

Long-range interactions of metastable helium atoms

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The electric dipole, quadrupole, and octupole polarizabilities of a helium atom and the associated dispersion coefficients for the interaction of two helium atoms are calculated for the atomic $n=2$ singlet and triplet states, respectively, $\text{He}(2^1S)$ and $\text{He}(2^3S)$. The calculations utilize highly accurate, variationally determined, wave functions composed of doubled basis sets. The effects of retardation due to the finite speed of light are calculated for the dispersion potentials of two $\text{He}(2^1S)$ or two $\text{He}(2^3S)$ atoms and for the atom-surface interaction potentials of a $\text{He}(2^1S)$ or a $\text{He}(2^3S)$ atom with a perfectly conducting or a dielectric wall. [S1050-2947(98)06208-8]

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The advent of doubled basis sets has made it possible to calculate precisely many properties of two-electron atomic systems [1–4]. We apply variational methods developed previously and demonstrated for the helium atom [5] to calculate nonrelativistic values of the electric dipole, quadrupole, and octupole polarizabilities and corresponding dispersion and retardation coefficients for the metastable $n=2$ singlet and triplet states, respectively, $\text{He}(2^1S)$ and $\text{He}(2^3S)$. The results are useful for studies of elastic and inelastic collisional processes of the atoms [6,7]. Additionally, potentials for the atom-wall interaction of a $\text{He}(2^1S)$ or a $\text{He}(2^3S)$ atom and a single perfectly conducting wall or a dielectric wall are calculated with the inclusion of retardation effects due to the finite speed of light. Our results for atom-wall interactions are germane to experiments involving atom-evanescent wave mirrors [8].

In this paper the notation of Ref. [9] is followed very closely; references to equations of Ref. [9] will be preceded by the symbol I. Atomic units are used throughout.

The dispersion interaction of two like atoms can be written as

$$U(R) = -C_6 R^{-6} - C_8 R^{-8} - C_{10} R^{-10}, \quad (1)$$

where the coefficients C_6 , C_8 , and C_{10} are the van der Waals coefficients, R is the interatomic distance, and

$$C_6 = (3/\pi)G(1,1), \quad (2)$$

$$C_8 = (15/\pi)G(1,2), \quad (3)$$

$$C_{10} = (28/\pi)G(1,3) + (35/\pi)G(2,2), \quad (4)$$

TABLE I. Values of the static polarizabilities $\alpha_1(0)$, $\alpha_2(0)$, and $\alpha_3(0)$ for the 2^1S and 2^3S states of He. Numbers in parentheses represent the estimated error in the last digit of the listed, extrapolated value.

State	$\alpha_1(0)$	$\alpha_2(0)$	$\alpha_3(0)$
2^1S	800.316 33(7)	7 106.053 7(5)	293 703.50(6)
2^3S	315.631 468(12)	2 707.877 3(3)	88 377.325 3(7)

with

$$G(l,m) = \int_0^\infty \alpha_l(i\omega)\alpha_m(i\omega)d\omega, \quad (5)$$

where $\alpha_l(i\omega)$ is the 2^l -pole dynamic polarizability function evaluated at imaginary frequency defined by Eqs. (6)–(9) of Ref. [5], and similarly for $\alpha_m(i\omega)$.

When the effects of retardation due to the finite speed of light are considered the potential $U(R)$, Eq. (1), can be replaced by [10,11]

$$V(R) = -C_6 f_6(R)R^{-6} - C_8 f_8(R)R^{-8} - C_{10} f_{10}(R)R^{-10}. \quad (6)$$

