Two-atom van der Waals forces with one atom excited: The identical-atom limit

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We compute the conservative van der Waals forces between two atoms, one of which is initially excited, in the limit of identical atoms. Starting with the perturbative calculation of the interaction between two dissimilar atoms, we show that a time-dependent approach in the weak-interaction approximation is essential in considering the identical-atom limit in the perturbative regime. In this limit we find that, at leading order, the van der Waals forces are fully resonant and grow linearly in time, being different upon each atom. The resultant net force upon the two-atom system is related to the directionality of spontaneous emission, which results from the violation of parity symmetry. The strength of this force is much greater than that found in dissimilar atoms, raising the possibility of its experimental detection. In contrast to the usual stationary van der Waals forces, the time-dependent conservative forces cannot be written as the gradients of the expectation values of the interaction potentials, but as the expectation values of the gradients of the interaction potentials only.

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I. INTRODUCTION

Dispersion forces between neutral atoms are the result of the coupling of the quantum fluctuations of the electromagnetic (EM) field in its vacuum state with the fluctuations of the atomic charges in stable or metastable states. Generically, the corresponding forces are known as van der Waals (vdW) forces [1–5]. In recent decades renewed interest has been drawn to the interaction between excited atoms. The interests are twofold. From a practical perspective, this is the kind of interaction between Rydberg atoms [6–12] which makes possible the coherent manipulation of their quantum states, facilitating the entanglement between separated quantum systems as well as the storage of quantum information [13–17]. On the other hand, from a fundamental perspective, the attention has focused on different aspects of the interaction, namely, its scaling behavior with the distance [18–24], the role of dissipation [22,23,25–27], its inherent time dependence [19,20,22,28–30], and the net forces induced by parity and time-reversal violation on a binary system [31,32].

Hereafter and for the sake of simplicity we will consider the interaction between a pair of two-level atoms, A and B, with resonance frequencies ω_A and ω_B, natural linewidths Γ_A and Γ_B, and ground and excited states labeled with subscripts $+$ and $-$, respectively, $|A_\pm, B_\pm\rangle$. In the case of dissimilar atoms, i.e., for $|\Delta_{AB}| = |\omega_A - \omega_B| \gg \Gamma_A, \Gamma_B$, it is possible to use quasi-stationary perturbation theory to compute the interaction. This is so because the excitation process can be considered adiabatic with respect to the rate at which the excitation is transferred between the atoms, Δ_{AB}. That is, denoting by Ω the Rabi frequency of the external exciting field, an adiabatic excitation holds for $|\Delta_{AB}| \gg \Omega$. It was shown in Ref. [22] that, for arbitrary values of Ω, the resultant resonant interaction contains a quasistationary term which oscillates in space with wavelength $c\pi/\omega_A$ and is exponentially attenuated in time at the rate Γ_A, and time-oscillating terms of frequency Δ_{AB} whose amplitude is proportional to $\Omega^2/(\Delta_{AB}^2 - \Omega^2)$. In the adiabatic limit the latter terms vanish [19], and the result is equivalent to that obtained using adiabatic time-dependent perturbation theory [22]. Other approaches based on Heisenberg's formalism [21,23,29] and Feynman's Lagrangian formalism between asymptotic states [28,30] lead to an equivalent quasistationary result. In the opposite limit, that is, for a sudden excitation with $\Omega \gg |\Delta_{AB}|$, quasistationary and time oscillating terms happen to be of the same order [20]. Either way, it was also found in Refs. [22,31] that a weak net force acts upon the center of mass of the two-atom system while excited.

As for the interaction of a binary system of identical two-level atoms, with one of them initially excited, neither quasistationary nor adiabatic approximations make physical sense for two reasons. In the first place, the system becomes degenerate, as the states $|A_+, B_-\rangle$ and $|A_-, B_+\rangle$ possess identical energies, and the quasi-stationary sates are instead the symmetric and antisymmetric Dicke's states, $(|A_+, B_-\rangle \pm |A_-, B_+\rangle)/\sqrt{2}$, respectively. This implies that the states in which only one of the atoms is excited are no longer quasistationary, and the use of stationary or time-adiabatic perturbation theory becomes unsuitable. Second, in contrast to the interaction between dissimilar atoms, the null value of Δ_{AB} makes an adiabatic excitation unfeasible with respect to the original detuning. On the contrary, a sudden excitation is suitable as long as its associated Rabi frequency Ω is much greater than the detuning between the stationary Dicke's states [2,33].

In this article we will show that, starting with a binary system of dissimilar atoms, the identical-atom limit upon the interaction of the excited system can be formulated in a consistent manner using time-dependent perturbation theory in the sudden excitation approximation. In order to keep

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the calculation perturbative, we will restrict ourselves to the weak-interaction regime, meaning that the observation time is small in comparison to the time it takes for the excitation to be transferred from the initially excited atom to the other, which is of the order of the inverse of the detuning between the Dicke states. We will show that, for two-level atoms, the van der Waals forces are dominated by fully resonant components which are different for each atom. Besides, in addition to the familiar off-resonant van der Waals force, a reciprocal semiresonant force arises. Interestingly, the time-dependent forces are not derived from the gradients of the expectation values of the interaction potentials, but from the expectation values of the gradients of the interaction potentials instead. The nonreciprocal components of the force result in a net force upon the system. In contrast to the net force found on dissimilar atoms [22,31], which is inversely proportional to the detuning between their resonant frequencies, it grows linearly in time for identical atoms. This finding invites one to think of its experimental verification in excited systems made of long lifetime identical atoms. Nonreciprocal forces are explained in terms of parity symmetry violation, and are related to the asymmetry in the probability of emission of photons from either atom [31,34]. The effect of the deexcitation upon the off-resonant van der Waals force will be also analyzed.

Our perturbative approach in the weak-interaction regime paves the way for its extension to the strong-interaction regime where the vdW interaction between identical atoms becomes nonperturbative, e.g., in binary systems of Rydberg atoms. It is in the strong regime where the nonreciprocal forces and the directional emission found here are expected to be experimentally accessible. Hence, advances in the control of individual atoms by optical tweezers and magneto-optical traps have allowed the design of protocols and algorithms for the deterministic loading of atomic arrays and the tracking of the atomic positions [35].

The article is organized as follows. In Sec. II we explain the formalism of our approach and discuss its adequacy for the problem addressed in this work. In Sec. III we perform the computation of the vdW forces between two dissimilar two-level atoms, one of which is suddenly excited. The origin of nonreciprocal forces is related to the directionality of spontaneous emission. In Sec. IV the identical-atom limit is considered in the weak-interaction regime. The conclusions are summarized in Sec. V together with a discussion on the extension of our results.

II. COMPUTATION OF VAN DER WAALS FORCES

Let us consider two atoms, A and B, located a distance R apart. Since we are ultimately interested in the identical-atom limit, $|\Delta_{AB}| \ll \Gamma_A$, $\Gamma_A \to \Gamma_B$, atom A is assumed to be suddenly excited with an external field of strength $\Omega \gg |\Delta_{AB}|$. From the calculation carried out in Ref. [22] we note that the effect of the external field on the vdW forces reduces to a delay time π/Ω in their expectation values. Also, in order not to confuse the recoil of the excited atom due to the absorption of an external field photon with that due to the vdW interaction, which is directed along the interatomic axis, we assume that the illumination of atom A is transverse to the interatomic axis. This is the situation considered in Ref. [20], where the calculation was restricted to quasi-resonant processes, and to observation times T such that $\Gamma_{A,B}T \ll 1$. Here we will go beyond those restrictions and we will evaluate all the contributions to the vdW forces on both atoms, at leading order in the coupling parameter.

A. Fundamentals of the approach

Let us consider a sudden excitation of atom A. The state of the system at time 0 is $|\Psi(0)\rangle = |A_+\rangle \otimes |B_-\rangle \otimes |0_\gamma\rangle$, where $(A, B)_\pm$ label the upper and lower internal states of the atoms A and B respectively, and $|0_\gamma\rangle$ is the electromagnetic (EM) vacuum state. At any given time $T > 0$ the state of the two-atom-EM field system can be written as $|\Psi(T)\rangle = \mathbb{U}(T)|\Psi(0)\rangle$, where $\mathbb{U}(T)$ denotes the time propagator in the Schrödinger representation,

$$\mathbb{U}(T) = \text{T-exp}\left\{-i\hbar^{-1}\int_0^T dt\, H\right\},$$
$$H = \mathcal{T} + H_A + H_B + H_{EM} + W. \quad (1)$$

