Resonance oscillations of nonreciprocal long-range van der Waals forces between atoms in electromagnetic fields

Yury Sherkunov[*]

Department of Physics, Loughborough University, Loughborough LE11 3TU, United Kingdom

We study theoretically the van der Waals interaction between two atoms out of equilibrium with an isotropic electromagnetic field. We demonstrate that at large interatomic separations, the van der Waals forces are resonant, spatially oscillating, and nonreciprocal due to resonance absorption and emission of virtual photons. We suggest that the van der Waals forces can be controlled and manipulated by tuning the spectrum of artificially created random light.

I. INTRODUCTION

The long-range dispersion interaction between atoms arising from quantum or thermal fluctuations of electromagnetic (EM) fields and atomic charges has been well understood for equilibrium systems since the pioneering works by Casimir and Polder [1] and Lifshitz with collaborators [2,3]. The situation is different for nonequilibrium systems, where, e.g., the long-distance dispersion interaction between excited- and ground-state atoms has been a subject of intense theoretical debate for nearly 50 years [4–13]. It has been predicted that at large interatomic separations, the magnitude of the interaction potential exhibits spatial oscillations [4,14]. In later works, it has been claimed that the potential monotonically decays as a function of interatomic separation [5,15]. However, the latter result seemed to be in contradiction with the long-distance interaction potential between an excited atom and metal or dielectric plate, which has been shown theoretically [16] and experimentally [17,18] to oscillate with the atom-plate distance.

The reason for the controversy is divergent energy denominators appearing in time-independent perturbation theory, which can be integrated by adding an infinitesimal imaginary part to the divergent denominators, with its sign determining whether the interaction potential oscillates with the distance or is monotonic. In conventional perturbation theory, there is no indication of the correct sign. However, using a dynamic theory with subsequent observation-time averaging, a third result for the interaction potential on the excited atom, which at a long-distance limit oscillates both in magnitude and sign, has been obtained for nonidentical atoms [8,9] and later generalized to the case of identical atoms using a quantum-electrodynamical approach [13].

Resolutions of the contradiction have been offered in a number of recent publications suggesting that both monotonic and oscillating behaviors are valid, but they describe different physical situations involving reversible and irreversible excitation exchange [10], or can appear in the same system, where the ground-state atom experiences the monotonic dispersion force, and the excited atom is subject to the oscillating force [11,12]. The latter implies the violation of the action-reaction theorem for the two atoms, but can be justified by taking into account photon emission by the excited atom [19].

Another potentially controversial nonequilibrium situation can occur in the system of two ground-state atoms out of equilibrium with an isotropic EM field, where the monotonic behavior of the van der Waals (vdW) force at large distances has been predicted [6,20–22], which seems to be in contrast to the oscillating force on a ground-state atom out of equilibrium with the EM field-dielectric plate system [6,23].

In this paper, we study the vdW interaction between two dissimilar atoms prepared in arbitrary initial states (ground or excited) out of equilibrium with the surrounding isotropic EM field and derive closed-form expressions for the energy shifts of each atom and related vdW forces using the Keldysh diagrammatic technique [24,25] formulated for few-body systems [26–28]. This method prescribes regularization rules of divergent energy denominators allowing us to avoid controversies associated with the standard time-independent perturbation theory [8]. We assume that the observation time is smaller than the lifetimes of the states of the atoms, implying that the atoms experience only virtual transitions, allowing us to apply a quasistationary version of the theory.

We found that in the long-distance regime, $R \gg \lambda$, where R is the interatomic distance and λ is a characteristic wavelength of atomic transitions, both atoms, in general, experience oscillating and monotonic components of the interaction potentials arising from resonance emission or absorption of virtual photons by one of the atoms, inducing spatial oscillations for its own potential and the monotonic component for the other atom. This implies unequal vdW potentials on each atom giving rise to nonreciprocity, which can be explained when a photon emitted by an excited atom or absorbed by a ground-state atom is taken into account restoring overall momentum balance similarly to what has been shown for a system of an excited-state atom and a ground-state one in vacuum [19]. In the latter case, as we show, the retarded vdW potential of the ground-state atom loses its oscillating component, while the vdW potential of the excited atom is purely oscillating in agreement with recent works [11,12].
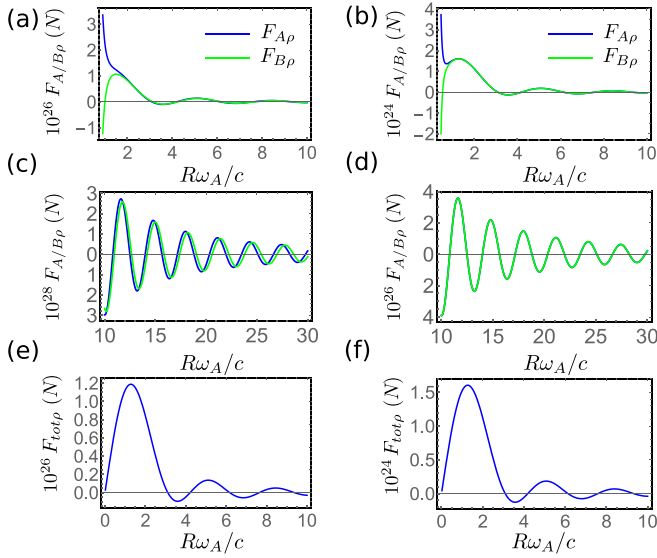
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[*]sherkunov@gmail.com

FIG. 1. Spatial oscillations of the vdW forces in the system of two ground-state atoms out of equilibrium with thermal EM field at $T = \omega_A$. (a) The vdW forces on atoms A, $\mathbf{F}_A$, and B, $\mathbf{F}_B$, projected onto the direction $\boldsymbol{\rho} = (\mathbf{R}_A - \mathbf{R}_B)/R$ as functions of interatomic separation R for the set of parameters corresponding to optical transitions $5\,^2S_{1/2} \to 5\,^2P_{3/2}$ for ^{87}Rb atoms (atom A) and $2\,^2S_{1/2} \to 2\,^2P_{3/2}$ for ^{40}K atoms (atom B) with optical transition frequencies $\omega_A = 1.59$ eV and $\omega_B = 1.61$ eV. (b) Same as (a), but for magnetically tuned transition frequencies $(\omega_A - \omega_B)/\omega_A = 10^{-4}$. (c)–(d) Long-distance behavior of $\mathbf{F}_{A\rho}$ and $\mathbf{F}_{B\rho}$ for the set of parameters of (a) and (b). (e)–(f) Net force on the two-atom system $\mathbf{F}_{\text{tot}} = (\mathbf{F}_A + \mathbf{F}_B)/2$ projected onto the direction $\boldsymbol{\rho}$ for the set of parameters of (a) and (b).

