Energy states of a hydrogenic atom placed between two metal slabs

V. V. Paranjape
Department of Physics, Lakehead University, Thunder Bay, Canada P7B 5E1

P. V. Panat
Department of Physics, University of Poona, Pune (M.S.), 411007, India

K. N. Pathak
Department of Physics, Panjab University, Chandigarh, 160014, India
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We have derived expressions for the self-energy of a hydrogenic atom placed in the cavity formed by two semi-infinite metal slabs. The excitations of the metal surfaces are described in terms of the surface plasmon modes and the atom is assumed to interact with these modes. The self-energies are obtained for the 10*S* and 14*S* excited states of the atom. The earlier calculations of the self-energy are based on the dipolar approximation but we show in this paper that when the gap size is sufficiently small, the multipolar interactions can exceed the dipolar contribution. We consider the case when the atom is placed at the center of the gap and obtain the variations in the self-energy when the gap size is varied. [S0163-1829(97)05008-X]

I. INTRODUCTION

For the understanding of various processes such as the scattering of atoms by a surface, surface catalysis, or the atomic adsorption, it is important to know the variation in the interaction energy of the atom as a function of the distance between the atom and the surface. Although the subject has been studied both experimentally and theoretically over the past century, there is a renewed interest in the field in the past decade as a result of advances in the mapping of a solid surface by means of the scanning tunneling and the atomic-force microscopes. These advances have made it possible to make more detailed measurements than possible before concerning the growth, orientation, and motion of atoms near a surface. With the increase in the quality of experiments, there is now a greater need to obtain numerical estimates of the self-energies of the atom near surfaces. It is the object of the present paper to propose an approximate method for obtaining the self-energy of a hydrogenic atom placed between two parallel metal plates. Numerical estimates of the self-energy of the atom are obtained when the atom is at the center of the gap and when gap width is varied. We also obtain the self-energy of the atom near the center of the gap.

A charge neutral atom is attracted to a metal surface through a force known as the van der Waals force. The potential energy that leads to this attraction is a consequence of correlated electronic polarizations induced in the atom and in the metal surface. The calculation of the interaction energy is a many-body problem because of the presence of a large number of electrons in the metal. It is convenient and usually sufficient to study the many-body aspects of the interaction by describing the excitations of metal electrons in terms of their collective motion called the ''plasmons.'' Making use of this description, Manson and Ritchie[1] have developed a self-energy formalism that allows one to obtain the position-dependent self-energy of a charged particle near a metal surface. The significant feature of this formalism is that the calculation of the self-energy takes into account the effect of the recoil of the electron as it emits a virtual plasmon. The effect that is strong near a metal surface makes the electron self-energy dependent on its velocity. Manson and Ritchie[1] were the first to propose this direct and relatively simple method for calculating the self-energy. Several authors have extended Manson and Ritchie's[1] approach to calculate the self-energies for specific situations. For example, Mahanty, Pathak, and Paranjape[2,3] have considered the effect of plasmon dispersion on the electron self-energy while others have applied the formalism to calculate the self-energy of the hydrogenic atom[4–7] in the vicinity of a metal surface. Manson and Ritchie's[1] method, although elegant, is based on the plasmon model, which treats the electron gas as a charge continuum. Hence the self-energy obtained using this approach is valid when the distance between the charged particle (or the atom) and the metal surface is greater than the interelectron distance of the metal electrons. The method has provided excellent results for the self-energy for distances greater than a few angstroms from the surface and provided a reasonable estimate for the self-energy at closer distances. Although we continue to use the plasmon model in this paper we propose a simpler and approximate method to calculate the self-energy than given by the Manson-Ritchie[1] formalism. Our approach does not take into account the atomic recoil and therefore the velocity-dependent effects are neglected. In spite of its approximate nature, our method gives the essential features of the self-energy reasonably well.

