

Dynamic polarizabilities and van der Waals coefficients of the 2^1S and 2^3S metastable states of helium

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Dynamic dipole and quadrupole polarizabilities of the 2^1S and 2^3S metastable states of He are calculated using our time-dependent gauge-invariant method and compared with previous theoretical results. Dispersion coefficients for the He(2^1S)-H₂ and He(2^3S)-H₂ systems, and their dependence on the intramolecular H—H distance are reported.

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I. INTRODUCTION

Collision processes involving excited atoms play an important role in many fields of physics including gas-laser physics, plasma physics, and upper-atmosphere physics [1]. Among these processes, an important one—which has been extensively studied—is the Penning-ionization process involving metastable helium. For the systems He(2^1S and 2^3S)-H₂, experimental data in the thermal [2–6] and superthermal [7] energy range have been reported. Qualitatively good agreement between experimental results and quantum-mechanical calculations based on an optical-potential model [8] and classical trajectory calculations [9,10] has shown that the dipole-dipole interaction is the major contribution to the autoionization process at large interatomic distances [11]. However, although reliable values for the dipole-dipole (C_6) dispersion coefficients are available for a large number of rare-gas diatomic systems in which atoms are in their ground state [12–15], very little is known for excited atoms. On the other hand, it is well known that dispersion coefficients may be obtained from the knowledge of frequency-dependent polarizabilities. Then, to be capable of calculating frequency-dependent polarizabilities for the low-lying excited states of rare gas is of importance.

The purpose of this paper is first to present accurate calculations of the dynamic (both real and imaginary frequencies) dipole and quadrupole polarizabilities for the two low-lying metastable states (2^1S and 2^3S) of He using our time-dependent gauge-invariant (TDGI) method [16–18]. When possible, our values are compared with the nearly exact results obtained by Glover and Weinhold [19,20] using explicitly correlated wave functions. Then, the frequency-dependent polarizabilities are used to compute the two-body dispersion coefficients corresponding to 1^1S , 2^1S , and 2^3S states, and by combining the present results with those obtained in a previous work [18], to ob-

tain the C_6 dispersion coefficients and their dependence on the intramolecular coordinate of H₂ for the He(2^1S and 2^3S)-H₂ systems.

Some methodological and computational details are given in Sec. II. Results are presented and discussed in Sec. III. Atomic units are used throughout the paper.

II. METHODOLOGICAL AND COMPUTATIONAL DETAILS

Our calculations are done by using a recently presented method based on a TDGI formalism for calculating static as well as dynamic linear and nonlinear polarizabilities [16,17]. This method, which has been first applied to systems in their ground state, is extended here to calculate polarizabilities of the 2^1S and 2^3S metastable excited states of He at frequencies below and above the first excitation threshold. Many of the theoretical details are similar to those described in Refs. [16,17] and [21], and therefore need not be repeated here. The fundamental ingredient to obtain accurate values of dynamic polarizabilities is to generate wave functions leading to accurate energies for the ground 1^1S and excited 2^1S , 2^3S , 2^1P , and 2^3P states, as well as accurate dipole-transition moments.

TABLE I. Comparison between calculated and experimental transition energies (ΔE), oscillator strengths (f_{ik}), and transition probability (A_{ki}) involving 2^1S , 2^1P , 2^3S , and 2^3P states of He. Experimental values (see Ref. [29]) are in parentheses.

Transition	ΔE (a.u.)	f_{ik}	A_{ki} (10^9 s ⁻¹)
$1^1S \rightarrow 2^1P$	0.778 879	0.2723	1.769
$1s^2 \ 1s2p$	(0.779 751)	(0.2762)	(1.799)
$2^1S \rightarrow 2^1P$	0.022 251	0.3801	0.002 015
$1s2s \ 1s2p$	(0.022 130)	(0.3764)	(0.001 976)
$2^3S \rightarrow 2^3P$	0.042 143	0.5408	0.010 28
$1s2s \ 1s2p$	(0.042 060)	(0.5391)	(0.010 22)