In this equation $\mathcal{T} = m_A|\dot{\mathbf{R}}_A|^2/2 + m_B|\dot{\mathbf{R}}_B|^2/2$ is the kinetic energy of the center of mass of the atomic system, with $m_{A,B}$ being the atomic masses and $\mathbf{R}_{A,B}$ the position vectors of the centers of mass of each atom. $H_A + H_B$ is the free Hamiltonian of the internal atomic states, $\hbar\omega_A|A_+\rangle\langle A_+| + \hbar\omega_B|B_+\rangle\langle B_+|$, while the Hamiltonian of the free EM field is $H_{EM} = \sum_{\mathbf{k},\boldsymbol{\epsilon}} \hbar\omega(a^\dagger_{\mathbf{k},\boldsymbol{\epsilon}}a_{\mathbf{k},\boldsymbol{\epsilon}} + 1/2)$, where $\omega = ck$ is the photon frequency, and the operators $a^\dagger_{\mathbf{k},\boldsymbol{\epsilon}}$ and $a_{\mathbf{k},\boldsymbol{\epsilon}}$ are the creation and annihilation operators of photons with momentum $\hbar\mathbf{k}$ and polarization $\boldsymbol{\epsilon}$, respectively. Finally, the interaction Hamiltonian in the electric dipole approximation reads $W = W_A + W_B$, with

$$W_{A,B} \simeq -\mathbf{d}_{A,B}\cdot\mathbf{E}(\mathbf{R}_{A,B}). \quad (2)$$

In this expression $\mathbf{d}_{A,B}$ are the electric dipole operators of each atom, and $\mathbf{E}(\mathbf{R}_{A,B})$ are the quantum electric field operators in Schrödinger's representation evaluated at the position of the center of mass of each atom. In terms of the EM vector potential,

$$\mathbf{A}(\mathbf{r}, t) = \sum_{\mathbf{k},\boldsymbol{\epsilon}} \sqrt{\frac{\hbar}{2\omega\mathcal{V}\epsilon_0}}[\boldsymbol{\epsilon}a_{\mathbf{k},\boldsymbol{\epsilon}}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} + \boldsymbol{\epsilon}^* a^\dagger_{\mathbf{k},\boldsymbol{\epsilon}}e^{-i(\mathbf{k}\cdot\mathbf{r}-\omega t)}],$$

the electric field $\mathbf{E}(\mathbf{R}_{A,B}) = -\partial_t\mathbf{A}(\mathbf{R}_{A,B}, t)|_{t=0}$ can be written as a sum over normal modes,

$$\mathbf{E}(\mathbf{R}_{A,B}) = \sum_{\mathbf{k}}[\mathbf{E}^{(-)}_{\mathbf{k}}(\mathbf{R}_{A,B}) + \mathbf{E}^{(+)}_{\mathbf{k}}(\mathbf{R}_{A,B})]$$
$$= i\sum_{\mathbf{k},\boldsymbol{\epsilon}} \sqrt{\frac{\hbar ck}{2\mathcal{V}\epsilon_0}}[\boldsymbol{\epsilon}a_{\mathbf{k},\boldsymbol{\epsilon}}e^{i\mathbf{k}\cdot\mathbf{R}_{A,B}} - \boldsymbol{\epsilon}^* a^\dagger_{\mathbf{k},\boldsymbol{\epsilon}}e^{-i\mathbf{k}\cdot\mathbf{R}_{A,B}}],$$

where $\mathcal{V}$ is a generic volume and $\mathbf{E}^{(\mp)}_{\mathbf{k}}$ denote the annihilationand creation electric field operators of photons of momentum $\hbar\mathbf{k}$, respectively. Strictly speaking, W includes an additional term in the electric dipole approximation which is referred to as the Röntgen term [36]. As argued in Ref. [37], that term is negligible since its contribution to Eq. (1) contains terms of orders $\dot{R}_{A,B}/c$ and $\mathbf{d}_{A,B}\cdot\mathbf{E}(\mathbf{R}_{A,B})/m_{A,B}$ smaller than the contribution of Eq. (2).

Next, considering W as a perturbation to the free Hamiltonians, the unperturbed time propagator for atom and free photon states is $\mathbb{U}_0(t) = \exp[-i\hbar^{-1}(\mathcal{T} + H_A + H_B + H_{EM})t]$. In terms of W and $\mathbb{U}_0$, $\mathbb{U}(T)$ admits an expansion in powers of W which can be developed out of the time-ordered exponential equation,

$$\mathbb{U}(T) = \mathbb{U}_0(T)\,\text{T-exp}\int_0^T (-i/\hbar)\mathbb{U}_0^\dagger(t)\,W\,\mathbb{U}_0(t)dt, \tag{3}$$

which can be written as a series in powers of W as $\mathbb{U}(T) = \mathbb{U}_0(T) + \sum_{n=1}^{\infty}\delta\mathbb{U}^{(n)}(T)$, with $\delta\mathbb{U}^{(n)}$ being the term of order W^n.

The system possesses a conserved total momentum [38,39], $[H, \mathbf{K}] = \mathbf{0}$,

$$\mathbf{K} = \mathbf{P}_A + \mathbf{P}_B + \mathbf{P}_\perp^\gamma, \tag{4}$$

where $\mathbf{P}_{A,B}$ are the canonical conjugate momenta of the centers of mass of each atom and $\mathbf{P}_\perp^\gamma = \sum_{\mathbf{k},\epsilon}\hbar\mathbf{k}\,a_{\mathbf{k},\epsilon}^\dagger a_{\mathbf{k},\epsilon}$ is the transverse EM momentum. Further, if the charges $\{q_i\}$ within the atoms are considered individually at positions $\{\mathbf{r}_i\}$, the canonical conjugate momenta can be written as

$$\mathbf{P}_A + \mathbf{P}_B = m_A\dot{\mathbf{R}}_A + m_B\dot{\mathbf{R}}_B + \sum_i q_i\mathbf{A}(\mathbf{r}_i), \tag{5}$$

where the first two terms are the kinetic momenta of the centers of mass of each atom, and the momentum within the summation symbol is referred to as longitudinal EM momentum [38], $\mathbf{P}_\parallel^\gamma = \sum_i q_i\mathbf{A}(\mathbf{r}_i)$. Lastly, in the electric dipole approximation, $\mathbf{P}_\parallel^\gamma$ reads [36], $\mathbf{P}_\parallel^\gamma \simeq -\mathbf{d}_A\times\mathbf{B}(\mathbf{R}_A) - \mathbf{d}_B\times\mathbf{B}(\mathbf{R}_B)$, where $\mathbf{B}(\mathbf{R}_{A,B}) = \boldsymbol{\nabla}_{A,B}\times\mathbf{A}(\mathbf{R}_{A,B})$.

Following Refs. [31,37], the force on each atom is computed applying the time derivative to the expectation value of the kinetic momenta of the centers of mass of each atom. Writing the latter in terms of the canonical conjugate momenta and the longitudinal EM momentum, in the electric dipole approximation, we arrive at

$$\begin{aligned}\langle\mathbf{F}_{A,B}\rangle_T &= \partial_T\langle m_{A,B}\dot{\mathbf{R}}_{A,B}\rangle_T\\ &= -i\hbar\partial_T\langle\Psi(0)|\mathbb{U}^\dagger(T)\boldsymbol{\nabla}_{A,B}\mathbb{U}(T)|\Psi(0)\rangle\\ &\quad+\partial_T\langle\Psi(0)|\mathbb{U}^\dagger(T)\mathbf{d}_{A,B}\times\mathbf{B}(\mathbf{R}_{A,B})\mathbb{U}(T)|\Psi(0)\rangle\\ &= -\langle\boldsymbol{\nabla}_{A,B}W_{A,B}\rangle_T + \partial_T\langle\mathbf{d}_{A,B}\times\mathbf{B}(\mathbf{R}_{A,B})\rangle_T,\end{aligned} \tag{6}$$

The first term on the right-hand side of the last equality is a conservative force along the interatomic axis, which we will refer to as vdW force. Note however that, in contrast to the stationary vdW forces computed in the adiabatic approximation, cf. Ref. [22], time-dependent conservative forces cannot be generally written as $\frac{-1}{2}\boldsymbol{\nabla}_{A,B}\langle W_{A,B}\rangle_T$. We will show later, including up to two-photon exchange processes, that the reason is the functional dissymmetry in the contribution of the two photons to the time-dependent terms. The second term is a nonconservative force equivalent to the time derivative of the longitudinal EM momentum at each atom, with opposite sign. We will show in a separate publication [40] that its contribution is only observable for $|\Delta_{AB}| \ll \omega_{A,B}$, being of the order of $\max(|\Delta_{AB}|, \Gamma_A)/\omega_A$ times smaller than the vdW conservative force. Hereafter we will neglect it and approximate $\langle\mathbf{F}_{A,B}\rangle_T \simeq -\langle\boldsymbol{\nabla}_{A,B}W_{A,B}\rangle_T$.

B. Discussion on the formalism

As in preceding works [22,31,37], we have formulated the vdW forces on a binary system in terms of the time-dependent expectation value of the time derivative of the kinetic momentum of each atom, which in the Schrödinger picture reads $\langle\mathbf{F}_{A,B}\rangle_T = \partial_T\langle\Psi(0)|\mathbb{U}^\dagger(T)m_{A,B}\dot{\mathbf{R}}_{A,B}\mathbb{U}(T)|\Psi(0)\rangle$; cf. Eq. (6).

However, previous approaches have formulated these forces in terms of the level shift of the two-atom wave function, $\delta\mathcal{E}$, assuming implicitly that the atomic state is stationary. Next, appealing to the conservative character of the vdW interaction—up to considering the longitudinal EM momentum—some authors have identified the vdW force with $-\boldsymbol{\nabla}\delta\mathcal{E}$, where $\delta\mathcal{E}$ was computed in the framework of stationary perturbation theory or linear response theory, regardless of whether or not the atomic state was metastable, e.g., Refs. [18,27].

