Two-atom van-der-Waals forces with one atom excited: the identical atoms limit I

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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 atoms 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. 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.

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 the last decades a renewed interest has been drawn on 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– 18]. 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 [19–25], the role of dissipation [23, 24, 26–28], its inherent time-dependence [20, 21, 23, 29–31], and the net forces induced by parity and time-reversal violation [32, 33].

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 quasistationary perturbation theory to compute the interaction. This is so because the excitation process can be taken 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. [23] that, for arbitrary values of Ω , the resultant resonant interaction contains a quasi-stationary 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 term vanishes [20], and the result is equivalent to that obtained using adiabatic timedependent perturbation theory [23]. Other approaches based on Heisenberg's formalism [22, 24, 30] and Feynman's Lagrangian formalism between asymptotic states [29, 31] lead to an equivalent quasi-stationary result. In the opposite limit, that is, for a sudden excitation with $\Omega \gg |\Delta_{AB}|$, quasi-stationary and time oscillating terms happen to be of the same order [21]. Either way, it was also found in Refs.[23, 32] 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 quasi-stationary nor adiabatic approximations make physical sense for two reasons. In the first place, the system becomes degenerate, as the states $|A_+, B_-\rangle$ and $|A_{-}, B_{+}\rangle$ posses identical energies, and the stationary sates are the symmetric and antisymmetric Dicke's states, $(|A_+, B_-\rangle \pm |A_-, B_+\rangle)/\sqrt{2}$, respectively. This implies that the use of stationary 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, 34].

In this article we will show that, starting with a binary system of dissimilar atoms, the identical atoms 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 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's states. We will show that, for two-level atoms, the van-

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der-Waals forces are dominated by fully-resonant components which grow linearly in time and are different upon each atom. Besides, in addition to the familiar offresonant van-der-Waals force, a reciprocal semi-resonant force arises. Interestingly, the time-dependent forces do not derive 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 non-reciprocal components of the force are explained in terms of parity symmetry violation, which generate an asymmetry in the probability of emission of photons from either atom. The effect of the de-excitation upon the off-

The article is organized as follows. In Sec.II we perform the computation of the vdW forces between two dissimilar two-level atoms, one of which is suddenly excited. The origin of non-reciprocal forces is related to the directionality of spontaneous emission. In Sec.III the identical atoms limit is considered in the weak-interaction regime. The conclusions are summarized in Sec.IV together with a discussion on the extension of our results.

resonant van-der-Waals force is also analyzed.

II. VDW INTERACTION OF TWO DISSIMILAR ATOMS AFTER A SUDDEN EXCITATION

Let us consider two atoms, A and B, located a distance R apart. Since we are ultimately interested in the identical atoms limit, $|\Delta_{AB}| \ll \Gamma_A$, $\Gamma_A \to \Gamma_B$, atom Ais assumed to be suddenly excited with an external field of strength $\Omega \gg |\Delta_{AB}|$. This is the situation considered in Ref.[21], where the calculation was restricted to quasiresonant 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.

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/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 > 0the 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,

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

In this equation $H_A + H_B$ 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

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

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

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

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,

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

where \mathcal{V} is a generic volume and $\mathbf{E}_{\mathbf{k}}^{(\mp)}$ denote the annihilation/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 Röntgen term [35]. As argued in Ref.[36], 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), with $m_{A,B}$ being the atomic masses.

