Long-range interaction of two metastable rare-gas atoms

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We present semiempirical calculations of long-range van der Waals interactions for two interacting metastable rare-gas atoms Ne through Xe. Dispersion coefficients $C_6$ are obtained for homonuclear molecular potentials asymptotically connecting to the $ns(3/2)_2 + ns(3/2)_2$ atomic states. The estimated uncertainty of the calculated $C_6$ dispersion coefficients is 4%.

Motivated by cold-collision studies of metastable rare-gas atoms [1–4] and prospects of achieving Bose-Einstein Condensation in these systems [5], we present calculations of long-range dispersion (van der Waals) coefficients for two atoms interacting in the $ns(3/2)_2$ atomic states ($n = 3$ for Ne, $n = 4$ for Ar, $n = 5$ for Kr, and $n = 6$ for Xe). The metastable states have long lifetimes, 43 sec for Xe [6], decaying to the ground $^1S_0$ state by a weak magnetic-quadrupole transition. With such a long lifetime the metastable atom behaves as an effective ground state in experiments. Compared to alkali-metal systems, an attractive feature of the noble gas atoms is the availability of isotopes with zero nuclear spin. The lack of hyperfine structure leads to a substantial simplification of molecular potentials, though some complexity arises due to the nonvanishing total electron angular momentum ($J_0$) of the metastable state. The anisotropy leads to fifteen distinct long-range molecular states connecting to the $ns(3/2)_2 + ns(3/2)_2$ asymptotic configuration.

Our theoretical treatment of long-range interactions is similar to recent high-precision calculations of van der Waals coefficients for alkali-metal atoms [7]. By using many-body methods and accurate experimental matrix elements for the principal transitions, leading dispersion coefficients $C_6$ were determined to an accuracy better than 1% for Na, K, and Rb, and of 1% for Cs and 1.5% for Fr. The semiempirical values of $C_6$ coefficients for metastable noble-gas atoms obtained here have an estimated uncertainty of 4%. The approach relies on the determination of dynamic polarizability functions. To construct the polarizabilities we combine experimental lifetime [8–15] and energy data of the excited states with accurate semiempirical dynamic polarizabilities of the ground states of noble-gas atoms [16]. The theoretical lifetimes and branching ratios [17] are adjusted to reproduce the measured static polarizabilities [18], which are known with a 2% uncertainty. We estimated the additional small contributions within the Dirac-Hartree-Fock framework. The resulting polarizabilities satisfy the Thomas-Reiche-Kuhn oscillator strength sum rule.

The Racah notation for atomic levels is used. The particle-hole states are labeled as $n\ell(K)_J$ or $n\ell'(K)_J$, where $n$ and $\ell$ are the principal and the orbital angular momentum quantum numbers of the valence electron and $K = J_0 + \ell$, where $J_0$ is the angular momentum of the core. The primed configurations converge to a Rydberg series limit with a hole in the $(n-1)p_{1/2}$ state, and the unprimed to a hole in the $(n-1)p_{3/2}$ state. The manifold of the lowest $ns$ valence states has four fine-structure states $ns'(1/2)_{0,1}$ and $ns(3/2)_{1,2}$, and the lowest $np$ manifold consists of ten states. We investigate here the molecular potentials asymptotically connecting to the $ns(3/2)_2$ atomic states.

We calculate the long-range molecular potentials in the framework of Rayleigh-Schrödinger perturbation theory. The basis functions are defined as products of atomic wavefunctions

$$|M_1M_2; \Omega\rangle = |ns(3/2)_2M_1\rangle_1 |ns(3/2)_2M_2\rangle_2,$$

where the index 1(2) describes the wavefunction located on the center 1(2) and $\Omega = M_1 + M_2$, $M_{1,2}$ being projections of the atomic total angular momentum on the internuclear axis. Due to the axial symmetry of a dimer $\Omega$ is a conserved quantum number. It takes values ranging from zero to four. The two-atom basis (1) is degenerate and the correct molecular wavefunctions are obtained by diagonalizing the molecular Hamiltonian

$$\hat{H} = \hat{H}_1 + \hat{H}_2 + \hat{V}(R).$$

In expression (2) $\hat{H}_k$ represent the Hamiltonians of the two non-interacting atoms, and $\hat{V}(R)$ is the interaction potential at an internuclear distance $R$. The energy of the $ns(3/2)_2$ metastable state is designated as $E^*$. Then in the model space (1) we have