On the other hand, applying time-dependent perturbation theory in the interaction picture, Berman's computation has led to the identification $\delta\mathcal{E} = \text{Re}\{i\hbar\partial_T\langle\Psi(0)|\mathbb{U}_0^\dagger(T)\mathbb{U}(T)|\Psi(0)\rangle\}$ [19,22] for a binary atomic system in an excited state. In this expression it is assumed that, subjected to some physical conditions, the time derivative of the probability amplitude provides time-independent terms in addition to fast oscillating secular terms that can be discarded. In the case of the setup addressed in Refs. [19,22], the physical conditions needed are the adiabatic excitation of the atomic system together with its quasistationary dynamics. Further, Refs. [19–23,32] have proved that a fully stationary treatment is insufficient to account for the dissipative effects that accompany the computation of the level shift in an excited metastable system.

More recently, the time-dependent approaches of Refs. [20,22,23] have computed the vdW forces on excited systems out of the expectation values of the interaction energies $\langle W_A\rangle$ and $\langle W_B\rangle$. In the quasistationary limit, or considering an adiabatic excitation otherwise, they have found that the forces on each atom differ in spatially oscillating terms, resulting in the apparent violations of the classical action-reaction principle and the conservation of total momentum, which is in clear contradiction with the invariance of the system under global spatial translation. This apparent contradiction was solved in Ref. [31], where it was shown that the *missing* momentum was carried by the virtual photons which mediate the interaction, i.e., $\partial_T\langle\mathbf{P}_\perp^\gamma\rangle + \langle\mathbf{F}_A\rangle + \langle\mathbf{F}_B\rangle = \mathbf{0}$. Moreover, it was proved that the momentum carried by the virtual photons while the atoms are excited is equivalent to the asymmetric momentum which is found along the interatomic axis in the emission of actual photons in the deexcitation process, causing the directionality of spontaneous emission. Both phenomena, namely, the nonreciprocity of the forces and the directionality of the emission, are thus closely related, and can be interpreted as a consequence of the absence of parity symmetry along the interatomic axis of a binary system which is asymmetrically excited.

As for the specific case of identical atoms, with one of them initially excited, an analogous reasoning applies and analogous results are to be expected. However, the fact that

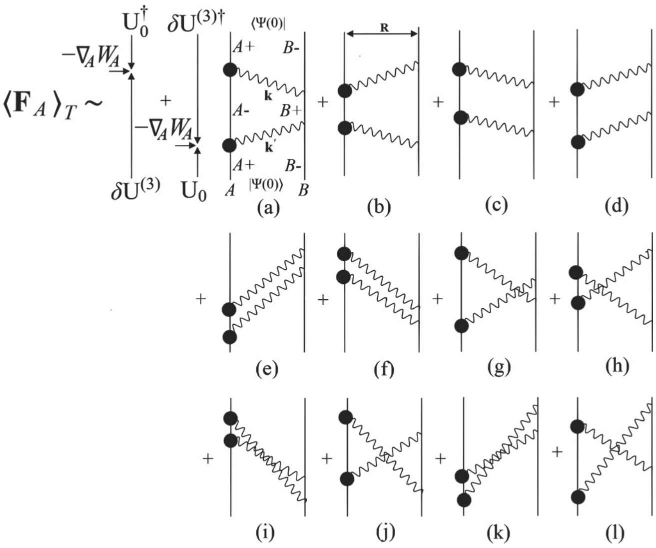

FIG. 1. Diagrammatic representation of twelve of the processes which contribute to $\langle \mathbf{F}_A \rangle_T$. Solid straight lines stand for propagators of atomic states, while wavy lines stand for photon propagators. In diagram (a), atomic and photon states are indicated explicitly. The atoms A and B are separated by a distance R along the horizontal direction, whereas time runs along the vertical. The big circles in black on the left of each diagram stand for the insertion of the Schrödinger operator $-\nabla_A W_A$ whose expectation value is computed. Each diagram contributes with two terms, one from each of the operators inserted. They are sandwiched between two time propagators, $U(T)$ and $U^\dagger(T)$ (depicted by vertical arrows), which evolve the initial state $|\Psi(0)\rangle$ towards the observation time at which $-\nabla_A W_A$ applies.

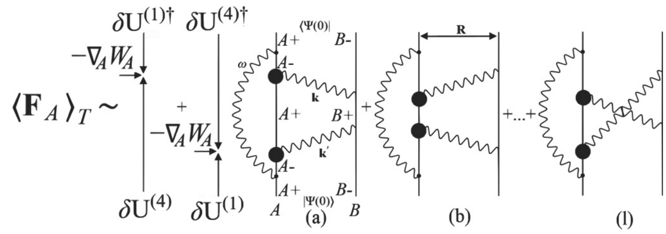

FIG. 2. Diagrammatic representation of processes which contribute to the fully off-resonant component of $\langle \mathbf{F}_A \rangle_T$. In contrast to the diagrams in Fig. 1, the self-interacting photon on atom A leads the spontaneous emission from the excited atom. The omitted diagrams are analogous to those in Fig. 1.

the system is degenerate implies that the vdW forces are explicitly time dependent, since no adiabatic approximation can be taken. Note that the stationary approach suggested in Ref. [27], based on degenerate stationary perturbation theory, is not appropriate. The reason is the nonstationary character of the state $|A_+, B_-\rangle$, whose dynamics comprises both the coherent transfer of the excitation between the atoms and the incoherent decay of the excited atoms. Further comments on this issue can be found in Sec. V.

III. vdW FORCES ON A BINARY SYSTEM OF DISSIMILAR ATOMS AFTER A SUDDEN EXCITATION

A perturbative development of Eq. (6) shows that, up to terms involving two-photon exchange processes, 24 diagrams contribute to $\langle \mathbf{F}_A \rangle_T$ for a two-level atom. They are depicted in Figs. 1 and 2. Note that those in Fig. 2 just differ with respect to those of Fig. 1 by the photon embracing the two exchanged photons, which accounts for the deexcitation of the system via spontaneous emission from atom A. For the sake of illustration we give below the expression of diagram (a) in Fig. 1, which contributes to $\langle \mathbf{F}_A \rangle_T$ in the form

$$\frac{1}{\hbar^3}\int_0^\infty \frac{\mathcal{V}k^2dk}{(2\pi)^3}\int_0^\infty \frac{\mathcal{V}k'^2dk'}{(2\pi)^3}\int_0^{4\pi} d\Theta \int_0^{4\pi} d\Theta' \Big\{\Big[i\langle A_+, B_-, 0_\gamma| e^{i\Omega_a^* T}|A_+, B_-, 0_\gamma\rangle \int_0^T dt \int_0^t dt' \int_0^{t'} dt''$$
$$\times\ \langle A_+, B_-, 0_\gamma| -\nabla_A[\mathbf{d}_A\cdot\mathbf{E}^{(-)}_{\mathbf{k}}(\mathbf{R}_A)]|A_-, B_-, \gamma_{\mathbf{k}}\rangle e^{-i\omega(T-t)}\langle A_-, B_-, \gamma_{\mathbf{k}}|\mathbf{d}_B\cdot\mathbf{E}^{(+)}_{\mathbf{k}}(\mathbf{R}_B)|A_-, B_+, 0_\gamma\rangle$$
$$\times\ e^{-i\Omega_b(t-t')}\langle A_-, B_+, 0_\gamma|\mathbf{d}_B\cdot\mathbf{E}^{(-)}_{\mathbf{k}'}(\mathbf{R}_B)|A_-, B_-, \gamma_{\mathbf{k}'}\rangle e^{-i\omega'(t'-t'')}\langle A_-, B_-, \gamma_{\mathbf{k}'}|\mathbf{d}_A\cdot\mathbf{E}^{(+)}_{\mathbf{k}'}(\mathbf{R}_A)|A_+, B_-, 0_\gamma\rangle e^{-i\Omega_a t''}\Big] + [k\leftrightarrow k']^\dagger\Big\}, \tag{7}$$

where it is implicit that the causality condition $T \gg R/c$ holds at the time of observation. In this equation $|A_+, B_-, 0_\gamma\rangle$ is the initial two-atom-EM-vacuum state, with atom A excited at time 0, $|\gamma_{\mathbf{k}}\rangle$ is a one-photon state of momentum $\mathbf{k}$ and frequency $\omega = ck$, the complex time exponentials are the result of the application of the free time-evolution operator $U_0(t) = e^{-i\hbar^{-1}H_0 t}$ between the interaction vertices $W_{A,B}$, with $\Omega_a = \omega_A - i\Gamma_A/2$ and $\Omega_b = \omega_B - i\Gamma_B/2$, where the dissipative imaginary terms account for radiative emission in the Weisskopf-Wigner approximation and $\omega_{A,B}$ include the contribution of the free-space Lamb shift. After integrating in time and solid angles, one arrives at

$$\frac{c^2\hbar^{-1}}{\pi^2\epsilon_0^2}\mathrm{Re}\int_0^\infty dk' k'^2 \nabla_A[\boldsymbol{\mu}_A\cdot \mathrm{Im}\mathbb{G}(k'R)\cdot\boldsymbol{\mu}_B]\int_0^\infty dk\, k^2 \boldsymbol{\mu}_B\cdot\mathrm{Im}\mathbb{G}(kR)\cdot\boldsymbol{\mu}_A\, e^{i\Omega_a^* T}\Bigg[\frac{e^{-i\Omega_a T}-e^{-i\omega T}}{(\omega'-\Omega_a)(\Omega_b-\Omega_a)(\omega-\Omega_a)}$$
$$-\frac{e^{-i\Omega_b T}-e^{-i\omega T}}{(\omega'-\Omega_a)(\Omega_b-\Omega_a)(\omega-\Omega_b)}+\frac{e^{-i\omega' T}-e^{-i\omega T}}{(\omega'-\Omega_a)(\omega'-\Omega_b)(\omega-\omega')}-\frac{e^{-i\Omega_b T}-e^{-i\omega T}}{(\omega'-\Omega_a)(\omega'-\Omega_b)(\omega-\Omega_b)}\Bigg], \tag{8}$$