Next, considering W as a perturbation to the free Hamiltonians, the unperturbed time propagator for atom and free photon states is $\mathbb{U}_0(t) =$ $\exp\left[-i\hbar^{-1}(H_A + H_B + H_{EM})t\right]$. 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,

$$\mathbb{U}(T) = \mathbb{U}_0(T) \,\mathcal{T} \exp \int_0^T (-i/\hbar) \mathbb{U}_0^{\dagger}(t) \,W \,\mathbb{U}_0(t) \mathrm{d}t, \quad (3)$$

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 [37, 38], $[H, \mathbf{K}] = \mathbf{0}$,

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

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

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

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 [37], $\mathbf{P}_{\parallel}^{\gamma} = \sum_{i} q_{i} \mathbf{A}(\mathbf{r}_{i})$. Lastly, in the electric dipole approximation, $\mathbf{P}_{\parallel}^{\gamma}$ reads [35], $\mathbf{P}_{\parallel}^{\gamma} \simeq -\mathbf{d}_{A} \times \mathbf{B}(\mathbf{R}_{A}) - \mathbf{d}_{B} \times \mathbf{B}(\mathbf{R}_{B})$, where $\mathbf{B}(\mathbf{R}_{A,B}) = \nabla_{A,B} \times \mathbf{A}(\mathbf{R}_{A,B})$.

Following Refs. [32, 36], 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, we arrive at

$$\langle \mathbf{F}_{A,B} \rangle_T = \partial_T \langle m_{A,B} \dot{\mathbf{R}}_{A,B} \rangle_T$$

$$= -i\hbar \partial_T \langle \Psi(0) | \mathbb{U}^{\dagger}(T) \nabla_{A,B} \mathbb{U}(T) | \Psi(0) \rangle$$

$$+ \partial_T \langle \Psi(0) | \mathbb{U}^{\dagger}(T) \mathbf{d}_{A,B} \times \mathbf{B}(\mathbf{R}_{A,B}) \mathbb{U}(T) | \Psi(0) \rangle$$

$$= - \langle \nabla_{A,B} W_{A,B} \rangle_T + \partial_T \langle \mathbf{d}_{A,B} \times \mathbf{B}(\mathbf{R}_{A,B}) \rangle_T,$$

$$(6)$$

The first term on the right hand side of last equality is a conservative force along the interatomic axis, which we will refer to as vdW force. Note hower that, in contrast to the stationary vdW forces computed in the adiabatic approximation -cf. Ref. [23], time-dependent conservative forces cannot be generally written as $\frac{-1}{2} \nabla_{A,B} \langle W_{A,B} \rangle_T$. We will show latter, including up to two-photon exchange processes, that the reason is the functional dissymmetry in the contribution of the two photons to the timedependent terms. The second term is a non-conservative force equivalent to the time derivative of the longitudinal EM momentum at each atom, with opposite sign. We will show in a separate publication [39] 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 $\langle \mathbf{F}_{A,B} \rangle_T \simeq - \langle \nabla_{A,B} W_{A,B} \rangle_T$.

A perturbative development of Eq.(6) shows that, up to terms involving two-photon exchange processes, twenty-four diagrams contribute to $\langle \mathbf{F}_A \rangle_T$ for a two-level atom. They are depicted in Fig.1 and Fig.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 de-excitation of the system via spontaneous emission from atom A.



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.

