Long-range interactions between excited helium and alkali-metal atoms

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The dispersion coefficients for the long-range interaction of the first four excited states of He, i.e., He($2\,^{1,3}S$) and He($2\,^{1,3}P$), with the low-lying states of the alkali-metal atoms Li, Na, K, and Rb are calculated by summing over the reduced matrix elements of the multipole transition operators. For the interaction between He and Li the uncertainty of the calculations is 0.1–0.5%. For interactions with other alkali-metal atoms the uncertainty is 1–3% in the coefficient C_5, 1–5% in the coefficient C_6, and 1–10% in the coefficients C_8 and C_{10}. The dispersion coefficients C_n for the interaction of He($2\,^{1,3}S$) and He($2\,^{1,3}P$) with the ground-state alkali-metal atoms and for the interaction of He($2\,^{1,3}S$) with the alkali-metal atoms in their first ^2P states are presented in this Brief Report. The coefficients for other pairs of atomic states are listed in the Supplemental Material.

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Cold and ultracold mixtures of multispecies atoms are an active topic of research in recent years. Ultracold mixtures of two atomic species are used to study the elastic and inelastic collisions between different species. Such mixtures also make it possible to sympathetically cool one species through collisional energy exchange with the other species [1–3]. In addition, weakly bound heteronuclear molecules can be created [4–6]. Heteronuclear polar molecules have attracted particular attention because their permanent dipole moments can be controlled with external fields. They are potential candidates for quantum simulation, quantum computing, and quantum metrology [7,8]. Most dual-species experiments involve combinations of alkali-metal atoms, such as Na-{K, Rb, Cs} [9], K-{Rb, Cs} [2,6,9], and Rb-Cs [9,10]. Recently, a few experiments have employed ultracold mixtures of alkali-metal and metastable noble gas atoms, such as ^{40}Ar*-^{87}Rb [11,12] and He*-^{87}Rb [12–14]. The latter mixture is promising for creating a dual-species Bose-Einstein condensate by significantly suppressing the Penning ionization [13–15], which causes the trap losses. The loss rate is determined primarily by the long-range interaction between the He* and ^{87}Rb atoms.

There are very few calculations of dispersion coefficients reported in the literature for the long-range interaction between low-lying excited states of He and alkali-metal atoms. The coefficients C_6 for the van der Waals interaction between the He($2\,^{1,3}S$) metastable states and the ground states of Li, Na, K, Rb, and Cs have been computed by Bell *et al.* [16] by means of the Casimir-Polder formula [17]. Dalgarno and Victor [18] have improved the calculation by more accurate representations of the dynamic dipole polarizabilities $\alpha_1(\omega)$ of the metastable states. For the alkali-metal atoms, these authors employ semiempirical representations of $\alpha_1(\omega)$ [19]. Using the configuration-interaction plus core-polarization (CICP) method [20], Spelsberg and Meyer [21] have calculated the coefficients C_6, C_8, and C_{10} for the interaction between He($2\,^{1,3}S$) and the ground states of Li, Na, and K.

In the present work, we compute the dispersion coefficients for the long-range interaction of the first four excited states of He, i.e., He($2\,^{1,3}S$) and He($2\,^{1,3}P$), with the low-lying states of the alkali-metal atoms Li, Na, K, and Rb by summing over the reduced matrix elements of the multipole transition operators [22–24]. For the symmetries of diatomic molecules we adopt the notations of Ref. [24]. Atomic units are used throughout the following sections.

A systematic formalism has been presented for the calculations of the long-range interaction between two heteronuclear atoms in arbitrary atomic states in Ref. [24], treating the interaction as a perturbation to the isolated atoms. In general, the long-range interaction between two heteronuclear atoms can be written in the form

$$V(R) = -\sum_{s=1}^{\infty} \frac{C_{2s+4}}{R^{2s+4}} - \sum_{s=1}^{\ell_a+\ell_b-1} \frac{C_{2s+3}}{R^{2s+3}} - \cdots , \qquad (1)$$

where ℓ_a and ℓ_b represent the quantum numbers of orbital angular momenta of atoms A and B, respectively; R is the distance between the two atoms; and C_n are the dispersion coefficients. The first term arises from the second order correction to the energy and is always present. The second term (the first-order correction) occurs only if each atom is in a state with nonzero angular momentum.

