Body-Assisted van der Waals Interaction between Excited Atoms

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We present a formula for the body-assisted van der Waals interaction potential between two atoms, one or both being prepared in an excited energy eigenstate. The presence of an arbitrary arrangement for a material environment is taken into account via the Green function. The resulting formula supports one of two conflicting findings recorded. The consistency of our formula is investigated by applying it for the case of two atoms in free space and comparing the resulting expression with the one found from the limiting Casimir-Polder potential between an excited atom and a small dielectric sphere.

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Dispersion interactions are understood as a result of the quantum description of the electromagnetic field. As the atoms under consideration are assumed to be ground-state atoms, transitions to excited energy eigenstates are involved in the interaction together with the emission of virtual photons from a continuous range of frequencies. In the case of excited atoms, transitions to lower-lying states can occur while releasing the energy difference in the form of real photons of certain discrete frequencies (see, e.g., Ref. [1] for atom-body interaction). In the two-atom case, the long-range potential was first found to have an oscillatory distance dependence with an amplitude falling off as r^{-2} [2,3]. Later on, the long-range potential was confirmed to fall off as r^{-2}, but without oscillations [4,5]. The difference between the two findings arises from the way the photon integrals have been treated. The two contradictory results are valid from a mathematical point of view and a possible oscillatory behavior of the retarded van der Waals (VDW) potential remains as an open question. A time-dependent calculation supports the oscillatory result [6]. On the experimental side, the vacuum-induced level shift (Casimir-Polder potential) of an excited barium ion in the presence of a mirror is observed to show an oscillatory distance dependence [7,8].

In this Letter, we first derive the body-assisted VDW interaction potential between two excited atoms, using fourth-order perturbation theory. As the background media is replaced by free space in our formula, the resulting expression for the long-range interatomic separation shows an oscillatory distance dependence, in agreement with Refs. [2,3], while it does not agree with the formula given in Refs. [4,5]. In order to facilitate a judgement about the two contradictory results, we will take a proper limit from the known atom-body Casimir-Polder (CP) potential for an excited atom and reduce it to the atom-atom VDW interaction, to see whether the outcome supports any of the above mentioned results.

Let us, first, derive the formula of the body-assisted VDW interaction between two excited atoms. A very detailed derivation of the formula for the case of ground-state atoms is given in Ref. [9]. In the case of excited atoms, the starting point is the same as for the ground-state atoms; hence, we refer the reader to the calculation in Ref. [9]. We only point out the differences which arise from the fact that one or both atoms might be in excited states here.

Consider two atoms A and B in the presence of an arbitrary arrangement of magnetoelectric bodies, located at positions $\boldsymbol{r}_A$ and $\boldsymbol{r}_B$, each being excited to an energy eigenstate, say $|k\rangle_A$ and $|l\rangle_B$, respectively. The VDW interaction potential resulting form the fourth-order perturbation, following a calculation similar to the one for the ground-state atoms in Ref. [9], leads to

$$
\begin{aligned}
U_{AB}(\boldsymbol{r}_A,\boldsymbol{r}_B) = {} & \frac{i\mu_0^2}{\hbar\pi}\sum_{\substack{m\neq k\\ n\neq l}}\frac{1}{\omega_A^{mk}+\omega_B^{nl}} \\
& \times \mathcal{P}\left\{\int_0^\infty d\omega\,\frac{\omega^4(\omega_A^{mk}+\omega_B^{nl}+\omega)}{(\omega+\omega_A^{mk})(\omega+\omega_B^{nl})}\right. \\
& \left. + \int_0^{-\infty} d\omega\,\frac{\omega^4(\omega_A^{mk}+\omega_B^{nl}-\omega)}{(\omega-\omega_A^{mk})(\omega-\omega_B^{nl})}\right\} \\
& \times [\boldsymbol{d}_A^{km}\cdot\mathbf{G}(\boldsymbol{r}_A,\boldsymbol{r}_B,\omega)\cdot\boldsymbol{d}_B^{ln}]^2
\end{aligned}
\qquad (1)
$$