We select for our investigation a hydrogenic atom since it represents the simplest atomic system in which numerical calculations are relatively easy. We describe the atom in terms of a valence electron in the presence of a single positive charge comprised of the charge of the nucleus and the charge of the core electrons. We also assume that the main interaction of the hydrogenic atom is with the surface plasmons and its interaction with the bulk plasmons is neglected on the assumption that the atomic wave functions would not

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overlap significantly with the metal electrons. In order to establish the theoretical framework for this paper we express the interaction Hamiltonian for a hydrogenic atom interacting with two parallel metal surfaces by straightforward extension of the Hamiltonian used for a single charged particle used in the previous publications.[8–11] The Manson and Ritchie[1] approach is mainly useful for atoms with a low atomic mass such as the positronium (or for an electron) in which the recoil effects are significant. On the other hand, the atoms considered in this work have significantly larger mass in comparison with a positronium atom and therefore the velocity-dependent effects[4] are weak and therefore neglected. In spite of the differences in the two approaches, both have many common features and when the recoil effects are neglected they would give the same self-energy. The main features of our approach are as follows: We show in this paper that the Coulomb energy between the valence electron and the positively charged atomic core is altered as a result of the interaction between the atom and the surface. We derive the renormalized Coulomb energy using a unitary transformation. The transformation, introduced by Platzman,[12] was used earlier by him to evaluate the self-energy of a bound polaron in which the electron is coupled simultaneously to an attractive Coulomb potential and to the potential induced by the three-dimensional polar modes of the lattice. The situation considered by Platzman[12] is similar to ours. In the present calculations the polar modes considered by Platzman[12] are replaced by the plasmon modes. Using Platzman's transformation, we are able to express the changes in the potential energy of the atom. Treating the change as a perturbation and using the first-order perturbation theory, we obtain the shifts in the self-energy of a hydrogenic atom as a function of position and as a function of the gap size.

The presence of two surfaces requires a change in the usual plasmon model. The surfaces that are in close proximity introduce a coupling between the uncoupled plasmon modes present in each of the surfaces.[8–11] The coupled plasmon excitations can now be expressed in terms of symmetric and antisymmetric plasmon modes. The use of the coupled modes in the calculation of the self-energy of a charged particle was reported in earlier publications.[8,9] The calculation of the self-energy of an atom, proposed in this paper, is an extension of this work. The proposed research is motivated by the intense research interest in the study of an electron or an atom, enclosed in restricted spaces such as cavities or placed in various heterostructures.

When the gap between the metals is large, an estimate for the self-energy of the atom is obtained, by assuming that the spread in the atomic wave functions is significantly smaller than the gap size. In this case, it is reasonable to make use of the dipolar interaction between the metal and the atom. On the other hand when the gap is comparable to the size of the atom, the dipolar approximation is inadequate and the multipolar interaction of the atom with the surfaces must be taken into account. It is the main feature of the present paper to obtain the self-energy of a hydrogenic atom taking into account multipolar interaction of the atom with the surfaces.

A first direct experimental observation of van der Waals interaction between a sodium atom moving between two metallic plates was achieved recently by Sandoghdar, Sukenik, Hinds, and Haroche.[13] The two metal plates are assumed to be parallel to each other and the separation between them is taken to be a few micrometers. These authors estimate experimentally the changes in the energy states of a sodium atom placed at the center of the gap. The energy shifts are obtained for excited states of the sodium atoms with the radial quantum numbers varying from $n=10$ to $n=13$. The experimental results analyzed by these authors make use of the dipolar approximation, which is adequate because of the large size of the gap considered in the experiments reported by Sandoghdar *et al.*[13]

The plan of the paper is as follows. In Sec. II, we develop the interaction Hamiltonian for a hydrogenic atom enclosed between two parallel metal surfaces. We introduce a unitary transformation in which the Coulomb interaction between the two charges is transformed to include the effect of the interaction of the atom to the surface. We derive the expression for the self-energy of the hydrogenic atom by treating the renormalized potential energy as a perturbation. In Sec. III, we apply the theory of the previous section to obtain the numerical values for the self-energy of the excited states of the sodium atom. The numerical results are also discussed. Concluding remarks are made in Sec. IV.

II. THEORY

We describe our system as follows. Let the metal slabs occupy the regions defined by $|z|>L$, where the center of the gap is selected as the origin of the z coordinate. The gap is therefore $2L$ and is assumed to be larger than the atom size. The center of mass of the atom is kept well within the gap so that the electron wave function does not excessively overlap the metal electrons.

