

## Long-range interactions of lithium atoms

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The long-range interactions of two atoms, of an atom and a dielectric wall, of an atom and a perfectly conducting wall, and of an atom between two perfectly conducting walls are calculated, including the effects of retardation, for Li using dynamic polarizabilities determined from highly correlated, variationally determined wave functions. [S1050-2947(97)02604-8]

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### I. INTRODUCTION

Long-range interactions between two ground state atoms and between a ground state atom and a surface are now measured using lasers and cold atoms or atomic beams. Photoassociation spectroscopy has yielded strict limits on the values of coefficients of dispersion forces between two Rb atoms [1], two Na atoms [2], and two Li atoms [3] in their ground states. Spectroscopy combined with deflection of alkali-metal atomic beams near surfaces [4] or reflection of Na atoms from surfaces in atomic fountains [5,6] have made it possible to measure the coefficients of atom-surface forces. The experiments are consistent with theoretical models for the interaction potentials, but accurate theoretical estimations of the potentials remain elusive for atoms other than H and He. The effects of retardation, due to the finite speed of light, cause the potentials to become weaker, approaching simple power laws for asymptotically large distances. The advent of highly correlated basis sets for Li using multiple nonlinear variational parameters [7] makes it possible to perform well-converged calculations of the dynamic electric polarizability functions, thereby enabling, as we will show, precise evaluation of long-range interaction potentials, including retardation, for two Li atoms, for a Li atom and a dielectric or perfectly conducting wall, and for a Li atom between two perfectly conducting walls.

### II. FORMULATION

#### A. Atom-atom interactions

The effect of retardation on the long-range induced dipole interactions of two atoms was investigated by Casimir and Polder [8] and the effects on higher induced multipoles by Au and Feinberg [9], Jenkins, Salam, and Thirunamachandran [10], and Power and Thirunamachandran [11]. In this paper the dipolar-dipolar and dipolar-quadrupolar interactions are considered, the higher multipolar interactions being negligible. We use the expression for the retarded dipolar-quadrupolar interaction of Power and Thirunamachandran [11] which differs from the approximate expression obtained by Au and Feinberg [9].

The interaction potential between two like atoms including the effects of retardation can be written [8,11]

$$V(R) = -\frac{C_6 f_6(R)}{R^6} - \frac{C_8 f_8(R)}{R^8}, \quad (1)$$

where  $R$  is the internuclear distance,

$$C_6 = \frac{3}{\pi} G(1,1), \quad (2)$$

$$C_8 = \frac{15}{\pi} G(1,2), \quad (3)$$

with

$$G(l_a, l_b) = \int_0^\infty \alpha_{l_a}(i\omega) \alpha_{l_b}(i\omega) d\omega \quad (4)$$

and the retardation coefficients are

$$f_6(R) = \frac{1}{\pi C_6} \int_0^\infty d\omega \exp(-2\alpha_{FS}\omega R) \alpha_1^2(i\omega) P_{11}(\omega\alpha_{FS}R), \quad (5)$$

where

$$P_{11}(x) = x^4 + 2x^3 + 5x^2 + 6x + 3 \quad (6)$$

and

$$f_8(R) = \frac{1}{3\pi C_8} \int_0^\infty d\omega \exp(-2\alpha_{FS}\omega R) \times \alpha_1(i\omega) \alpha_2(i\omega) P_{12}(\omega\alpha_{FS}R), \quad (7)$$

where

$$P_{12}(x) = \frac{1}{2}x^6 + 3x^5 + \frac{27}{2}x^4 + 42x^3 + 81x^2 + 90x + 45, \quad (8)$$

and  $\alpha_{FS} = 1/137.0359895$  is the fine structure constant. We use atomic units throughout.

The functions  $\alpha_l(i\omega)$  appearing in Eqs. (2)–(7) are the dynamic electric multipole polarizability functions at imaginary frequency defined by expressions (6)–(9) of [12].