where $\boldsymbol{\mu}_A = \langle A_-|\mathbf{d}_A|A_+\rangle$, $\boldsymbol{\mu}_B = \langle B_-|\mathbf{d}_B|B_+\rangle$, and $\mathbb{G}(kR)$ is the dyadic Green's function of the electric field induced at $\mathbf{R}$ by an electric dipole of frequency $\omega = ck$ placed at the origin. It reads

$$\mathbb{G}(kR) = \frac{k\, e^{ikR}}{-4\pi}[\alpha/kR + i\beta/(kR)^2 - \beta/(kR)^3], \tag{9}$$

where the tensors α and β read $\alpha = \mathbb{I} - \hat{\mathbf{R}}\hat{\mathbf{R}}$, $\beta = \mathbb{I} - 3\hat{\mathbf{R}}\hat{\mathbf{R}}$, with $\hat{\mathbf{R}} = \mathbf{R}/R$.

Operating in an analogous fashion with the rest of the terms derived from the diagrams of Figs. 1 and 2, upon integration in k and k' in the complex plane, using the identity $\nabla_B = -\nabla_A = -\nabla_{\mathbf{R}}$, we arrive at

$$\begin{aligned}\langle \mathbf{F}_A\rangle_T = &-\frac{2\omega_A^4 e^{-\Gamma_A T}}{c^4\epsilon_0^2\hbar\Delta_{AB}}[\boldsymbol{\mu}_A\cdot \mathrm{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B]-\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B]]\\
&+\frac{2\omega_B^4 e^{-(\Gamma_A+\Gamma_B)T/2}}{c^4\epsilon_0^2\hbar\Delta_{AB}}[\boldsymbol{\mu}_A\cdot \mathrm{Re}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Re}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B]-\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B]]\\
&\times\cos(\Delta_{AB}T)-\frac{2\omega_B^4 e^{-(\Gamma_A+\Gamma_B)T/2}}{c^4\epsilon_0^2\hbar\Delta_{AB}}[\boldsymbol{\mu}_A\cdot \mathrm{Re}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B]\\
&+\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Re}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B]]\sin(\Delta_{AB}T)\\
&+\frac{2\omega_A^4 e^{-\Gamma_A T}}{c^4\epsilon_0^2\hbar(\omega_A+\omega_B)}[\boldsymbol{\mu}_A\cdot \mathrm{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B]-\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B]]\\
&-\frac{2\omega_B^2 e^{-(\Gamma_A+\Gamma_B)T/2}}{c^3\epsilon_0^2\hbar}[\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Re}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B]\cos(\Delta_{AB}T)-\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B]\sin(\Delta_{AB}T)]\\
&\times\int_0^\infty\frac{dq}{\pi}\frac{(q^2-k_Ak_B)q^2\boldsymbol{\mu}_A\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_B}{(q^2+k_A^2)(q^2+k_B^2)}+\frac{4\omega_A\omega_B(1-2e^{-\Gamma_AT})}{c^3\epsilon_0^2\hbar}\int_0^\infty\frac{dq}{\pi}\frac{q^4\boldsymbol{\mu}_A\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_B}{(q^2+k_A^2)(q^2+k_B^2)}\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_B].\end{aligned}\tag{10}$$

In this equation, negligible and unobservable terms have been discarded. These are off-resonant terms whose integrands are attenuated in time as e^{-cTq} and whose contribution is $(R/cT)^3 \ll 1$ times smaller, and fast oscillating secular terms of frequency $\omega_A+\omega_B$ which average to zero upon observation. The origin of the terms in Eq. (10) is as follows. The first three terms, which scale as $\sim 1/\Delta_{AB}$, are fully resonant and involve the evaluation of the two residues associated to simple poles in k and k' in the integrals stemming from diagram (a). The fourth term, which scales as $\sim 1/(\omega_A+\omega_B)$, fully resonant too, results from the two resonant photons of diagram (g), which contains a two-photon intermediate state. The semiresonant terms, which entail evaluating the residue associated to a simple pole in k or k' only, oscillate in time at frequency Δ_{AB}. They stem from diagrams (c,d,e,f). Finally, the last term is the result of the addition of the off-resonant contributions coming from the twelve diagrams of Figs. 1 and 2 together. The discarded fast oscillating terms, resonant and semi-resonant, are associated with diagrams (i,k) and (i,j,k,l), respectively, which contain two-photon intermediate states. In the far field we can approximate

$$\langle \mathbf{F}_A\rangle_T \approx \frac{k_A^7 e^{-\Gamma_AT}(\boldsymbol{\mu}_A\cdot\alpha\cdot\boldsymbol{\mu}_B)^2}{8\pi^2\epsilon_0^2\hbar\Delta_{AB}}\left[\frac{\cos(2k_AR)}{(k_AR)^3}+\frac{\sin(2k_AR)}{(k_AR)^2}\right]\hat{\mathbf{R}}.$$

Analogous diagrams hold for $\langle \mathbf{F}_B\rangle_T$, but for the evaluation of the operator $-\nabla_B W_B$ at atom B; see Figs. 3 and 4:

$$\begin{aligned}\langle \mathbf{F}_B\rangle_T = &\frac{2\omega_A^4 e^{-\Gamma_A T}}{c^4\epsilon_0^2\hbar\Delta_{AB}}[\boldsymbol{\mu}_A\cdot \mathrm{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_A]+\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_A]]\\
&-\frac{2\omega_B^2\omega_A^2 e^{-(\Gamma_A+\Gamma_B)T/2}}{c^4\epsilon_0^2\hbar\Delta_{AB}}[\boldsymbol{\mu}_A\cdot \mathrm{Re}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_A]+\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_A]]\\
&\times\cos(\Delta_{AB}T)-\frac{2\omega_B^2\omega_A^2 e^{-(\Gamma_A+\Gamma_B)T/2}}{c^4\epsilon_0^2\hbar\Delta_{AB}}[\boldsymbol{\mu}_A\cdot \mathrm{Re}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_A]\\
&-\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_A]]\sin(\Delta_{AB}T)\\
&-\frac{2\omega_A^4 e^{-\Gamma_A T}}{c^4\epsilon_0^2\hbar(\omega_A+\omega_B)}[\boldsymbol{\mu}_A\cdot \mathrm{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_A]+\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_A]]\\
&+\frac{2\omega_A^2 e^{-(\Gamma_A+\Gamma_B)T/2}}{c^3\epsilon_0^2\hbar}[\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_A]\cos(\Delta_{AB}T)+\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_A]\sin(\Delta_{AB}T)]\\
&\times\int_0^\infty\frac{dq}{\pi}\frac{(q^2-k_Ak_B)q^2\boldsymbol{\mu}_A\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_B}{\left(q^2+k_A^2\right)\left(q^2+k_B^2\right)}-\frac{4\omega_A\omega_B(1-2e^{-\Gamma_AT})}{c^3\epsilon_0^2\hbar}\int_0^\infty\frac{dq}{\pi}\frac{q^4\boldsymbol{\mu}_A\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_B}{\left(q^2+k_A^2\right)\left(q^2+k_B^2\right)}\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_A].\end{aligned}\tag{11}$$

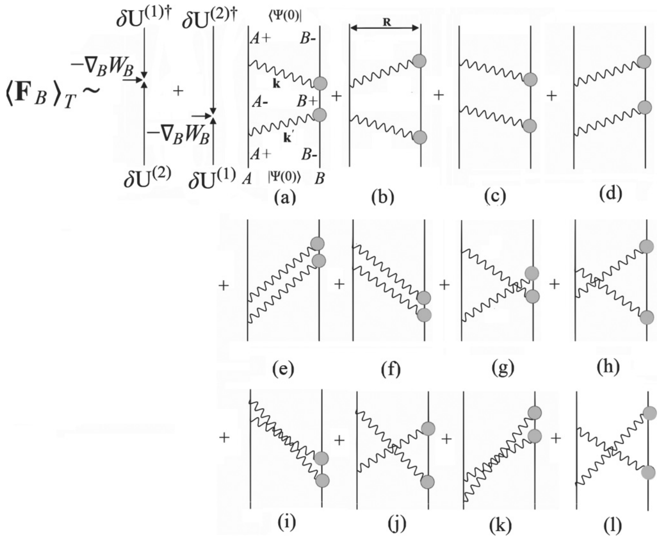

FIG. 3. Diagrammatic representation of twelve of the processes which contribute to $\langle \mathbf{F}_B \rangle_T$. The big circles in gray on the right of each diagram stand for the insertion of the Schrödinger operator $-\nabla_B W_B$ whose expectation value is computed.