Then, we focus on the case of two ground-state atoms with close transition frequencies, ω_A and ω_B, surrounded by a thermal EM field, whose photon density does not change much within $\omega_A - \omega_B$. At small interatomic separation $R \ll \lambda$, the interaction is reciprocal, repulsive, and nonresonant, and the vdW forces decay as R^{-7}, as shown in Figs. 1(a) and 1(b). However, at large separations, $R \gg \lambda$, the system of two atoms becomes nonreciprocal and the vdW forces on each atom are codirectional, almost equal, and resonant. They decay as R^{-2} and oscillate with R almost in-phase [see Figs. 1(c) and 1(d)] giving rise to the sizable oscillating net force and negligible interatomic force. The former reaches its maximum in the intermediate regime $R \sim \lambda$ [see Figs. 1(e) and 1(f)], when the forces on each atom become codirectional and almost equal.

As an example, we numerically calculate the vdW forces in the system of ^{87}Rb and ^{40}K ground-state atoms out of equilibrium with thermal EM field at temperature close to the dominant transition frequencies of the atoms with and without magnetic field inducing Zeeman splitting. We find that the magnitude of the net force on the atomic system can be within experimentally available values.

The vdW forces discussed in this paper can also be induced using artificially created fluctuating light fields [29]. We found that the vdW forces not only can be dramatically enhanced, but also controlled and manipulated by applying light fields with tailored spectral properties. As we show in Fig. 2, in the short-distance regime, the interaction becomes resonantly
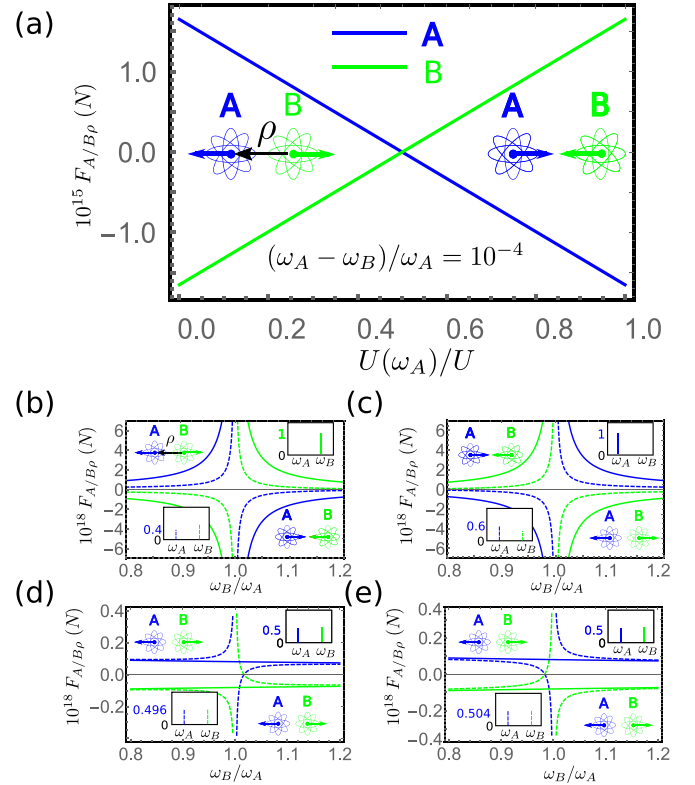

FIG. 2. Enhancement of the short-distance van der Waals forces on two ground-state atoms by artificially created random light for magnetically tuned optical transitions $5\,^2S_{1/2} \to 5\,^2P_{3/2}$ of the ^{87}Rb atom (atom A) and $2\,^2S_{1/2} \to 2\,^2P_{3/2}$ of ^{40}K atom (atom B). The atoms separated by a distance $R = 0.3\lambda$ are out of equilibrium with artificial random light with narrow spectral energy densities peaked at ω_A and ω_B (see insets). (a) The vdW forces as functions of photon energy density $U(\omega_A)/U$, where $U = U(\omega_A) + U(\omega_B)$, for the transition frequencies $(\omega_A - \omega_B)/\omega_A = 10^{-4}$. (b)–(e) The vdW forces as functions of ω_B/ω_A for a set of photon energy densities shown in insets. Insets: Photon energy densities, $U(\omega_A)$ and $U(\omega_B)$, normalized to $U = 0.6 \times 10^{-3}$ J/m^3.

enhanced provided the energy densities of the external EM field, $U(\omega_A)$ and $U(\omega_B)$, at frequencies ω_A and ω_B are not equal. Adjusting the ratio $U(\omega_A)/U(\omega_B)$ would allow one not only to control the magnitudes of the vdW forces, which scale linearly with $U(\omega_A)$ and $U(\omega_B)$, but also to change their direction, switching the interaction from repulsive to attractive. In the large-distance regime, adjusting the spectral densities $U(\omega_A)$ and $U(\omega_B)$ would allow one to control the amplitudes of the oscillating components of the vdW forces on each atom, and even make the interaction monotonic, as shown in Fig. 3. It would also allow us to control the net force on the system, provided the transition frequencies ω_A and ω_B are not too close.

II. MODEL

We consider two dissimilar two-level atoms, A and B, characterized by resonance transition frequencies ω_A and ω_B and line widths γ_A and γ_B, such that $|\omega_A - \omega_B| \gg \gamma_A, \gamma_B$, located at positions $\mathbf{R}_A$ and $\mathbf{R}_B$ and interacting with isotropic
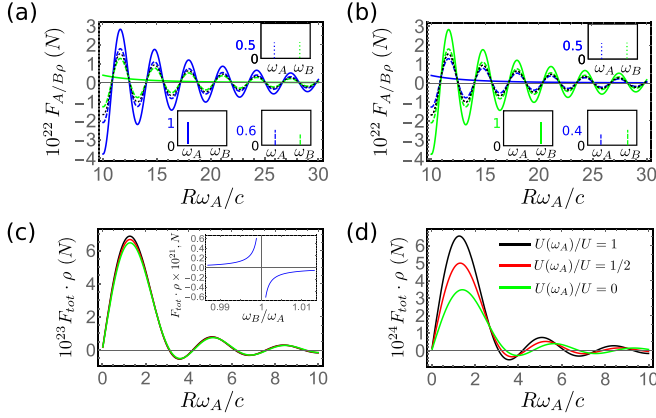
FIG. 3. (a)–(b) Long-distance van der Waals forces on atoms A and B (see caption of Fig. 2) as functions of interatomic separation R for $(\omega_A - \omega_B)/\omega_A = 10^{-4}$ and $U(\omega_A)/U$ $[U(\omega_B)/U]$ shown in insets. (c) Net force on the system of the two atoms as a function of R for $(\omega_A - \omega_B)/\omega_A = 10^{-2}$ for the set of photon energy densities shown in (d). Inset: Net force as a function of ω_B/ω_A. (d) Same as (c), but for $(\omega_A - \omega_B)/\omega_A = 10^{-1}$.