The dispersion coefficients are evaluated for diatomic molecular states according to the nondegenerate and degenerate perturbation theories. For a simple nondegenerate system where both atoms A and B are spherically symmetric, for example, C_6 can be represented in the form

$$C_6 = \sum_{n_a n_b} \frac{2|\langle \psi_{0_a} \| \sum_i r_i \mathbf{C}^1(\hat{\mathbf{r}}_i) \| \psi_{n_a}\rangle|^2}{3} \times \frac{|\langle \psi_{0_b} \| \sum_j r_j \mathbf{C}^1(\hat{\mathbf{r}}_j) \| \psi_{n_b}\rangle|^2}{E_{n_a} + E_{n_b} - E_{0_a} - E_{0_b}} \qquad (2)$$

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where $\mathbf{C}^1(\hat{\mathbf{r}})$ is the spherical tensor of rank 1; ψ_{0_a} and ψ_{n_a} are the wave function of the initial state and the n_ath intermediate eigenfunction for the atom A, respectively; E_{0_a} and E_{n_a} are their corresponding eigenenergies; the sum i runs over all the electrons in the atom A; the definitions of symbols ψ_{0_b}, ψ_{n_b}, E_{0_b}, E_{n_b} and j for the atom B are similar to those of their counterpart symbols for the atom A. In the degenerate case, the zeroth-order wave functions are determined by diagonalizing the leading term of the first-order correction in the degenerate space. They are then used to calculate the dispersion coefficients by summing over intermediate states represented by atomic physical states and pseudostates.

For atomic He and Li the energy spectra and reduced matrix elements of the multipole transition operators are the same as those used for calculating the dispersion coefficients for the low-lying states of He and Li [25–31]. They were calculated using Hylleraas basis functions. For a two-electron system, the basis functions have the form

$$r_1^i\, r_2^j\, r_{12}^k\, e^{-\alpha r_1-\beta r_2}\mathcal{Y}_{\ell_1\ell_2}^{LM}(\hat{\mathbf{r}}_1,\hat{\mathbf{r}}_2), \qquad (3)$$

TABLE I. Dispersion coefficients for the interaction of He($2\,^{1,3}S$) with the $n\ ^2S$ ground states of the alkali-metal atoms. Numbers in square brackets in the second, third, and fourth columns denote powers of 10.

System	C_6 (a.u.)	C_8 (a.u.)	C_{10} (a.u.)
LiHe($2\,^1S$)$\,^2\Sigma$	3.504[3]	2.633[5]	3.018[7]
Hylleraas	3.502[3]	2.632[5]	3.016[7]
Ref. [18]	3.500[3]		
Ref. [21]	3.495[3]	2.627[5]	2.990[7]
No core effect	3.499[3]	2.628[5]	3.013[7]
NaHe($2\,^1S$)$\,^2\Sigma$	3.598[3]	3.019[5]	3.606[7]
Ref. [18]	3.660[3]		
Ref. [21]	3.574[3]	2.997[5]	3.558[7]
No core effect	3.567[3]	2.994[5]	3.579[7]
KHe($2\,^1S$)$\,^2\Sigma$	5.984[3]	5.805[5]	7.588[7]
Ref. [18]	5.940[3]		
Ref. [21]	5.845[3]	5.690[5]	7.600[7]
No core effect	5.816[3]	5.668[5]	7.439[7]
RbHe($2\,^1S$)$\,^2\Sigma$	6.509[3]	6.846[5]	9.207[7]
Ref. [18]	6.440[3]		
No core effect	6.235[3]	6.614[5]	8.954[7]
LiHe($2\,^3S$)$\,^{2,4}\Sigma$	2.090[3]	1.326[5]	1.280[7]
Hylleraas	2.089[3]	1.325[5]	1.279[7]
Ref. [18]	2.090[3]		
Ref. [21]	2.083[3]	1.321[5]	1.272[7]
No core effect	2.086[3]	1.323[5]	1.278[7]
NaHe($2\,^3S$)$\,^{2,4}\Sigma$	2.178[3]	1.553[5]	1.570[7]
Ref. [18]	2.220[3]		
Ref. [21]	2.159[3]	1.540[5]	1.553[7]
No core effect	2.157[3]	1.540[5]	1.560[7]
KHe($2\,^3S$)$\,^{2,4}\Sigma$	3.523[3]	3.055[5]	3.485[7]
Ref. [18]	3.480[3]		
Ref. [21]	3.419[3]	2.993[5]	3.534[7]
No core effect	3.406[3]	2.983[5]	3.426[7]
RbHe($2\,^3S$)$\,^{2,4}\Sigma$	3.832[3]	3.634[5]	4.294[7]
Ref. [18]	3.760[3]		
No core effect	3.641[3]	3.512[5]	4.191[7]

TABLE II. Dispersion coefficients for the interaction of He($2\,^{1,3}S$) with the first ^2P states of the alkali-metal atoms. Numbers in square brackets denote powers of 10.