[compare with Eq. (45) in Ref. [9]] with $\boldsymbol{d}_A^{km} = \langle k|\hat{\boldsymbol{d}}_A|m\rangle$ and ω_A^{km} denoting, respectively, the electric dipole moments and frequencies of the atomic transitions. All geometric and magnetoelectric properties of the environmental media are contained in the Green tensor $\mathbf{G}$ via the frequency-dependent relative electric permittivity $\varepsilon(\boldsymbol{r},\omega)$ and relative magnetic permeability $\mu(\boldsymbol{r},\omega)$. The Green tensor is the unique solution to the inhomogeneous Helmholtz differential equation

$$
\nabla\times\frac{1}{\mu(\boldsymbol{r},\omega)}\nabla\times\mathbf{G}(\boldsymbol{r},\boldsymbol{r}',\omega)-\varepsilon(\boldsymbol{r},\omega)\frac{\omega^2}{c^2}\mathbf{G}(\boldsymbol{r},\boldsymbol{r}',\omega)=\mathbf{I}\delta(\boldsymbol{r}-\boldsymbol{r}'),
\qquad (2)
$$

with the boundary condition

$$\mathbf{G}(\boldsymbol{r},\boldsymbol{r}',\omega)=\mathbf{0}\quad\text{for }|\boldsymbol{r}-\boldsymbol{r}'|\to\infty.\tag{3}$$

Further, it obeys the Schwartz reflection principle,

$$\mathbf{G}(\boldsymbol{r},\boldsymbol{r}',\omega)=\mathbf{G}^{*}(\boldsymbol{r},\boldsymbol{r}',-\omega^{*}),\tag{4}$$

and the Onsager reciprocal relation,

$$\mathbf{G}(\boldsymbol{r},\boldsymbol{r}',\omega)=\mathbf{G}^{\top}(\boldsymbol{r}',\boldsymbol{r},\omega).\tag{5}$$

The integrands in Eq. (1), recalling the general properties the Green tensor $\mathbf{G}$ as a response function, are analytic for $m>k$ and $n>l$, in the upper half of the complex-frequency plane including the real axis. The letter $\mathcal{P}$ just before the curly brackets stands for the principal value and makes particular sense for $m<k$ and/or $n<l$. Equation (1) can be simplified by using contour-integral techniques. For the first integral, we may use Cauchy's theorem and replace the integral by a contour integral along infinitesimal half-circles around the possible poles at $\omega=-\omega_A^{mk}=\omega_A^{km}$ and $\omega=-\omega_B^{nl}=\omega_B^{ln}$, an infinitely large quarter-circle in the first quadrant and along the positive imaginary axis, introducing a purely imaginary frequency, $\omega=iu$. The integral along the infinitely large quarter-circle vanishes due to the limiting behavior of the Green tensor [10]

$$\lim_{|\omega|\to\infty}\frac{\omega^2}{c^2}\mathbf{G}(\boldsymbol{r},\boldsymbol{r}',\omega)=-\mathbf{I}\delta(\boldsymbol{r}-\boldsymbol{r}').\tag{6}$$

The result becomes

$$\begin{aligned}\mathcal{P}\int_0^\infty d\omega\,&\frac{\omega^4(\omega_A^{mk}+\omega_B^{nl}+\omega)}{(\omega+\omega_A^{mk})(\omega+\omega_B^{nl})}\mathcal{G}(\boldsymbol{r}_A,\boldsymbol{r}_B,\omega)\\&=\int_0^\infty du\,\frac{iu^4(\omega_A^{mk}+\omega_B^{nl}+iu)}{(iu+\omega_A^{mk})(iu+\omega_B^{nl})}\mathcal{G}(\boldsymbol{r}_A,\boldsymbol{r}_B,iu)\\&\quad+i\pi\frac{\omega_A^{km}\omega_B^{ln}}{\omega_B^{ln}-\omega_A^{km}}[\Theta(k-m)(\omega_A^{km})^3\mathcal{G}(\boldsymbol{r}_A,\boldsymbol{r}_B,\omega_A^{km})\\&\quad-\Theta(l-n)(\omega_B^{ln})^3\mathcal{G}(\boldsymbol{r}_A,\boldsymbol{r}_B,\omega_B^{ln})],\end{aligned}\tag{7}$$