and unpolarized EM field modeled, in dipole approximation, by the Hamiltonian ($\hbar = c = 1$)

$$H_{\text{int}} = - \sum_{j=A,B} \int \psi_j^{\dagger}(\mathbf{r})\mathbf{d} \cdot \mathbf{E}(\mathbf{r})\psi_j(\mathbf{r})d^3\mathbf{r}. \tag{1}$$

Here, $\psi_j(\mathbf{r}) = \sum_{i=g,e} \phi_i^j(\mathbf{r} - \mathbf{R}_j)b_{ji}$ is the field operator of atom j,

$$\mathbf{E}(\mathbf{r}) = i \sum_{\mathbf{k}\mu} \sqrt{\frac{2\pi|\mathbf{k}|}{V}} \mathbf{e}_{\mathbf{k}\mu}(\alpha_{\mathbf{k}\mu}e^{i\mathbf{k}\mathbf{r}} - \alpha_{\mathbf{k}\mu}^{\dagger}e^{-i\mathbf{k}\mathbf{r}}) \tag{2}$$

is the electric field operator, and $\mathbf{d} = e\mathbf{r}$ is the operator of dipole moment, where b_{ji} is the annihilation operator of the ground ($i = g$) or excited ($i = e$) state of atom j described by the wave function $\phi_i^j(\mathbf{r} - \mathbf{R}_j)$, $\alpha_{\mathbf{k}\mu}$ is the annihilation operator of a photon with momentum $\mathbf{k}$ and polarization index μ, $\mathbf{e}_{\mathbf{k}\mu}$ is the unit polarization vector, and V is the quantization volume.

At the initial time t_0, the atoms are prepared in their initial states i with probabilities p_i^A and p_i^B and are out of equilibrium with the EM field. We assume that within the observation time $1/|\omega_A - \omega_B| \ll t_f < \gamma_A^{-1}, \gamma_B^{-1}$, the atoms stay in their initial states and do not equilibrate with the EM field, allowing us to match the vdW potentials with the energy shifts of the initial states of each atom [11], and calculate them from the density matrices of atom j,

$$\rho_j(\mathbf{r},t;\mathbf{r}',t') = \text{Tr}[\hat{\psi}_j^{\dagger}(\mathbf{r}',t')\hat{\psi}_j(\mathbf{r},t)], \tag{3}$$

where $\hat{\psi}$ is in the Heisenberg picture, using the adiabatic hypothesis [25], with the help of the Keldysh Green's function method [25–28].

III. VAN DER WAALS POTENTIAL OF AN ATOM IN A GENERIC SURROUNDING

First we consider a more general situation when atom A is prepared in an arbitrary state and surrounded by EM field

and/or arbitrary magneto-dielectric bodies and calculate its energy shifts. As shown in Refs. [26,27], the density matrix of atom A is given by the following equation, provided the atom does not change its initial state, i.e., the condition $|\omega_A - \omega_B| \gg \gamma_A, \gamma_B$ is fulfilled (see Appendix A):

$$\rho_A(X,X') = \rho_A^0(X,X')e^{-i\langle M_{11}^A\rangle(t-t_0)}e^{i\langle M_{22}^A\rangle(t'-t_0)}, \tag{4}$$

where we use $X = \{\mathbf{r},t\}$. Here

$$\rho_A^0(X,X') = \phi_i^A(\mathbf{r} - \mathbf{R}_A)\phi_i^{A*}(\mathbf{r}' - \mathbf{R}_A)e^{-i\epsilon_i^A(t-t')} \tag{5}$$

is the density matrix of noninteracting atom A in state i with bare energy ϵ_i^A and $M_{11}^A = M_{22}^{A*}$ is the self-energy of atom A,

$$\langle M_{11}^A\rangle = \int d^3\mathbf{r}d^3\mathbf{r}' \phi_i^A(\mathbf{r} - \mathbf{R}_A)$$
$$\times M_{11}^A(\mathbf{r},\mathbf{r}',\epsilon_i^A)\phi_i^{A*}(\mathbf{r}' - \mathbf{R}_A), \tag{6}$$

with

$$M_{11}^A(X,X') = i \sum_{\nu\nu'} g_{A11}^0(X,X')d^\nu d^{\nu'} D_{11}^{\nu\nu'}(X',X), \tag{7}$$

expressed in terms of the atomic propagator g_{A11}^0 and causal photonic Green's tensor $D_{11}^{\nu\nu'}$. The former is defined in terms of vacuum averages $g_{A11}^0(X,X') = -i\langle \hat{T}\psi_j(X)\psi_j^{\dagger}(X')\rangle_{\text{vac}}$ and, in the energy domain, takes the form

$$g_{A11}^0(\mathbf{r},\mathbf{r}',E) = \sum_{i=g,e} \frac{\phi_i^A(\mathbf{r} - \mathbf{R}_A)\phi_i^{A*}(\mathbf{r} - \mathbf{R}_A)}{(E - \epsilon_i^A + i0)}. \tag{8}$$

The latter is defined as

$$D_{11}^{\nu\nu'}(X,X') = -i\langle \hat{T}\hat{E}^\nu(X)\hat{E}^{\nu'}(X')\rangle, \tag{9}$$

where $\hat{T}$ is the time-ordering operator, and $\nu = \{x,y,z\}$.

As it follows from Eq. (4), the energy shift of atom A induced by the EM field and, thus, the corresponding vdW potential is determined by the real part of the self-energy, $U_A = \Delta\epsilon_i^A = \text{Re}\langle M_{11}^A\rangle$, while the corresponding line width is equal to its imaginary part $\gamma_i^A = \text{Im}\langle M_{11}^A\rangle$.