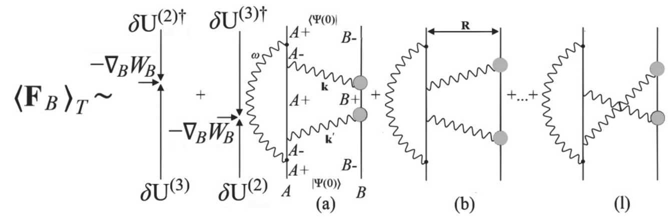

FIG. 4. Diagrammatic representation of processes which contribute to the fully off-resonant component of $\langle \mathbf{F}_B \rangle_T$. In contrast to the diagrams in Fig. 3, the self-interacting photon on atom A leads the spontaneous emission from the excited atom. The omitted diagrams are analogous to those in Fig. 3.

In the far field, its approximate expression is

$$\langle \mathbf{F}_B \rangle_T \approx -\frac{k_A^7 e^{-\Gamma_A T} (\boldsymbol{\mu}_A \cdot \alpha \cdot \boldsymbol{\mu}_B)^2}{8\pi^2 \epsilon_0^2 \hbar \Delta_{AB}} \frac{\hat{\mathbf{R}}}{(k_A R)^3}.$$

Note that, as anticipated after Eq. (6), the conservative vdW forces cannot be written in the form $-\nabla_{\mathbf{R}} \langle W_{A,B} \rangle_T / 2$ due to the functional dissymmetry of the time-dependent terms proportional to $\text{Re}\mathbb{G}(k_{A,B}R)\text{Im}\mathbb{G}(k_{A,B}R)$; for comparison, see also the expressions for $\langle W_{A,B} \rangle_T$ in the Appendix.

Comparing Eqs. (10) and (11), we observe that the only term which is common to both expressions is that involving off-resonant photons, which is proportional to $(2e^{-\Gamma_A T} - 1)$. That implies that it changes sign at an observation time $T \approx \ln 2/\Gamma_A$. As for the rest of the terms, some fully resonant and semiresonant terms, either stationary or oscillating at frequency Δ_{AB}, differ in sign. Those terms constitute nonreciprocal forces and amount to a net force on the two-atom system. The stationary nonreciprocal forces were shown in Ref. [31] to result from the excess of momenta stored in the virtual photons which mediate the resonant interaction in the processes depicted by diagrams (a) and (g). In addition, the slowly oscillating non-reciprocal forces arise after a sudden excitation only, and their associated momentum variation is supplied by the resonant photons of diagrams (a,c,d,e,f). Assuming that $|\Delta_{AB}| \ll \omega_{A,B}$ for the oscillating forces to be observable, the net force on the atomic system reads

$$\begin{aligned}
\langle \mathbf{F}_A + \mathbf{F}_B \rangle_T \simeq{}& \frac{8e^{-\Gamma_A T} k_A^4}{\epsilon_0^2 \hbar} \frac{\omega_B}{\omega_A^2 - \omega_B^2} \boldsymbol{\mu}_A \cdot \text{Im}\mathbb{G}(k_A R) \cdot \boldsymbol{\mu}_B \nabla_{\mathbf{R}} [\boldsymbol{\mu}_B \cdot \text{Im}\mathbb{G}(k_A R) \cdot \boldsymbol{\mu}_A] \\
&+ \frac{2e^{-(\Gamma_A+\Gamma_B)T/2} k_B^2}{\epsilon_0^2 \hbar \Delta_{AB}} \{\boldsymbol{\mu}_A \cdot \text{Re}\mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_B \nabla_{\mathbf{R}} [k_B^2 \boldsymbol{\mu}_B \cdot \text{Re}\mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_A - k_A^2 \boldsymbol{\mu}_A \cdot \text{Re}\mathbb{G}(k_A R) \cdot \boldsymbol{\mu}_B] \\
&- \boldsymbol{\mu}_A \cdot \text{Im}\mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_B \nabla_{\mathbf{R}} [k_B^2 \boldsymbol{\mu}_B \cdot \text{Im}\mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_A + k_A^2 \boldsymbol{\mu}_A \cdot \text{Im}\mathbb{G}(k_A R) \cdot \boldsymbol{\mu}_B]\} \cos(\Delta_{AB} T) \\
&- \frac{2e^{-(\Gamma_A+\Gamma_B)T/2} k_B^2}{\epsilon_0^2 \hbar \Delta_{AB}} \{\boldsymbol{\mu}_A \cdot \text{Re}\mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_B \nabla_{\mathbf{R}} [k_B^2 \boldsymbol{\mu}_B \cdot \text{Im}\mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_A + k_A^2 \boldsymbol{\mu}_A \cdot \text{Im}\mathbb{G}(k_A R) \cdot \boldsymbol{\mu}_B] \\
&+ \boldsymbol{\mu}_A \cdot \text{Im}\mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_B \nabla_{\mathbf{R}} [k_B^2 \boldsymbol{\mu}_B \cdot \text{Re}\mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_A - k_A^2 \boldsymbol{\mu}_A \cdot \text{Re}\mathbb{G}(k_A R) \cdot \boldsymbol{\mu}_B]\} \sin(\Delta_{AB} T) \\
&- \frac{2e^{-(\Gamma_A+\Gamma_B)T/2}}{c^3 \epsilon_0^2 \hbar} \int_0^\infty \frac{dq}{\pi} \frac{(q^2 - k_A k_B) q^2 \boldsymbol{\mu}_A \cdot \mathbb{G}(iqR) \cdot \boldsymbol{\mu}_B}{(q^2 + k_A^2)(q^2 + k_B^2)} [\nabla_{\mathbf{R}} [\omega_B^2 \boldsymbol{\mu}_A \cdot \text{Re}\mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_B - \omega_A^2 \boldsymbol{\mu}_A \cdot \text{Re}\mathbb{G}(k_A R) \cdot \boldsymbol{\mu}_B] \\
&\times \cos(\Delta_{AB} T) - \nabla_{\mathbf{R}} [\omega_B^2 \boldsymbol{\mu}_A \cdot \text{Im}\mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_B + \omega_A^2 \boldsymbol{\mu}_A \cdot \text{Im}\mathbb{G}(k_A R) \cdot \boldsymbol{\mu}_B] \sin(\Delta_{AB} T)],
\end{aligned} \tag{12}$$

where the first nonoscillating term coincides with the net force for the case of an adiabatic excitation [22]. Its asymptotic form in the far field reads

$$\langle \mathbf{F}_A + \mathbf{F}_B \rangle_T \approx \frac{k_A^7 e^{-\Gamma_A T} (\boldsymbol{\mu}_A \cdot \alpha \cdot \boldsymbol{\mu}_B)^2}{8\pi^2 \epsilon_0^2 \hbar \Delta_{AB}} \left[\frac{\cos(2k_A R) - 1}{(k_A R)^3} + \frac{\sin(2k_A R)}{(k_A R)^2}\right] \hat{\mathbf{R}}.$$

In what follows we study the directionality of one-photon spontaneous emission and show its relationship with the net force. Directionality is provided by the asymmetry in the emission rate of one of the resonant exchanged photons of the diagrams (a,c,d,e,f) depicted in Fig. 5. Hence, the resultant formula for the directional emission rate as a function of the solid angle,

$d\Gamma_{\rm dir}/d\Theta$, is not invariant under parity inversion. Hence, the asymmetry is maximum along the interatomic axis. The evaluation of the one-photon emission diagrams in Fig. 5 yields

$$\frac{d\Gamma_{\rm dir}}{d\Theta}=\frac{\boldsymbol{\mu}_A\cdot(\mathbb{I}-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\cdot\boldsymbol{\mu}_B}{2(\pi\epsilon_0\hbar)^2}\begin{cases}\frac{e^{-(\Gamma_A+\Gamma_B)T/2}k_B^5}{\Delta_{AB}}[\cos(\Delta_{AB}T)[\cos(k_BR\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Re}\,\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B-\sin(k_BR\cos\theta)\\ \times\boldsymbol{\mu}_A\cdot{\rm Im}\,\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B]-\sin(\Delta_{AB}T)[\cos(k_BR\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Im}\,\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B\\ +\sin(k_BR\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Re}\,\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B]]\\ -\frac{e^{-\Gamma_AT}k_A^5}{\Delta_{AB}}[\cos(k_AR\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Re}\,\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B-\sin(k_AR\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Im}\,\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B]\\ -e^{-(\Gamma_A+\Gamma_B)T/2}k_B^3[\cos(\Delta_{AB}T)\cos(k_BR\cos\theta)-\sin(\Delta_{AB}T)\sin(k_BR\cos\theta)]\\ \times\int_0^\infty c\frac{dq}{\pi}\frac{q^2(q^2-k_Ak_B)}{(q^2+k_A^2)(q^2+k_B^2)}\boldsymbol{\mu}_A\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_B\quad\text{for }\cos\theta\in(0,1],\\ \\ \frac{e^{-(\Gamma_A+\Gamma_B)T/2}k_B^2k_A^3}{\Delta_{AB}}[\cos(\Delta_{AB}T)[\cos(k_AR\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Re}\,\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B-\sin(k_AR\cos\theta)\\ \times\boldsymbol{\mu}_A\cdot{\rm Im}\,\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B]-\sin(\Delta_{AB}T)[\cos(k_AR\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Im}\,\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B\\ +\sin(k_AR\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Re}\,\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B]]\\ -\frac{e^{-\Gamma_AT}k_A^5}{\Delta_{AB}}[\cos(k_AR\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Re}\,\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B-\sin(k_AR\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Im}\,\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B]\\ -e^{-(\Gamma_A+\Gamma_B)T/2}k_A^3[\cos(\Delta_{AB}T)\cos(k_AR\cos\theta)-\sin(\Delta_{AB}T)\sin(k_AR\cos\theta)]\\ \times\int_0^\infty c\frac{dq}{\pi}\frac{q^2(q^2-k_Ak_B)}{(q^2+k_A^2)(q^2+k_B^2)}\boldsymbol{\mu}_A\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_B\quad\text{for }\cos\theta\in[-1,0).\end{cases}\tag{13}$$

In this equation parity symmetry is manifestly broken by the difference between the terms defined in each interval of $\cos\theta$. In particular, those applicable to $\cos\theta\in(0,1]$ contribute to $\mathbf{F}_A$ upon integration of Eq. (14), while those for $\cos\theta\in[-1,0)$ contribute to $\mathbf{F}_B$, respectively.