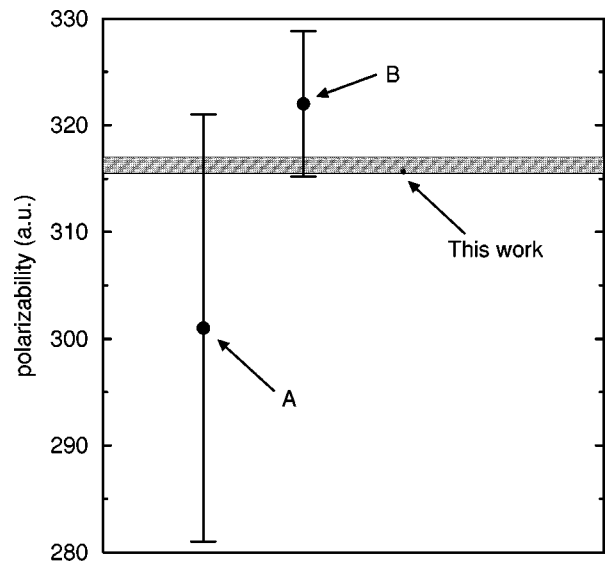


FIG. 1. Comparison of values of the static polarizability $\alpha_1(0)$ (in atomic units) for $\text{He}(2^3S)$. The point A is the measurement of Crosby and Zorn [20] and the point B is that of Ekstrom *et al.* [18] determined in combination with measurements from Molof *et al.* [19]. The small dot above the arrow labeled “This work” represents the present calculation, which lies within the upper and lower bounds of Glover and Weinhold (shaded region) [21].

TABLE II. Comparison of static multipole polarizabilities $\alpha_1(0)$, $\alpha_2(0)$, and $\alpha_3(0)$ for He(2^1S). For the experimental value numbers in parentheses give the quoted error.

Author (year)	Ref.	$\alpha_1(0)$	$\alpha_2(0)$	$\alpha_3(0)$
Crosby and Zorn (1977) Expt.	[20]	729(88)		
Chung and Hurst (1966)	[25]	801.9		
Drake (1972)	[26]	800.2		
Chung (1977)	[27]	801.10		
Glover and Weinhold (1977)	[28]	803.31 \pm 6.61 ^a		
Lamm and Szabo (1980), ECA	[29]	790.8		
Rérat <i>et al.</i> (1993)	[30]	803.25	6 870.9	
Chen (1995)	[31]	800.34		
This work		800.316 33(7)	7 106.053 7(5)	293 703.50(6)

^aBounded theoretical value.

The coefficient $f_{10}(R)$ will not be considered in this paper as it is likely that the practical consequences of retardation on the C_{10} term will be negligible.

Expressions for the retardation coefficients, $f_6(R)$ and $f_8(R)$, as integrals involving the dynamic electric dipole polarizabilities, are given in Eqs. I-(5) and I-(7).

The form (6) intrinsically includes certain relativistic effects, so that when $f_6(R)$ and $f_8(R)$ are expanded in powers of the fine structure constant $\alpha_{fs} = 1/137.035 989 5$ for small distances

$$V(R) \sim -R^{-6}[C_6 - \alpha_{fs}^2 R^2 W_4] - R^{-8}[C_8 - \alpha_{fs}^2 R^2 W_6], \quad (7)$$

where

$$W_4 = \frac{1}{\pi} \int_0^\infty d\omega \omega^2 \alpha_1^2(i\omega) \quad (8)$$

and

$$W_6 = \frac{3}{\pi} \int_0^\infty d\omega \omega^2 \alpha_1(i\omega) \alpha_2(i\omega). \quad (9)$$

The relativistic origin of the coefficient W_4 has been discussed by Power and Zienau [12], see also [13]. The coefficient W_6 of the factor α_{fs}^2/R^6 in Eq. (7) corresponds to the

theory of Power and Thirunamachandran [11] and is equal to the coefficient $W_{LL;4,2}$ in the theory of Meath and Hirschfelder based on the Breit-Pauli Hamiltonian [13]. As the distance increases retardation arising from the finite speed of light becomes important and the potential approaches its asymptotic form, see Eqs. I-(13) and I-(14),

$$V(R) \sim -K_7 R^{-7} - K_9 R^{-9}, \quad (10)$$

with

$$K_7 = \frac{23}{4\pi} \frac{\alpha_1^2(0)}{\alpha_{fs}} = 250.81 \alpha_1^2(0),$$

$$K_9 = \frac{531}{16\pi} \frac{\alpha_1(0) \alpha_2(0)}{\alpha_{fs}} = 1 447.6 \alpha_1(0) \alpha_2(0). \quad (11)$$