Using Eqs. (5), (4), and (7), we find the vdW potential experienced by atom A,

$$U_A = \text{Re}\left[\frac{i}{2\pi}\int_{-\infty}^{\infty} \frac{d_A^\nu d_A^{\nu'} D_{11}^{\nu'\nu}(\omega,\mathbf{R}_A,\mathbf{R}_A)}{\omega - \epsilon_{\bar{i}}^A + \epsilon_i^A + i0} d\omega\right], \tag{10}$$

where $\bar{i}$ stands for the state of atom A, opposite to i, and d_A is the transition matrix element of the dipole moment. We use a property of the photon Green's tensor, $D_{11}^{\nu\nu'}(\mathbf{r},\mathbf{r}', -\omega) = D_{11}^{\nu'\nu}(\mathbf{r}',\mathbf{r},\omega)$, which follows from its definition (9) [27] and rewrite Eq. (10) as $U_A = -\text{Re}[\frac{i}{2\pi}\int_0^{\infty} \tilde{\alpha}_{Ai}^{\nu\nu'}(\omega)D_{11}^{\nu'\nu}(\omega,\mathbf{R}_A,\mathbf{R}_A)d\omega]$, where $\tilde{\alpha}_{Ag/e}(\omega) = d_A^\nu d_A^{\nu'}(\frac{1}{\pm\omega_A-\omega-i0} + \frac{1}{\pm\omega_A+\omega-i0})$ is related to the polarizability of atom A,

$$\alpha_{Ag/e}(\omega) = \left(\frac{d_A^\nu d_A^{\nu'}}{\pm\omega_A - \omega - i0} + \frac{d_A^\nu d_A^{\nu'}}{\pm\omega_A + \omega + i0}\right), \tag{11}$$

as $\tilde{\alpha}_{Ag/e}(\omega) = \alpha_{Ag/e}(\omega) + 2\pi i d_A^\nu d_A^{\nu'}\delta(\omega \pm \omega_A)$. Note that for $\omega > 0$, the difference between $\tilde{\alpha}$ and α is significant only if

atom A is in its excited state, leading to

$$U_A = -\operatorname{Re}\left[\frac{i}{2\pi}\int_0^\infty \alpha_{Ai}^{\nu\nu'}(\omega)D_{11}^{\nu'\nu}(\omega,\mathbf{R}_A,\mathbf{R}_A)d\omega\right]$$
$$+ \operatorname{Re}\left[d_A^\nu d_A^{\nu'} D_{11}^{\nu'\nu}(\omega_A,\mathbf{R}_A,\mathbf{R}_A)p_e^A\right]. \tag{12}$$

Equation (12) represents the general formula describing the interaction of a two-level atom prepared in an arbitrary state, excited or ground, with the EM field described by Green's function D_{11}, provided that the observation time is small compared to the lifetime of the atom's initial state.

To check the result, we calculate the Casimir-Polder force experienced by atom A prepared in an arbitrary state, ground or excited, positioned near a dispersive and absorbing medium. We suppose that the medium is kept at temperature T and is at thermal equilibrium with the electromagnetic field. For the initial stage of the atom-field interaction, $t < \gamma_A^{-1}$, the atom does not change its initial state, and the interaction potential experienced by the atom can be evaluated with the help of Eq. (12) with $D_{11} = D_r - i\rho_{\mathrm{ph}}$ (see Appendix B) and the photonic density matrix, ρ_{ph}, given by the fluctuation-dissipation theorem ($\omega > 0$) [30]:

$$\rho_{\mathrm{ph}}^{\nu\nu'}(\omega,\mathbf{r},\mathbf{r}') = -2N(\omega)\operatorname{Im}D_r^{\nu\nu'}(\omega,\mathbf{r},\mathbf{r}'), \tag{13}$$

where $N(\omega) = (e^{\omega/T} - 1)^{-1}$ is the average number of photons with frequency ω, yielding the result of Ref [23]:

$$U_i^A = U_{\mathrm{eq}} + U_{\mathrm{neq}},$$
$$U_{\mathrm{eq}} = -\operatorname{Re}\left[\frac{i}{2\pi}\int_0^\infty [2N(\omega)+1]\alpha_{Ai}^{\nu\nu'}(\omega)\right.$$
$$\left.\times D_r^{\nu'\nu}(\omega,\mathbf{R}_A,\mathbf{R}_A)d\omega\right],$$
$$U_{\mathrm{neq}} = -\operatorname{Re}[d_A^\nu d_A^{\nu'} D_r^{\nu'\nu}(\omega_A,\mathbf{R}_A,\mathbf{R}_A)$$
$$\times \{N(\omega_A)p_g^A - [N(\omega_A)+1]p_e^A\}], \tag{14}$$

where the equilibrium potential describing the Casimir-Polder interaction of a thermalized atom can be evaluated as $U_{\mathrm{eq}} = T\sum_{m=0}^\infty (1 - \frac{1}{2}\delta_{m0})\alpha_A^{\nu'\nu}(i\xi_m)D_r^{\nu\nu'}(i\xi_m)$, with $\xi_m = 2\pi mT$ the Matsubara frequency. Here, we used the property of the polarizability, $\operatorname{Re}[\frac{i}{2\pi}\alpha_A(\omega)] = \delta(\omega - \omega_A)(p_e^A - p_g^A)/2$, which follows from its definition (11).

IV. VAN DER WAALS INTERACTION BETWEEN TWO ATOMS SURROUNDED BY ISOTOPIC EM FIELD

A. General case

Now we consider the interaction between two atoms, A and B, prepared in arbitrary states and embedded in the isotopic EM field. We assume that the optical Stark shift induces by the free EM field as well as the Lamb shift due to free EM vacuum fluctuations are taken into account in the atomic transition frequencies and suppose, without loss of generality, averaging over all possible directions of dipole matrix elements, so that $d_{A/B}^\nu d_{A/B}^{\nu'} = \delta_{\nu\nu'}|d_{A/B}|^2/3$, where δ is the Kronecker symbol. The interaction potential on the atoms is given by Eq. (12) with the scattering part of the photon Green's function D_{11},

satisfying the equation (see Appendix B):