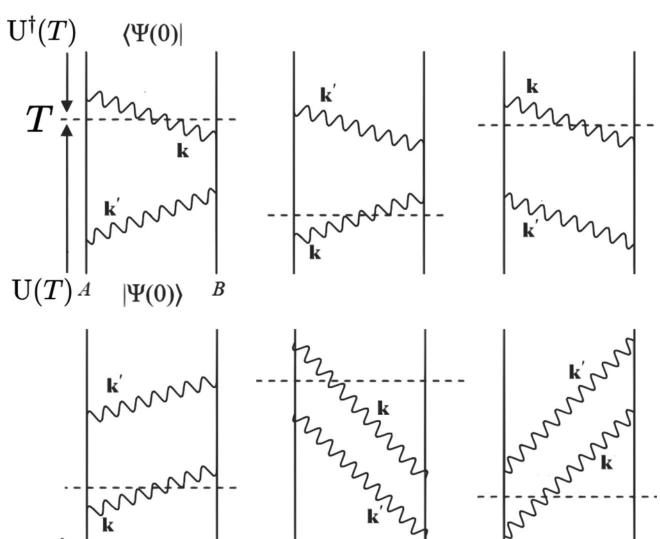

FIG. 5. Diagrammatic representation of the processes which contribute to the one-photon directional emission rate, $d\Gamma_{\rm dir}/d\Theta$. The time evolution of the initial state, both from the bottom and from the top of each diagram, is indicated in diagram (a) with vertical arrows that meet at the observation time T, where the horizontal dotted line cuts each diagram along the final state, $|\Psi(T)\rangle=\mathbb{U}(T)|\Psi(0)\rangle$. That final state contains the emitted photon and the ground states of both atoms

Under the condition $|\Delta_{AB}|\ll\omega_{A,B}$ and considering $\omega_A\simeq\omega_B$ for simplicity, we can write the time derivative of the transverse EM momentum as

$$\langle\dot{\mathbf{P}}_\perp^\gamma\rangle_T\simeq\hbar k_A\int_0^{4\pi}d\Theta\,\hat{\mathbf{k}}\frac{d\Gamma_{\rm dir}}{d\Theta}.\tag{14}$$

Straight integration of this equation leads to Eq. (12)—up to two-photon emission terms—except for a negative sign in front, proving that $\langle\mathbf{F}_A+\mathbf{F}_B\rangle_T=-\langle\dot{\mathbf{P}}_\perp^\gamma\rangle_T$, in agreement with the conservation of the momentum $\mathbf{K}$ defined in Eq. (4). Physically this implies that, while excited, the atomic system accelerates as a whole whereas the virtual photons which mediate the vdW interaction store a corresponding momentum in the opposite direction. That momentum is ultimately carried by the real photon which is emitted in the deexcitation process. On average, that momentum determines the directionality of the spontaneous emission from the binary system.

IV. THE IDENTICAL-ATOM LIMIT IN THE WEAK-INTERACTION REGIME

We proceed to take the identical-atom limit upon the equations obtained in the previous section. That is, we consider $\omega_B\to\omega_A=\omega_0$, $\Gamma_B\to\Gamma_A=\Gamma_0$, $\mu_A=\mu_B$. Note that, in order for the perturbative computations of Sec. III to remain valid in this limit, the observation time T must be small in comparison to the time that it takes for the excitation to be transferred from atom A to atom B, i.e., $k_0^2\boldsymbol{\mu}_A\cdot{\rm Re}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B\lesssim\hbar\epsilon_0/T$ [2,33]. This is the weak-interaction regime, which implies that the original atomic states are quasistationary despite the degeneracy of the system. In this limit, the vdW forces read

$$\begin{aligned}\langle \mathbf{F}_A\rangle_T =& -\frac{2\omega_0^4 e^{-\Gamma_0 T}}{c^4\epsilon_0^2\hbar}T\,[\boldsymbol{\mu}_A\cdot \mathrm{Re}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B]+\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Re}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B]]\\ &-\frac{2e^{-\Gamma_0 T}}{c^4\epsilon_0^2\hbar}\frac{\partial}{\partial\omega}[\omega^4\boldsymbol{\mu}_A\cdot\mathrm{Re}\mathbb{G}(kR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Re}\mathbb{G}(kR)\cdot\boldsymbol{\mu}_B]-\omega^4\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(kR)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(kR)\cdot\boldsymbol{\mu}_B]]_{\omega=\omega_0}\\ &+\frac{\omega_0^3 e^{-\Gamma_0 T}}{c^4\epsilon_0^2\hbar}[\boldsymbol{\mu}_A\cdot\mathrm{Re}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Re}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B]-\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B]]\\ &-\frac{2\omega_0^2 e^{-\Gamma_0 T}}{c^3\epsilon_0^2\hbar}\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathrm{Re}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B]\int_0^\infty\frac{dq}{\pi}\frac{(q^2-k_0^2)q^2\boldsymbol{\mu}_A\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_B}{\left(q^2+k_0^2\right)^2}\\ &+\frac{4\omega_0^2(1-2e^{-\Gamma_0 T})}{c^3\epsilon_0^2\hbar}\int_0^\infty\frac{dq}{\pi}\frac{q^4\boldsymbol{\mu}_A\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_B}{(q^2+k_0^2)^2}\nabla_{\mathbf{R}}[\boldsymbol{\mu}_A\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_B], \end{aligned}\tag{15}$$

$$\begin{aligned}\langle \mathbf{F}_B\rangle_T =& -\frac{2\omega_0^4 e^{-\Gamma_0 T}}{c^4\epsilon_0^2\hbar}T\,[\boldsymbol{\mu}_A\cdot \mathrm{Re}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Im}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_A]-\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Re}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_A]]\\ &+\frac{2e^{-\Gamma_0 T}\omega_0^2}{c^4\epsilon_0^2\hbar}\bigg[\frac{\partial}{\partial\omega}[\omega^2\boldsymbol{\mu}_A\cdot\mathrm{Re}\mathbb{G}(kR)\cdot\boldsymbol{\mu}_B]_{\omega=\omega_0}\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Re}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_A]\\ &+\frac{\partial}{\partial\omega}[\omega^2\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(kR)\cdot\boldsymbol{\mu}_B]_{\omega=\omega_0}\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Im}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_A]\bigg]\\ &-\frac{\omega_0^3 e^{-\Gamma_0 T}}{c^4\epsilon_0^2\hbar}[\boldsymbol{\mu}_A\cdot\mathrm{Re}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Re}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_A]+\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Im}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_A]]\\ &+\frac{2\omega_0^2 e^{-\Gamma_0 T}}{c^3\epsilon_0^2\hbar}\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Re}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_A]\int_0^\infty\frac{dq}{\pi}\frac{(q^2-k_0^2)q^2\boldsymbol{\mu}_A\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_B}{\left(q^2+k_0^2\right)^2}\\ &-\frac{4\omega_0^2(1-2e^{-\Gamma_0 T})}{c^3\epsilon_0^2\hbar}\int_0^\infty\frac{dq}{\pi}\frac{q^4\boldsymbol{\mu}_A\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_B}{\left(q^2+k_0^2\right)^2}\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_A], \end{aligned}\tag{16}$$

with asymptotic values in the far field

$$\begin{aligned}\langle\mathbf{F}_A\rangle_T &\approx -\frac{k_0^7 e^{-\Gamma_0 T}(\boldsymbol{\mu}_A\cdot\alpha\cdot\boldsymbol{\mu}_B)^2 T}{8\pi^2\epsilon_0^2\hbar}\left[\frac{\cos(2k_0R)}{(k_0R)^2}-\frac{\sin(2k_0R)}{(k_0R)^3}\right]\hat{\mathbf{R}},\\ \langle\mathbf{F}_B\rangle_T &\approx -\frac{k_0^7 e^{-\Gamma_0 T}(\boldsymbol{\mu}_A\cdot\alpha\cdot\boldsymbol{\mu}_B)^2 T}{8\pi^2\epsilon_0^2\hbar}\frac{\hat{\mathbf{R}}}{(k_0R)^2}.\end{aligned}$$