We write the Hamiltonian representing a hydrogenic atom placed within the gap as follows:

$$H=\frac{p_n^2}{2m_n}+\frac{p_e^2}{2m_e}+H_{\text{metal}}+H_{\text{int}}-\frac{e^2}{|\mathbf{r}_1-\mathbf{r}_2|}, \tag{1}$$

where

$$H_{\text{metal}}=\sum_{\alpha,\boldsymbol{\kappa}}\hbar\omega_{\alpha,k}[a^{\dagger}_{\alpha,\mathbf{k}}a_{\alpha,\mathbf{k}}+(1/2)]. \tag{2}$$

Following Sols and Ritchie[8] we express H_{int}, the Hamiltonian representing the interaction between the atom and the metal surfaces, as

$$H_{\text{int}}=-e\sum_{\alpha,\mathbf{k}}\Gamma_{\alpha,k}(z_1)\exp(i\mathbf{k}\cdot\mathbf{R}_1)(a^{\dagger}_{\alpha,-\mathbf{k}}+a_{\alpha,\mathbf{k}})$$
$$+e\sum_{\alpha,\mathbf{k}}\Gamma_{\alpha,\mathbf{k}}(z_2)\exp(i\mathbf{k}\cdot\mathbf{R}_2)(a^{\dagger}_{\alpha,-\mathbf{k}}+a_{\alpha,\mathbf{k}}). \tag{3}$$

In Eqs. (1)–(3), $-e$ is the charge of the valence electron, e is the effective charge of the rest of the atom, $\alpha=\pm1$ where the plus sign refers to the symmetric and the minus sign to the antisymmetric mode of the surface plasmon, $\mathbf{r}_1\equiv(\mathbf{R}_1,z_1)$ and $\mathbf{r}_2\equiv(\mathbf{R}_2,z_2)$ are, respectively, the three-dimensional coordinates of the electron and the ion, which are assumed to be

within the gap so that $-L<z<+L$ where z stands for both z_1 and z_2 and the factor $\Gamma_{\alpha,k}(z)$ is the coupling constant defined as follows:

$$\Gamma_{\alpha,k}(z)=g_{\alpha,k}(z)N_{\alpha,k}, \tag{4}$$

where

$$g_{\alpha,k}(z)=e^{-kz}+(\alpha)e^{kz}, \tag{5}$$

$$N^2_{\alpha,k}=\pi\frac{\hbar\omega_{\alpha,k}}{2Ak(e^{2kL}+\alpha)}, \tag{6}$$

and

$$2\omega^2_{k,\alpha}=\omega_p^2(1+\alpha e^{-2kL}). \tag{7}$$

$\omega_{\alpha,k}$ represents the two coupled plasmon modes. When $2kL\ll 1$ the effect of the coupling is large and two distinct plasmon modes, one symmetric and the other antisymmetric, are given by Eq. (7). The coupling decreases when $2kL\gg 1$ and the two coupled mode frequencies merge into the common value $(\omega_p^2/2)^{1/2}$.

We now introduce the Platzman[12] transformation to rewrite the Hamiltonian. The unitary transformation is defined by

$$U=\exp\left(\sum_{\mathbf{k},\alpha}(f_{\alpha,\mathbf{k}}a_{\alpha,\mathbf{k}}-f^*_{\alpha,\mathbf{k}}a^\dagger_{\alpha,\mathbf{k}})\right), \tag{8}$$

where $f_{\alpha,\mathbf{k}}$ is a parameter whose value is determined by the need that the transformed Hamiltonian can be given a simple interpretation. It is straightforward to obtain the relationship

$$U^{-1}a_{\alpha,\mathbf{k}}U\rightarrow a_{\alpha,\mathbf{k}}+f_{\alpha,\mathbf{k}}. \tag{9}$$

The transformed Hamiltonian H_t is defined by

$$H_t=U^{-1}HU. \tag{10}$$

We now ascribe a value to $f_{\alpha,\mathbf{k}}$ such that in the transformed Hamiltonian terms linear in $a_{\alpha,\mathbf{k}}$ and $a^\dagger_{a,\mathbf{k}}$ are eliminated. This requires that the value of $f_{\alpha,\mathbf{k}}$ be

$$f_{\alpha,\mathbf{k}}=\frac{e}{\hbar\omega_{\alpha,k}}[\exp(-i\mathbf{k}\cdot\mathbf{R}_1)\Gamma_{\alpha,k}(z_1) - \exp(-i\mathbf{k}\cdot\mathbf{R}_2)\Gamma_{\alpha,k}(z_2)]. \tag{11}$$