$$D_{11}^{\nu\nu'}(\omega,\mathbf{R}_A,\mathbf{R}_A) = -[2N(\omega)+1]\alpha_B^{\nu_1\nu_2}(\omega)D_r^{0\nu\nu_1}(\omega,\mathbf{R}_A,\mathbf{R}_B)$$
$$\times D_r^{0\nu_2\nu'}(\omega,\mathbf{R}_B,\mathbf{R}_A) + 2N(\omega)\operatorname{Re}$$
$$\times \left[\alpha_B^{\nu_1\nu_2}(\omega)D_r^{0\nu\nu_1}(\omega,\mathbf{R}_A,\mathbf{R}_B)\right.$$
$$\left.\times D_r^{0\nu_2\nu'}(\omega,\mathbf{R}_B,\mathbf{R}_A)\right] + 2iN(\omega)\operatorname{Im}$$
$$\times [\alpha_B^{\nu_1\nu_2}(\omega)]D_r^{0\nu\nu_1}(\omega,\mathbf{R}_A,\mathbf{R}_B)$$
$$\times \left[D_r^{0\nu_2\nu'}(\omega,\mathbf{R}_B,\mathbf{R}_A)\right]^*$$
$$-2\operatorname{Im}\left[\alpha_B^{\nu_1\nu_2}(\omega)\right]p_e^B D_r^{0\nu\nu_1}(\omega,\mathbf{R}_A,\mathbf{R}_B)$$
$$\times \left[D_r^{0\nu_1\nu'}(\omega,\mathbf{R}_B,\mathbf{R}_A)\right]^*. \tag{15}$$

Under these assumptions, with the help of Eqs. (15) and (12), we find that apart from the usual equilibrium potential rapidly decaying with interatomic separation [2,3,31]

$$U_A^{\mathrm{eq}} = \operatorname{Re}[\frac{i}{\pi}\int_0^\infty d\omega[N(\omega)+1/2]\alpha_A(\omega)\alpha_B(\omega)$$
$$\times \left[D_r^0(\omega,\mathbf{R}_A - \mathbf{R}_B)\right]^2 \tag{16}$$

describing the interaction between atoms thermalized with EM field, atom A experiences the nonequilibrium resonant potential

$$U_A^{\mathrm{neq}} = \frac{2|d_A|^2|d_B|^2}{9(\omega_A^2 - \omega_B^2)}\left(\omega_A\{N(\omega_B)p_g^B - [N(\omega_B)+1]p_e^B\}\right.$$
$$\times \left|D_r^0(\omega_B,\mathbf{R}_A - \mathbf{R}_B)\right|^2 - \omega_B\{N(\omega_A)p_g^A$$
$$- [N(\omega_A)+1]p_e^A\}$$
$$\left.\times \operatorname{Re}\left\{\left[D_r^0(\omega_A,\mathbf{R}_A - \mathbf{R}_B)\right]^2\right\}\right), \tag{17}$$

disappearing with the equilibration between the atoms and the EM field. Indeed, assuming that the EM field is thermal, i.e., obeys Bose-Einstein distribution, and the probabilities to find each atom in a specific state are described by Boltzmann distribution, $p_g^j = e^{\omega_j/T}(e^{\omega_j/T}+1)^{-1}$ and $p_e^j = p_g^j e^{-\omega_j/T}$, the nonequilibrium potential vanishes.

Using the same procedure, we find that the nonequilibrium vdW potential for atom B,

$$U_B^{\mathrm{neq}} = -\frac{2|d_A|^2|d_B|^2}{9(\omega_A^2 - \omega_B^2)}\left(\omega_B\{N(\omega_A)p_g^A - [N(\omega_A)+1]p_e^A\}\right.$$
$$\times \left|D_r^0(\omega_A,\mathbf{R}_B - \mathbf{R}_A)\right|^2 - \omega_A\{N(\omega_B)p_g^B$$
$$- [N(\omega_B)+1]p_e^B\}$$
$$\left.\times \operatorname{Re}\left\{\left[D_r^0(\omega_B,\mathbf{R}_B - \mathbf{R}_A)\right]^2\right\}\right). \tag{18}$$

is not, in general, equal to U_A^{neq}. Moreover, in the long-distance regime, $R \gg \lambda$, they both contain oscillating and monotonic in R terms, which can be seen by substituting $\operatorname{Re}[(D_r^0)^2]$ and $|D_r^0|^2$ [30],

$$|D_r^0(\omega,R)|^2 = \frac{2\omega^4}{R^2}[1 + 1/(\omega R)^2 + 3/(\omega R)^4], \tag{19}$$

$$\operatorname{Re}\left\{\left[D_r^0(\omega,R)\right]^2\right\} = \frac{2\omega^4}{R^2}\{\cos(2\omega R)$$

$$\times [1 - 5/(\omega R)^2 + 3/(\omega R)^4]$$
$$+ \sin(2\omega R)[3/(\omega R)^3 - 1/(\omega R)]\}, \quad (20)$$

into Eqs. (17) and (18),

$$U_A^{\text{neq}} = \frac{4|d_A|^2|d_B|^2\omega_A\omega_B}{9R^2(\omega_A^2 - \omega_B^2)} \big(\omega_B^3 \{ N(\omega_B)p_g^B - [N(\omega_B) + 1]p_e^B \}$$
$$- \omega_A^3 \{ N(\omega_A)p_g^A - [N(\omega_A) + 1]p_e^A \} \cos(2\omega_A R) \big),$$
$$(21)$$

$$U_B^{\text{neq}} = -\frac{4|d_A|^2|d_B|^2\omega_A\omega_B}{9R^2(\omega_A^2 - \omega_B^2)} \big(\omega_A^3 \{ N(\omega_A)p_g^A - [N(\omega_A) + 1]p_e^A \}$$
$$- \omega_B^3 \{ N(\omega_B)p_g^B - [N(\omega_B) + 1]p_e^B \} \cos(2\omega_B R) \big). \quad (22)$$

The origin of these components depends on which atom takes part in the resonance processes: for the potential on atom A, the oscillations are due to *its* spontaneous (stimulated) emission of virtual quanta or *its* resonant absorption of an external photon, while the monotonic component is due to the resonance processes involving *atom B* and vice versa. However, in the short-distance regime, $R \ll \lambda$, the oscillations disappear and Eqs. (17) and (18) give us monotonic and equal vdW potentials:

$$U_{A/B}^{\text{neq}} = \frac{4|d_A|^2|d_B|^2}{3R^6(\omega_A^2 - \omega_B^2)} \big(\{ \omega_A N(\omega_B)p_g^B - [N(\omega_B) + 1]p_e^B \}$$
$$- \omega_B \{ N(\omega_A)p_g^A - [N(\omega_A) + 1]p_e^A \} \big). \quad (23)$$

If atom A is excited and atom B is in its ground state and the external EM field is absent, the long-distance vdW potential of the excited atom in Eq. (21) exhibits spatial oscillations both in sign and magnitude supporting the results of Ref. [9] and the one of the ground-state atom in Eq. (22) is monotonic in agreement with Refs [11,12]. In this case, the asymmetry leading to nonreciprocal vdW forces violating the action-reaction theorem has been attributed to a net transfer of linear momentum to the quantum fluctuations of the EM field due to spontaneous emission by the excited atom [19].

