Long-range interactions of sodium atoms

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Long-range interaction potentials between two and three sodium atoms, between a sodium atom and a perfectly conducting wall, and for a sodium atom between two perfectly conducting walls, are calculated with an electric-dipole oscillator strength distribution constructed from combinations of experimental and theoretical energy levels, oscillator strengths, and photoionization cross section data, constrained by accurate values of oscillator strength sum rules. The leading dispersion coefficients for Na-Na and Na-Na-Na and the Lennard-Jones coefficient for the Na-wall system are determined. For Na-Na and for the Na-wall systems, the retarded (Casimir) potentials are also calculated. [S1050-2947(97)03605-6]

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I. INTRODUCTION

The scattering of atoms at ultralow temperatures is sensitive to the interactions at very large interatomic distances. Approximate calculations of the coefficients of the leading terms in the representation of the interactions in inverse powers of the internuclear distance R have been carried out for alkali metals [1–9]. It is often assumed that the errors in the predicted values are no more than 5%, but, except for lithium, the error estimates are uncertain.

The leading term is the van der Waals interaction, $-C_6/R^6$. It arises from a dipole-dipole interaction and it can be expressed formally in terms of the dynamic electric-dipole polarizabilities of the atoms. For sodium, a reliable empirical construction of the dynamic polarizability can be created from the available experimental and theoretical data on discrete oscillator strengths and photoionization cross sections and its accuracy can be assessed by the use of sum rules. The dynamic polarizability can be used also to determine the coefficient of the leading term of the three-body Na-Na-Na interaction and the leading term of Na-wall interactions. The modifying effects of retardation are readily included.

II. OSCILLATOR STRENGTH SUM RULES

If $|0\rangle$ and $|n\rangle$ represent, respectively, the eigenfunctions of the ground state and the nth excited state of Na, then the electric-dipole absorption oscillator strength is defined by

$$f_n = \frac{2}{3}(E_n - E_0)\left|\left\langle 0\left|\sum_{i=1}^{N}\mathbf{r}_i\right|n\right\rangle\right|^2, \quad (1)$$

where $\mathbf{r}_i$ is the position vector of the ith electron, E_n is the eigenvalue of state n, N is the number of electrons, and all quantities are expressed in a.u.

Summing over all the excited discrete states and integrating over all the excited continuum states, we obtain

$$S(0) = \mathsf{S}_n f_n = N = 11, \quad (2)$$

$$S(-1) = \mathsf{S}_n f_n / (E_n - E_0) = \frac{2}{3}\left\langle 0\left|\left(\sum_{i=1}^{N}\mathbf{r}_i\right)^2\right|0\right\rangle \quad (3)$$

and

$$S(-2) = \mathsf{S}_n f_n / (E_n - E_0)^2 = \alpha(0), \quad (4)$$

where $\alpha(0)$ is the static electric-dipole polarizability.

The dynamic electric-dipole polarizability at frequency ω is given by

$$\alpha(\omega) = \mathsf{S}_n \frac{f_n}{(E_n - E_0)^2 - \omega^2}. \quad (5)$$

The coefficient C_6 of the van der Waals interaction between a pair of atoms with polarizability $\alpha(\omega)$ is given by

$$C_6 = \frac{3}{\pi}\int_0^\infty d\omega[\alpha(i\omega)]^2, \quad (6)$$

the coefficient C_9 of the leading R^{-9} term of the three-body interaction is given by

$$C_9 = \frac{3}{\pi}\int_0^\infty d\omega[\alpha(i\omega)]^3, \quad (7)$$

and the coefficient C_3 of the R^{-3} term of the Lennard-Jones interaction between an atom and a perfectly conducting wall is given by

$$C_3 = \frac{1}{4\pi}\int_0^\infty d\omega\, \alpha(i\omega). \quad (8)$$

When the effects of retardation are included [10], the leading atom-atom interaction term is modified to

$$V(R) = \frac{-C_6}{R^6}F_6(R), \quad (9)$$

where

$$F_6 = \frac{1}{\pi C_6} \int_0^\infty d\omega \; \alpha(i\omega)^2 \exp(-2\alpha_{fs}\omega R) P(\omega\alpha_{fs}R),$$
$$(10a)$$

$$P(x) = x^4 + 2x^3 + 5x^2 + 6x + 3 \qquad (10b)$$

and α_{fs} is the fine-structure constant, and the leading atom-wall interaction term to