An expression for the potential $V_{AtD}(R, \epsilon)$ for the interaction [14,15] of an atom and a dielectric wall was presented in Eq. I-(15), where R is the atom-wall distance and ϵ is the dielectric constant of the wall. The expression is a double integral that can be evaluated with knowledge of the function $\alpha_1(i\omega)$. For small distances $V_{AtD}(R, \epsilon)$ has the limiting form

$$V_{AtD}(R, \epsilon) \sim -\frac{C_3}{R^3} \frac{\epsilon - 1}{\epsilon + 1}, \quad (12)$$

TABLE III. Comparison of static multipole polarizabilities $\alpha_1(0)$, $\alpha_2(0)$, and $\alpha_3(0)$ for He(2^3S).

Author (year)	Ref.	$\alpha_1(0)$	$\alpha_2(0)$	$\alpha_3(0)$
Crosby and Zorn (1977) Expt.	[20]	301(20)		
Ekstrom <i>et al.</i> (1995) Expt.	[18,19]	322(6.8)		
Bishop and Pipin (1993)	[32]	315.631	2 707.85	88 377.2
Rérat <i>et al.</i> (1993)	[30]	315.92	2 662.02	
Glover and Weinhold (1977)	[28]	316.24 \pm 0.78 ^a		
Drake (1972)	[26]	315.608		
Chung (1977)	[27]	315.63		
Chen and Chung (1996), B Spline	[23]	315.63	2 707.89	88 377.4
Chen and Chung (1996), Slater	[23]	315.611	2 707.81	88 356.2
Chung and Hurst (1966)	[25]	315.63		
Chen (1995)	[31]	315.633		
This work		315.631 468(12)	2 707.877 3(3)	88 377.325 3(7)

^aBounded theoretical value.

TABLE IV. Values of C_6 , C_8 , and C_{10} for the interaction of two He atoms.

System	C_6	C_8	C_{10}
2^1S-2^1S	11 241.052(5)	817 250.5(4)	108 167 630(54)
2^3S-2^3S	3 276.680 0(3)	210 566.55(6)	21 786 760(5)

where

$$C_3 = \frac{1}{4\pi} \int_0^\infty d\omega \alpha_1(i\omega). \quad (13)$$

As the separation increases retardation becomes important and the potential approaches its asymptotic form,

$$V_{\text{AID}}(R, \epsilon) \sim -\frac{K_4}{R^4} \frac{\epsilon - 1}{\epsilon + 1} \phi(\epsilon), \quad (14)$$

where $\phi(\epsilon)$ is given in Eq. I-(21) and

$$K_4 = 3\alpha_1(0)/(8\pi\alpha_{\text{fs}}) = 16.357\alpha_1(0). \quad (15)$$

For a perfectly conducting wall $V_{\text{AID}}(R, \epsilon)$ reduces to $V_{\text{AUM}}(R)$, where

$$V_{\text{AUM}}(R) \equiv V_{\text{AID}}(R, \infty) = -C_3 f_3(R) R^{-3} \quad (16)$$

and the retardation coefficient $f_3(R)$ is an integral involving $\alpha_1(i\omega)$ and is given in Eq. I-(26). For small distances $V_{\text{AUM}}(R) \sim -C_3/R^3$ and for asymptotically large distances $V_{\text{AUM}}(R) \sim -K_4/R^4$. Table II of Ref. [16] summarizes the various limits of $V_{\text{AID}}(R, \epsilon)$.

It has been shown that double basis sets work well for calculations involving S states of helium [3]. The basis set used here was constructed as in Ref. [5] with basis set functions expressed using Hylleraas coordinates

$$\{\chi_{ijk}(\gamma, \beta) = r_1^i r_2^j r_{12}^k e^{-\gamma r_1 - \beta r_2}\}. \quad (17)$$

The explicit form for the wave function is

$$\Psi(\mathbf{r}_1, \mathbf{r}_2) = \sum_{ijk} [a_{ijk}^{(1)} \chi_{ijk}(\gamma_1, \beta_1) + a_{ijk}^{(2)} \chi_{ijk}(\gamma_2, \beta_2)] \pm \text{exchange}, \quad (18)$$

and $i+j+k \leq \Omega$. The convergence of the eigenvalues is studied as Ω is progressively enlarged. Finally, a complete