B. Two ground-state atoms in isotopic EM field

Next, we consider two ground-state atoms out of equilibrium with the external EM field. For the short-distance regime $R \ll \lambda$ the nonequilibrium vdW potentials can be found from Eqs. (23):

$$U_{A/b}^{\text{neq}} = \frac{4|d_A|^2|d_B|^2[\omega_A N(\omega_B) - \omega_B N(\omega_A)]}{3R^6(\omega_A^2 - \omega_B^2)}, \quad (24)$$

and can be related to the field-assisted vdW forces acting along the direction $\boldsymbol{\rho} = (\mathbf{R}_A - \mathbf{R}_B)/R$, $\mathbf{F}_A = -\nabla_A U_A^{\text{neq}}$ and $\mathbf{F}_B = -\nabla_B U_B^{\text{neq}}$,

$$\mathbf{F}_A = -\mathbf{F}_B$$
$$= \frac{8|d_A|^2|d_B|^2[\omega_A N(\omega_B) - \omega_B N(\omega_A)]\boldsymbol{\rho}}{R^7(\omega_A^2 - \omega_B^2)}. \quad (25)$$

In the large distance regime, $R \gg \lambda$, we find

$$U_A^{\text{neq}} = \frac{4|d_A|^2|d_B|^2\omega_A\omega_B}{9R^2(\omega_A^2 - \omega_B^2)}$$
$$\times [\omega_B^3 N(\omega_B) - \omega_A^3 N(\omega_A)\cos(2\omega_A R)], \quad (26)$$

$$U_B^{\text{neq}} = -\frac{4|d_A|^2|d_B|^2\omega_A\omega_B}{9R^2(\omega_A^2 - \omega_B^2)}$$
$$\times [\omega_A^3 N(\omega_A) - \omega_B^3 N(\omega_B)\cos(2\omega_B R)], \quad (27)$$

which leads to

$$\mathbf{F}_A = -\frac{8|d_A|^2|d_B|^2 N(\omega_A)\omega_A^5\omega_B\boldsymbol{\rho}}{9R^2(\omega_A^2 - \omega_B^2)}\sin(2\omega_A R), \quad (28)$$

$$\mathbf{F}_B = -\frac{8|d_A|^2|d_B|^2 N(\omega_B)\omega_A\omega_B^5\boldsymbol{\rho}}{9R^2(\omega_A^2 - \omega_B^2)}\sin(2\omega_B R). \quad (29)$$

C. Two ground-state atoms in thermal EM field

In the case of a thermal EM field at temperature $T \gg |\omega_A - \omega_B|$, so that $N = N(\omega_A) \approx N(\omega_B)$, the short-distance forces described by Eq. (25) are repulsive, have equal magnitudes, and are nonresonant [see Figs. 1(a) and 1(b)]. Consequently, the net force is absent. However, in the large-distance case, the forces given by Eqs. (28) and (29) are resonant, have the same direction and amplitude, and show spatial oscillations almost in-phase [see Figs. 1(c) and 1(d)], giving rise to a spatially oscillating net force $\mathbf{F}_{\text{tot}} = (\mathbf{F}_A + \mathbf{F}_B)/2$, as shown in Figs. 1(e) and 1(f). In the intermediate regime $R \sim \lambda$, in which the interaction crosses over from mutual monotonic repulsion to spatial oscillations, the net force reaches its maximum with its direction toward the atom with smaller transition frequency, as we show in Figs. 1(e) and 1(f). At the same time, the vdW forces on each atom become almost equal in direction and magnitude. Note that in the long-distance regime, the equilibrium contribution to the field-assisted vdW force, $\mathbf{F}_A = -\mathbf{F}_B = -\frac{4T|d_A|^2|d_B|^2}{\omega_A\omega_B R^7}\boldsymbol{\rho}$ [30], can be neglected.

As an example, we consider a system of ^{87}Rb and ^{40}K atoms prepared in $5\,^2S_{1/2}$ and $2\,^2S_{1/2}$ ground states, respectively, out of equilibrium with the thermal EM field at temperatures comparable with the quasiresonant transition energies for $5\,^2S_{1/2} \rightarrow 5\,^2P_{3/2}$ of the ^{87}Rb atoms, $\omega_A = 1.59$ eV, and $2\,^2S_{1/2} \rightarrow 2\,^2P_{3/2}$ of the ^{40}K atoms, $\omega_B = 1.61$ eV, and we calculate the net vdW force numerically [Fig. 1(e)]. However, the magnitude of the net force appears to be too small to be detected experimentally. Applying an external magnetic field would result in Zeeman shifts of the atomic energy levels allowing one to tune the transition frequencies and enhance the resonant net force. For the relative detuning $\delta\omega = |\omega_A - \omega_B|/\omega_A = 10^{-4}$ limited by the Doppler broadening $\Delta\omega \approx 10^{-5}\omega_A$, we found that the maximum value of the net force $F_{\text{tot}}^{\text{max}} \approx 10^{-23}N$ [see Fig. 1(f)], which is within experimentally achievable values [32].

D. Two ground-state atoms in artificial random EM field

The forces discussed in this paper can be induced not only by a thermal EM field, but also by using artificially created random isotopic light in a small cavity [29]. This would allow us to not only enhance the vdW forces compared with the thermal light, but also control and manipulate their direction and magnitude. To demonstrate this point, we consider two ground-state atoms in a small cavity filled with random light characterized by energy density, $U(\omega)$, peaked at ω_A and ω_B (see insets of Fig. 2). We calculate the vdW forces on the atoms numerically, for a cavity of volume $V = (50 \,\mu\text{m})^3$ and

the light generated by a laser with the power $P = 0.5$ mW corresponding to the total energy density of random light in the cavity $U = U(\omega_A) + U(\omega_B) \approx 6 \times 10^{-4}$ J/m^3 [29]. As in the previous example, we choose a system of ^{87}Rb and ^{40}K atoms prepared in $5\,^2S_{1/2}$ and $2\,^2S_{1/2}$ respectively.