The retardation coefficients are dimensionless and can be expanded for small  $R$  as

$$C_6 f_6(R) \sim C_6 - \alpha_{FS}^2 R^2 W_4, \quad (9)$$

with

$$W_4 = \frac{1}{\pi} \int_0^\infty d\omega \omega^2 \alpha_1^2(i\omega) \quad (10)$$

and

$$C_8 f_8(R) \sim C_8 - \alpha_{\text{FS}}^2 R^2 W_6, \quad (11)$$

with

$$W_6 = \frac{3}{\pi} \int_0^\infty d\omega \omega^2 \alpha_1(i\omega) \alpha_2(i\omega). \quad (12)$$

The coefficients  $W_4$  and  $W_6$  can also be derived from an analysis in perturbation theory of the orbit-orbit term arising from the Breit interaction in the Pauli approximation [13]. Expanding Power and Thirunamachandran's result for  $-C_8 f_8(R)$  for small  $R$ , according to Eq. (11), we find a value of  $W_6$  a factor of  $\frac{3}{2}$  times larger than the value of  $W_6$  resulting from the theory of Au and Feinberg [see Eq. (4.21) of [14]]. This resolves the discrepancy of  $\frac{3}{2}$  found in [14], between the value of  $W_{LL,4;2}$  for H from Johnson, Epstein, and Meath [15], who evaluated terms from the Breit interaction in the Pauli approximation [13], and the value of  $W_6$  evaluated for H in [14] using the Au and Feinberg formulation.

For asymptotically large  $R$ , the retardation coefficients have the limits

$$f_6(R) \rightarrow \frac{23}{4\pi\alpha_{\text{FS}}R} \frac{\alpha_1^2(0)}{C_6}, \quad R \rightarrow \infty \quad (13)$$

and

$$f_8(R) \rightarrow \frac{531}{16\pi\alpha_{\text{FS}}R} \frac{\alpha_1(0)\alpha_2(0)}{C_8}, \quad R \rightarrow \infty. \quad (14)$$

### B. Atom-wall interactions

Expressions for the interaction potential of an atom and a dielectric wall, including the effects of retardation, have been given by Dzyaloshinskii, Lifshitz, and Pitaevskii [16], Parsegian [17], and Tikochinsky and Spruch [18]. For a wall with a dielectric constant  $\epsilon$ , the potential can be written [18]

$$V_{\text{AtD}}(R, \epsilon) = -\frac{\alpha_{\text{FS}}^3}{2\pi} \int_0^\infty d\xi \xi^3 \alpha_1(i\xi) \times \int_1^\infty dp \exp(-2\xi R p \alpha_{\text{FS}}) H(p, \epsilon), \quad (15)$$

where

$$H(p, \epsilon) = \frac{s-p}{s+p} + (1-2p^2) \frac{s-\epsilon p}{s+\epsilon p}, \quad (16)$$

$$s = (\epsilon - 1 + p^2)^{1/2}, \quad (17)$$

and  $R$  is now the atom-wall distance. We follow the notation of [19] and the subscripts At, D, and M denote, respectively, an atom, a dielectric wall, and a perfectly conducting (i.e., metal) wall.

For asymptotically large distances,

TABLE I. The coefficients  $W_4$  and  $W_6$  for two Li atoms. Numbers in parentheses represent theoretical uncertainty due to the finite basis set size.

$W_4$	$W_6$	Reference
3.214(2)	219.9(2)	Present
2.9312		Easa and Shukla [23]
3.233		Margoliash and Meath [22]

$$V_{\text{AtD}}(R, \epsilon) \rightarrow V_{\text{AtD}}^\infty(R, \epsilon) = -\frac{K_4}{R^4} \frac{\epsilon-1}{\epsilon+1} \phi(\epsilon), \quad (18)$$

where

$$K_4 = 3\alpha_1(0)/(8\pi\alpha_{\text{FS}}) = 16.36\alpha_1(0) \quad (19)$$

and

$$\phi(\epsilon) = \frac{\epsilon+1}{2(\epsilon-1)} \int_0^\infty \frac{dp}{(p+1)^4} H(p+1, \epsilon). \quad (20)$$