$$V_W(R) = -\frac{C_3}{R^3} F_3(R), \qquad (11)$$

where

$$F_3 = \frac{1}{4\pi C_3} \int_0^\infty d\omega \; \alpha(i\omega)\exp(-2\alpha_{fs}\omega R)Q(\alpha_{fs}\omega R)$$
$$(12)$$

and

$$Q(x) = 2x^2 + 2x + 1. \qquad (13)$$

For asymptotically large distances, the retarded interactions have the forms

$$V(R) \sim \frac{-23}{4\pi} \frac{1}{\alpha_{fs}} \frac{\alpha^2(0)}{R^6}, \qquad (14)$$

and

$$V_W(R) \sim \frac{-3}{8\pi} \frac{1}{\alpha_{fs}} \frac{\alpha(0)}{R^4}. \qquad (15)$$

The corresponding limits of the retardation coefficients are

$$F_6(R) \rightarrow \frac{23}{4\pi\alpha_{fs}R} \frac{\alpha^2(0)}{C_6}, \qquad (16)$$

and

$$F_3(R) \rightarrow \frac{3}{8\pi\alpha_{fs}R} \frac{\alpha(0)}{C_3}. \qquad (17)$$

The potential for an atom located between two parallel perfectly conducting walls has been given by Barton [11] and by Zhou and Spruch [12]. We can write the potential $U(z,L)$ as the sum of two double integrals involving $\alpha(i\omega)$,

$$U(z,L) = -\frac{1}{\pi L^3} \int_0^\infty dt \frac{t^2\cosh(2tz/L)}{\sinh t} \int_0^{t/\alpha_{fs}L} d\omega \; \alpha(i\omega)$$
$$+ \frac{\alpha_{fs}^2}{\pi L} \int_0^\infty d\omega \; \omega^2\alpha(i\omega) \int_{\alpha_{fs}L\omega}^\infty dt \frac{e^{-t}}{\sinh t}, \qquad (18)$$

where L is the wall separation and z is the distance of the atom from the midpoint. The coefficient of the wall-atom-wall interaction for small distances L can be obtained from Eq. (18) by letting $\alpha_{fs}\rightarrow 0$ (which corresponds to letting the speed of light become infinite), giving

$$U(z,L) = \frac{-1}{\pi L^3} T(z/L) \int_0^\infty ds \; \alpha(is), \qquad (19)$$

where

$$T(z/L) = \int_0^\infty dt \frac{t^2\cosh(2tz/L)}{\sinh t}, \qquad (20)$$

in agreement with the result of Zhou and Spruch [12]. Using expression (8) for the coefficient C_3, we may reduce the potential (19) to the form

$$U(z,L) \sim \frac{-4}{L^3} T(z/L)C_3. \qquad (21)$$

The asymptotic limit of $U(z,L)$ for large L is proportional to $\alpha(0)$ and is given by Barton [11], Hinds [14], and Zhou and Spruch [12] as

$$U(z,L) \sim \frac{\pi^3\alpha(0)}{\alpha_{fs}L^4}\left[\frac{1}{360} - \frac{3-2\cos^2(\pi z/L)}{8\cos^4(\pi z/L)}\right]. \qquad (22)$$

Experimental evidence of retardation effects on Na-wall interactions have been obtained by Kasevich *et al.* [15] for a dielectric wall and by Sukenik *et al.* [13] for Na atoms in the presence of two parallel metallic walls. Kasevich *et al.* inferred an interaction decreasing as L^{-4} and Sukenik *et al.* demonstrated that the measured deflection of Na atoms was consistent with the potential $U(z,L)$ they calculated using discrete transitions to represent $\alpha(i\omega)$.

We determine $\alpha(i\omega)$ subject to oscillator strength sum rules. The sum rule (2) we know exactly. The sum rule (4) is known to high accuracy from a measurement of the static polarizability [16] to be 162.7 ± 0.8, where the uncertainty is a combination of systematic and statistical errors. Because the dynamic polarizability obeys the relationship

$$\frac{2}{\pi} \int_0^\infty d\omega \; \alpha(i\omega) = S(-1) \qquad (23)$$

the sum rule (3) is a critical parameter in the construction of $\alpha(\omega)$. Its value is less certain. Using many-body perturbation theory to third order, Johnson and Fritzsche [17] obtained 15.58 for $S(-1)$ and using the configuration-interaction wave functions of Müller [18], Müller [19] obtained 15.64. We will argue that despite this close agreement, they are too large.