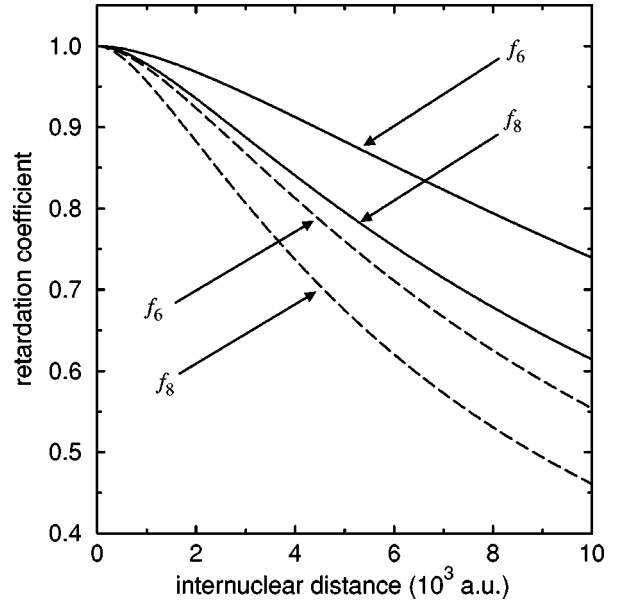


FIG. 2. Dimensionless retardation coefficients for He(2^1S) (solid line) and He(2^3S) (dashed line) as a function of the internuclear distance in atomic units.

optimization is performed with respect to the two sets of nonlinear parameters γ_1, β_1 , and γ_2, β_2 by first calculating the derivatives analytically in

$$\frac{\partial E}{\partial \gamma} = 2 \left\langle \Psi \left| H \left| \frac{\partial \Psi}{\partial \gamma} \right. \right. \right\rangle - 2E \left\langle \Psi \left| \frac{\partial \Psi}{\partial \gamma} \right. \right\rangle, \quad (19)$$

where γ represents any nonlinear parameter, E is the trial energy, H is the Hamiltonian, and $\langle \Psi | \Psi \rangle = 1$ is assumed, and then locating the zeros of the derivatives by Newton's method. These techniques yield much improved convergence relative to single basis set calculations. The method of the evaluation of the two-electron integrals in Hylleraas coordinates can be found in Ref. [17].

The expressions for the dynamic dipole polarizabilities, Eqs. (6)–(9) of Ref. [5], were evaluated using the wave functions determined by the variational method. Values of the static polarizabilities are given in Table I for He(2^1S) and He(2^3S). The polarizabilities given in Table I are extrapolated results, with the convergence studied as in Refs. [3] and [5], and the estimated extrapolation error in the last digit is given in parentheses with the listed values. The largest basis set sizes used consisted of 616 functions for the S states, 910 functions for the P states, 931 functions for the D states, and 1092 functions for the F states. The converged results are

TABLE V. Comparison of C_6 , C_8 , and C_{10} for the He(2^1S)-He(2^1S) system.

Author (year)	Ref.	C_6	C_8	C_{10}
Glover and Weinhold (1977)	[21]	11 330 ± 630 ^a		
Rérat <i>et al.</i> (1993)	[30]	11 360	812 500	
Victor <i>et al.</i> (1968)	[33]	11 300		
Lamm and Szabo (1980), ECA	[29]	10 980		
Chen (1995)	[34]	11 244	817 360	108 184 000
This work		11 241.052(5)	817 250.5(4)	108 167 630(54)

^aBounded theoretical value.

TABLE VI. Comparison of C_6 , C_8 , and C_{10} for the He(2^3S)-He(2^3S) system.