$$(\hat{H}_1 + \hat{H}_2)|M_1M_2; \Omega\rangle = 2E^* |M_1M_2; \Omega\rangle.$$  

The residual electrostatic potential $\hat{V}(R)$ is defined as the full Coulomb interaction energy in the dimer excluding interactions of the atomic electrons with their parent nuclei.
The multipole interactions (\(L = 1\) for dipole-dipole, and \(L = 2\) for quadrupole-quadrupole interactions) are given by [19]

\[ V_{\LL}(R) = \frac{1}{R^{2L+1}} \sum_{\mu = -L}^{L} \frac{(2L)!}{(L-\mu)!(L+\mu)!} \left( T_{\mu}^{(L)} \right)_{1} \left( T_{-\mu}^{(L)} \right)_{2}, \]

with the multipole spherical tensors

\[ T_{\mu}^{(L)} = -|e| \sum_{i} r_{i}^{L} C_{\mu}^{(L)}(\mathbf{r}_{i}), \]

where the summation is over atomic electrons, \(\mathbf{r}_{i}\) is the position vector of electron \(i\), and \(C_{\mu}^{(L)}(\mathbf{r}_{i})\) are reduced spherical harmonics [20]. In the following we write \(d_{\mu} = T_{\mu}^{(1)}\) and \(Q_{\mu} = T_{\mu}^{(2)}\).

The lowest-order contribution to the term energies arises from the quadrupole-quadrupole interaction \(\hat{V}_{qq}\), which varies as \(1/R^5\). However, the corresponding \(C_{6}\) coefficients are only of the order \(10^{-4} - 10^{-2}\) a.u. [2], and the dominant contribution appears in the second order in \(\hat{V}(R)\), arising from the dipole-dipole interaction \(\hat{V}_{dd}\). The second-order dipole interaction is proportional to \(1/R^6\), and the associated dispersion coefficient \(C_{6}\) is of the order of \(10^4 - 10^5\) a.u. Applying the formalism of degenerate perturbation theory in second order [21], we obtain an effective Hamiltonian within the two-atom basis Eq. (1)

\[ \langle m|H_{\text{eff}}^{(2)}|n\rangle = 2E^{*}\delta_{mn} + \langle m|\hat{V}_{qq}|n\rangle + \sum_{\Psi_{i}} \langle m|\hat{V}_{dd}|\Psi_{i}\rangle \langle \Psi_{i}|\hat{V}_{dd}|n\rangle \frac{2E^{*} - E_{i}}{2E^{*} - E_{i}}. \]

The intermediate molecular state \(|\Psi_{i}\rangle\) with unperturbed energy \(E_{i}\) runs over a complete set of two-atom states, excluding the model-space states Eq. (1). The formalism of the generalized Bloch equation [22] would allow the inclusion in the model space of the other three atomic states in the ns manifold and would account for the mixing of the different fine-structure levels; but such a large model space is not necessary for \(R > 10\) a.u.. The position of the avoided level crossing can be estimated from \(R_{l,c}\approx 2C_{6}/(E_{ns(3/2)_{1}} - E_{ns(3/2)_{2}}) \sim 10\) a.u.. The \(\Omega = 4\) molecular term is unique and being unaffected by avoided crossings, the region of applicability is extended to \(R \sim n\) a.u., before the electronic clouds start to overlap. The effect of the quadrupole-quadrupole interaction on the term energy can be disregarded at values of \(R \ll C_{6}/C_{5} \sim 10^{3}\) a.u.. The quadrupole-quadrupole correction is discussed by Doery et al. [3].

Using the Wigner-Eckart theorem, we can represent the matrix element of the dipole-dipole term in the effective second-order Hamiltonian as

\[
\sum_{\Psi_{i}} \frac{\langle M_{1}M_{2};\Omega|\hat{V}_{dd}|\Psi_{i}\rangle \langle \Psi_{i}|\hat{V}_{dd}|M'_{1}M'_{2};\Omega \rangle}{2E^{*} - E_{i}} - \frac{1}{R^{6}} \sum_{J_{a}J_{b}} C_{6}^{J_{a}J_{b}} (-1)^{J_{a}+J_{b}} \frac{Z}{3} \sum_{I_{\mu}} w_{I}^{1} w_{I_{\mu}} A_{I_{\mu}}^{J_{a}J_{b}}(M_{1}, M'_{1}) A_{-\mu}^{J_{b}}(\Omega - M_{1}, \Omega - M'_{1}) \frac{2E^{*} - E_{i}}{2E^{*} - E_{i}}.
\]