III. OSCILLATOR STRENGTH DISTRIBUTION

Sodium has the configuration $(1s^2 2s^2 2p^6 3s)^2S$. As we shall demonstrate, the major contributor to the summations arising from the excitation of the valence $3s$ electron is the resonance $3s$-$3p$ transition. Empirical values of the oscillator strength may be derived from the measured radiative lifetimes of the $3p\,^2P$ state [20], from the measured linewidth [21] and from the coefficient of the leading long-range interaction term in electronically excited states of Na_2 [22]. Recent theoretical calculations [23,24] are consistent with the experiments in indicating an oscillator strength of 0.962 within a probable uncertainty of ±0.002. The uncertainty is

FIG. 1. The adopted photoionization cross sections.

TABLE I. Contributions to the sum rules $S(k)$ and their totals compared to empirical or theoretical values.

	$S(0)$	$S(-1)(e^2/a_0)$	$S(-2)/a_0^3$
$1s$	1.53	⋯	⋯
$2s$	1.25	0.19	0.040
$2p$	7.18	2.29	0.930
$3s$	1.04	12.6	161.7
Total	11	15.1	162.7
Experimental			162.7±0.8
Theoretical	11	15.6	
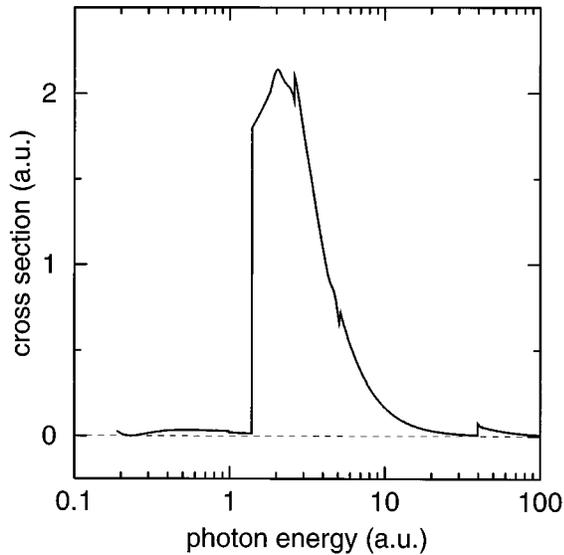
greater than the difference between the oscillator strengths of the $3\,^2S$-$3\,^2P_{1/2}$ and $3\,^2S$-$3\,^2P_{3/2}$ transitions [23]. The contributions to the sum rules (3) and (4) with $f=0.962$ are, respectively, 12.44 and 160.8. The small contributions from the higher transitions $3\,^2S$-$n\,^2P$ can be calculated from model potential calculations [25,26]. They are in order 0.018, 0.12, and 0.87.

Several sources exist for the continuum oscillator strengths corresponding to ejection of the $3s$ electron [26–37]. At low transition energies the many-body correlated calculations of Saha, Froese-Fischer, and Langhoff [38] may be the most reliable and we adopt them for energies from threshold to the excitation of the $2p$ shell at 31 eV. We extended them to higher energies using the results of Verner *et al.* [37]. The contributions to the three sum rules are 0.065, 0.054, and 0.090, respectively, giving total valence shell contributions of 1.04, 12.6, and 161.7.

We consider next transitions involving excitation and ionization of the K shell electrons. The threshold for ejection of a $1s$ electron is 1079.1 eV. The resonance structure of the K-shell absorption spectrum has been studied by Tuilier, Laporte, and Estera [39], but they do not give absolute cross sections. We adopt the cross sections calculated by Verner *et al.* [37]. The transitions make negligible contributions to $S(-1)$ and $S(-2)$ but contribute 1.53 to $S(0)$.

A confirmation of this estimate of the $1s$ contribution to $S(0)$ can be obtained by recognizing that the K-shell electrons are dominated by their interaction with each other and with the nucleus and are little affected by the outer $2s$ and $2p$ shell electrons. The sum rule $S(0)=2$ that the $1s$ transitions would otherwise obey is modified, because the $2p$ orbital is occupied and must be excluded from the sum. We calculated the $1s$-$2p$ oscillator strength using Hartree-Fock orbitals [40] and obtain a value of 0.18. There are two $1s$ electrons and, hence, $S(0)$ for the K shell is predicted to be approximately 1.64, in good agreement with our explicit calculation of 1.53.