[$\Theta(x)$, unit step function], where $\mathcal{G}(\boldsymbol{r}_A,\boldsymbol{r}_B,\omega)$ is used as an abbreviation for $[\boldsymbol{d}_A^{km}\cdot\mathbf{G}(\boldsymbol{r}_A,\boldsymbol{r}_B,\omega)\cdot\boldsymbol{d}_B^{ln}]^2$. In a similar manner, for the second integral in Eq. (1), we find

$$\begin{aligned}\mathcal{P}\int_0^{-\infty} d\omega\,&\frac{\omega^4(\omega_A^{mk}+\omega_B^{nl}-\omega)}{(\omega-\omega_A^{mk})(\omega-\omega_B^{nl})}\mathcal{G}(\boldsymbol{r}_A,\boldsymbol{r}_B,\omega)\\&=\int_0^\infty du\,\frac{iu^4(\omega_A^{mk}+\omega_B^{nl}-iu)}{(iu-\omega_A^{mk})(iu-\omega_B^{nl})}\mathcal{G}(\boldsymbol{r}_A,\boldsymbol{r}_B,iu)\\&\quad+i\pi\frac{\omega_A^{km}\omega_B^{ln}}{\omega_B^{ln}-\omega_A^{km}}[\Theta(k-m)(\omega_A^{km})^3\mathcal{G}^*(\boldsymbol{r}_A,\boldsymbol{r}_B,\omega_A^{km})\\&\quad-\Theta(l-n)(\omega_B^{ln})^3\mathcal{G}^*(\boldsymbol{r}_A,\boldsymbol{r}_B,\omega_B^{ln})],\end{aligned}\tag{8}$$

where Eq. (4) is used. Now, by combining Eqs. (1), (7), and (8), and making use of Eq. (5), the two-atom interaction potential, after being split into the off-resonant and resonant parts, can be written as follows:

$$U(\boldsymbol{r}_A,\boldsymbol{r}_B)=U^{or}(\boldsymbol{r}_A,\boldsymbol{r}_B)+U^{r}(\boldsymbol{r}_A,\boldsymbol{r}_B),\tag{9}$$

$$\begin{aligned}U^{or}(\boldsymbol{r}_A,\boldsymbol{r}_B)=&-\frac{\hbar\mu_0^2}{2\pi}\int_0^\infty du u^4\\&\times\mathrm{tr}[\boldsymbol{\alpha}_A^k(iu)\cdot\mathbf{G}(\boldsymbol{r}_A,\boldsymbol{r}_B,iu)\cdot\boldsymbol{\alpha}_B^l(iu)\cdot\mathbf{G}(\boldsymbol{r}_B,\boldsymbol{r}_A,iu)],\end{aligned}\tag{10}$$

$$\begin{aligned}U^{r}(\boldsymbol{r}_A,\boldsymbol{r}_B)=&-\mu_0^2\sum_{m<k}(\omega_A^{km})^4\mathrm{Re}[\boldsymbol{d}_A^{km}\cdot\mathbf{G}(\boldsymbol{r}_A,\boldsymbol{r}_B,\omega_A^{km})\\&\cdot\boldsymbol{\alpha}_B^l(\omega_A^{km})\cdot\mathbf{G}(\boldsymbol{r}_B,\boldsymbol{r}_A,\omega_A^{km})\cdot\boldsymbol{d}_A^{mk}]\\&-\mu_0^2\sum_{n<l}(\omega_B^{ln})^4\mathrm{Re}[\boldsymbol{d}_B^{ln}\cdot\mathbf{G}(\boldsymbol{r}_B,\boldsymbol{r}_A,\omega_B^{ln})\\&\cdot\boldsymbol{\alpha}_A^k(\omega_B^{ln})\cdot\mathbf{G}(\boldsymbol{r}_A,\boldsymbol{r}_B,\omega_B^{ln})\cdot\boldsymbol{d}_B^{nl}].\end{aligned}\tag{11}$$

In Eqs. (10) and (11), $\boldsymbol{\alpha}_A^k(\omega)$ is the electric polarizability tensor of atom A in the kth energy eigenstate, defined as

$$\begin{aligned}\alpha_A^k(\omega)&=\frac{2}{\hbar}\lim_{\epsilon\to0+}\sum_m\frac{\omega_A^{mk}\boldsymbol{d}_A^{km}\boldsymbol{d}_A^{mk}}{(\omega_A^{mk})^2-\omega^2-i\epsilon\omega}\\&=\frac{2\mathbf{I}}{3\hbar}\lim_{\epsilon\to0+}\sum_m\frac{\omega_A^{mk}|\boldsymbol{d}_A^{km}|^2}{(\omega_A^{mk})^2-\omega^2-i\epsilon\omega}\equiv\alpha_A^k(\omega)\mathbf{I}.\end{aligned}\tag{12}$$

Needless to say, in the case where atom B is in its ground state ($l=0$), the second term on the right-hand side of Eq. (11) vanishes.