For He, the basis set used consists of $13s$, $7p$, and $6d$ primitive Gaussian orbitals based on the van Duijneveldt ($10s$) primitive set [22,23] augmented by an even-tempered ($7p,6d$) polarization set and $3s$ diffuse functions. Exponents of the $3s$ orbitals are 0.049 069, 0.022 304, and 0.010 138, respectively. Using this basis set, full configuration-interaction (CI) calculations were carried out to obtain the ground and the low-lying excited states by means of the multireference second-order many-body perturbation through the CIPSI algorithm [24,25]. A comparison of our energies with the “exact” nonrelativistic values [26–28] illustrates how good our wave functions are: 2^1S : $-2.145\,916$ (exact: $-2.145\,974$), 2^3S : $-2.175\,229$ (exact: $-2.175\,229$), 2^1P : $-2.123\,666$ (exact: $-2.123\,843$), and 2^3P : $-2.133\,086$ (exact: $-2.133\,163$). As an additional test of the accuracy and the completeness of our wave functions, we calculated the absorption oscillator strengths

TABLE II. Dynamic dipole polarizability $\alpha_1(\omega)$ of the 2^1S state of He. Comparison with the rigorous bounds results of Glover and Weinhold [19,20]. We think there must be a typing error on this value marked with an asterisk. All results are in a.u.

$\hbar\omega$	TDGI results	Rigorous bounds [19,20]
0	803.25	803.31±6.61
0.001	804.84	804.90±6.63
0.002	809.66	809.68±6.68
0.003	817.81	817.81±6.76
0.004	829.51	829.46±6.87
0.005	845.07	844.96±7.03
0.006	864.93	864.74±7.22
0.007	889.69	889.38±7.47
0.008	920.14	919.68±7.77
0.009	957.38	956.71±8.14
0.010	1002.85	1011.91*±8.60
0.011	1058.59	1057.28±9.15
0.012	1127.45	1125.63±9.84
0.013	1213.58	1211.02±10.71
0.014	1323.18	1319.56±11.81
0.015	1466.01	1460.78±13.26
0.016	1658.27	1650.45±15.22
0.017	1928.94	1916.66±17.99
0.018	2335.48	2314.75±22.18
0.019	3010.05	2970.69±29.21
0.020	4339.60	4246.89±43.32
0.021	8149.49	7785.88±85.30
0.025	-2700.55	-2707.52±93.61
0.030	-877.88	-881.51±22.42
0.035	-474.98	-477.48±13.19
0.040	-301.61	-303.61±10.09
0.045	-206.30	-208.08±8.77
0.050	-146.21	-147.95±8.30
0.055	-104.49	-106.35±8.39
0.060	-73.05	-72.23±9.04
0.065	-47.24	-50.04±10.41
0.070	-23.65	-27.60±12.97
0.075	1.46	-4.78±17.98
0.080	35.23	24.01±29.18
0.085	103.15	78.79±64.32
0.090	529.0	631.0±591.0

f_{ik} for the dipole transitions between lower (i) and upper (k) states as well as the Einstein spontaneous-transition probability A_{ki} .

As can be seen in Table I, there is an excellent agreement between our theoretical values for ΔE , f_{ik} , and A_{ki} for the $2^1S \rightarrow 2^1P$ and $2^3S \rightarrow 2^3P$ transitions and the experimental ones [29]. Such an agreement is important since the more these properties are accurately described the more the results on computed dynamic dipole polarizabilities are expected to be good. In order to obtain realistic potential surfaces for triatomic van der Waals systems consisting of a metastable atom and a stable diatom, the knowledge of the dependence of the atom-diatom dispersion coefficients on the diatomic internal distance is needed [18,30–33]. For the He(2^1S and 2^3S)-H₂ systems, calculations of the dispersion coefficients are based on the so-called Casimir-Polder [34] formula which expresses dispersion coefficients in terms of imaginary-frequency integrals of the dynamic polarizabilities for the noninteracting systems. Note that by us-

TABLE III. Dynamic dipole polarizability $\alpha_1(\omega)$ of the 2^3S state of He. Comparison with the rigorous bounds results of Glover and Weinhold [19,20]. All results are in a.u.

$\hbar\omega$	TDGI results	Rigorous bounds [19,20]
0	315.92	316.24±0.78
0.0025	317.00	317.33±0.78
0.0050	320.31	320.63±0.79
0.0075	325.97	326.30±0.80
0.0100	334.25	334.59±0.83
0.0125	345.56	345.90±0.86
0.0150	360.50	360.75±0.90
0.0175	379.96	380.32±0.96
0.0200	405.28	405.65±1.03
0.0225	438.51	438.89±1.13
0.0250	482.95	483.34±1.25
0.0275	544.17	544.57±1.43
0.0300	632.41	632.82±1.70
0.0325	768.67	769.05±2.10
0.0350	1003.68	1003.93±2.82
0.0375	1499.53	1499.16±4.39
0.0400	3205.28	3199.13+10.27
0.0450	-2097.0	-8388.0±6339.0
0.0500	-726.27	-724.20±5.89
0.0550	-417.20	-416.44±3.05
0.0600	-281.78	-281.32±2.14
0.0650	-206.21	-205.84±1.74
0.0700	-158.17	-157.82±1.56
0.0750	-124.94	-124.58±1.51
0.0800	-100.49	-100.11±1.55
0.0850	-81.55	-81.15±1.71
0.0900	-66.11	-65.71±2.03
0.0950	-52.74	-52.38±2.63
0.1000	-40.07	-39.91±3.79
0.1050	-26.03	-26.50±6.40
0.1090	-10.17	-11.93±11.88
0.1095	-7.49	-9.82±12.77
0.1125	16.98	10.98±25.63
0.1150	74.32	61.24±67.87
0.1170	450.0	1193.0±1192.0