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,

$$\frac{1}{\hbar^{3}} \int_{0}^{\infty} \frac{\mathcal{V}k^{2} \mathrm{d}k}{(2\pi)^{3}} \int_{0}^{\infty} \frac{\mathcal{V}k^{\prime 2} \mathrm{d}k^{\prime}}{(2\pi)^{3}} \int_{0}^{4\pi} \mathrm{d}\Theta \int_{0}^{4\pi} \mathrm{d}\Theta^{\prime} \Big\{ \Big[i\langle A_{+}, B_{-}, 0_{\gamma} | e^{i\Omega_{a}^{*}T} | A_{+}, B_{-}, 0_{\gamma} \rangle \int_{-\infty}^{T} \mathrm{d}t \int_{-\infty}^{t} \mathrm{d}t^{\prime} \int_{-\infty}^{t^{\prime}} \mathrm{d}t^{\prime\prime} \\
\times \langle A_{+}, B_{-}, 0_{\gamma} | - \nabla_{A} [\mathbf{d}_{A} \cdot \mathbf{E}_{\mathbf{k}^{\prime}}^{(-)}(\mathbf{R}_{A})] | A_{-}, B_{-}, \gamma_{k^{\prime}} \rangle e^{-i\omega^{\prime}(T-t)} \langle A_{-}, B_{-}, \gamma_{k^{\prime}} | \mathbf{d}_{B} \cdot \mathbf{E}_{\mathbf{k}^{\prime}}^{(+)}(\mathbf{R}_{B}) | A_{-}, B_{+}, 0_{\gamma} \rangle \\
\times e^{-i\Omega_{b}(t-t^{\prime})} \langle A_{-}, B_{+}, 0_{\gamma} | \mathbf{d}_{B} \cdot \mathbf{E}_{\mathbf{k}}^{(-)}(\mathbf{R}_{B}) | A_{-}, B_{-}, \gamma_{k} \rangle e^{-i\omega(t^{\prime}-t^{\prime\prime})} \langle A_{-}, B_{-}, \gamma_{k} | \mathbf{d}_{A} \cdot \mathbf{E}_{\mathbf{k}}^{(+)}(\mathbf{R}_{A}) | A_{+}, B_{-}, 0_{\gamma} \rangle e^{-i\Omega_{a}t^{\prime\prime}} \Big] \\
+ [k \leftrightarrow k^{\prime}]^{\dagger} \Big\},$$
(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, $|\gamma_k\rangle$ is a one-photon state of momentum **k** and frequency $\omega = ck$, the complex timeexponentials are the result of the application of the free time-evolution operator $U_0(t) = e^{-i\hbar^{-1}H_0t}$ 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. After integrating in time and solid angles, one arrives at

$$\frac{e^{2}\hbar^{-1}}{\pi^{2}\epsilon_{0}^{2}}\operatorname{Re}\int_{0}^{\infty}\mathrm{d}k'k'^{2}\boldsymbol{\nabla}_{A}[\boldsymbol{\mu}_{A}\cdot\operatorname{Im}\mathbb{G}(k'R)\cdot\boldsymbol{\mu}_{B}]\int_{0}^{\infty}\mathrm{d}k\,k^{2}\boldsymbol{\mu}_{B}\cdot\operatorname{Im}\mathbb{G}(kR)\cdot\boldsymbol{\mu}_{A}\,e^{i\Omega_{a}^{*}T}\Big[\frac{e^{-i\Omega_{a}T}-e^{-i\omega T}}{(\omega'-\Omega_{a})(\Omega_{b}-\Omega_{a})(\omega-\Omega_{a})}-\frac{e^{-i\Omega_{b}T}-e^{-i\omega T}}{(\omega'-\Omega_{a})(\Omega_{b}-\Omega_{a})(\omega-\Omega_{b})}\Big]$$

$$(8)$$



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.

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 **R** by an electric dipole of frequency $\omega = ck$ placed at the origin. It reads

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

where the tensors α and β read $\alpha = \mathbb{I} - \mathbf{RR}/R^2$, $\beta = \mathbb{I} - 3\mathbf{RR}/R^2$.

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 $\nabla_B = -\nabla_A = -\nabla_R$, we arrive at