In the simplest case of two isotropic atoms in an infinitely extended free space, the required Green tensor ($\mathbf{G}\to\mathbf{G}^{(0)}$) is given as [11]

$$\mathbf{G}^{(0)}(\boldsymbol{r}_A,\boldsymbol{r}_B,\omega)=\frac{-c^2\mathrm{e}^{i\omega l/c}}{4\pi\omega^2l^3}[p(-il\omega/c)\mathbf{I}-q(-il\omega/c)\mathbf{e}\mathbf{e}],\tag{13}$$

with $l=|\boldsymbol{r}_B-\boldsymbol{r}_A|$, $\mathbf{e}=(\boldsymbol{r}_B-\boldsymbol{r}_A)/l$, and

$$p(x)=1+x+x^2,\tag{14}$$

$$q(x)=3+3x+x^2.\tag{15}$$

Choosing the Cartesian coordinates system such that its origin corresponds to the location of atom A while $\boldsymbol{r}_B=(r,0,0)$, the only nonzero matrix elements of the Green tensor are as follows:

$$G_{xx}^{(0)}(\boldsymbol{r}_A,\boldsymbol{r}_B,\omega)=\frac{c^2}{2\pi\omega^2r^3}(1-ir\omega/c)\mathrm{e}^{i\omega r/c},\tag{16}$$

$$\begin{aligned}G_{yy}^{(0)}(\boldsymbol{r}_A,\boldsymbol{r}_B,\omega)&=G_{zz}^{(0)}(\boldsymbol{r}_A,\boldsymbol{r}_B,\omega)\\&=\frac{-c^2}{4\pi\omega^2r^3}(1-ir\omega/c-r^2\omega^2/c^2)\mathrm{e}^{i\omega r/c}.\end{aligned}\tag{17}$$

Substitution of these into Eqs. (10) and (11) leads to

$$U^{or}(r) = \frac{-\hbar}{16\pi^3\varepsilon_0^2 r^6}\int_0^\infty du\,\alpha_A^k(iu)\alpha_B^0(iu)f(ru/c), \quad (18)$$

$$U^r(r) = \frac{-1}{24\pi^2\varepsilon_0^2 r^6}\sum_{m<k}|\boldsymbol{d}_A^{km}|^2\alpha_B^0(\omega_A^{km}) \times [(3-5\eta_m^2+\eta_m^4)\cos(2\eta_m)+(6\eta_m-2\eta_m^3)\sin(2\eta_m)], \quad (19)$$

where $\eta_m = r\omega_A^{km}/c$, atom B is assumed to be in its ground state, and

$$f(x) = e^{-2x}(3+6x+5x^2+2x^3+x^4). \quad (20)$$

Equation (18) is exactly the well known result for the off-resonant part of the VDW interaction potential in free space (see, e.g., Ref. [4]). The resonant part, Eq. (19), is in agreement with Ref. [3] in the retarded limit, while it does not agree with the finding of Refs. [4,5]. The difference arises from the ways the perturbative calculation was accomplished; the frequency integrals in Refs. [4,5] were performed such that the poles on the real axis were treated by addition or subtraction of small pure-imaginary frequencies, whereas in Ref. [3], the integrals were treated as principal value integrals. If one imitates the calculations in Ref. [4] in order to generalize its result to the inclusion of material background, for the resonant part of the VDW interaction potential between an excited atom A and a ground-state atom B, one ends up with

$$U^r(\boldsymbol{r}_A,\boldsymbol{r}_B) = -\frac{\mu_0^2}{3}\sum_{m<k}(\omega_A^{km})^4 \times |\boldsymbol{d}_A^{km}|^2\alpha_B^0(\omega_A^{km})\sum_{i,j}|G_{ij}(\boldsymbol{r}_B,\boldsymbol{r}_A,\omega_A^{km})|^2 \quad (21)$$

[for an equivalent formula for two-level atoms see Eq. (68) in Ref. [12] or Eq. (3) in Ref. [13]]. Now, the interaction potential in free space, according to this formula, can be obtained using the matrix elements (16) and (17). The result, as expected, meets the one given in Refs. [4,5],

$$U^r(r) = \frac{-\mu_0^2}{24\pi^2 r^2}\sum_{m<k}(\omega_A^{km})^4 \times |\boldsymbol{d}_A^{km}|^2\alpha_B^0(\omega_A^{km})(1+\eta_m^{-2}+3\eta_m^{-4}). \quad (22)$$

A possible way to judge the two contradictory results, Eq. (19) and Eq. (22), may be attacking the problem via a sufficiently different approach. To this end, we start with the known CP interaction formula of an excited atom with a macroscopic body. The atom-atom interaction can be found by taking a proper limit of a small dielectric body (a homogeneous sphere here) and replacing it with a second atom.