TABLE IV. Dynamic quadrupole polarizability of the 2^1S and 2^3S states of He. All results are in a.u. $C_{zz,zz}$ is defined [49] as $C_{zz,zz} = 2/3\hbar \sum_{m \neq g} |\langle g | \hat{\theta}_{zz} | m \rangle|^2 / \omega_{mg} = 1/3\alpha_2$.

$\hbar\omega$	$C_{zz,zz}$ (2^1S)	$C_{zz,zz}$ (2^3S)
0	2290.3	887.34
0.005	2296.9	888.97
0.010	2317.0	893.07
0.015	2351.1	900.14
0.020	2400.9	910.51
0.025	2468.1	923.80
0.030	2555.6	940.91
0.035	2667.7	961.92
0.040	2810.0	987.40
0.045	2991.6	1018.1
0.050	3225.6	1054.8
0.055	3531.8	1098.5
0.060	3944.5	1151.2
0.065	4522.8	1214.4
0.070	5383.5	1292.1
0.075	6785.9	1387.5
0.080	9460.5	1507.5
0.085	16 548.0	1662.6
0.090	90 882.0	1868.1
0.095	-22 973.0	2153.9
0.100	-9525.4	2577.6
0.105	-5639.2	3267.9

ing our TDGI method the dispersion coefficients for seven specific H—H bond distances varying from 1.4 to 6.0 a.u. have already been calculated in a previous work [18].

III. RESULTS AND DISCUSSION

There is a vast amount of theoretical [19,20,35–45] and experimental data [46] for the static dipole polarizabilities α_1 of the 2^1S and 2^3S metastable states of helium. To our knowledge, none are so accurate and so reliable as the rigorous upper- and lower-bound results obtained by Glover and Weinhold [19,20]. They also give the rigorous bounds for dynamic dipole polarizability at real $[\alpha_1(\omega)]$ and at imaginary frequencies $[\alpha_1(i\omega)]$ for a wide

range of energies. We shall use these rigorous bounds as a criterion to estimate the quality of our TDGI results (Tables II and III). A first point to emphasize is that our results for the static polarizabilities (803.25 and 315.92 a.u. for 2^1S and 2^3S , respectively) are in excellent agreement with the rigorous bounds of Glover and Weinhold [19,20] (803.31 ± 6.61 and 316.24 ± 0.78 a.u.), and also agree with the experimental values (728.8 ± 87.7 and 301 ± 20 a.u.) determined by the electric-deflection time-of-flight method [26]. For the dynamic polarizabilities at frequencies up to the second excitation threshold, TDGI results are always compatible with the rigorous bounds, except for the 2^1S metastable state near the resonance within the range 0.019–0.021 a.u.

In contrast to the dipole polarizabilities very little is known about quadrupole polarizabilities ($\alpha_2 = 3C_{zz,zz}$) of excited states of He. For the 2^3S state, earlier theoretical values for the static quadrupole polarizability component $C_{zz,zz}$ range from 887.7 to 947.6 a.u. [47] in the extended Coulomb approximation (ECA) and quantum-defect orbital (QDO) methods, while multiconfiguration self-consistent-field (MCSCF) calculations done by Konowalow and Lengsfeld [48] give a value of 914.4 a.u. (note that we have divided their results by a factor 3 to adopt Bishop's convention [49]).

For the 2^1S state, the values obtained by Lamm and Szabo [47] are 2346 and 2425 a.u. depending on the approximation used. Calculated dynamic quadrupole polarizabilities are given in Table IV. Our static $C_{zz,zz}$ values (2290.3 a.u. for 2^1S and 887.34 a.u. for 2^3S) agree within 2% with the ECA value of Lamm and Szabo [47]. For the dynamic values we found no published data to compare with [50].