System	C_6 (a.u.)	C_8 (a.u.)	C_{10} (a.u.)
LiHe($2\,^1S$)$\,^2\Sigma$	2.048[3]	2.483[6]	2.600[8]
Hylleraas	2.050[3]	2.481[6]	
LiHe($2\,^1S$)$\,^2\Pi$	2.663[3]	5.222[5]	3.298[7]
Hylleraas	2.663[3]	5.219[5]	
NaHe($2\,^1S$)$\,^2\Sigma$	8.845[3]	5.392[6]	6.368[8]
NaHe($2\,^1S$)$\,^2\Pi$	6.364[3]	9.699[5]	5.672[7]
KHe($2\,^1S$)$\,^2\Sigma$	9.756[3]	7.974[6]	1.219[9]
KHe($2\,^1S$)$\,^2\Pi$	9.242[3]	9.847[5]	7.951[7]
RbHe($2\,^1S$)$\,^2\Sigma$	1.298[4]	1.012[7]	1.652[9]
RbHe($2\,^1S$)$\,^2\Pi$	1.174[4]	1.078[6]	9.601[7]
LiHe($2\,^3S$)$\,^{2,4}\Sigma$	−2.190[3]	1.063[6]	1.230[8]
Hylleraas	−2.187[3]	1.062[6]	
LiHe($2\,^3S$)$\,^{2,4}\Pi$	7.605[2]	1.571[5]	1.034[7]
Hylleraas	7.611[2]	1.570[5]	
NaHe($2\,^3S$)$\,^{2,4}\Sigma$	1.969[3]	2.252[6]	3.142[8]
NaHe($2\,^3S$)$\,^{2,4}\Pi$	2.919[3]	2.535[5]	1.804[7]
KHe($2\,^3S$)$\,^{2,4}\Sigma$	−3.189[3]	3.776[6]	6.189[8]
KHe($2\,^3S$)$\,^{2,4}\Pi$	2.995[3]	3.147[5]	2.696[7]
RbHe($2\,^3S$)$\,^{2,4}\Sigma$	−3.007[3]	4.801[6]	8.430[8]
RbHe($2\,^3S$)$\,^{2,4}\Pi$	3.851[3]	3.491[5]	3.297[7]

where $\mathcal{Y}_{\ell_1\ell_2}^{LM}(\hat{\mathbf{r}}_1,\hat{\mathbf{r}}_2)$ are the coupled spherical harmonics. Except for some truncations made to avoid near linear dependence, all terms with $i+j+k \leqslant \Omega$ are included, where Ω is an integer. For the Li atom the basis functions have a similar form:

$$r_1^i\, r_2^j\, r_3^k\, r_{12}^m\, r_{13}^n\, r_{23}^p\, e^{-\alpha r_1-\beta r_2-\gamma r_3}\mathcal{Y}_{(\ell_1\ell_2)\ell_{12},\ell_3}^{LM}(\hat{\mathbf{r}}_1,\hat{\mathbf{r}}_2,\hat{\mathbf{r}}_3). \qquad (4)$$

The nonlinear parameters α, β, and γ are variationally optimized by the power method.

The reduced matrix elements of the transition operators and energy spectra for the alkali-metal atoms have been used for calculating the dispersion coefficients for the interaction with ground-state H and He [24]. The spectra of the valence electrons have been generated by the CICP method with large mixed Laguerre-type and Slater-type orbital basis sets. Moreover, the core effect on the valence electrons has been represented by a semiempirical polarization potential, and the contribution of the core excitations to the dispersion coefficients has been estimated by approximate oscillator strength distributions. For details on the calculations for alkali-metal atoms the reader is referred to Refs. [24,32–37].