According to the findings of Refs. [1,14], the CP Potential of an isotropic atom A prepared in an energy eigenstate $|k\rangle$ and located at a position $\boldsymbol{r}_A$ is given as

$$U(\boldsymbol{r}_A) = U^{or}(\boldsymbol{r}_A) + U^r(\boldsymbol{r}_A), \quad (23)$$

where U^{or} and U^r are, respectively, the off-resonant and resonant parts of the potential

$$U^{or}(\boldsymbol{r}_A) = \frac{\hbar\mu_0}{2\pi}\int_0^\infty du\,u^2\alpha_A^k(iu)\mathrm{tr}\mathbf{G}^{(1)}(\boldsymbol{r}_A,\boldsymbol{r}_A,iu), \quad (24)$$

$$U^r(\boldsymbol{r}_A) = -\frac{\mu_0}{3}\sum_{m<k}(\omega_A^{km}|\boldsymbol{d}_A^{km}|)^2\mathrm{tr}[\mathrm{Re}\mathbf{G}^{(1)}(\boldsymbol{r}_A,\boldsymbol{r}_A,\omega_A^{km})], \quad (25)$$

with $\mathbf{G}^{(1)}$ being the scattering part of the Green tensor.

Let us consider the atom at a distance r from the center of a homogeneous dielectric sphere of radius a ($r > a$). The equiposition Green tensor required in Eqs. (24) and (25) can be extracted from Ref. [15] given for a more general case of a spherical magnetoelectric multilayer. However, we adopt a simplified version from Ref. [16]. Choosing the spherical coordinates system such that its origin coincides with the center of the sphere, the scattering part of the equiposition Green tensor reads

$$\mathbf{G}^{(1)}(\boldsymbol{r},\boldsymbol{r},\omega) = \sum_{i=r,\theta,\phi}G_{ii}^{(1)}(\boldsymbol{r},\boldsymbol{r},\omega)\boldsymbol{e}_i\boldsymbol{e}_i, \quad (26)$$

with $\boldsymbol{e}_r$, $\boldsymbol{e}_\theta$, and $\boldsymbol{e}_\phi$ being the unit vectors pointing the directions of radial distance r, polar angle θ, and azimuthal angle ϕ, respectively. The matrix elements of the Green tensor (26) are as follows:

$$G_{rr}^{(1)} = \frac{ic}{4\pi\omega r^2}\sum_{n=1}^\infty n(n+1)(2n+1)B_n^N(\omega)[h_n^{(1)}(r\omega/c)]^2, \quad (27)$$

$$G_{\theta\theta}^{(1)} = G_{\phi\phi}^{(1)} = \frac{i\omega}{8\pi c}\sum_{n=1}^\infty(2n+1)\left\{B_n^M(\omega)[h_n^{(1)}(r\omega/c)]^2 + \frac{c^2B_n^N(\omega)}{\omega^2r^2}[zh_n^{(1)}(z)]'^2_{z=r\omega/c}\right\}, \quad (28)$$

where $h_n^{(1)}(z)$ denotes the spherical Hankel function of the first kind, prime indicates differentiation with respect to the argument, and

$$B_n^M(\omega) = -\frac{[z_0j_n(z_0)]'j_n(z_1)-[z_1j_n(z_1)]'j_n(z_0)}{[z_0h_n^{(1)}(z_0)]'j_n(z_1)-[z_1j_n(z_1)]'h_n^{(1)}(z_0)}, \quad (29)$$

$$B_n^N(\omega) = -\frac{\varepsilon(\omega)[z_0j_n(z_0)]'j_n(z_1)-[z_1j_n(z_1)]'j_n(z_0)}{\varepsilon(\omega)[z_0h_n^{(1)}(z_0)]'j_n(z_1)-[z_1j_n(z_1)]'h_n^{(1)}(z_0)}, \quad (30)$$

with $j_n(z)$ being the spherical Bessel function of the first kind, $z_0 = a\omega/c$, $z_1 = \sqrt{\varepsilon(\omega)}z_0$. Substitution of $\mathbf{G}^{(1)}$ from Eq. (26) together with (27) and (28) into Eqs. (24) and (25) leads to