Likewise, the net force upon the atomic system is

$$\begin{aligned}\langle\mathbf{F}_A+\mathbf{F}_B\rangle_T =& -\frac{4e^{-\Gamma_0 T}}{c^4\epsilon_0^2\hbar}\bigg\{\omega_0^4 T\,[\boldsymbol{\mu}_A\cdot\mathrm{Re}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Im}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_A]]-\omega_0^4\frac{\partial}{\partial\omega}[\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(kR)\cdot\boldsymbol{\mu}_B]_{\omega=\omega_0}\\ &\times\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Im}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_A]+\frac{\omega_0^4}{2}[\boldsymbol{\mu}_A\cdot\mathrm{Re}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B\frac{\partial}{\partial\omega}[\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Re}\mathbb{G}(kR)\cdot\boldsymbol{\mu}_A]]_{\omega=\omega_0}\\ &-\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B\frac{\partial}{\partial\omega}[\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Im}\mathbb{G}(kR)\cdot\boldsymbol{\mu}_A]]_{\omega=\omega_0}]-\frac{5\omega_0^3}{2}\boldsymbol{\mu}_A\cdot\mathrm{Im}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B\\ &\times\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Im}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_A]+\omega_0^3\boldsymbol{\mu}_A\cdot\mathrm{Re}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B\nabla_{\mathbf{R}}[\boldsymbol{\mu}_B\cdot\mathrm{Re}\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_A]\bigg\},\end{aligned}\tag{17}$$

which contains fully resonant terms only. The leading terms in Eqs. (15)-(17) are the first ones on the right hand side of each equation, which scale linearly with T. Its asymptotic expression reads

$$\langle\mathbf{F}_A+\mathbf{F}_B\rangle_T \approx -\frac{k_0^7 e^{-\Gamma_0 T}(\boldsymbol{\mu}_A\cdot\alpha\cdot\boldsymbol{\mu}_B)^2 T}{8\pi^2\epsilon_0^2\hbar}\left[\frac{1+\cos(2k_0R)}{(k_0R)^2}-\frac{\sin(2k_0R)}{(k_0R)^3}\right]\hat{\mathbf{R}}.$$

As for the one-photon directional emission rate, taking the identical-atom limit on Eq. (13), we arrive at

$$\frac{d\Gamma_{\rm dir}}{d\Theta}=-\frac{\boldsymbol{\mu}_A\cdot(\mathbb{I}-\hat{\mathbf{k}}\otimes\hat{\mathbf{k}})\cdot\boldsymbol{\mu}_B e^{-\Gamma_0 T}}{2(\pi\epsilon_0\hbar)^2}\begin{cases} Tk_0^5[\cos(k_0R\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Im}\,\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B+\sin(k_0R\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Re}\,\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B] \\ +5c^{-1}k_0^4[\cos(k_0R\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Re}\,\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B-\sin(k_0R\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Im}\,\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B] \\ -c^{-1}k_0^5R\cos\theta[\cos(k_0R\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Im}\,\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B \\ +\sin(k_0R\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Re}\,\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B] \\ +k_0^5[\cos(k_0R\cos\theta)\frac{\partial}{\partial\omega}[\boldsymbol{\mu}_A\cdot{\rm Re}\,\mathbb{G}(kR)\cdot\boldsymbol{\mu}_B]_{\omega=\omega_0} \\ -\sin(k_0R\cos\theta)\frac{\partial}{\partial\omega}[\boldsymbol{\mu}_A\cdot{\rm Im}\,\mathbb{G}(kR)\cdot\boldsymbol{\mu}_B]_{\omega=\omega_0}] \\ +k_0^3\cos(k_0R\cos\theta)\int_0^\infty c\frac{dq}{\pi}\frac{q^2\left(q^2-k_0^2\right)}{(q^2+k_0^2)(q^2+k_0^2)}\boldsymbol{\mu}_A\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_B \quad \text{for } \cos\theta\in(0,1], \\ \\ Tk_0^5[\cos(k_0R\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Im}\,\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B+\sin(k_0R\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Re}\,\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B] \\ +2c^{-1}k_0^4[\cos(k_0R\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Re}\,\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B-\sin(k_0R\cos\theta)\boldsymbol{\mu}_A\cdot{\rm Im}\,\mathbb{G}(k_0R)\cdot\boldsymbol{\mu}_B] \\ +k_0^5[\cos(k_0R\cos\theta)\frac{\partial}{\partial\omega}[\boldsymbol{\mu}_A\cdot{\rm Re}\,\mathbb{G}(kR)\cdot\boldsymbol{\mu}_B]_{\omega=\omega_0} \\ -\sin(k_0R\cos\theta)\frac{\partial}{\partial\omega}[\boldsymbol{\mu}_A\cdot{\rm Im}\,\mathbb{G}(kR)\cdot\boldsymbol{\mu}_B]_{\omega=\omega_0}] \\ +k_0^3\cos(k_0R\cos\theta)\int_0^\infty c\frac{dq}{\pi}\frac{q^2\left(q^2-k_0^2\right)}{(q^2+k_0^2)(q^2+k_0^2)}\boldsymbol{\mu}_A\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_B \quad \text{for } \cos\theta\in[-1,0). \end{cases} \tag{18}$$

The nonreciprocal terms of Eq. (18), i.e., those which are not common for $\cos\theta\in(0,1]$ and $\cos\theta\in[-1,0)$ and those proportional to $\sin(k_0R\cos\theta)$, are in correspondence with those in Eq. (17), except for a two-photon emission contribution neglected in Eq. (18), such that

$$\langle\mathbf{F}_A+\mathbf{F}_B\rangle_T=-\langle\dot{\mathbf{P}}^{\gamma}_{\perp}\rangle_T\simeq-\hbar k_0\int_0^{4\pi}d\Theta\hat{\mathbf{k}}\frac{d\Gamma_{\rm dir}}{d\Theta}. \tag{19}$$

Two-photon emission terms together with those terms proportional to ω_0^3 in Eq. (17) are indeed negligible in comparison to the term linear in T. In Fig. 6 we represent the net force on a binary system of identical atoms as a function of the interatomic distance, once normalized as indicated. For simplicity, the dipole moments are chosen isotropic, $\mu^x_{A,B}=\mu^y_{A,B}=\mu^z_{A,B}$. Note that, in order to preserve the perturbative nature of our calculation, the following inequality must be satisfied, $\cos(k_0R)\lesssim k_0R/\Gamma_0T$. Considering the lower bound value of this inequality, $\Gamma_0T\sim1$, it implies for isotropic dipoles $k_0R\gtrsim1$, as indicated with the vertical straight line in Fig. 6. For comparison, we represent the net force on a binary system of dissimilar atoms—see Eq. (12) and Ref. [31]—which is of the order of $1/\Delta_{AB}T$ times smaller. The force on dissimilar atoms presents a maximum at $k_0R\approx1.3$, which coincides approximately with the value at which the force on the identical atoms vanishes for the first time.

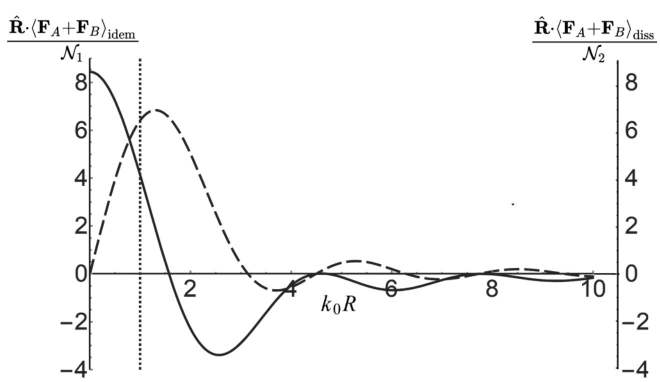

FIG. 6. Graphical representation of the net force on a binary system of identical atoms according to Eq. (17) as a function of k_0R, $\langle\mathbf{F}_A+\mathbf{F}_B\rangle_{\rm idem}$: solid curve, normalized to $\mathcal{N}_1=\frac{|\boldsymbol{\mu}_A|^2|\boldsymbol{\mu}_B|^2\omega_0^7T}{10^{-3}c^7\hbar\epsilon_0^2}$; and net force on a binary system of dissimilar atoms according to Eq. (12) and Ref. [31], $\langle\mathbf{F}_A+\mathbf{F}_B\rangle_{\rm diss}$: dashed curve, normalized to $\mathcal{N}_2=\frac{|\boldsymbol{\mu}_A|^2|\boldsymbol{\mu}_B|^2\omega_A^7}{10^{-3}c^7\hbar\epsilon_0^2\Delta_{AB}}$, with $\omega_A\approx\omega_0$.

V. DISCUSSION AND CONCLUSIONS

In the first place, starting with the perturbative time-dependent computation of the dipole-dipole interaction between two dissimilar atoms, up to two-photon exchange processes, with one of the atoms suddenly excited, we have shown that the dipole-dipole forces contain two components. These are the conservative forces identifiable with the ordinary van der Waals forces, and nonconservative forces which derive from the time variation of the longitudinal EM momentum. In contrast to previous quasistationary computations we find that, generally, the time-dependent vdW forces cannot be written as the gradients of the expectation values of the interaction potentials, but as the expectation values of the gradients of the interaction potentials only. As for the nonconservative forces, they will be computed in a separate publication [40].

Second, we have taken the identical-atom limit upon the perturbative expressions for the vdW forces on dissimilar atoms. That compels us to constrain ourselves to the weak-interaction regime. We find that, at leading order, the van der Waals forces are fully resonant and grow linearly in time, being different on each atom. Besides, in addition to the familiar off-resonant vdW forces, which change direction at $T=\ln 2/\Gamma_0$, semiresonant reciprocal forces arise; see Eqs. (15) and (16).