The transformed Hamiltonian contains two main changes; H_{metal} is altered and the change can be expressed as a shift in the zero-point energy of the surface plasmons. The changes in the kinetic energies of the electron and the nucleus also occur but they lead to the velocity dependent effect on the self-energy which we neglect. The transformed Hamiltonian is expressed by

$$H_t=\frac{p_n^2}{2m_n}+\frac{p_e^2}{2m_e}-\frac{e^2}{|\mathbf{r}_1-\mathbf{r}_2|}+\sum_{\alpha,k}\hbar\omega_{a,k} \times[a^\dagger_{\alpha,\mathbf{k}}a_{\alpha,\mathbf{k}}+f^*_{\alpha,\mathbf{k}}f_{\alpha,\mathbf{k}}+(1/2)]. \tag{12}$$

Substituting the value of $f_{\alpha,\mathbf{k}}$ from Eq. (11) into Eq. (12) gives H_t as

$$H_t=\frac{p_n^2}{2m_n}+\frac{p_e^2}{2m_e}-\frac{e^2}{|\mathbf{r}_1-\mathbf{r}_2|}+\sum_{\alpha,\mathbf{k}}\hbar\omega_{\alpha,k}[a^\dagger_{\alpha,\mathbf{k}}a_{\alpha,\mathbf{k}}+(1/2)] + V_1(z_1)+V_2(z_2)+V_3(z_1,z_2,\mathbf{R}_1,\mathbf{R}_2), \tag{13}$$

where

$$V_1(z_1)=-e^2\sum_{\alpha,k}\frac{|\Gamma_{\alpha,k}(z_1)|^2}{\hbar\omega_{\alpha,k}}, \tag{14}$$

$$V_2(z_2)=-e^2\sum_{\alpha,k}\frac{|\Gamma_{\alpha,k}(z_2)|^2}{\hbar\omega_{\alpha,k}}, \tag{15}$$

and

$$V_3(z_1,z_2,\mathbf{R}_1,\mathbf{R}_2)=2e^2\sum_{\alpha,k}\exp[i\mathbf{k}\cdot(\mathbf{R}_1-\mathbf{R}_2)] \times\Gamma_{\alpha,k}(z_1)\Gamma_{\alpha,k}(z_2)/\hbar\omega_{\alpha,k}. \tag{16}$$

$V_1(z_1)$ and $V_2(z_2)$ are respectively the self-energies of the electron and of the nucleus arising from its interaction with the metal surface. $V_1(z_1)$, as expected, depends on the separation of the electron from the surface and is independent of its planar coordinate. It represents the self-energy of the valence electron obtained by summing the potential energy arising from the infinite set of images formed by the electron in the metal. Similarly $V_2(z_2)$, for the nucleus, has the same interpretation as $V_1(z_1)$. $V_3(z_1,z_2,\mathbf{R}_1,\mathbf{R}_2)$ corresponds to the interaction energy of the electron interacting with the images of the nucleus and the energy of the nucleus interacting with the images of the electron. It depends not only on the separation of the charges from the surface but also on their planar separation. The effective potential energy of the atom can now be expressed as $V(z_1,z_2,\mathbf{R}_1,\mathbf{R}_2)$ such that

$$V(z_1,z_2,\mathbf{R}_1,\mathbf{R}_2)=\frac{-e^2}{|\mathbf{r}_1-\mathbf{r}_2|}+V_1(z_1)+V_2(z_2) + V_3(z_1,z_2,\mathbf{R}_1,\mathbf{R}_2), \tag{17}$$

where the first term on the right-hand side is the bare Coulomb energy between the two charges and the last three terms arise as a result of the interaction of the charges with the two surfaces.

We now define the z coordinate of the nucleus as $z_2=Z$, which corresponds to the approximate position of the center of mass of the atom and define the electron coordinate with respect to the nucleus as $z_1=Z+z$.