Cross sections for the ejection of electrons from the $2s$ shell have been calculated by De Araújo and Petrini [41,42]

at photon energies from 86.7 eV to 250 eV and less accurately by Verner *et al.* [37] and Jain and Mathur [36] at higher energies. The calculations of Isenberg *et al.* [35] included the resonance structures arising from the transitions $(1s\,^2 2s\,^2 2p\,^6 3s)\,^2S$ to $(1s\,^2 2s 2p\,^6 3snp)\,^2P$. The resonance structures and their influence have been further explored by Craig and Larkins [43] and by De Araújo and Petrini [41,42], who demonstrated the importance of the shake-up processes. Measurements in the energy region above 71 eV in which the $2s$ electron can be ejected have been carried out by Wolff *et al.* [44], Codling, Hawley, and West [45], and Cubaynes *et al.* [46].

The oscillator strengths of the transitions into the $(2s2p\,^6 nl)\,^2P$ resonance states of Na are not large. The theoretical cross sections [47] and the measured cross sections [44,45] for $nl=3p$ of about 3×10^{-17} cm^2 at the peak and a width of about 0.2 eV yield an integrated oscillator strength of only 0.003. Most of the oscillator strength of the $2s$ transitions lies in the background continuum. The theoretical calculations of the nonresonant cross sections have been carried out at different levels of approximation and there are discrepancies between them. We adopt the calculations of De Araújo and Petrini [42] for photon energies between 86.7 eV and 209 eV, extrapolated smoothly to threshold at 71 eV and
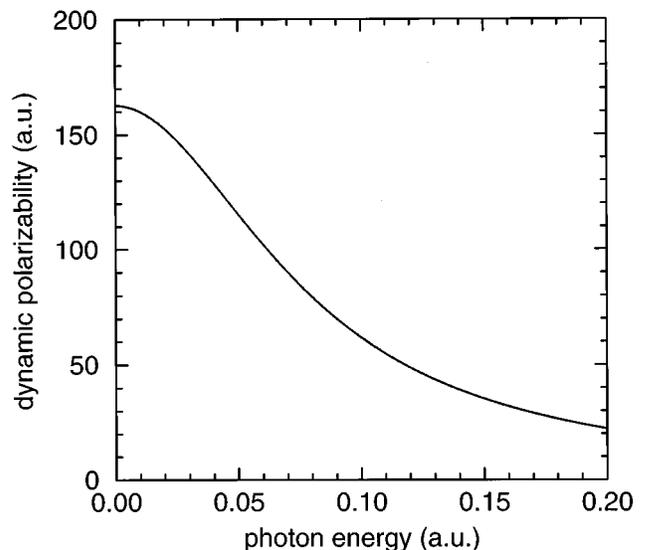
FIG. 2. The adopted dynamic polarizability at imaginary frequency Eq. (25).

TABLE II. The coefficients (in atomic units) and retardation coefficients (dimensionless) for the atom-atom interaction, column 2, and the atom-wall interaction, column 3, for Na. The last line gives the asymptotic values of F_6 and F_3 calculated, respectively, using Eqs. (16) and (17). Numbers in square brackets represent powers of ten.

R/a_0	C_6 1561 F_6	C_3 1.889 F_3
1.0[2]	9.984[−1]	8.658[−1]
1.5[2]	9.968[−1]	8.356[−1]
2.0[2]	9.949[−1]	8.116[−1]
2.5[2]	9.927[−1]	7.911[−1]
3.0[2]	9.902[−1]	7.727[−1]
5.0[2]	9.778[−1]	7.113[−1]
7.0[2]	9.626[−1]	6.606[−1]
1.0[3]	9.364[−1]	5.967[−1]
1.5[3]	8.882[−1]	5.122[−1]
2.0[3]	8.387[−1]	4.468[−1]
2.5[3]	7.904[−1]	3.950[−1]
3.0[3]	7.446[−1]	3.531[−1]
7.0[3]	4.825[−1]	1.852[−1]
1.0[4]	3.721[−1]	1.347[−1]
1.5[4]	2.651[−1]	9.193[−1]
2.0[4]	2.044[−1]	6.957[−1]
2.5[4]	1.658[−1]	5.590[−1]
3.0[4]	1.392[−1]	4.670[−1]
7.0[4]	6.056[−1]	2.011[−1]
1.0[5]	4.247[−1]	1.408[−1]
1.0[6]	4.254[−3]	1.409[−3]
Asymptotic		
1.0[6]	4.254[−3]	1.417[−3]

