay between the following size-dependent factors: size quantization, electron screening, and the electron-cloud spillout outside the boundary of the positive background charge. As a consequence, such assumptions as the independent-electron approximation, the infinite-barrier model, and the approximation of a quasicontinuous distribution of electronic states are not able to describe the response properties accurately.

As is well known, the random-phase approximation (RPA) has been highly successful in describing collective excitations in solids and nuclei. In a series of interesting papers,<sup>6</sup> Lushnikov and Simonov (LS) applied this method to metallic particles. However, their results for the polarizability and the position of the plasma-resonance peak do not agree with observations on small clusters.

In this paper,<sup>22</sup> we calculate cluster polarizabilities and resonance frequencies by using the RPA and accurately taking into account the density distribution of the valence electrons, employing the recently developed Thomas-Fermi statistical description of metal clusters (Ref. 23, below referred to as I). The latter has already been successful in treating such cluster properties as relative stabilities ("magic numbers"), ionization potentials, and diamagnetism. The analytical evaluation presented below leads to very good agreement with experimental data.

The contents of the paper are as follows. In Sec. II we give a detailed derivation of the formula for the collective resonance frequency. This result is used in Sec. III to calculate the static electric polarizability.

## II. RESONANCE FREQUENCY

### A. Cluster in an external field

Consider a cluster in an external potential  $v(\mathbf{r})\exp(i\omega t)$ . The effective field inside the particle,  $V(\mathbf{r},\omega)\exp(i\omega t)$ , differs from the external field because of screening by the conduction electrons and satisfies the following integral equation:

$$V(\mathbf{r},\omega) = v(\mathbf{r}) + e^2 \int \frac{1}{|\mathbf{r}-\mathbf{r}_1|} \Pi(\mathbf{r}_1, \mathbf{r}_2, \omega) V(\mathbf{r}_2, \omega) d^3r_1 d^3r_2, \quad (1)$$

where  $\Pi(\mathbf{r}, \mathbf{r}_1, \omega)$  is the irreducible polarization operator of the electron gas.<sup>24</sup> In the RPA, this operator is given by<sup>24,25</sup> ( $\hbar=1$ )

$$\begin{aligned} \Pi_0(\mathbf{r}, \mathbf{r}_1, \omega) = & \sum_{\lambda, \lambda'} \phi_\lambda(\mathbf{r}) \phi_{\lambda'}^*(\mathbf{r}) \frac{n_\lambda - n_{\lambda'}}{\varepsilon_\lambda - \varepsilon_{\lambda'} - \omega} \\ & \times \phi_{\lambda'}^*(\mathbf{r}_1) \phi_{\lambda'}(\mathbf{r}_1), \end{aligned} \quad (2)$$

where  $\varepsilon_\lambda$  and  $\phi_\lambda$  are the single-electron energies and wave functions, respectively, and  $n_\lambda$  are the occupation numbers.

Self-supporting oscillations of the electron system correspond to nontrivial solutions of Eq. (1) with no external field present [ $v(\mathbf{r})=0$ ]. Combining (1) and (2) then gives

$$\begin{aligned} V^r(\mathbf{r}, \omega_r) = & e^2 \int d^3r_1 \frac{1}{|\mathbf{r}-\mathbf{r}_1|} \\ & \times \sum_{\lambda, \lambda'} \phi_\lambda(\mathbf{r}_1) \phi_{\lambda'}^*(\mathbf{r}_1) \frac{n_\lambda - n_{\lambda'}}{\varepsilon_\lambda - \varepsilon_{\lambda'} - \omega_r} V_{\lambda\lambda'}^r; \end{aligned} \quad (3)$$

the frequency  $\omega_r$  for which such a nontrivial solution exists is in fact the resonance frequency. It turns out (see below) that the result depends only on the density distribution of the valence electrons.

Note that  $V_{\lambda\lambda'}^r$  is markedly different from zero only if  $\lambda$  and  $\lambda'$  are neighboring single-electron levels. This can be shown by semiclassical arguments.<sup>6,25</sup> In a more qualitative way, one can reason as follows. We are looking for the eigenfield  $V^r$  of dipole oscillations of the electron system. In the macroscopic case, the electric field inside a polarized particle is uniform,<sup>26</sup> and the electric potential is linear in the coordinates. We expect that the functional form of  $V^r$  will be similar. On the other hand, the single-electron potential is harmonic-oscillator-like in shape (see, e.g., Fig. 1, I, and Ref. 1), and it is known that in such a potential only neighboring levels give nonvanishing matrix elements of a linear function of the coordinates. The conclusion is that the major contribution to the right-hand side of Eq. (3) will come from states with  $|\varepsilon_\lambda - \varepsilon_{\lambda'}| \sim \Delta$ , the single-particle level spacing.