The dipole weights \(w_{I}^{1} = w_{I}^{-1} = w_{I}^{0} = 1\), and \(w_{I}^{1} = 2\). \(J_{a}\) and \(J_{b}\) are the corresponding total angular momenta of intermediate atomic states of atoms 1 and 2, and

\[ A_{I_{\mu}}^{J_{a}J_{b}}(M_{1}, M'_{1}) = \begin{pmatrix} 2 & 1 & I \\ -M_{1} & 1 & \mu \\ -m & \lambda & M'_{1} \end{pmatrix}, \]

where \(m = \frac{1}{2}(M_{1} + M_{1}' + \lambda - \mu)\). The intermediate (uncoupled) dispersion coefficients are

\[ C_{6}^{J_{a}J_{b}} = \frac{3}{\pi} \int_{0}^{\infty} dw S_{J_{a}}(iw)S_{J_{b}}(iw). \]

The reduced dynamic dipole polarizability \(S_{I}(iw)\) of purely imaginary argument is defined as the sum over atomic states \(|\alpha_{I}IM_{I}\rangle\) with total angular momentum \(I\) and energy \(E_{\alpha_{I}}\)

\[ S_{I}(iw) = \sum_{\alpha_{I}} \frac{(E^{*} - E_{\alpha_{I}})|\langle ns(3/2)_{2}|d|\alpha_{I}I\rangle|\langle \alpha_{I}I||d||ns(3/2)_{2}\rangle|}{(E^{*} - E_{\alpha_{I}})^{2} + \omega^{2}}. \]
Here $\alpha_I$ stands for all quantum numbers of the intermediate state, except for the total angular momentum $I$, and $\langle i||d||j \rangle$ are the reduced electric-dipole matrix elements, defined by the Wigner-Eckart theorem. Three sums with $I = 1, 2, 3$ are allowed by electric-dipole selection rules.

We proceed now to construct the dipole polarizability functions $S_I(i\omega)$, Eq. (9), evaluate the uncoupled dispersion coefficients (8), and set and diagonalize the second-order effective Hamiltonian (5).

The functions $S_I(i\omega)$ satisfy several sum rules. In particular, the static tensor dipole polarizability $\alpha_{zz}(M)$ of the $ns(3/2)_2$ state may be expressed as

$$\alpha_{zz}(M) = -2 \sum_I (-1)^I \left( \begin{array}{cc} 2 & I \\ -M & 0 \end{array} \right)^2 \times S_I(0).$$

The static tensor dipole polarizabilities $\alpha_{zz}(M = 1)$ and $\alpha_{zz}(M = 2)$ of metastable noble-gas atoms Ne through Xe have been measured by Molof et al. [18] to within an error of 2%. In the present calculations the values of $S_I(0)$ are adjusted to reproduce these experimental values. In addition, as $\omega \to \infty$, the reduced polarizabilities satisfy the nonrelativistic Thomas-Reiche-Kuhn (TRK) sum rule

$$\sum_n f_{kn} = \frac{2}{15} \sum_I (-1)^{I+1} S_I(i\infty) = N,$$

$N$ being the number of electrons in the atom. Our constructed polarizabilities satisfy the sum rule.

It is instructive to consider the action of a one-particle operator on the reference particle-hole Slater determinant $ns(3/2)_2$ in the independent electron approximation. Such an operator can (i) annihilate the reference particle-hole pair; (ii) promote a valence electron from $ns_{1/2}$ state to another valence state $np_{1/2,3/2}$, the state of the $(n-1)p_{3/2}$ hole being unchanged; (iii) deexcite the $(n-1)p_{3/2}$ hole into some other hole state, the valence state remaining the same; and (iv) create another particle-hole pair in addition to the reference pair. According to such a classification it is convenient to break the polarizability function, Eq. (9), into three contributions $S^k_I$ corresponding to a number $k$ of particle-hole pairs in the intermediate state $|\alpha_I I\rangle$

$$S_I = S^0_I + S^1_I + S^2_I.$$ 

Since an electric-dipole transition from $ns(3/2)_2$ to the closed-core state $^1S_0$ is prohibited by the angular selection rules, $S^0_I \equiv 0$.