extrapolated to higher energies using the results of Verner *et al.* [37]. The corresponding contribution to the oscillator strength sum rule $S(0)$ is 1.25.
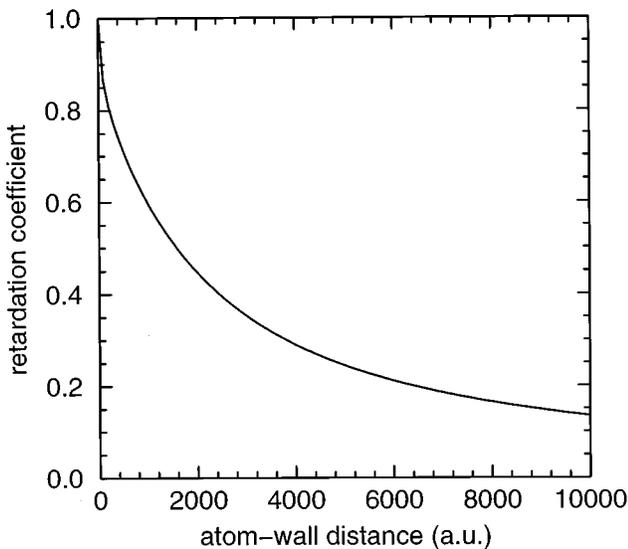
FIG. 3. The dimensionless retardation coefficient $F_3(R)$, Eq. (12), as a function of the atom-wall distance.
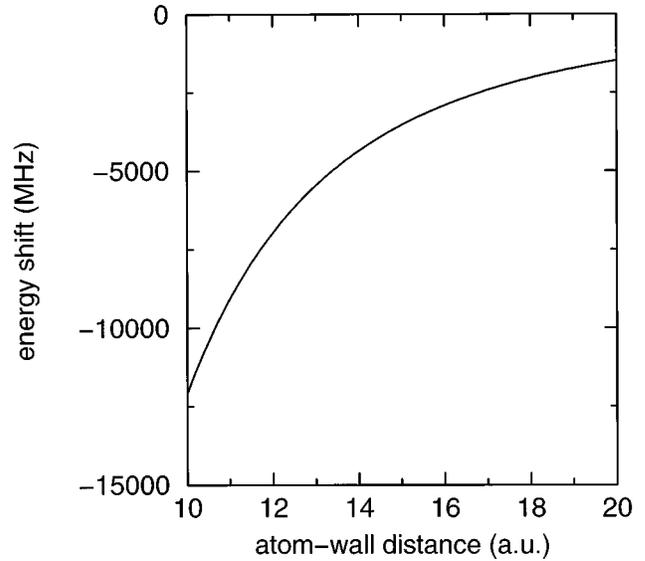
FIG. 4. The energy shift near the wall arising from the atom-wall interaction potential.

We assess its reliability by using the same arguments that we applied earlier, although the underlying assumptions are less secure than for the $1s$ shell.

We anticipate that the contribution to $S(0)$ of the $2s$ shell will be 2 minus the $2s$-$2p$ transition oscillator strength. For the oscillator strength we calculate 0.437 yielding $S(0)$ = 1.13, close to the value of 1.25 obtained from the photo-ionization cross sections.

The oscillator strength of 1.22 excluded from the $1s$ and $2s$ shells by the occupied $2p$ shell must be recovered by excitations of the $2p$ electrons for which we conclude $S(0)$ must equal 7.18.

The adopted $2s$ photoionization cross sections yield for the other two sum rules $S(-1)=0.19$ and $S(-2)=0.04$.