It is known<sup>27</sup> that even in clusters with as little as eight atoms, this spacing is small:  $\Delta \sim 0.1-0.3$  eV, whereas the resonance frequency is  $\omega_r \gtrsim 2$  eV. This allows us to expand the right-hand side of Eq. (3) in powers of  $(\varepsilon_\lambda - \varepsilon_{\lambda'})/\omega_r$ . The zeroth-order term can be shown to vanish. The first-order term, after some manipulation,<sup>6</sup> can be brought into the following form:

$$\begin{aligned} & \left[ 1 - \frac{4\pi n(\mathbf{r})e^2}{m\omega_r^2} \right] V^r(\mathbf{r}) \\ & = - \frac{e^2}{m\omega_r^2} \int d^3r_1 V^r(\mathbf{r}_1) \nabla_1 \frac{1}{|\mathbf{r}-\mathbf{r}_1|} \cdot \nabla_1 n(\mathbf{r}_1), \end{aligned} \quad (4)$$

where  $n(\mathbf{r})$  is the number density of valence electrons in the particle and  $m$  is the electron mass (from now on, we do not explicitly indicate the frequency dependence of  $V^r$ ).

LS solved this equation only for the case  $n(\mathbf{r})=n\Theta(R-r)$  and obtained the classical surface-plasmon frequency,  $\omega_r = \omega_s$ . However, as stressed in the Introduction, the spillover of the electron cloud outside the edge of the positive background is an essential feature which must not be neglected. (The strong effect on the diffuseness of the electron-density profile on the red shift of the absorption line was indicated by calculations employing the hydrodynamic model,<sup>28</sup> and by the recent density-functional calculations of the resonance absorption.<sup>13,21</sup>)

### B. Solution of the integral equation

We now solve Eq. (4), taking into account the realistic shape of the charge distribution. We consider spherical clusters (i.e., the "magic-number" ones with spherical closed shells<sup>1</sup>). The valence-electron density distribution is then a function of the radial coordinate only:  $n(\mathbf{r})=n(r)$ . The positive ion background of the cluster is treated as a uniform sphere of radius  $R = a_0 r_s N_a^{1/3}$  ( $a_0$  is the Bohr radius,  $r_s$  is the bulk Wigner-Seitz density parameter, and  $N_a$  is the number of atoms in the cluster).

The results of the Thomas-Fermi statistical approach to the electronic structure of metal clusters developed in I will be used for the valence-electron number density. In contrast to the familiar atomic case which requires a numerical solution of the Thomas-Fermi differential equation, it turns out that the presence of the extended positive cluster background (instead of the central point charge in an atom) allows one to linearize the equation and to obtain an accurate solution analytically. Figure 1 illustrates results of the calculation carried out in I; it shows valence-electron densities and single-electron potentials found for some Na clusters.

To solve Eq. (4), we look for the dipole eigenfield in the form  $V^r(r_1) = f(r_1)\cos\theta_1$  [we have chosen  $\mathbf{r}$  to lie along the  $z_1$  axis in Eq. (4)]. Expanding  $1/|\mathbf{r}-\mathbf{r}_1|$  in Legendre polynomials and integrating over the angles, we arrive at the following equation:

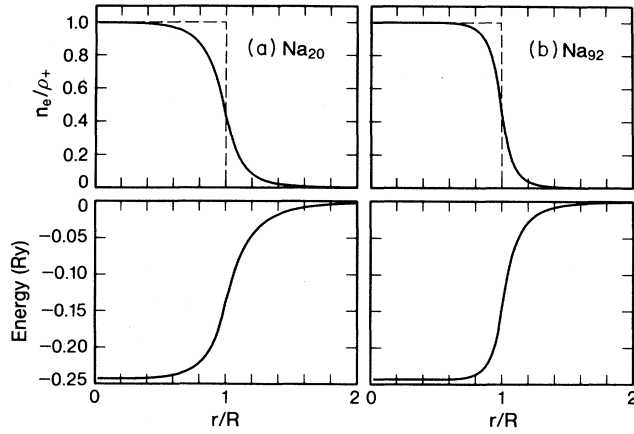


FIG. 1. Density distribution of the valence electrons and single-electron potentials in (a)  $\text{Na}_{20}$  and (b)  $\text{Na}_{92}$ , as given by the Thomas-Fermi statistical method (Ref. 23). The electron density  $g(r) = n(r)/\rho_+$  and the radial distance are expressed in units of the density and radius of the positive background (denoted by the dotted line). The amount of the electron spill-out outside the background boundary determines the collective resonance frequency and the static electric polarizability of a cluster.

$$\begin{aligned} \frac{1}{r^2} \int_0^r dr_1 r_1^2 g'(r_1) f(r_1) - 2r \int_r^\infty dr_1 \frac{1}{r_1} g'(r_1) f(r_1) \\ = \left[ 3g(r) - \left[ \frac{\omega_r}{\omega_s} \right]^2 \right] f(r). \end{aligned} \quad (5)$$

We have written  $n(r) = \rho_+ g(r)$ , where  $\rho_+$  is the number density of the uniform positive background and  $g(r)$  is the dimensionless electron-cloud-shape factor; we also used the fact that  $(4\pi\rho_+ e^2/3m)^{1/2}$  is the classical surface-plasmon frequency,  $\omega_s$ .