The sum $S^1_I$ is separated into contributions from the intermediate states in the lowest $np$ fine-structure multiplet $(S^1_I)_{np}$, and the rest of the sum $(S^1_I)^J$

$$S^1_I = (S^1_I)_{np} + (S^1_I)^J.$$ 

The first term is the dominant contribution. We calculate $(S^1_I)_{np}$ using experimental values of transition energies and decay rates, and adjusted branching ratios. The rest of the sum over valence states (including bound and continuum states) $(S^1_I)^J$ is estimated in the Dirac-Hartree-Fock (DHF) approximation. The metastable state $|vh; J\rangle$ in lowest order is represented as a combination of the $|n\rangle = ns_{1/2}$ particle state and the hole state $|h\rangle = (n-1)p_{3/2}$, coupled to the total angular momentum $J = 2$, $|(n-1)p_{3/2}^{-1}ns_{1/2}\rangle$. The lowest-order energy of such a state is $\varepsilon^* = \varepsilon_v - \varepsilon_h$, $\varepsilon_i$ being the energy of the DHF orbital $|i\rangle$. The intermediate state is represented as $|ma; I\rangle$, a particle state $m$ coupled with hole state $a$ to the total momentum $I$. Explicitly,

$$(S^1_I)^J(i\omega) = (-1)^J+1|J||J| \sum_{ma} \left\{ \delta_{ha} \frac{(\varepsilon_v - \varepsilon_m) \langle v||d||m \rangle^2}{(\varepsilon_v - \varepsilon_m)^2 + \omega^2} \left( \begin{array}{cc} J & 1 \\ jm & jh \end{array} \right)^2 \right\}

+ \delta_{mv} \frac{(\varepsilon_v - \varepsilon_h) \langle h||d||a \rangle^2}{(\varepsilon_v - \varepsilon_h)^2 + \omega^2} \left( \begin{array}{cc} J & 1 \\ ja & jh \end{array} \right)^2,$$

where $|K\rangle \equiv 2K + 1$, the summation is performed over the core orbitals $a$ and excited states $m$, excluding states of the lowest $np$ multiplet, and $J = 2$. The first sum is associated with excitation of the valence electron (case (ii)) while the second sum with deexcitation of the hole state (case (iii)). To arrive at this result we disregarded the coupling between levels within the same fine-structure multiplet. For example, for Ne the $[(2p_{3/2})^{-1}4p_{1/2}]^2$, $[(2p_{3/2})^{-1}4p_{3/2}]^2$, and $[(2p_{1/2})^{-1}4p_{3/2}]^2$ states are summed over independently, even though the correct lowest-order wave-function is a linear combination of them. This approximation corresponds to a disregard of the small difference between the
the reference data. To reduce the consequent errors, the experimental values of the static polarizability, accurate to 2%, were chosen as

\[ \frac{1}{\alpha_g} = \frac{1}{\alpha_g + \frac{1}{2}J \alpha_g} \]

which would introduce an uncertainty of 4-5% in the static polarizabilities, and 8-10% inaccuracy in the values of \( C_6 \). The most accurate measurements of \( C_6 \) for Ne \([8,9]\), Ar \([10]\) and Kr \([11]\), and less than 3% for Xe \([12–14]\). However, the branching ratios \( B \) are not established to the same precision. The relevant lifetimes have been measured to within an error less than 1% for Ne \([8,9]\), Ar \([10]\) and Kr \([11]\), and less than 3% for Xe \([12–14]\). However, the branching ratios \( B \) are not established to the same precision. The relevant lifetimes have been measured to within an error less than 1

\[ S_I^2 = (S_I^2)_{\text{core}} + (S_I^2)_{\text{ctr}}. \]

The first term is associated with the dynamic polarizability \( \alpha_g(i\omega) \) of the closed-shell ground state \( ^1S_0 \) and the second term is a corrective counter term.

\[ (S_I^2)_{\text{core}} (i\omega) = (-1)^{f+J+1} \frac{1}{2} \alpha_g (i\omega). \]

We use the semiempirical dynamic polarizabilities for the ground states of noble-gas atoms of Kumar and Meath \([16]\). We separate the sum \( S_I^2 \) over the core excited states into two contributions

\[ \sum_{n} (\alpha_g) \]
of the C approximately 4%, reflecting the 2% experimental error in the static dipole tensor polarizabilities [18]. The values of the form of the small corrections due to the quadrupole-quadrupole interaction results in parameterization of term energies in becomes increasingly marked, from 6.5% in Ne to 16% in Xe.

\[ \text{The calculated dispersion coefficients } C_6 \text{ for various molecular symmetries are listed in Table III. Since the region close to } \omega = 0 \text{ contributes the most to the values of the integral in Eq. (8), the uncertainty in the values of } C_6 \text{ is approximately 4%, reflecting the 2% experimental error in the static dipole tensor polarizabilities [18]. The values of the } C_6 \text{ coefficients grow monotonically from Ne to Xe, due to the reduction in the energy separations between the metastable states and the } np \text{-manifold. For heavier systems the anisotropy in } C_6, \text{ arising from relativistic effects becomes increasingly marked, from 6.5% in Ne to 16% in Xe.}