At small interatomic separations, $R \ll \lambda$, the vdW forces on each atom are equal in magnitude; however, their direction depends on the ratios of $U(\omega_A)/U(\omega_B)$ and ω_A/ω_B as shown in Fig. 2. For $U(\omega_A) = 0$, when all the photonic energy density is concentrated at the frequency ω_B, the interaction is repulsive provided $\omega_A > \omega_B$ and attractive for $\omega_A < \omega_B$ [see Figs. 2(a)–2(c)]. As the ratio $U(\omega_A)/U(\omega_B)$ increases, the magnitude of the forces decreases linearly taking their minimum at $U(\omega_A) = U(\omega_B)$. Further increase of $U(\omega_A)/U(\omega_B)$ is accompanied by the linear increase of the magnitudes of the forces; however, the interaction becomes attractive for $\omega_A > \omega_B$ and repulsive otherwise. For $U(\omega_A) \neq U(\omega_B)$, the interaction demonstrates resonance behavior at $\omega_A \approx \omega_B$, as shown in Figs. 2(b) and 2(c); however, as $U(\omega_A)$ approaches $U(\omega_B)$, the forces become repulsive independently of the ratio ω_A/ω_B and nonresonant, as shown in Figs. 2(d) and 2(e), in agreement with Eq. (25). Note that the amplitudes of the forces induced by artificial random light can be up to nine orders of magnitude greater than the ones induced by thermal light discussed above.

At large interatomic separations, $R \gg \lambda$, the vdW forces on each atom are resonant, have the same direction, and oscillate with the interatomic separation almost in phase; however, their amplitudes depend on the ratio $U(\omega_A)/U(\omega_B)$, as shown in Figs. 3(a) and 3(b). At $U(\omega_B) = 0$, the force on atom B loses its oscillating component and drops with the interatomic distance as R^{-3}, in agreement with [6], while the oscillation amplitude of the force on atom A takes its maximum value [see Fig. 3(a)]. As the ratio $U(\omega_B)/U(\omega_A)$ increases, the oscillation amplitude of atom A decreases, while it increases for atom B to equalize at $U(\omega_A) = U(\omega_B)$, in agreement with Eqs. (28) and (29). Further increase in $U(\omega_B)/U(\omega_A)$ leads to the decrease of the oscillation component of atom A, which disappears at $U(\omega_A) = 0$, as shown in Fig. 3(b). Again, as in the case of the thermal EM field, the artificial random radiation generates a net spatially oscillating force on the system of two atoms, which takes its maximum at $R \sim \lambda$. However, in the vicinity of the resonance $\omega_A = \omega_B$ [see inset of Fig. 3(c)], the net force is determined by the total energy density $U = U(\omega_A) + U(\omega_B)$, but not by $U(\omega_A)$ and $U(\omega_B)$, as shown in Fig. 3(c). Thus, to control the net force, one has to detune from the resonance, as we demonstrate in Fig. 3(d).

V. CONCLUSIONS

Finally, we comment on the disagreement with previously found monotonic long-distance vdW potential between atoms out of equilibrium with an EM field [6,20–22] where the interaction potential was *a priori* assumed equal for each atom and interpolated from the calculations for the atom with vanishing absorption rate. However, as we show in this work, this procedure is not sufficient if the absorption rates of both atoms are not small.

In this paper, we presented a formula for the vdW potential in the system of an atom surrounded by arbitrary magneto-

dielectric bodies and an isotropic EM field. We applied this formula to the case of two atoms prepared in arbitrary states out of equilibrium with EM field. We found, that in the long-distance regime, the vdW potentials have both monotonic and oscillating behavior with interatomic distance and, in general, unequal for each atom resulting in the net resonant spatially oscillating force. We suggest that the vdW forces can be controlled and manipulated with the help of artificially created random light with tailored spectral properties. In the particular case of a system with an excited-state atom and a ground-state one in an EM vacuum, our results are in agreement with the recent findings reported in Refs. [11,12,19].

APPENDIX A: DERIVATION OF EQ. (4)

In the interaction picture, the Keldysh Green's functions for atom j,

$$G_{ll'}^{j}(X,X') = -i \langle T_c \psi_{jl}(X) \psi_{jl'}^{\dagger}(X') S_c(t_f,t_0) \rangle, \quad \text{(A1)}$$

and EM field,

$$D_{ll'}^{\nu\nu'}(X,X') = -i \langle T_c E_l^{\nu}(X) E_{l'}^{\nu'}(X') S_c(t_f,t_0) \rangle, \quad \text{(A2)}$$

where $X = \{\mathbf{r},t\}$ and $\nu = x,y,z$ describes the projection on the corresponding axis, are defined on the Keldysh contour, which goes in time from $-\infty$ to ∞ for $l = 1$ and from ∞ to $-\infty$ for $l = 2$ determining the (anti-)chronological ordering T_c [25,27]. The time-evolution operator, $S_c(t_f,t_0) = T_c \exp[i \sum_{l=1,2} (-1)^l \int_{t_0}^{t_f} dt\, H_{\text{int},l}(t)]$, can be expanded in H_{int} enabling one to construct the perturbation series for the density matrix of atom j, $\rho_j = iG_{12}^j$. Applying the exact Wick's theorem to the atomic operators [26,27] $T_c \psi_{jl}(X) \psi_{jl'}^{\dagger}(X') =: \psi_{jl}(X) \psi_{jl'}^{\dagger}(X') : + i g_{j,ll'}^0(X,X')$, where $: \cdots :$ means normal ordering and the atomic propagator is determined in terms of vacuum average $g_{jll'}^0(X,X') = -i \langle T_c \psi_{jl}(X) \psi_{jl'}^{\dagger}(X') \rangle_{\text{vac}}$, we find the perturbation series, as shown in Fig. 4(a), where the first Feynman diagram describes noninteracting atom j, the second and third diagrams correspond to the elastic scattering of EM field on atom j, and the fourth diagram describes spontaneous emission or resonant absorption of a photon. Under the condition $t \ll \gamma_j^{-1}$, we can neglect the fourth term.