Direct integration of Eq. (20) yields

$$\phi(\epsilon) = \frac{\epsilon+1}{\epsilon-1} \left[ \frac{1}{3} + \epsilon + \frac{4 - (\epsilon+1)\epsilon^{1/2}}{2(\epsilon-1)} + A(\epsilon) + B(\epsilon) \right], \quad (21)$$

where

$$A(\epsilon) = -\frac{\text{arcsinh}[(\epsilon-1)^{1/2}]}{2(\epsilon-1)^{3/2}} [1 + \epsilon + 2\epsilon(\epsilon-1)^2] \quad (22)$$

and

$$B(\epsilon) = \frac{\epsilon^2}{(\epsilon+1)^{1/2}} [\text{arcsinh}(\epsilon^{1/2}) - \text{arcsinh}(\epsilon^{-1/2})], \quad (23)$$

in agreement with Dzyaloshinskii *et al.* [16]. Approximations to  $V_{\text{AtD}}^\infty(R, \epsilon)$  will be considered in Sec. III below.

The potential for the interaction of an atom and a perfectly conducting wall follows by letting  $\epsilon \rightarrow \infty$  in Eq. (15), giving [8]

$$V_{\text{AtM}}(R) \equiv V_{\text{AtD}}(R, \infty) = -\frac{C_3 f_3(R)}{R^3}, \quad (24)$$

where the coefficient is

$$C_3 = \frac{1}{4\pi} \int_0^\infty d\omega \alpha_1(i\omega), \quad (25)$$

and the retardation coefficient is

$$f_3(R) = \frac{1}{8C_3\pi\alpha_{\text{FS}}R} \int_0^\infty dx e^{-x} \alpha_1(ix/2\alpha_{\text{FS}}R) [\frac{1}{2}x^2 + x + 1]. \quad (26)$$

Equation (26) approaches for asymptotically large distances the form

TABLE II. The dimensionless retardation coefficients  $f_6(R)$  and  $f_8(R)$  for the atom-atom interaction. The dispersion coefficients  $C_6$  and  $C_8$  from [12] are also given.

$R$	$C_6$	$C_8$
	1393.39(16) $f_6(R)$	83 425.8(4.2) $f_8(R)$
15	1.0000	1.0000
20	1.0000	0.9999
25	0.9999	0.9999
30	0.9999	0.9999
50	0.9997	0.9997
70	0.9995	0.9994
100	0.9991	0.9988
150	0.9980	0.9974
200	0.9966	0.9955
250	0.9950	0.9933
300	0.9931	0.9907
500	0.9833	0.9775
700	0.9708	0.9608
1000	0.9489	0.9319
1500	0.9076	0.8791
2000	0.8641	0.8256
2500	0.8208	0.7743
3000	0.7789	0.7263
5000	0.6341	0.5709
7000	0.5253	0.4627
10000	0.4113	0.3555
15000	0.2970	0.2528
20000	0.2304	0.1947
25000	0.1875	0.1579
30000	0.1578	0.1326
50000	0.0961	0.0805
70000	0.0689	0.0577
100000	0.0484	0.0405

$$f_3(R) \rightarrow \frac{3}{8\pi} \frac{\alpha_1(0)}{\alpha_{\text{FS}} C_3}, \quad (27)$$

giving

$$V_{\text{AtM}}(R) \rightarrow V_{\text{AtM}}^\infty(R) \equiv -K_4/R^4. \quad (28)$$

The interaction potential for an atom between two parallel, perfectly conducting walls has been given by Barton [20] and by Zhou and Spruch [21]. It can be expressed as

$$V_{\text{MAtM}}(z, L) = T_2(L) - T_1(z, L), \quad (29)$$

where

$$T_1(z, L) = \frac{1}{\pi L^3} \int_0^\infty dt \frac{t^2 \cosh(2zt/L)}{\sinh t} \int_0^{t/\alpha_{\text{FS}} L} ds \alpha_1(is) \quad (30)$$

and

$$T_2(L) = \frac{\alpha_{\text{FS}}^2}{\pi L} \int_0^\infty ds s^2 \alpha_1(is) \int_{\alpha_{\text{FS}} L s}^\infty dt \frac{e^{-t}}{\sinh t}, \quad (31)$$