Several theoretical calculations of the cross sections for the ejection of electrons from the $2p$ shell of sodium have been carried out [47,35,36,48–52] and measurements have been reported from the threshold to 250 eV [38–40] in which the influence of shake up and conjugate shake-up satellite lines has been explored. Double ionization has also been investigated [53].
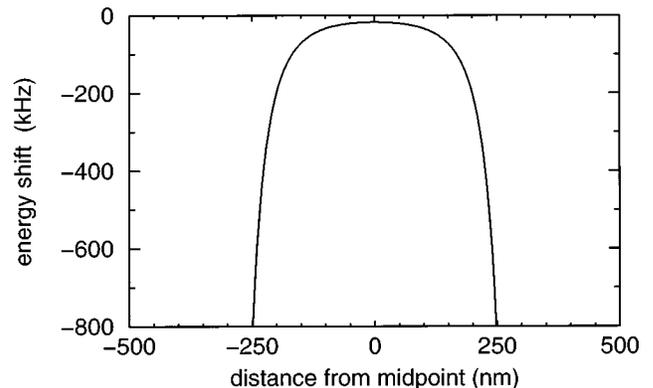
FIG. 5. The energy shift arising from the wall-atom-wall inter-action potential, Eq. (18), as a function of the distance z of the atom from the midpoint for a wall spacing $L=0.7$ μm.

FIG. 6. Same as Fig. 5 for $L=1$ μm.
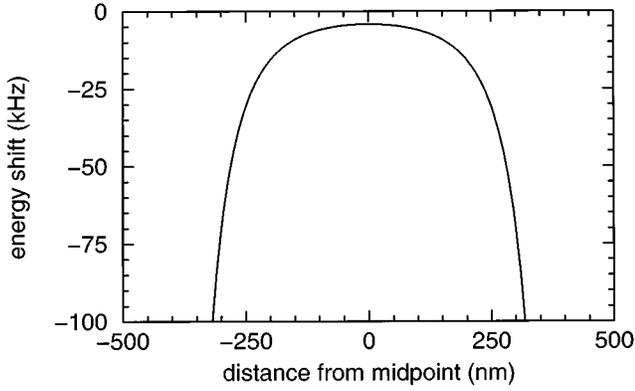
FIG. 7. Same as Fig. 6 for $L=2$ μm.

We adopt, as an initial estimate, the theoretical cross sections at photon energies from threshold to 200 eV presented by Liu and Liu [50] and we extend the cross sections to higher energies using the calculations of Verner *et al.* [37]. If we assume the contributions to the summations from the resonances arising from transitions of the $2p$ electron into the Rydberg states ($1s^2 2s^2 2p^5 3snl$) are negligible, we obtain a contribution to $S(0)$ of 7.19.

The adopted photoionization cross sections are shown in Fig. 1 and the contributions to the summations from excitation of the $1s$, $2s$, $2p$, and $3s$ electrons are summarized in Table I.

IV. CALCULATIONS

The continuum oscillator strength distribution is given in terms of the photoionization cross section $\sigma(E)$ by

$$\frac{df}{dE} = \frac{\sigma(E)}{2\pi^2\alpha} \quad E > 0.189 \tag{24}$$

and the dynamic dipole polarizability at imaginary frequency is given by
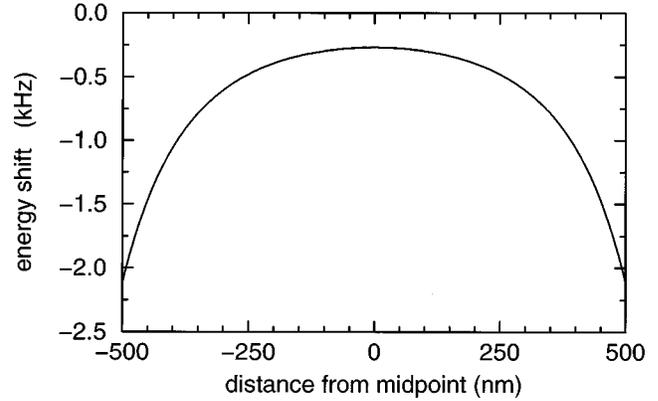
$$\alpha(i\omega) = \sum_n \frac{f_n}{(E_n - E_0)^2 + \omega^2} + \int \frac{df/dE}{E^2 + \omega^2}\, dE. \tag{25}$$

The dynamic polarizability corresponding to our choice of oscillator strengths is illustrated in Fig. 2.