Summing up all relevant reducible bubble diagrams giving rise to atom-EM field interactions, we arrive at the density matrix of atom j described by the Feynman diagrams depicted in Fig. 4(b) [26,27]:

$$\rho_j(X,X') = \rho_j^0(X,X')$$

$$+ \int dX_1 dX_2 \rho_j^0(X,X_1) M_{22}^j(X_1,X_2) g_{j22}(X_2,X')$$

$$+ \int dX_1 dX_2 g_{j11} M_{11}^j \rho_j^0$$

$$+ \int dX_1 dX_2 dX_3 dX_4 g_{j11} M_{11}^j \rho_j^0 M_{22}^j g_{j22},$$

$$g_{jll'} = g_{jll'}^0 + \sum_{l_1,l_2} \int dX_1 dX_2 g_{jll_1}^0 M_{l_1l_2}^j g_{jl_2l'}, \quad \text{(A3)}$$

where we omit obvious arguments and ρ^0 and $M_{11}^j = M_{11}^{j*}$ are given by Eqs. (5) and (7), respectively. Keeping in mind that
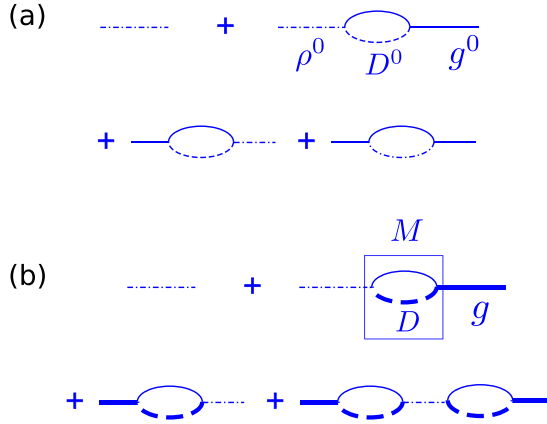
FIG. 4. (a) Feynman diagrams describing the dressing of the density matrix of atom j, ρ_j^0 (dashed-dotted line) by electromagnetic field (first three diagrams) and field-induced transition (last diagram) up to the second-order perturbation theory. The bare propagator of atom j, $g_{jll'}^0$, is shown as a thin solid line and the bare photon Green's tensor $D_{ll'}^0$ is shown as a thin dashed line. The summation over Keldysh indices $l = 1,2$ is assumed in each vertex. (b) Summation of Feynman diagrams for the density matrix. Thick lines correspond to the total Green's functions.

atom j does not change its initial state i during the interaction with the EM field, we factorize the density matrix

$$\rho_j(X,X') = \phi_i^j(\mathbf{r}-\mathbf{R}_A)f(t)\phi_i^{j*}(\mathbf{r}'-\mathbf{R}_A)f^*(t') \quad (A4)$$

in terms of the wave functions of noninteracting atom j, where $f(t)$ obeys the equation [26,27]

$$i\frac{\partial f(t)}{\partial t} - \epsilon_i^j f(t) = \int_{t_0}^{\infty} dt' \langle M_{11}^j(t,t')\rangle f(t'), \quad (A5)$$

and $\langle M_{11}^j(t,t')\rangle$ is given by Eq. (6). Equation (A5) can be solved in the pole approximation,

$$f(t) = e^{-i\epsilon_i^j t}\, e^{-i\langle M_{11}^j(\epsilon_i^j)\rangle(t-t_0)}, \quad (A6)$$

which, along with Eq. (A4), yields Eq. (4).

APPENDIX B: DERIVATION OF EQ. (15)

In the presence of atom B, the photon Green's functions (A2) can be calculated in the lowest orders of perturbation theory [26,27],

$$D_{ll'}^{\nu\nu'}(X,X') = D_{ll'}^{0\nu\nu'}(X,X') + \int dX_1 dX_2 D_{ll_1}^{0\nu\nu_1}(X,X_1)$$
$$\times \Pi_{Bl_1l_2}^{\nu_1\nu_2}(X_1,X_2)D_{0l_2l'}^{\nu_2\nu'}(X_2,X'), \quad (B1)$$

with the polarization operators

$$\Pi_{Bll'}^{\nu\nu'}(X_1,X_2) = (-1)^{l+l'}d^\nu d^{\nu'}\big[g_{Bll'}^0(X_1,X_2)\rho_B^0(X_2,X_1)$$
$$+ \rho_B^0(X_1,X_2)g_{Bl'l}^0(X_2,X_1)\big], \quad (B2)$$

where summation over repeating indices is assumed. However, only three Green's functions are linearly independent, allowing us to express the energy shifts in terms of the retarded, D_r, and advanced, $D_a = (D_r)^*$, Green's function, and the photon density matrix $\rho_{\mathrm{ph}}^{\nu\nu'} = iD_{12}^{\nu\nu'}$ satisfying the equations [25,27]

$$D_{11} = D_r - i\rho_{\mathrm{ph}}, \quad (B3)$$

$$D_r^{\nu\nu'}(\omega,\mathbf{R}_A,\mathbf{R}_A) = D_r^{0\nu\nu'}(\omega,\mathbf{R}_A,\mathbf{R}_A)$$
$$+ D_r^{0\nu\nu_1}(\omega,\mathbf{R}_A,\mathbf{R}_B)\Pi_{Br}^{\nu_1\nu_2}(\omega)$$
$$\times D_r^{0\nu_2\nu'}(\omega,\mathbf{R}_B,\mathbf{R}_A), \quad (B4)$$

$$\rho_{\mathrm{ph}} = \rho_{\mathrm{ph}}^0 + D_r^0\Pi_r\rho_{\mathrm{ph}}^0 + \rho_{\mathrm{ph}}^0\Pi_a D_a^0 - iD_r^0\Pi_{12}D_a^0, \quad (B5)$$

where the free photon density matrix for an isotropic and unpolarized EM field with occupation numbers $N(\omega)$ obeys the fluctuation-dissipation relation $\rho_{\mathrm{ph}}^{0\nu\nu'}(\omega,\mathbf{r},\mathbf{r}') = -2N(\omega)\,\mathrm{Im}\,D_r^{0\nu\nu'}(\omega,\mathbf{r},\mathbf{r}')$ $(\omega > 0)$ [6,30] and the polarization operators obey the equations

$$\Pi_r = \Pi_{11} + \Pi_{12}, \quad (B6)$$

$$\Pi_a = \Pi_{11} + \Pi_{21}. \quad (B7)$$

Direct calculations with the help of Eqs. (11) and (B2) reveals

$$\Pi_{Br}^{\nu_1\nu_2}(\omega) = -\alpha_B^{\nu_1\nu_2}(\omega), \quad \Pi_a^{\nu_1\nu_2} = \Pi_r^{\nu_2\nu_1*}, \quad (B8)$$

$$\Pi_{B12}^{\nu_1\nu_2}(\omega) = -2\,\mathrm{Im}[\alpha_B^{\nu_1\nu_2}(\omega)]p_e^B. \quad (B9)$$

Thus, plugging Eqs. (B8) and (B9) into Eqs. (B3)–(B5) leads to Eq. (15).

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