Author (year)	Ref.	C_6	C_8	C_{10}
Glover and Weinhold (1977)	[21]	3 289±90 ^a		
Victor <i>et al.</i> (1968)	[33]	3 290		
Lamm and Szabo (1980), ECA	[29]	3 300		
Rérat <i>et al.</i> (1993)	[30]	3 279	208 600	
Bishop and Pipin (1993)	[32]	3 276.677 0	210 563.99	21 786 484
Chen (1995)	[34]	3 276.1	210 520	21 783 800
Chen and Chung (1996), <i>B</i> spline	[23]	3 276.10	210 518	21 783 800
Chen and Chung (1996), Slater	[23]	3 275.90	210 507	21 780 200
This work		3 276.680 0(3)	210 566.55(6)	21 786 760(5)

^aBounded theoretical value.TABLE VII. The dimensionless retardation coefficients $f_6(R)$ and $f_8(R)$ for the atom-atom interaction. The dispersion coefficients C_6 and C_8 are also given. In the last line, labeled ‘‘Asymptotic’’ the values calculated using the asymptotic forms $f_6 \sim K_7/(RC_6)$ and $f_8 \sim K_9/(RC_8)$ are given in, respectively, columns 2,4 and columns 3,5, with K_7 and K_9 given in Eq. (11).

R	He(2^1S)-He(2^1S)		He(2^3S)-He(2^3S)	
	C_6	C_8	C_6	C_8
	112 41.052(5)	817 250.5(4)	3 276.680 0(3)	210 566.55(6)
	$f_6(R)$	$f_8(R)$	$f_6(R)$	$f_8(R)$
10	0.999998	0.999996	0.999995	0.999992
15	0.999996	0.999992	0.999988	0.999982
20	0.999993	0.999986	0.999980	0.999969
25	0.999989	0.999978	0.999968	0.999952
30	0.999984	0.999968	0.999955	0.999931
50	0.999958	0.999913	0.999879	0.999812
70	0.999919	0.999833	0.999770	0.999638
100	0.999840	0.999666	0.999548	0.999281
150	0.999655	0.999271	0.999034	0.998446
200	0.999408	0.998742	0.998358	0.997337
250	0.999106	0.998088	0.997533	0.995980
300	0.998750	0.997318	0.996573	0.994397
500	0.996857	0.993223	0.991568	0.986162
700	0.994325	0.987790	0.985052	0.975560
1000	0.989563	0.977772	0.973189	0.956657
1500	0.979675	0.957749	0.949690	0.920633
2000	0.968032	0.935332	0.923467	0.882350
2500	0.955170	0.911784	0.895919	0.843974
3000	0.941459	0.887865	0.867915	0.806627
5000	0.882570	0.795572	0.759993	0.675026
7000	0.822962	0.714364	0.666219	0.572701
10000	0.739435	0.614288	0.554000	0.460880
15000	0.622323	0.492124	0.424424	0.342480
20000	0.530963	0.407029	0.340027	0.270048
25000	0.459732	0.345261	0.282000	0.221929
30000	0.403520	0.298792	0.240136	0.187921
50000	0.266133	0.191769	0.149197	0.115667
70000	0.196376	0.140118	0.107696	0.083251
100000	0.140107	0.099381	0.075824	0.058519
	Asymptotic			
100000	0.142912	0.100738	0.076257	0.058760

compared with some previous calculations and experiments in Tables II and III. Ekstrom *et al.* [18] determined the He(2^3S) polarizability by combining their measured Na polarizability with the Molof *et al.* [19] measurement of the Na polarizability relative to the polarizability of He(2^3S). For the triplet state the experimental values of Ref. [20] and of Refs. [18,19] and the bounds of Glover and Weinhold [21] are compared with our calculated polarizability in Fig. 1.

The dynamic polarizability functions were constructed using the largest basis sets of each symmetry and used to evaluate the atom-atom dispersion constants and retardation coefficients. Our results for the dispersion constants are given in Table IV, with the estimated convergence errors given in parentheses, and the results are compared to other calculations in Tables V and VI. The retardation coefficients are given in Table VII and Fig. 2. The dispersion and retardation coefficients may be useful in studies of elastic and inelastic collisional processes of metastable helium atoms [6,7,22], which at low temperatures are sensitive to the interatomic potential energy [7].