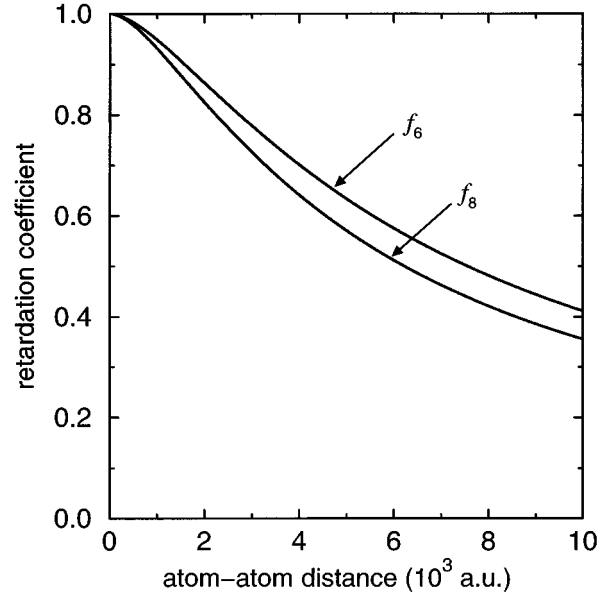


FIG. 1. Dimensionless retardation coefficients  $f_6(R)$  and  $f_8(R)$  for two Li atoms.

where  $L$  is the interwall distance and  $z$  is the distance of the atom from the midpoint. For small values of  $L$ , the potential is [21]

$$V_{\text{MAtM}}(z, L) \rightarrow -\frac{4}{L^3} T(z/L) C_3, \quad (32)$$

TABLE III. Values of  $-R^3 V_{\text{AtD}}(R, \epsilon)$ , where  $V_{\text{AtD}}(R, \epsilon)$  is the atom-wall potential, for values of  $\epsilon$  corresponding to fused silica and BK-7 glass in, respectively, the second and third columns, and in the fourth column values of  $-R^3 V_{\text{AtM}}(R)$  for a perfectly conducting wall.

$R$	Fused silica $\epsilon = 2.123$	BK-7 glass $\epsilon = 2.295$	Perfect $\epsilon = \infty$
10	0.5360	0.5859	1.5007
15	0.5323	0.5819	1.4937
20	0.5289	0.5782	1.4871
25	0.5259	0.5749	1.4810
30	0.5230	0.5717	1.4753
50	0.5130	0.5608	1.4551
70	0.5045	0.5515	1.4380
100	0.4933	0.5392	1.4157
150	0.4772	0.5215	1.3836
200	0.4629	0.5060	1.3551
250	0.4500	0.4919	1.3289
300	0.4381	0.4788	1.3042
500	0.3974	0.4344	1.2160
700	0.3644	0.3983	1.1393
1000	0.3244	0.3546	1.0398
1500	0.2741	0.2996	0.9049
2000	0.2368	0.2589	0.7981
2500	0.2081	0.2276	0.7118
3000	0.1853	0.2026	0.6409
5000	0.1276	0.1395	0.4526