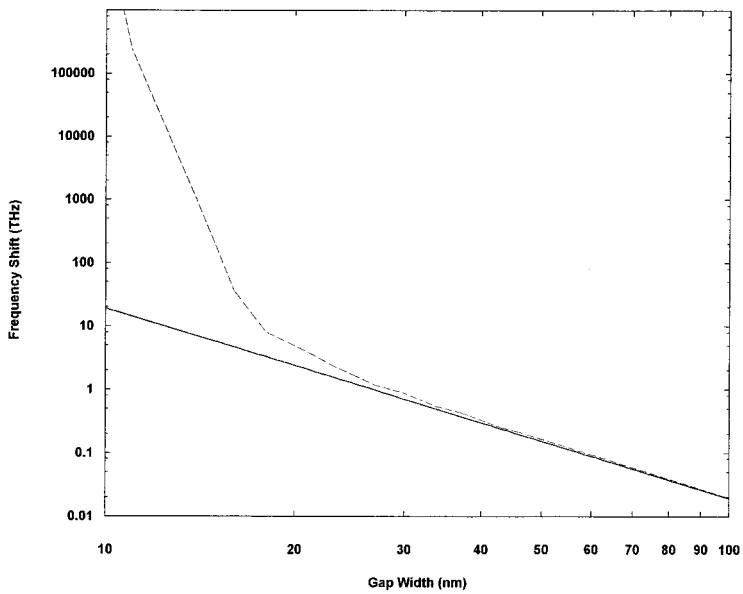

FIG. 1. The variation of the self-energy expressed in terms of a frequency shift as a function of the gap width for the 10S state of the sodium atom placed at the center of the gap. The self-energy given by the full line uses the dipolar approximation while the dashed line contains all multipolar interactions.

Substituting Eqs. (4)–(7) into Eqs. (14)–(16) and completing the angular integration, we write

$$V_1(Z'+z') = \frac{-e^2}{L}\int_0^\infty \left[\frac{\cosh^2\{(k'L')[(Z'+z')/L']\}}{2\cosh(k'L')} + \frac{\sinh^2\{(k'L')[(Z'+z')/L']\}}{2\sinh(k'L')}\right] \times \exp(-k'L')d(k'L'), \tag{18}$$

where k', L', and z' are dimensionless quantities defined in terms of a, which is related to the spread of the electron wave function and has the dimension of length. The precise value of a will depend on the state of the atom and will be defined later. Thus $k'=ka$, $L'=(L/a)$, $Z'=(Z/a)$, and $z'=(z/a)$. Similarly

$$V_2(Z') = \frac{-e^2}{L}\int_0^\infty \left[\frac{\cosh^2[(k'L')(Z'/L')]}{2\cosh(k'L')} + \frac{\sinh^2[(k'L')(Z'/L')]}{2\sinh(k'L')}\right] \times \exp(-k'L')d(k'L') \tag{19}$$

and

$$V_3(z_1',z_2',|\mathbf{R}_1-\mathbf{R}_2|) = \frac{2e^2}{L}\int_0^\infty J_0[(k'L')(|\mathbf{R}_1-\mathbf{R}_2|/L')] \times \left\{\frac{\cosh[(k'L')(z_1'/L')]\cosh[(k'L')(z_2'/L')]}{2\cosh(k'L')}\right.$$

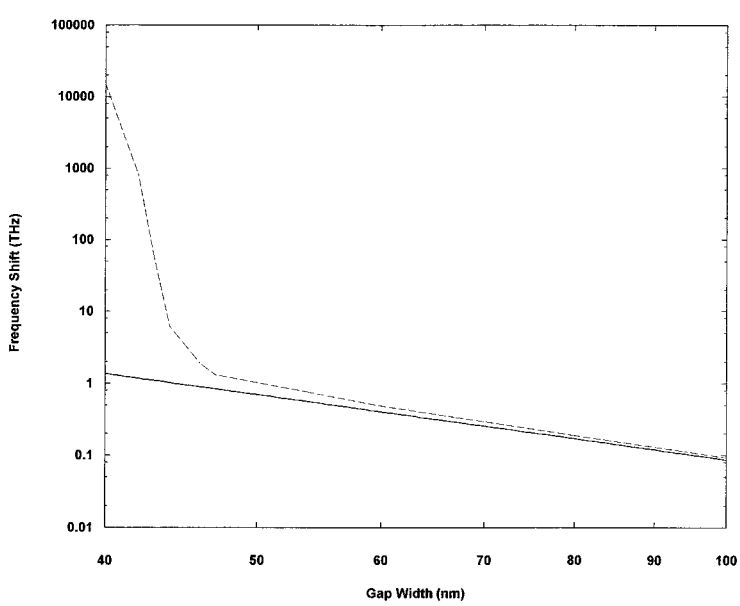

FIG. 2. The variation of the self-energy expressed as a frequency shift as a function of the gap width for the 14S state of the sodium atom placed at the center of the gap. The self-energy given by the full line uses the dipolar approximation while the dashed line contains all multipolar excitations.