Table I presents the dispersion coefficients for the interaction of He($2\,^{1,3}S$) with the ground-state alkali-metal atoms. Table II addresses the interaction between He($2\,^{1,3}S$) and the first ^2P excited states of the alkali-metal atoms, i.e., Li($2\,^2P$), Na($3\,^2P$), K($4\,^2P$), and Rb($5\,^2P$) whereas Table III refers to the interaction between He($2\,^{1,3}P$) and the ground-state alkali-metal atoms. Dispersion coefficients C_n for the interaction between He($2\,^{1,3}S$) and the first ^2S and ^2D excited states of the alkali-metal atoms and those for the interaction between He($2\,^{1,3}P$) and the first ^2P excited states of the alkali-metal atoms are listed in the Supplemental Material [38].

For the Li atom we calculate the dispersion coefficients using both the CICP transition data and the transition data

TABLE III. Dispersion coefficients for the interaction of He($2\,^{1,3}P$) with the ground states of the alkali-metal atoms. Numbers in square brackets denote powers of 10.

System	C_6 (a.u.)	C_8 (a.u.)	C_{10} (a.u.)
LiHe($2\,^1P$)$^2\Sigma$	6.123[3]	7.851[5]	1.019[8]
Hylleraas	6.120[3]	7.847[5]	1.018[8]
LiHe($2\,^1P$)$^2\Pi$	2.605[3]	8.148[4]	5.676[6]
Hylleraas	2.604[3]	8.143[4]	5.672[6]
NaHe($2\,^1P$)$\,^2\Sigma$	5.981[3]	8.510[5]	1.179[8]
NaHe($2\,^1P$)$\,^2\Pi$	2.633[3]	1.013[5]	7.649[6]
KHe($2\,^1P$)$\,^2\Sigma$	1.114[4]	1.543[6]	2.330[8]
KHe($2\,^1P$)$\,^2\Pi$	4.575[3]	2.518[5]	2.200[7]
RbHe($2\,^1P$)$\,^2\Sigma$	1.223[4]	1.768[6]	2.772[8]
RbHe($2\,^1P$)$\,^2\Pi$	5.008[3]	3.175[5]	2.887[7]
LiHe($2\,^3P$)$\,^{2,4}\Sigma$	7.153[3]	5.617[5]	6.620[7]
Hylleraas	7.148[3]	5.614[5]	6.616[7]
LiHe($2\,^3P$)$^{2,4}\Pi$	2.609[3]	6.790[4]	4.464[6]
Hylleraas	2.607[3]	6.785[4]	4.461[6]
NaHe($2\,^3P$)$\,^{2,4}\Sigma$	6.318[3]	6.167[5]	7.754[7]
NaHe($2\,^3P$)$\,^{2,4}\Pi$	2.452[3]	8.484[4]	6.053[6]
KHe($2\,^3P$)$\,^{2,4}\Sigma$	1.534[4]	1.166[6]	1.571[8]
KHe($2\,^3P$)$\,^{2,4}\Pi$	5.198[3]	2.193[5]	1.779[7]
RbHe($2\,^3P$)$\,^{2,4}\Sigma$	1.738[4]	1.371[6]	1.887[8]
RbHe($2\,^3P$)$\,^{2,4}\Pi$	5.833[3]	2.832[5]	2.349[7]

computed by the Hylleraas-type basis functions. For the Li($2\,^2S$) and Li($3\,^2S$) states the coefficients C_6, C_8, and C_{10} are computed using the correlated basis sets. For the Li($2\,^2P$) and Li($3\,^2D$) states the coefficients up to C_8 are computed using the Hylleraas-type basis sets, because only the intermediate states related to the corresponding dipole and quadrupole transitions have been generated with the Hylleraas-type basis sets. The results of Dalgarno and Victor [18] and Spelsberg and Meyer [21] are also included in Table I for comparison. These four calculations agree within 0.3% for the Li($2\,^2S$) ground state. For the He($2\,^{1,3}S$)-Na($3\,^2S$) system the C_6 coefficient calculated by Dalgarno and Victor [18] is 2% larger than our result. Moreover, their C_6 values are 1% smaller than our values for the systems He($2\,^{1,3}S$)-K($4\,^2S$) and He($2\,^1S$)-Rb($5\,^2S$), and 2% smaller for the He($2\,^3S$)-Rb($5\,^2S$) system.