The multipole expansion of the second-order interaction energy between a pair of neutral S -state atoms is given by

$$\Delta E = -C_6 R^{-6} - C_8 R^{-8} - C_{10} R^{-10} \dots,$$

where the C_n 's are the van der Waals or dispersion coefficients. The C_6 and C_8 dispersion coefficients for the interaction between the ground (1^1S) and excited states

TABLE V. van der Waals C_6 and C_8 coefficients (in a.u.) for He in the 1^1S , 2^1S , and 2^3S states. The rigorous bounds results are given in parentheses for C_6 .

C_6	1^1S	2^1S	2^3S
1^1S	1.4593 (1.4597 \pm 0.0055)	42.12 (41.47 \pm 1.70)	29.19 (29.00 \pm 0.51)
2^1S		1.136×10^4 ($[1.133 \pm 0.063] \times 10^4$)	5.866×10^3 ($[5.767 \pm 0.339] \times 10^3$)
2^3S			3.279×10^3 ($[3.289 \pm 0.090] \times 10^3$)
C_8	1^1S	2^1S	2^3S
1^1S	13.883	3263	1689
2^1S		8.125×10^5	4.068×10^5
2^3S			2.086×10^5

TABLE VI. Calculated C_6 dispersion coefficients for $\text{He}(2^1S)\text{-H}_2$ and $\text{He}(2^3S)\text{-H}_2$ as a function of the interhydrogen distance R .

R	1.4	2.4	3.0	3.4	3.8	4.4	6.0
$\text{He}(2^1S)\text{-H}_2$							
C_6^{\parallel}	188.7	406.7	501.5	513.4	484.4	412.9	292.1
C_6^{\perp}	135.8	222.8	255.7	266.0	268.2	265.7	258.0
C_6	153.4	284.1	337.6	348.5	340.3	314.8	269.4
$\text{He}(2^3S)\text{-H}_2$							
C_6^{\parallel}	129.1	274.4	336.6	343.8	324.4	277.1	197.5
C_6^{\perp}	93.0	151.6	173.8	180.7	182.0	180.5	175.3
C_6	105.0	192.5	228.1	235.1	229.5	212.7	182.7

(2^1S and 2^3S) of He were calculated by combining our $\alpha_1(i\omega)$ and $\alpha_2(i\omega)$ TDGI values through a simple numerical integration using (see [51] and Table IV),

$$C_6 = \frac{3\hbar}{\pi} \int_0^{\infty} \alpha_1^A(i\omega) \alpha_1^B(i\omega) d\omega,$$

$$C_8 = \frac{15\hbar}{\pi} \int_0^{\infty} [\alpha_2^A(i\omega) \alpha_1^B(i\omega) + \alpha_1^A(i\omega) \alpha_2^B(i\omega)] d\omega.$$

For the ground and low-lying S states of He, C_6 and C_8 values are given in Table V. For C_6 our values are compatible with the very tight error bounds obtained by Glover and Weinhold [20]. Our best estimate of C_6 for (1^1S-1^1S), (2^1S-2^1S), and (2^3S-2^3S) are different from rigorous calculations by 0.03%, 0.26%, and 0.3%, while our (2^1S-2^3S) result is too high by 1.7% with respect to the rigorous bounds mean value of [20]. For C_8 the largest difference between the values in Table V and the results given by Proctor and Stwalley [52] is 2.7%. As expected, in all cases the results obtained for systems consisting of excited atoms are fairly large with respect to those obtained with atoms in their ground state since excited rare-gas atoms lose their “closed-shell” character.

Table VI presents TDGI calculations of the C_6 dispersion coefficients (both parallel and perpendicular) for the

systems $\text{He}(2^1S)\text{-H}_2$ and $\text{He}(2^3S)\text{-H}_2$ as a function of the interhydrogen $R_{\text{H-H}}$ distance. This is a calculation from first principles of this quantity involving low-lying metastable states of He. C_6 dispersion coefficients for both systems are obtained using accurate dipole polarizabilities at seven H—H distances ranging from 1.4 to 6.0 a.u. calculated in [18]. A maximum for the C_6 coefficients occurs at values of R similar to those at which the polarizabilities of H_2 reach their maximum value. It should be noted that for all distances the C_6 [$\text{He}(2^1S)\text{-H}_2$] coefficient is roughly 50% higher than the C_6 [$\text{He}(2^3S)\text{-H}_2$] value. These R -dependent coefficients may be used for representing model potential functions.

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