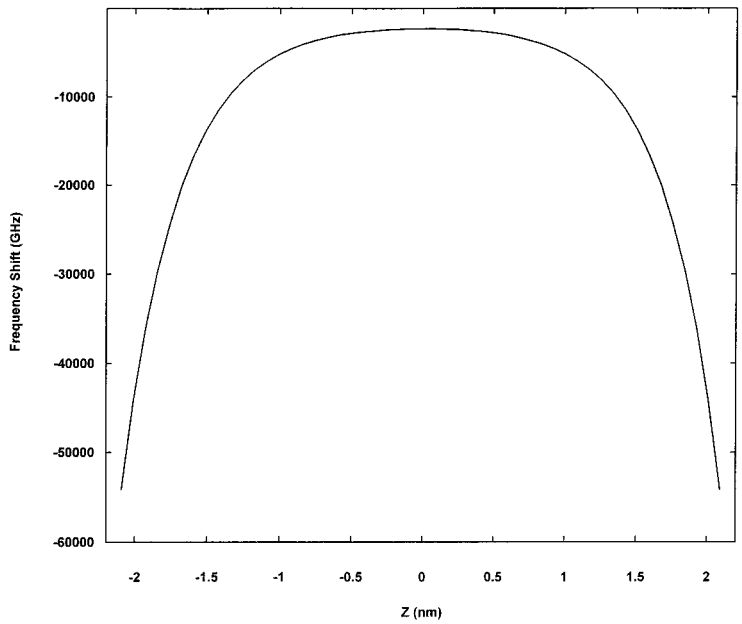

FIG. 3. The variation for the self-energy expressed in terms of a frequency shift for the 14*S* state of the sodium atom as a function of the distance near the center of the gap.

$$+\frac{\sinh[(k'L')(z_1'/L)]\sinh[(k'L')(z_2'/L')]}{2\sinh(k'L')}\Bigg\}$$

$$\times\exp(-k'L')dk'L'. \tag{20}$$

In Eq. (20), J_0 is the Bessel function of order zero.

As remarked earlier, $V_2(Z')$ is the self-energy of the nucleus and hence corresponds to the result obtained by Sols and Ritchie[8] when the dynamical effects from their work are removed. For confirmation, we perform the integration in Eq. (19) to get

$$V_2(Z)=-(e^2/8L)[2\psi\{1\}-\psi\{(1/2)+(Z/2L)\}$$
$$-\psi\{(1/2)-(Z/2L)\}], \tag{21}$$

where

$$\psi(y)=-\int_0^\infty \frac{e^{-yt}}{1-e^{-t}}\,dt. \tag{22}$$

We can also obtain the corresponding expression for $V_1(Z+z)$, the self-energy of the electron.

The shift ΔE in the energy of the atom is obtained by using the first-order perturbation theory. We note that the self-energy contains second-order contributions in the interaction potential but our use of the transformed Hamiltonian allows us to express the self-energy as a first-order change in the perturbation. For the nth radial quantum state of the atom the shift in the energy is given by

$$\Delta E_n=\langle n|V(z_1,z_2,\mathbf{R}_1,\mathbf{R}_2)|n\rangle. \tag{23}$$

For a given nS state, the exponential decrease in the wave function is proportional to $\exp(-2r/na_0)$ where a_0 is the Bohr radius. The value of a that was introduced in Eqs. (18)–(20) can now be specified as $a=na_0/2$. The definition of a is made purely for computational purposes in obtaining the average value of the self-energy in Eq. (23) for a given state of the atom.

We now consider the limiting case for the self-energy using the dipolar approximation. Assume that the nucleus is at the center of the gap so that $Z=0$. In the dipolar approximation, we expand V's in Eqs. (18)–(20) in powers of z and and of $|\mathbf{R}_1-\mathbf{R}_2|$ and retain up to quadratic terms in z and $|\mathbf{R}_1-\mathbf{R}_2|$. Substituting the approximated expressions into Eq. (23) and after some lengthy algebra we get the well-known result[11] used in the analysis of Sandoghdar *et al.*,[13]

$$\Delta E_n\cong-\left(\frac{7}{48}\right)\left(\frac{e^2}{L^3}\right)\zeta(3)\langle n|r^2|n\rangle, \tag{24}$$

where $\zeta(3)\cong1.2$ is the Riemann zeta function. In Eq. (24) the effect of the quantum defect is introduced by replacing n by $n^*=(n+1.35)$. The expression (24) is a reasonable approximation if the gap is much larger than the size of the atom. In the experiments reported by Sandoghdar *et al.*[13] the gap is of the order of a micrometer, which is more than 500 times the size of the tenth quantum state of the sodium atom. Thus the use of the approximate result given by Eq. (24) is appropriate in the work of Sandoghdar *et al.*[13]