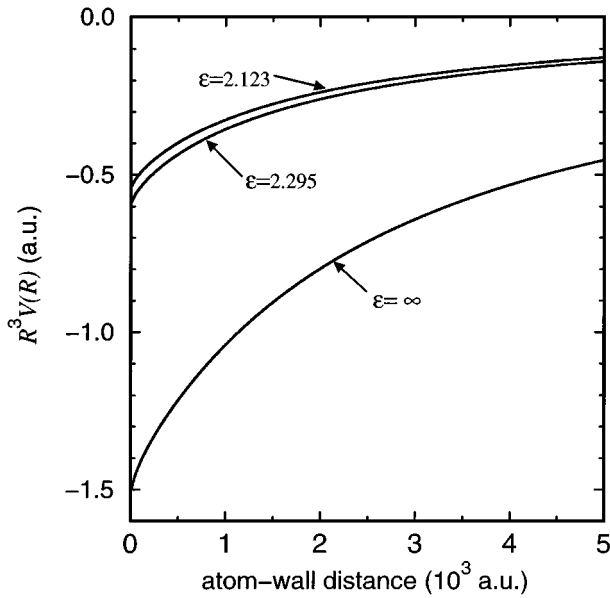


FIG. 2. Values of  $R^3 V_{AtD}(R, \epsilon)$ , for values of  $\epsilon$  corresponding to fused silica ( $\epsilon=2.123$ ), BK-7 glass ( $\epsilon=2.295$ ), and for a perfectly conducting wall ( $\epsilon=\infty$ ).

where

$$T(z/L) = \int_0^\infty dt \frac{t^2 \cosh(2tz/L)}{\sinh t} \quad (33)$$

and  $C_3$  is defined in Eq. (25). For asymptotically large values of  $L$ , the potential is

$$V_{MAIM}^\infty(z, L) = \frac{\pi^3 \alpha_1(0)}{\alpha_{FS} L^4} \left[ \frac{1}{360} - \frac{3 - 2\cos^2(\pi z/L)}{8\cos^4(\pi z/L)} \right]. \quad (34)$$

### III. CALCULATIONS

The calculations of the wave functions and the polarizability response functions have been described previously in, respectively, [7] and [12]. We briefly summarize the procedures.

The basis set for the lithium atom is constructed in Hyllebras coordinates [7]

$$\{\phi_{t, \mu_t}(\alpha_t, \beta_t, \gamma_t) = r_1^{j_1} r_2^{j_2} r_3^{j_3} r_{12}^{j_{12}} r_{23}^{j_{23}} r_{31}^{j_{31}} e^{-\alpha_t r_1 - \beta_t r_2 - \gamma_t r_3}\}, \quad (35)$$

where  $\mu_t$  denotes a sextuple of integer powers  $j_1, j_2, j_3, j_{12}, j_{23}$ , and  $j_{31}$ , and index  $t$  labels different sets of nonlinear parameters  $\alpha_t, \beta_t$ , and  $\gamma_t$ . Except for some truncations, all terms are included such that

$$j_1 + j_2 + j_3 + j_{12} + j_{23} + j_{31} \leq \Omega. \quad (36)$$

The wave function is expanded from the multiple basis sets

TABLE IV. The dimensionless function  $\phi(\epsilon)$ .

$\epsilon$	$\phi(\epsilon)$
1	$\frac{23}{30}$
1.5	0.761364
2	0.760757
2.123	0.760970
2.295	0.761425
4	0.770171
7	0.787334
9	0.797062
13	0.812791
16	0.822186
20	0.832501
50	0.874337
100	0.902534
500	0.950261
1000	0.963647
5000	0.982986
10000	0.987836
50000	0.994478
$1 \times 10^{11}$	0.999996

$$\Psi(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) = \mathcal{A} \sum_t \sum_{\mu_t} a_{t, \mu_t} \phi_{t, \mu_t}(\alpha_t, \beta_t, \gamma_t) \times (\text{angular function})(\text{spin function}). \quad (37)$$

A complete optimization is performed with respect to all the nonlinear parameters. The screened hydrogenic wave function is also included explicitly in the basis set.