The effects of core excitations on the dispersion coefficients are ignored in the calculations of Spelsberg and Meyer [21]. To enable a comparison, we also list in Table I our C_n values which exclude contributions of the core excitations. The discrepancy between the two calculations without core effects is less than 0.5% except for the C_{10} values for the He($2\,^{1,3}S$)-K($4\,^3S$) system. The C_{10} values of Spelsberg and Meyer are 2–3% larger than our results for the He($2\,^{1,3}S$)-K($4\,^3S$) system. This means that the octupole contributions are overestimated in their calculations. The overestimation is confirmed by comparing their values for the static multipole polarizabilities with other accurate calculations [32,33,39–43] (see Table I in the Supplemental Material [38]). For the static dipole polarizability of the ground-state K the spread of all four calculations is within 2.5%, and the four quadrupole polarizabilities are almost the same. However, the static octupole polarizability of Spelsberg and Meyer (1.914×10^5 a.u.) is 7.7% larger than the value obtained by the other three calculations (the CICP data set, the one-electron model potential approach by Marinescu *et al.* [40], and the relativistic many-body perturbation theory by Porsev and Derevianko [43]). It is also demonstrated that the core effect becomes more important for heavier atoms.

The uncertainty of the present calculations can be estimated from the uncertainties of the dispersion coefficients for interaction of the alkali-metal atoms with the ground states of H and He [24]. For the systems He($2\,^{1,3}S$)-Li($2\,^2S$), He($2\,^{1,3}S$)-Li($2\,^2P$), and He($2\,^3S$)-Li($3\,^2D$) [38] the accuracy of the calculations is 0.1%. For the system He($2\,^1S$)-Li($3\,^2D$) [38], the C_6 values of the Hylleraas and CICP methods deviate by less than 0.1% and the discrepancy of the C_8 values is 0.3%. For the $^2\Sigma$ state of the system He($2\,^1P$)-Li($3\,^2S$) [38] the discrepancy between the two methods is 0.7% for C_8. For both the $^2\Sigma$ and $^2\Pi$ states of the system He($2\,^3P$)-Li($3\,^2S$) [38] the discrepancy of the C_8 and C_{10} values between the two methods is 2%. For the system He($2\,^1P$)-Li($2\,^2P$) [38] the two methods agree within 0.5%. The C_6 coefficient deviates between the two methods by 90% for the second $^{2,4}\Sigma^+$ state of the He($2\,^3P$)-Li($2\,P$) system [38]. The reason for this large difference is that the negative contribution of -1.530×10^4 a.u. of the dipole transition pairs He($2\,^3P$)-He($2\,^3S$) and Li($2\,^2P$)-Li($2\,^2S$) as well as He($2\,^3P$)-He($3\,^3S$) and Li($2\,^2P$)-Li($2\,^2S$) almost cancels the positive contribution of 1.535×10^4 a.u. of all other dipole transition pairs. No such strong cancellation occurs for other dispersion coefficients. Another unexpected feature is that the C_6 coefficient is about 19 times smaller than the corresponding C_5 coefficient. In summary, the results from the Hylleraas-type calculations are recommended rather than those from the CICP calculations.

For the long-range interaction between He and other alkali-metal atoms, the uncertainties are 1–3% for C_5, 1–5% for C_6, and 1–10% for C_8 and C_{10}. Generally speaking, the uncertainties of the dispersion coefficients for the ground states of the alkali-metal atoms are smaller than those for the excited states, especially in the cases where the He atom is in one of the He($2\,^{1,3}P$) states.

In conclusion, dispersion coefficients have been calculated for the long-range interaction of the first four excited states of He, i.e., He($2\,^{1,3}S$) and He($2\,^{1,3}P$), with the low-lying states of the alkali-metal atoms Li, Na, K, and Rb by summing over the reduced matrix elements of the multipole transition operators [22–24]. For He and Li atoms the reduced matrix elements have been previously generated with Hylleraas-type basis functions [25–31]. For the alkali-metal atoms the transition arrays of the valence electrons have been previously computed by the CICP method, where the effect of core excitations has been taken into account by approximately constructing the oscillator strength distributions of the atomic cores [24,32–37]. For systems involving a He metastable state He($2\,^{1,3}S$) and the ground state of an alkali-metal atom, our results are more accurate than previously published values [18,21]. Dispersion coefficients for systems involving a He($2\,^{1,3}P$) state or excited state of an alkali-metal atom have been computed. These coefficients enable the construction of accurate long-range potentials for the corresponding atom-atom collisions.

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