We have obtained the changes in the self-energy of the electron without making the dipolar approximation in Eq. (23). The theoretical expression is therefore valid even when gap widths are comparable to but approximately greater than the atomic size. The numerical results with and without the dipolar approximation for the excited states of the sodium atom at presented and discussed in the following section.

III. RESULTS AND DISCUSSION

We have evaluated numerically the changes in the self-energy of the electron using Eq. (23), which includes multipolar interaction of the atom with the surface for the two excited states of the sodium atom corresponding to the radial quantum numbers $n=10$ and 14. We have considered the case in which the center of mass of the atom is located at the center of the gap. The gap width between the two metal slabs is allowed to vary. The changes in the self-energy expressed

as a change in the frequency shift $\Delta\nu=(\Delta E/h)$ are shown in Fig. 1 for $n=10$ and in Fig. 2 for $n=14$ with the doted lines using Eq. (23) and the full lines using the dipolar approximation. For $n=10$ the changes in the self-energy using the dipolar approximation and considering the multipolar interactions are close to each other when the gap width exceeds 40 nm but when the gap is smaller than 40 nm the self-energy given by multipolar interaction exceeds significantly the value obtained using the dipolar approximation. For $n=14$ the departures between the two results occur when the gap is below 60 nm. Clearly these results follow from the fact that the effect of the multipolar excitations would be stronger for the atom at the $n=14$ excited state than for the $n=10$ excited state. It should be noticed that the size of the atom for $n=10$ or for $n=14$ is still smaller than the gap widths considered in Figs. 1 and 2. Although the figures show the self-energy for extremely small gaps, we should be cautious with the use of perturbative theory for widths below 15 nm for the case of the 10*S* state of the atom and for widths less than 30 nm for the 14*S* state. For these small gaps the perturbation theory may not be fully justified. In Fig. 3, we have plotted the variation of the self-energy as the position of the center of the atom is varied within 2 nm from the midpoint position, for the gap of 42 nm and for the 14*S* state of the sodium atom. The potential energy is nearly constant within a distance of about 1 nm on both sides of the center. This feature is similar to the one obtained by Sandoghdar *et al.*[13] for the 10*S* state of the sodium atom for the gap of a micrometer with the important difference that the region of approximately constant potential energy in the calculation of Sandoghdar *et al.* is spread over 0.1 μm, which is considerably larger than the values obtained in our calculations. The ratio of the flat potential energy region to the total gap width is approximately (1/10) in the calculation of Sandoghdar *et al.*[13] while in our calculations the flat region is only approximately (1/20) of the total width.

IV. CONCLUDING REMARKS

Our numerical results show that the potential energy of the atom is attractive toward the surface and that it decreases as the width of the gap is reduced. The decrease in the self-energy follows the L^{-3} dependence on the gap based on the dipolar approximation but the dependence is much stronger at smaller widths when multipolar interactions become important. The expression for the potential energy given by Eq. (17) contains in addition to the Coulomb energy three more terms. The first two terms refer to the potential energies of the electron and the nucleus in the presence of the surface and depend on the separation of each from the surface. Both these terms give rise to an attractive force towards the surface. As the gap is decreased the binding potential energy of the surface could exceed the binding potential between the electron and the nucleus. The third term in Eq. (17) depends on the separation between the electron and the nucleus in the plane parallel to the surface and also on the separations of the charges from the surface. The latter potential-energy term can produce a repulsion between the electron and the nucleus and the repulsion increases as the width is decreased. At a sufficiently small width the force of repulsion can in principle exceed the Coulombic attraction. This will cause the atom to ionize within the gap. This qualitative picture may be approximately correct but such conclusions are not evident in our calculations since the ionization is expected to occur at extremely small gap widths where our theory would break down mainly because the perturbative treatment will not hold.

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