$$\langle \mathbf{F}_{A} \rangle_{T} = -\frac{2\omega_{A}^{A}e^{-\Gamma_{A}T}}{c^{4}\epsilon_{0}^{2}\hbar\Delta_{AB}} \Big[\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{B} \boldsymbol{\nabla}_{\mathbf{R}} \big[\boldsymbol{\mu}_{B} \cdot \operatorname{Re}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{A} \Big] - \boldsymbol{\mu}_{A} \cdot \operatorname{Im}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{B} \boldsymbol{\nabla}_{\mathbf{R}} \big[\boldsymbol{\mu}_{B} \cdot \operatorname{Im}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{A} \Big] \\ + \frac{2\omega_{B}^{A}e^{-(\Gamma_{A}+\Gamma_{B})T/2}}{c^{4}\epsilon_{0}^{2}\hbar\Delta_{AB}} \Big[\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{B} \boldsymbol{\nabla}_{\mathbf{R}} \big[\boldsymbol{\mu}_{B} \cdot \operatorname{Re}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{A} \Big] \\ - \frac{2\omega_{A}^{A}e^{-(\Gamma_{A}+\Gamma_{B})T/2}}{c^{4}\epsilon_{0}^{2}\hbar\Delta_{AB}} \Big[\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{B} \boldsymbol{\nabla}_{\mathbf{R}} \big[\boldsymbol{\mu}_{B} \cdot \operatorname{Im}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{A} \Big] \\ - \frac{2\omega_{A}^{A}e^{-(\Gamma_{A}+\Gamma_{B})T/2}}{c^{4}\epsilon_{0}^{2}\hbar\Delta_{AB}} \Big[\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{B} \boldsymbol{\nabla}_{\mathbf{R}} \big[\boldsymbol{\mu}_{B} \cdot \operatorname{Im}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{A} \Big] \\ + \frac{2\omega_{A}^{A}e^{-\Gamma_{A}T}}{c^{4}\epsilon_{0}^{2}\hbar\Delta_{AB}} \Big[\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{B} \boldsymbol{\nabla}_{\mathbf{R}} \big[\boldsymbol{\mu}_{B} \cdot \operatorname{Im}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{A} \Big] \\ + \frac{2\omega_{A}^{A}e^{-\Gamma_{A}T}}{c^{4}\epsilon_{0}^{2}\hbar(\omega_{A}+\omega_{B})} \Big[\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{B} \boldsymbol{\nabla}_{\mathbf{R}} \big[\boldsymbol{\mu}_{B} \cdot \operatorname{Re}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{A} \Big] \\ - \frac{2\omega_{B}^{2}e^{-(\Gamma_{A}+\Gamma_{B})T/2}}{c^{4}\epsilon_{0}^{2}\hbar(\omega_{A}+\omega_{B})} \Big[\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{B} \boldsymbol{\nabla}_{\mathbf{R}} \big[\boldsymbol{\mu}_{B} \cdot \operatorname{Re}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{B} \Big] \cos(\Delta_{AB}T) \\ - \frac{2\omega_{B}^{2}e^{-(\Gamma_{A}+\Gamma_{B})T/2}}{c^{4}\epsilon_{0}^{2}\hbar(\omega_{A}+\omega_{B})} \Big[\boldsymbol{\nabla}_{\mathbf{R}} \big[\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{B} \big] \cos(\Delta_{AB}T) - \boldsymbol{\nabla}_{\mathbf{R}} \big[\boldsymbol{\mu}_{A} \cdot \operatorname{Im}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{B} \boldsymbol{\nabla}_{\mathbf{R}} \big[\boldsymbol{\mu}_{B} \cdot \operatorname{Im}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{A} \big] \Big] \\ - \frac{2\omega_{B}^{2}e^{-(\Gamma_{A}+\Gamma_{B})T/2}}{c^{3}\epsilon_{0}^{2}\hbar} \Big[\boldsymbol{\nabla}_{\mathbf{R}} \big[\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{B} \big] \cos(\Delta_{AB}T) - \boldsymbol{\nabla}_{\mathbf{R}} \big[\boldsymbol{\mu}_{A} \cdot \operatorname{Im}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{B} \big] \sin(\Delta_{AB}T) \Big] \\ \times \int_{0}^{\infty} \frac{dq}{\pi} \frac{dq}{(q^{2}+k_{A})(q^{2}+k_{B}^{2})} \mathbf{\nabla}_{\mathbf{R}} \big[\boldsymbol{\mu}_{B} \cdot \mathbb{G}(iqR) \cdot \boldsymbol{\mu}_{A} \big]$$
 (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 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 ~ $1/\Delta_{AB}$, 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 $1/(\omega_A + \omega_B)$, 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 to diagrams (i,k) and (i,j,k,l), respectively, which contain two-photon intermediate states.