The dynamic polarizabilities are evaluated using effective oscillator strengths and transition energies obtained from the diagonalization of the Hamiltonian in a basis set of  $S$  sym-

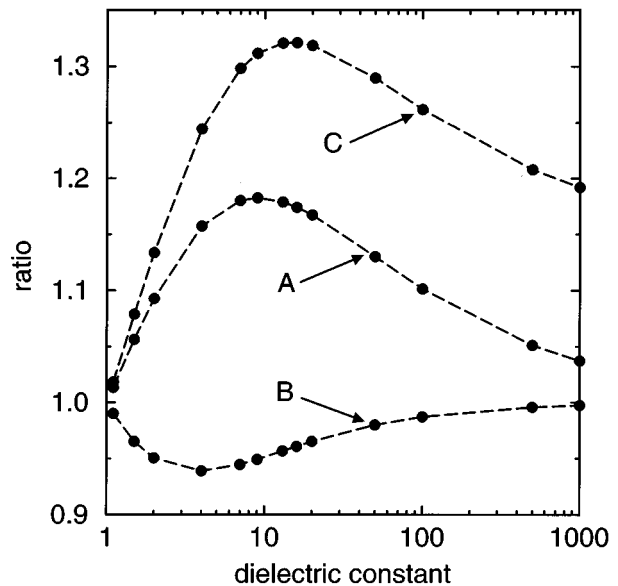


FIG. 3. Ratio of several approximations given by Spruch and Tikochinsky to the exact atom-wall potential calculated in the present work. The symbols A, B, and C represent, respectively, values from Eqs. (38), (39), and (40).

TABLE V. The coefficient  $C_3$  for the Li atom-wall interaction calculated in the present work compared to values calculated from Eq. (42) (direct), using matrix elements given by various authors, or from pseudo-oscillator strength distribution data of dimension  $M$  tabulated by various authors (osc. str.).

$C_3$	Method	Reference
1.518(2)	osc. str.	Present
1.518 000 51(3)	direct	Yan and Drake [7]
1.518 000	direct	King [24]
1.49	osc. str. ( $M=11$ )	Stacey and Dalgarno [26]
1.52	osc. str. ( $M=10$ )	Margoliash and Meath [22]

metry for the ground state and of  $P$  and  $D$  symmetry, respectively, for the intermediate states corresponding to the dipole and quadrupole polarizabilities. The basis sets were the size 919 set from [12] for the  $S$  symmetry and the size 1846 sets from [12] for the  $P$  and the  $D$  symmetries. A detailed discussion of the evaluation of  $\alpha_l(i\omega)$  can be found in [12]. The static polarizabilities have the values  $\alpha_1(0) = 164.111(2)$  and  $\alpha_2(0) = 1423.266(5)$  [12].

Values of the coefficient  $W_4$  for two Li atoms have been determined by Margoliash and Meath [22] and by Easa and Shukla [23]. Using our functions  $\alpha_1(i\omega)$  and  $\alpha_2(i\omega)$ , we determined the coefficients  $W_4$  and  $W_6$  using, respectively, Eqs. (10) and (12), and the results are compared with previous results in Table I. We also calculated the coefficients  $f_6(R)$  and  $f_8(R)$  using Eqs. (5) and (7) at various values of  $R$ . The results are given in Table II and Fig. 1. The values of the dipole-dipole potential  $-C_6 f_6(R)/R^6$  are in agreement with, but are more accurate than, those given in Ref. [14], calculated using a model potential method. The values of the dipole-quadrupole potential  $-C_8 f_8(R)/R^8$  replace those given in Ref. [14], which were calculated using the expression of Au and Feinberg as discussed above. The dipole-quadrupole potential is usually of secondary importance due to its  $1/R^8$  power law behavior.

Using the polarizability function  $\alpha_1(i\omega)$  we evaluated  $V_{AtD}(R, \epsilon)$  for values of  $\epsilon = 2.123$  and  $2.295$  corresponding to, respectively, fused silica and BK-7 glass. The values are listed in Table III and illustrated in Fig. 2 for values of  $R$  up to  $5000a_0$ . For larger values of  $R$ , the potential can be obtained from Eq. (18). The values of  $\phi(\epsilon)$  from our calculations are listed in Table IV and they are in agreement with the representative values given in Fig. 10 of Ref. [16].