The corresponding value of $S(-1)$ is 15.1. In a many-body perturbation theory calculation in which all contributions to third order in single-particle excitations were included, Johnson and Fritzsche [17] obtained $S(-1) = 15.58$ and using a multireference configuration-interaction method that recovered 95% of the correlation energy [18], Mueller [19] obtained $S(-1) = 15.64$. Despite the close agreement between the two *ab initio* calculations, we believe that the higher-order correlations are not negligible and that the empirical value of 15.1 that we obtain is a more accurate estimate of $S(-1)$ given that our oscillator strength distribution closely satisfies the adjacent sum rules $S(0)$ and $S(-2)$ and the major contribution which arises from the $3s$-$3p$ transition is accurately known.

For the interaction coefficients, we obtain $C_6 = 1561$, $C_9 = 189\,200$, and $C_3 = 1.889$. The model potential calculations
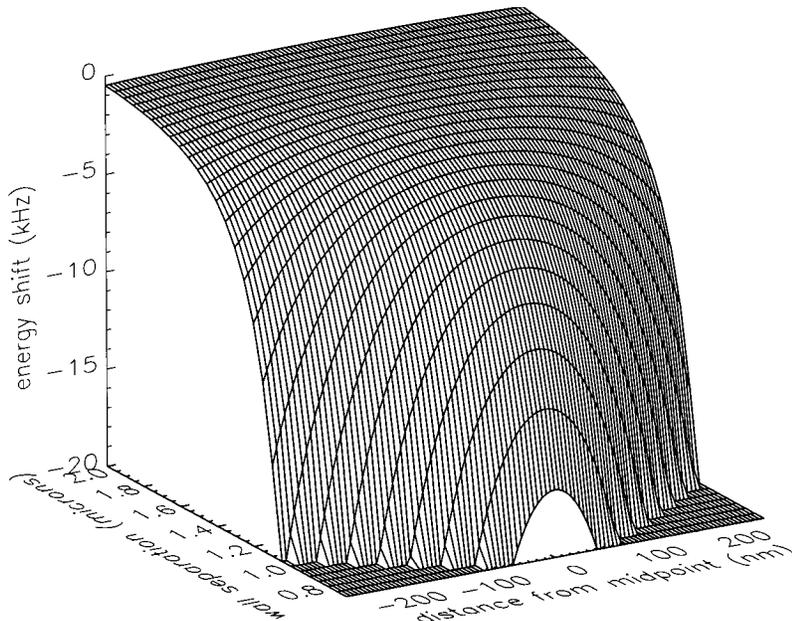
FIG. 8. The energy shift arising from the wall-atom-wall interaction potential, Eq. (18), as a function of the wall separation L and the distance z of the atom from the midpoint.

[9] modified to achieve consistency with the more recent measurements of $\alpha(0)$ [16] yielded $C_6 = 1539$ [54] and $C_9 = 187\,700$ [55]. The retardation coefficients $F_6(R)$ and $F_3(R)$ obtained from the empirical dynamic polarizability are listed in Table II, along with the values of C_6 and C_3. The potentials can be obtained using the data in Table II with Eqs. (9) and (11). The potential for Na_2 is somewhat more precise than that given by Marinescu, Babb, and Dalgarno [56]. For the atom-wall case, the coefficient $F_3(R)$ and the potential $V_W(R)$ are shown, respectively, in Figs. 3 and 4. The potential for wall-atom-wall interaction was evaluated as a function of L and z. Values of the corresponding energy shifts at $L = 0.7$, 1, and 2 μm are shown, respectively, in Figs. 5, 6, and 7. Sukenik *et al.* [13] and Marksteiner *et al.* [57] evaluated the first term of Eq. (18) for Na, incorporating only the discrete transitions in the oscillator strength distribution. The energy shifts at 1 μm agree well with, and are slightly more accurate, than those calculated by Sukenik *et al.* [13], and are in qualitative agreement with those given by Marksteiner *et al.* for dielectric walls separated by 1.65 μm. There is a dramatic increase in the potential $U(z,L)$ as the wall separation decreases or as the atom approaches either wall, which is demonstrated in Fig. 8.

Note added in proof. C. F. Fischer, M. Godefroid, and P. Jönsson (private communication) have calculated $S(-1)$ withj a core polarization model with additional correlations from the core [Phys. Rev. A **53**, 4021 (1996)]. They find with single and double substitutions that $S(-1) = 15.47$.

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