Differentiating Eq. (5) with respect to  $r$ , we obtain another equation:

$$\begin{aligned} \frac{2}{r^3} \int_0^r dr_1 r_1^2 g'(r_1) f(r_1) + 2 \int_r^\infty dr_1 \frac{1}{r_1} g'(r_1) f(r_1) \\ = \left[ \left[ \frac{\omega_r}{\omega_s} \right]^2 - 3g(r) \right] f'(r). \end{aligned} \quad (6)$$

These two equations can now be used to find  $\omega_r$ . Notice that the function  $g'$  is strongly peaked in the neighborhood of  $r_1 = R$ : the density of the electron cloud is close to  $\rho_+$  for  $r_1 < R$ , and decreases rapidly for  $r_1 > R$ , see I and Fig. 1. This allows us to expand the integrands about this point and keep only the linear terms in  $r_1 - R$ . It is also convenient to set  $r = R$ . Carrying out the integration, we find that Eqs. (5) and (6) simplify considerably. They become, respectively,

$$\left[ 1 - \left[ \frac{\omega_r}{\omega_s} \right]^2 \right] f(R) = 3Rg f'(R) \quad (7)$$

and

$$(3q - 1)f(R) = \frac{1}{2}R \left[ \left[ \frac{\omega_r}{\omega_s} \right]^2 - 3g(R) \right] f'(R), \quad (8)$$

where

$$q = \frac{1}{R} \int_R^\infty g(r) dr. \quad (9)$$

In deriving Eqs. (7) and (8), we have set  $g(0) = 1$ , i.e.,  $n(0) = \rho_+$ . This is well justified, because even for a cluster as small as  $\text{Na}_8$  the electron density at the center differs by less than 1% from the positive background density (see I), and this difference decreases with increasing cluster size.

We also have employed the following normalization condition: from the definition of  $g(r)$ , it is easily shown that

$$3 \int_0^\infty g(r) r^2 dr = R^3. \quad (10)$$

Integrating by parts, and again carrying out an expansion about the point  $r = R$ , we obtain

$$\int_0^\infty g(r) dr = R. \quad (11)$$

Equations (7) and (8) are easily solved for the ratio of the resonance frequency to the classical surface-plasmon frequency:

$$2 \left[ \frac{\omega_r}{\omega_s} \right]^2 = 3g(R) + 1 - \{ [3g(R) - 1]^2 + 24q(1 - 3q) \}^{1/2}. \quad (12)$$

We see that the shift of the resonance frequency with respect to its classical value is completely determined by the density profile of the valence electrons,  $g(r)$ . This is a general result, independent of the precise form of  $g(r)$ .

If we use the step-function profile, corresponding to the large-particle limit, then  $q = 0$  and we recover the macroscopic result  $\omega_r = \omega_s$ . To obtain more realistic results it is therefore essential to take into account the spill-out of the electron cloud outside the boundary of the positive background charge.

Figure 1 shows the distribution of electron density in clusters according to the statistical method I. In the region  $r \geq R$ , the function  $g(r)$  is given by

$$g(r) = \frac{30^3}{k^6} \left[ \frac{r}{R} \right]^{-3/2} \left[ \frac{r}{R} - D \right]^{-6}, \quad (13)$$

where

$$k = \left[ \frac{12}{\pi} \right]^{1/3} r_s^{1/2} N_e^{1/3}, \quad (14)$$

$$D = 1 - 6.1/k - 1.2/k^2 + O(1/k^3),$$

and  $N_e$  is the number of valence electrons in the cluster (for details see I). Note that since  $g(r)$  decreases rapidly, the dominant contribution to  $q$ , Eq. (9), comes from the region close to the edge of the positive background. The details of the behavior of the spillout tail at very large  $r$ ,

where the Thomas-Fermi equation is formally inapplicable and the function  $g(r)$  probably decreases even faster than given by Eq. (13), are therefore insignificant for our present purposes.

For a given material and cluster size, Eqs. (12), (9), and (13) completely determine the resonance frequency.

### C. Resonance frequencies of selected clusters

In Table I we present calculated resonance frequencies of some spherical Na and K clusters. Recent experiments<sup>18</sup> have shown that for Na<sub>8</sub> the ratio of the resonance frequency to its classical value is  $0.75 \pm 0.02$ ; the theory is in excellent agreement with these data.