Chen and Chung [23] calculated the coefficients W_4 and W_6 for He(2^1S) and their results are compared with ours in Table VIII; their published value of W_6 was multiplied by the factor $\frac{3}{2}$ to correspond to the theory of Ref. [11] and the agreement is very good.

For the atom-wall interactions the values of the coefficients C_3 can be obtained from the alternate expression

$$C_3 = \frac{1}{12} \left\langle 0 \left| \left(\sum_{i=1}^N \mathbf{r}_i \right)^2 \right| 0 \right\rangle, \quad (20)$$

which follows from integration of Eq. (13), where N is the number of electrons and $|0\rangle$ is accordingly the 2^1S or the 2^3S wave function. Since high-precision matrix elements

TABLE VIII. The coefficients W_4 and W_6 appearing in the expansion of the atom-atom interaction potential at small distances; see Eq. (7).

	Ref.	W_4	W_6
He(2^1S)-He(2^1S)	This work	3.912 7(5)	555.86(5)
He(2^3S)-He(2^3S)	Chen and Chung [23]	3.3006	314.18 ^a
	This work	3.305 2(5)	314.44(5)

^aMultiplied by the factor $\frac{3}{2}$ to correspond to the theory of Ref. [11].

TABLE IX. For $\text{He}(2^1S)$, values of $-R^3V_{\text{AtD}}(R, \epsilon)$, where $V_{\text{AtD}}(R, \epsilon)$ is the atom-wall potential, for values of ϵ corresponding to several types of dielectric, columns 2–4, and in column 5 values of $-R^3V_{\text{AtM}}(R)$ for a perfectly conducting wall. The coefficient $C_3(2^1S)$ is 2.67121.

R	$\epsilon=2.123$	$\epsilon=2.295$	$\epsilon=\infty$
10	0.95339	1.04221	2.65990
15	0.95029	1.03882	2.65455
20	0.94739	1.03564	2.64938
25	0.94463	1.03262	2.64439
30	0.94200	1.02975	2.63956
50	0.93244	1.01928	2.62159
70	0.92395	1.00999	2.60532
100	0.91253	0.99750	2.58320
150	0.89577	0.97916	2.55042
200	0.88087	0.96286	2.52098
250	0.86726	0.94797	2.49381
300	0.85463	0.93415	2.46833
500	0.81082	0.88624	2.37768
700	0.77415	0.84615	2.29898
1000	0.72765	0.79532	2.19547
1500	0.66478	0.72660	2.04896
2000	0.61386	0.67095	1.92470
2500	0.57109	0.62421	1.81635
3000	0.53433	0.58405	1.72023
5000	0.42573	0.46540	1.41942
7000	0.35344	0.38641	1.20473
10000	0.28060	0.30682	0.97660
15000	0.20730	0.22670	0.73525
20000	0.16342	0.17873	0.58540
25000	0.13443	0.14702	0.48434
30000	0.11395	0.12463	0.41205
50000	0.07032	0.07692	0.25591
70000	0.05067	0.05543	0.18478
100000	0.03565	0.03899	0.13013

TABLE X. For $\text{He}(2^3S)$, values of $-R^3V_{\text{AtD}}(R, \epsilon)$, where $V_{\text{AtD}}(R, \epsilon)$ is the atom-wall potential, for values of ϵ corresponding to several types of dielectric, columns 2–4, and in column 5 values of $-R^3V_{\text{AtM}}(R)$ for a perfectly conducting wall. The coefficient $C_3(2^3S)$ is 1.90092.