$$U^{or}(\boldsymbol{r}_A) = \frac{\hbar\mu_0 c}{8\pi^2 r^2}\sum_{n=1}^{\infty}(2n+1)\int_0^{\infty} du u \alpha_A^k(iu) \times \Big(B_n^N(iu)\{n(n+1)[h_n^{(1)}(z)]^2 + [zh_n^{(1)}(z)]'^2\} - \frac{r_A^2 u^2}{c^2}B_n^M(iu)[h_n^{(1)}(z)]^2\Big)_{z=iru/c}, \tag{31}$$

$$U^{r}(\boldsymbol{r}_A) = \frac{\mu_0 c}{12\pi r^2}\sum_{m<k}\omega_A^{km}|\boldsymbol{d}_A^{km}|^2\sum_{n=1}^{\infty}(2n+1) \times \mathrm{Im}\Big(B_n^N(\omega_A^{km})\{n(n+1)[h_n^{(1)}(z)]^2 + [zh_n^{(1)}(z)]'^2\} + \frac{r^2(\omega_A^{km})^2}{c^2}B_n^M(\omega_A^{km})[h_n^{(1)}(z)]^2\Big)_{z=r\omega_A^{km}/c}. \tag{32}$$

Equations (31) and (32) are valid as long as the atom is far enough from the surface of the sphere such that the description of the molecular structure of the sphere in a macroscopic manner is valid. However, we are interested in the limiting case of a small sphere, $a \ll r$. Following a discussion similar to the one given in Ref. [17], it can be shown that, in the summands in Eqs. (31) and (32), it is enough to retain only the $n = 1$ terms for which, Eqs. (29) and (30) reduce to

$$B_1^M(\omega) \simeq 0, \tag{33}$$

$$B_1^N(\omega) = \frac{2i}{3}\frac{\varepsilon(\omega)-1}{\varepsilon(\omega)+2}\left(\frac{a\omega}{c}\right)^3. \tag{34}$$

Substitution of these results for B_1^M and B_1^N and the explicit forms of the spherical Hankel function $h_1^{(1)}(z)$ into Eqs. (31) and (32) leads, after some simplifications, to

$$U^{or}(\boldsymbol{r}_A) = \frac{-\hbar a^3}{4\pi^2\varepsilon_0 r^6}\int_0^{\infty} du \alpha_A^k(iu)\frac{\varepsilon(iu)-1}{\varepsilon(iu)+2}f(ru/c), \tag{35}$$

$$U^{r}(\boldsymbol{r}_A) = \frac{-a^3}{6\pi\varepsilon_0 r^6}\sum_{m<k}|\boldsymbol{d}_A^{km}|^2\frac{\varepsilon(\omega_A^{km})-1}{\varepsilon(\omega_A^{km})+2} \times [(3-5\eta_m^2+\eta_m^4)\cos(2\eta_m) + (6\eta_m - 2\eta_m^3)\sin(2\eta_m)]. \tag{36}$$

Now, let us consider a sphere to which the Clausius-Mossotti relation applies, so that

$$\frac{\varepsilon(\omega)-1}{\varepsilon(\omega)+2} = \frac{\alpha_s(\omega)}{4\pi a^3\varepsilon_0}, \tag{37}$$

with α_s being the electric polarizability of the sphere. Making use of Eq. (37) in Eq. (35) leads back to formula (18), being the off-resonant part of the VDW interaction potential between an excited atom A and a ground-state atom in free space, where the electric polarizability of the sphere is just replaced by the ground-state polarizability of atom B.

Recovering the off-resonant part of the two-atom interaction potential via the limiting procedure mentioned above, also makes obtaining the resonant part via the same approach adequately reliable. This can be done by making use of Eq. (37) in Eq. (36). The result coincides with Eq. (19), which was based on a perturbative calculation, as the electric polarizability of the sphere is replaced by that of a ground-state atom B. Hence, it is evident that the photon integrals must be treated as principal-value integrals as in Refs. [2,3] and our formula, Eq. (11), is the correct one for the resonant VDW interaction potential between excited atoms.

In summary, in this Letter, we presented a new formula for the medium-assisted VDW interaction potential between two excited atoms. It can be a generalization of the findings of Refs. [2,3] to the presence of the arbitrary material environment. The consistency of the formula was confirmed by comparing its result for the case of two atoms in free space with the result obtained via a limiting approach from atom-body CP potential. The free-space result is of oscillatory distance-dependence behavior, in agreement with Refs. [2,3] for retarded atom-atom separations.

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