The resultant net force on the two-atom system is related to the directionality of spontaneous emission, which results from the violation of parity symmetry and is in agreement with total momentum conservation; see Eqs. (17), (18), and (19).

We may explain now why a stationary approach is not suitable for this computation. In the first place, note that level shifts $\delta\mathcal{E}$ can be only attributed to (quasi)stationary states upon which the identification of $\delta\mathcal{E}$ in the time-exponential factor of their wave functions can be made; cf. Ref. [22]. In the present case, that identification applies to the symmetric and antisymmetric Dicke states, $|\pm\rangle = (|A_+, B_-\rangle \pm |A_-, B_+\rangle)/\sqrt{2}$, with $\delta\mathcal{E}_{+,-} = \mp\hbar\Omega_R$, respectively, and $\Omega_R = k_0^2\boldsymbol{\mu}_A \cdot \text{Re}\mathbb{G}(k_0R) \cdot \boldsymbol{\mu}_B$ being the Rabi frequency of the excitation transfer. In first approximation, reciprocal forces on the atoms in Dicke's states can be computed, $\mathbf{F}_{A,B} = \pm\hbar\nabla\Omega_R$. However, the state $|A_+, B_-\rangle$ is not stationary, both because it is an excited state and also because it oscillates at frequency Ω_R between the symmetric and antisymmetric states. That is, if $|\Psi(0)\rangle = |A_+, B_-\rangle = (|+\rangle + |-\rangle)/\sqrt{2}$, at leading order in perturbation theory one finds $|\Psi(T)\rangle = e^{-i\omega_0 t}(e^{i\Omega_R t}|+\rangle + e^{-i\Omega_R T}|-\rangle)/\sqrt{2}$. Therefore, its level shift is not a well-defined quantity and, at best, one only could make the identification $\delta\mathcal{E}_\Psi = \langle\Psi(T)|W|\Psi(T)\rangle = (\delta\mathcal{E}_+ + \delta\mathcal{E}_-)/2 = 0$. Hence, at any time, $\langle\Psi(T)|W_{A,B}|\Psi(T)\rangle = 0$. On the other hand, it is known from the works of Refs. [19–23,31,32] that the correct calculation of the resonant interactions between excited atoms, even in the adiabatic limit, requires the appropriate introduction of the dissipative factors $e^{-\Gamma t}$ within the framework of time-dependent perturbation theory. Hence, it can be shown that it is by this means that the Rabi frequency becomes complex, cf. Refs. [41–43], $\tilde{\Omega}_R = k_0^2\boldsymbol{\mu}_A \cdot \mathbb{G}(k_0R) \cdot \boldsymbol{\mu}_B$, yielding in that case $\langle\Psi(T)|W_{A,B}|\Psi(T)\rangle \sim \text{Im}\{\tilde{\Omega}_R\}\sin(2\text{Re}\{\tilde{\Omega}_R\}T)$ which, for $\text{Re}\{\tilde{\Omega}_R\}T \ll 1$, renders the leading order term of the resonant interactions in Eqs. (15)–(17).

Thus, beyond the weak-interaction regime the calculation of the vdW forces between identical atoms becomes nonperturbative as a result of degeneracy. That implies that nonperturbative time-evolution propagators are to be computed [41,42]. Their calculation will be addressed in a separate publication, together with a proposal for the experimental observation of the net force on a binary system of Rydberg atoms.

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APPENDIX: INTERACTION ENERGIES

In this Appendix we compile the expressions for the interaction energies on each atom. Their diagrammatic representations are analogous to those in Figs. 1–4, except for the replacement of the operators $-\nabla_A W_A$ and $-\nabla_B W_B$ at the observation time T with W_A and W_B, respectively:

$$\begin{aligned}
\langle W_A\rangle_T =\ & \frac{2\omega_A^4 e^{-\Gamma_A T}}{c^4\epsilon_0^2\hbar\Delta_{AB}}[[\boldsymbol{\mu}_A\cdot\text{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B]^2 - [\boldsymbol{\mu}_A\cdot\text{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B]^2]\\
& - \frac{2\omega_B^4 e^{-(\Gamma_A+\Gamma_B)T/2}}{c^4\epsilon_0^2\hbar\Delta_{AB}}[[\boldsymbol{\mu}_A\cdot\text{Re}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B]^2 - [\boldsymbol{\mu}_A\cdot\text{Im}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B]^2]\cos(\Delta_{AB}T)\\
& + \frac{4\omega_B^4 e^{-(\Gamma_A+\Gamma_B)T/2}}{c^4\epsilon_0^2\hbar\Delta_{AB}}[\boldsymbol{\mu}_A\cdot\text{Re}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B\boldsymbol{\mu}_B\cdot\text{Im}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_A]\sin(\Delta_{AB}T)\\
& - \frac{2\omega_A^4 e^{-\Gamma_A T}}{c^4\epsilon_0^2\hbar(\omega_A+\omega_B)}[[\boldsymbol{\mu}_A\cdot\text{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B]^2 - [\boldsymbol{\mu}_A\cdot\text{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B]^2]\\
& + \frac{2\omega_B^2 e^{-(\Gamma_A+\Gamma_B)T/2}}{c^3\epsilon_0^2\hbar}[\boldsymbol{\mu}_A\cdot\text{Re}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B\cos(\Delta_{AB}T) - \boldsymbol{\mu}_A\cdot\text{Im}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B\sin(\Delta_{AB}T)]\\
& \times\int_0^\infty\frac{dq}{\pi}\frac{(q^2-k_Ak_B)q^2}{(q^2+k_A^2)(q^2+k_B^2)}\boldsymbol{\mu}_B\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_A - \frac{4\omega_A\omega_B(2e^{-\Gamma_A T}-1)}{c^3\epsilon_0^2\hbar}\int_0^\infty\frac{dq}{\pi}\frac{q^4[\boldsymbol{\mu}_A\cdot\mathbb{G}(iqR)\cdot\boldsymbol{\mu}_B]^2}{(q^2+k_A^2)(q^2+k_B^2)},
\end{aligned}\tag{A1}$$

$$\begin{aligned}
\langle W_B\rangle_T =\ & \frac{2\omega_A^4 e^{-\Gamma_A T}}{c^4\epsilon_0^2\hbar\Delta_{AB}}[[\boldsymbol{\mu}_A\cdot\text{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B]^2 + [\boldsymbol{\mu}_A\cdot\text{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B]^2]\\
& - \frac{2\omega_A^2\omega_B^2 e^{-(\Gamma_A+\Gamma_B)T/2}}{c^4\epsilon_0^2\hbar\Delta_{AB}}[\boldsymbol{\mu}_A\cdot\text{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B\boldsymbol{\mu}_A\cdot\text{Re}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B + \boldsymbol{\mu}_A\cdot\text{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B\boldsymbol{\mu}_A\cdot\text{Im}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_B]\\
& \times\cos(\Delta_{AB}T) - \frac{2\omega_A^2\omega_B^2 e^{-(\Gamma_A+\Gamma_B)T/2}}{c^4\epsilon_0^2\hbar\Delta_{AB}}[\boldsymbol{\mu}_A\cdot\text{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B\boldsymbol{\mu}_B\cdot\text{Im}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_A\\
& - \boldsymbol{\mu}_A\cdot\text{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B\boldsymbol{\mu}_B\cdot\text{Re}\mathbb{G}(k_BR)\cdot\boldsymbol{\mu}_A]\sin(\Delta_{AB}T)\\
& - \frac{2\omega_A^4 e^{-\Gamma_A T}}{c^4\epsilon_0^2\hbar(\omega_A+\omega_B)}[[\boldsymbol{\mu}_A\cdot\text{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B]^2 + [\boldsymbol{\mu}_A\cdot\text{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B]^2]\\
& + \frac{2\omega_A^2 e^{-(\Gamma_A+\Gamma_B)T/2}}{c^3\epsilon_0^2\hbar}[\boldsymbol{\mu}_A\cdot\text{Re}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B\cos(\Delta_{AB}T) + \boldsymbol{\mu}_A\cdot\text{Im}\mathbb{G}(k_AR)\cdot\boldsymbol{\mu}_B\sin(\Delta_{AB}T)]
\end{aligned}$$

$$\times \int_0^\infty \frac{dq}{\pi} \frac{(q^2 - k_A k_B) q^2}{(q^2 + k_A^2)(q^2 + k_B^2)} \boldsymbol{\mu}_B \cdot \mathbb{G}(iqR) \cdot \boldsymbol{\mu}_A - \frac{4\omega_A \omega_B (2e^{-\Gamma_A T} - 1)}{c^3 \epsilon_0^2 \hbar} \int_0^\infty \frac{dq}{\pi} \frac{q^4 [\boldsymbol{\mu}_A \cdot \mathbb{G}(iqR) \cdot \boldsymbol{\mu}_B]^2}{(q^2 + k_A^2)(q^2 + k_B^2)}. \quad \text{(A2)}$$

Straight comparison with Eqs. (10) and (11) reveals that $-\nabla_{A,B} \langle W_{A,B} \rangle_T / 2 \neq -\langle \nabla_{A,B} W_{A,B} \rangle_T / 2 = \langle \mathbf{F}_{A,B} \rangle_T$, up to two-photon exchange processes.

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