R	$\epsilon=2.123$	$\epsilon=2.295$	$\epsilon=\infty$
10	0.67644	0.73946	1.88963
15	0.67336	0.73608	1.88428
20	0.67047	0.73292	1.87912
25	0.66773	0.72992	1.87413
30	0.66512	0.72707	1.86931
50	0.65566	0.71671	1.85142
70	0.64728	0.70755	1.83525
100	0.63606	0.69527	1.81333
150	0.61966	0.67733	1.78095
200	0.60516	0.66146	1.75197
250	0.59196	0.64703	1.72529
300	0.57977	0.63370	1.70030
500	0.53788	0.58789	1.61162
700	0.50336	0.55016	1.53484
1000	0.46046	0.50328	1.43436
1500	0.40433	0.44195	1.29413
2000	0.36074	0.39433	1.17825
2500	0.32560	0.35593	1.08034
3000	0.29654	0.32419	0.99640
5000	0.21739	0.23769	0.75412
7000	0.17046	0.18640	0.60138
10000	0.12784	0.13981	0.45715
15000	0.08946	0.09785	0.32318
20000	0.06848	0.07490	0.24852
25000	0.05536	0.06055	0.20136
30000	0.04641	0.05076	0.16904
50000	0.02810	0.03074	0.10257
70000	0.02012	0.02201	0.07350
100000	0.01411	0.01543	0.05154

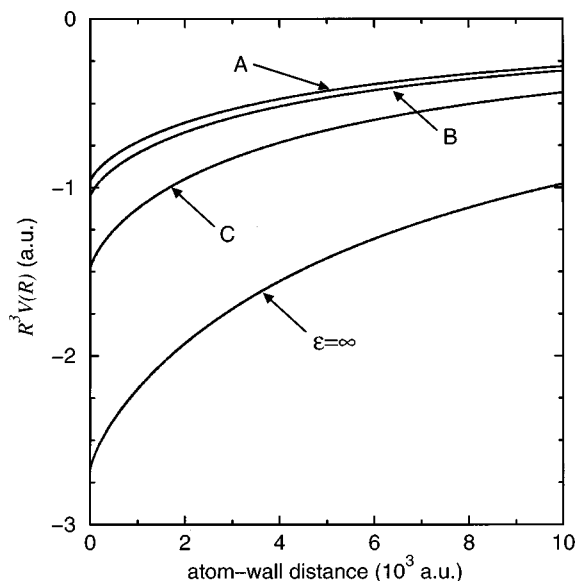


FIG. 3. In atomic units, potentials $R^3V(R)$ for $\text{He}(2^1S)$ atom-wall interactions. The labels A and B correspond, respectively, to dielectric constants ϵ of 2.123 and 2.295.

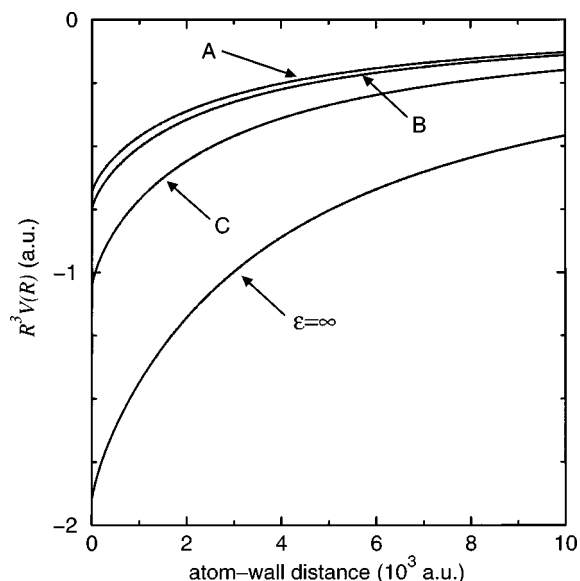


FIG. 4. In atomic units, potentials $R^3V(R)$ for $\text{He}(2^3S)$ atom-wall interactions. The labels A and B correspond respectively, to dielectric constants ϵ of 2.123 and 2.295.

are available [4,24] Eq. (20) was used to obtain the coefficients $C_3(2^1S) = 2.671\ 212\ 717\ 025$ and $C_3(2^3S) = 1.900\ 924\ 084\ 097$.

The dynamic dipole polarizabilities were used to evaluate the potentials for various dielectric walls. Results for He(2^1S) are given in Table IX and Fig. 3 and those for He(2^3S) are given in Table X and Fig. 4. The dielectric materials represented in the tables correspond to fused silica ($\epsilon = 2.123$) and BK-7 glass ($\epsilon = 2.295$). The tabulated potentials may be helpful in planning and analyzing experiments

with atom-evanescent wave mirrors; see, for example, Ref. [8].

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