## III. STATIC ELECTRIC POLARIZABILITY

The static electric polarizability of a particle can in principle be calculated from Eq. (1) with  $\omega=0$  and  $v(\mathbf{r})$  representing a uniform electric field. LS carried out such a calculation under the assumption that one can use the following quasicontinuum approximation:

$$n_\lambda - n_{\lambda'} = \frac{dn}{d\varepsilon_F} (\varepsilon_\lambda - \varepsilon_{\lambda'}). \quad (15)$$

This calculation led, as mentioned in the Introduction, to the conclusion that the static polarizability is decreased compared to its classical value. Evidently, for small clusters such an expansion is not valid.

Unfortunately, a direct evaluation of  $\Pi(r, r_1, \omega=0)$  is very difficult. It turns out, however, that an alternative approach, based on well-known sum rules (see, e.g., Refs. 29 and 30) can be used to calculate the static electric polarizability of a cluster, if the collective resonance frequency is known.

### A. Sum rules

Consider the following sums:

$$\begin{aligned} W_1 &= \sum_n (\omega_n - \omega_0) |\langle 0|x|n \rangle|^2, \\ W_{-1} &= \sum_n (\omega_n - \omega_0)^{-1} |\langle 0|x|n \rangle|^2, \end{aligned} \quad (16)$$

TABLE I. Calculated resonance frequencies [Eq. (12)] and static electric polarizabilities [Eq. (18)] of small spherical Na ( $r_s=3.96$ ) and K ( $r_s=4.87$ ) clusters, expressed in terms of their respective macroscopic values (experimental data from Ref. 10).

Cluster	$\omega_r/\omega_s$	$\alpha/R^3$	Expt.
Na <sub>8</sub>	0.74	1.81	1.77±0.03
Na <sub>20</sub>	0.79	1.62	1.68±0.10
Na <sub>40</sub>	0.81	1.51	1.61±0.03
Na <sub>58</sub>	0.83	1.46	
Na <sub>92</sub>	0.84	1.40	
K <sub>8</sub>	0.76	1.74	1.79±0.09
K <sub>20</sub>	0.80	1.57	1.66±0.13
K <sub>40</sub>	0.83	1.46	
K <sub>58</sub>	0.84	1.42	
K <sub>92</sub>	0.86	1.36	

where the summation is over all the quantum states  $|n\rangle$  of the valence-electron cloud in the cluster.

According to the well-known Thomas-Reiche-Kuhn sum rule for oscillator strengths, the first sum is  $W_1 = N_e/2m$ , where  $N_e$  is the number of valence electrons. The second sum is  $W_{-1} = \alpha_0/2e^2$ , where  $\alpha_0$  is the static polarizability of the system (this is easily seen by writing down the perturbation-theory expression for the energy shift of the system in an applied constant electric field). Therefore we have  $W_1/W_{-1} = N_e e^2/m\alpha_0$ .

As described in the Introduction, the collective resonance dominates the cluster spectra. Therefore the major contribution to the sums (16) comes from the neighborhood of this resonance frequency,  $\omega_r$ , and the ratio of the sums is simply  $W_1/W_{-1} = \omega_r^2$ . This leads to the following well-known relation between  $\alpha_0$  and  $\omega_r$ :

$$\alpha_0 = \frac{N_e e^2}{m \omega_r^2}. \quad (17)$$

If we substitute the classical metal-sphere surface-plasmon frequency,  $\omega_s$ , into this equation, we obtain  $\alpha_0 = R^3$ . The fact that the polarizability is greater than its classical value is seen to be related to the red shift in the position of the resonance.<sup>31</sup>

### B. Polarizability of selected clusters

We can rewrite Eq. (17) as follows:

$$\frac{\alpha_0}{R^3} = \left( \frac{\omega_s}{\omega_r} \right)^2, \quad (18)$$

and use Eq. (12) to calculate the static polarizability.

Table I presents the results of the calculation for several small sodium and potassium clusters together with the available experimental data; very good agreement is found.

## IV. CONCLUSION

We have calculated analytically the surface-plasma-resonance frequencies and static electric polarizabilities of small metal clusters. It was demonstrated that in considering cluster-response properties it is essential to take into consideration electron screening, size quantization, and the correct density distribution of the valence electrons.

The random-phase approximation was used to calculate the resonance frequency. The latter is red shifted with respect to its classical value, and is determined by the amount of the electron-cloud spill-out outside the boundary of the positive background. The recently developed<sup>23</sup> statistical Thomas-Fermi description of metal clusters was used for the density profile of the valence electrons.

From our results for the resonance frequency, and general sum rules, the static electric polarizability was deter-

mined. The theory is in very good agreement with experimental data.

This approach can be extended to the case of non-spherical clusters, and to a calculation of the lifetime of the collective resonance. However, the latter problem requires a knowledge of the widths of the discrete single-electron levels.

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