Long-range dispersion coefficients for two interacting metastable Ne atoms were evaluated recently by Doery et al. [3]. The \( C_6 \) coefficients were calculated from the diagonalization of the molecular dipole-dipole Hamiltonian in the model space containing the lowest \( np \)-manifold, so limiting the intermediate states to the lowest \( np \)-manifold states in the present formulation. Experimental values of decay rates and branching ratios were used to deduce the electric-dipole matrix elements. The precision of the calculated values of \( C_6 \) is about 8-10% due to the large uncertainty in the branching ratios. The values of \( C_6 \) from Ref. [3] for different molecular symmetries vary between 1951 and 1956 a.u., exhibiting much less anisotropy than the present results which range between 1877 and 1999 a.u.. The difference can be traced to the anisotropy in the static dipole polarizabilities. Indeed, utilizing input data from Ref. [3] we obtain \( \alpha_{zz}(M = 1) = 192 \) and \( \alpha_{zz}(M = 2) = 189 \) a.u. if we include only the \( np \) manifold as in Ref. [3]. While \( \alpha_{zz}(M = 1) \) agrees with the experimental value [18] 192(4), the \( \alpha_{zz}(M = 2) \) is overestimated by three standard deviations compared to the experimental value 180(3) a.u..

The accuracy of the dispersion coefficients could be improved by applying relativistic all-order many-body methods [23,24] to calculate transition amplitudes between \( ns - np \) manifolds. Such ab initio calculations are intrinsically more challenging than for alkali-metal atoms; the accurate experimental lifetimes would provide an excellent gauge of accuracy.

Our values of \( C_6 \) coefficients will be useful in studies of cold collisions of metastable rare-gas atoms [1–4]. For example, we can estimate the rate coefficient for Penning ionization by ignoring spin-polarization and assuming that every trajectory that surmounts the angular momentum barrier leads to ionization [1,25]. The corresponding rate coefficient is given by [25]

\[ k = 6.35 \times 10^{-9} \frac{C_6^{1/3} T^{1/6}}{\mu^{1/2}} \text{ cm}^3 \text{ s}^{-1}, \]

where \( \mu \) is the reduced mass measured in units of the electron mass and \( T \) is the temperature. Combined with short-range potentials [3,26] a number of other properties could be determined. For example, scattering lengths of elastic collisions could be found, providing input for mean-field equations describing dilute quantum gases.

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**TABLE I.** Input data for Ar and Xe calculations. Lifetimes \( \tau \) for Ar are from Volz and Schmoranzer, Ref. [10], and for Xe from Iinoue et al., Ref. [12], except where noted. Branching ratios \( B \) to the \( ns(3/2)_{2} \) level are adjusted to reproduce experimental tensor dipole polarizabilities, as discussed in the text.

<table>
<thead>
<tr>
<th>state</th>
<th>( \tau, \text{ ns} )</th>
<th>( B(%) )</th>
<th>( \tau, \text{ ns} )</th>
<th>( B(%) )</th>
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<tr>
<td>( np^1(1/2)_{1} )</td>
<td>27.85(7)</td>
<td>17.66</td>
<td>43.5(1.5)^{b}</td>
<td>5.15</td>
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<tr>
<td>( np^1(3/2)_{2} )</td>
<td>29.01(7)</td>
<td>10.59</td>
<td>38.1(1.3)^{c}</td>
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<tr>
<td>( np^1(3/2)_{1} )</td>
<td>29.83(8)</td>
<td>1.84</td>
<td>49(2)</td>
<td>1.16</td>
</tr>
<tr>
<td>( np(3/2)_{2} )</td>
<td>28.52(7)</td>
<td>68.55</td>
<td>31(1)</td>
<td>65.42</td>
</tr>
<tr>
<td>( np(3/2)_{1} )</td>
<td>29.62(7)</td>
<td>14.97</td>
<td>37(1)</td>
<td>7.45</td>
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TABLE II. Intermediate dispersion coefficients $C_6^{J_aJ_b}$, a.u., multiplied by a factor $10^{-3}$.

<table>
<thead>
<tr>
<th></th>
<th>$C_6^{11}$</th>
<th>$C_6^{21}$</th>
<th>$C_6^{22}$</th>
<th>$C_6^{31}$</th>
<th>$C_6^{32}$</th>
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<td>Ne</td>
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<tr>
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<td>36.91</td>
<td>47.74</td>
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<td>98.31</td>
</tr>
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</table>

TABLE III. Dispersion coefficients $C_6$ in a.u. for the interaction of two metastable $ns(3/2)_2$ noble-gas atoms.

<table>
<thead>
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<th>Ar</th>
<th>Kr</th>
<th>Xe</th>
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