Three approximations  $V'_{AtD}$ ,  $V''_{AtD}$ , and  $V'''_{AtD}$  for  $V_{AtD}$  were obtained by Spruch and Tikochinsky by imposing the requirements that the interaction be exact for  $\epsilon \approx \infty$  and for  $\epsilon \approx 1$ , see Eqs. (4.5), (4.9), and (4.12) of [19]. Expressing the approximations as ratios to the exact potential at very large distances, we have

$$\frac{V'_{AtD}}{V_{AtD}} = \frac{F(\epsilon)}{\epsilon + \frac{37}{23}}, \quad (38)$$

$$\frac{V''_{AtD}}{V_{AtD}} = \frac{F(\epsilon)}{\epsilon + \frac{30}{23}\epsilon^{1/2} + \frac{7}{23}}, \quad (39)$$

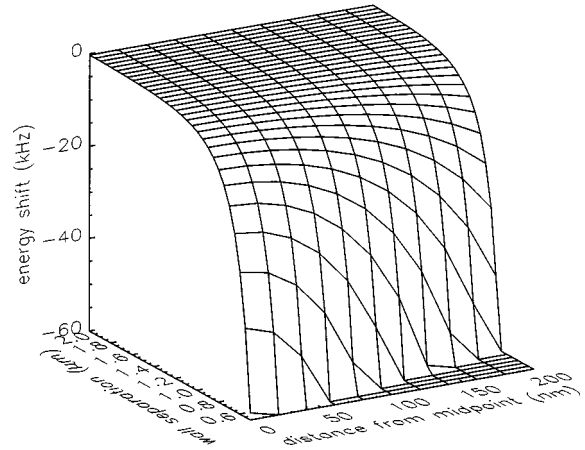


FIG. 4. The energy shift arising from the wall-atom-wall potential for various values of the wall separation  $L$  and the distance of the atom from the midpoint  $z$ . Only the values for  $z > 0$  are shown as the energy shift is symmetric about the  $z=0$  plane.

and

$$\frac{V'''_{AtD}}{V_{AtD}} = \frac{\frac{23}{20}F(\epsilon)}{\epsilon + 2}, \quad (40)$$

where

$$F(\epsilon) \equiv \frac{\epsilon + 1}{\phi(\epsilon)}. \quad (41)$$

We calculated the ratios appearing in Eqs. (38)–(40) using our values of  $\phi(\epsilon)$  and the results are presented in Fig. 3. Our results indicate that the second approximation defined by Eq. (39) is the most accurate, differing by about 6% at most from the exact value of the potential. The third approximation was developed for small values of  $\epsilon$  where it is seen to be somewhat less accurate than the second approximation.

The interaction potential for a Li atom and a perfectly conducting wall was evaluated from Eqs. (24)–(26). The value for the coefficient  $C_3$  is in excellent agreement with previous determinations, listed in Table V, particularly with those calculated from the alternative expression

$$C_3 = \frac{1}{12} \left\langle 0 \left| \left( \sum_{i=1}^N \mathbf{r}_i \right)^2 \right| 0 \right\rangle, \quad (42)$$

which follows from integration of Eq. (25), where  $N$  is the number of electrons. Note that only the ground state wave function  $|0\rangle$  is required to evaluate Eq. (42). For Table V we used expectation values given by King [24] and Yan and Drake [7]. The values of  $R^3 V(R)$  were calculated from Eq. (26) and values are listed in Table III. The present calculations of the potential values are in agreement with, but are more accurate than, those given in Ref. [25].

The potential  $V_{MAtM}(z, L)$  was evaluated using Eq. (29) for a range of wall separations  $L$  and distances  $z$  of the atom

from the midpoint. Values of the energy shift arising from the potential for values of  $L$  and  $z$  that might be realized in an experiment are given in Fig. 4.

The expressions involving dielectric walls in this paper were obtained under the approximation  $\epsilon(\omega) \approx \epsilon(0) \equiv \epsilon$ , where  $\epsilon(\omega)$  is the frequency-dependent dielectric function of the wall. This is an excellent approximation for  $R \sim \infty$ , but at smaller  $R$  it could lead to significant error if resonances play a role.

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