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,



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.

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 $\operatorname{Re}\mathbb{G}(k_{A,B}R)\operatorname{Im}\mathbb{G}(k_{A,B}R)$ –for comparison, see also the expressions for $\langle W_{A,B} \rangle_T$ in the Appendix.



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.

Comparing equations (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 $T \approx \log 2/\Gamma_A$. As for the rest of the terms, some fully resonant and semi-resonant terms, either stationary or oscillating at frequency Δ_{AB} , differ in sign. Those terms constitute non-reciprocal forces and amount to a net force on the two-atom system. The stationary non-reciprocal forces were shown in Ref.[32] 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

$$\langle \mathbf{F}_{A} + \mathbf{F}_{B} \rangle_{T} \simeq \frac{8e^{-\Gamma_{A}T}k_{A}^{4}}{\epsilon_{0}^{2}\hbar} \frac{\omega_{B}}{\omega_{A}^{2} - \omega_{B}^{2}} \boldsymbol{\mu}_{A} \cdot \operatorname{Im}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{B} \nabla_{\mathbf{R}} \left[\boldsymbol{\mu}_{B} \cdot \operatorname{Im}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{A}\right]$$

$$+ \frac{2e^{-(\Gamma_{A} + \Gamma_{B})T/2}k_{B}^{2}}{\epsilon_{0}^{2}\hbar\Delta_{AB}} \left\{\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{B} \nabla_{\mathbf{R}} \left[k_{B}^{2}\boldsymbol{\mu}_{B} \cdot \operatorname{Re}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{A} - k_{A}^{2}\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{B}\right]$$

$$- \boldsymbol{\mu}_{A} \cdot \operatorname{Im}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{B} \nabla_{\mathbf{R}} \left[k_{B}^{2}\boldsymbol{\mu}_{B} \cdot \operatorname{Im}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{A} + k_{A}^{2}\boldsymbol{\mu}_{A} \cdot \operatorname{Im}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{B}\right] \right\} \cos\left(\Delta_{AB}T\right)$$

$$- \frac{2e^{-(\Gamma_{A} + \Gamma_{B})T/2}k_{B}^{2}}{\epsilon_{0}^{2}\hbar\Delta_{AB}} \left\{\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{B} \nabla_{\mathbf{R}} \left[k_{B}^{2}\boldsymbol{\mu}_{B} \cdot \operatorname{Im}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{A} + k_{A}^{2}\boldsymbol{\mu}_{A} \cdot \operatorname{Im}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{B}\right] \right\} \sin\left(\Delta_{AB}T\right)$$

$$- \frac{2e^{-(\Gamma_{A} + \Gamma_{B})T/2}}{\epsilon_{0}^{2}\hbar\Delta_{AB}} \left\{\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{A} - k_{A}^{2}\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{B}\right\} \right\} \sin\left(\Delta_{AB}T\right)$$

$$- \frac{2e^{-(\Gamma_{A} + \Gamma_{B})T/2}}{\epsilon^{3}\epsilon_{0}^{2}\hbar} \int_{0}^{\infty} \frac{dq}{\pi} \frac{(q^{2} - k_{A}k_{B})q^{2}\boldsymbol{\mu}_{A} \cdot \mathbb{G}(iqR) \cdot \boldsymbol{\mu}_{B}}{(q^{2} + k_{A}^{2})(q^{2} + k_{B}^{2})} \left[\nabla_{\mathbf{R}}[\omega_{B}^{2}\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{B} - \omega_{A}^{2}\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{B} \right]$$

$$\times \cos(\Delta_{AB}T) - \nabla_{\mathbf{R}}[\omega_{B}^{2}\boldsymbol{\mu}_{A} \cdot \operatorname{Im}\mathbb{G}(k_{B}R) \cdot \boldsymbol{\mu}_{B} + \omega_{A}^{2}\boldsymbol{\mu}_{A} \cdot \operatorname{Im}\mathbb{G}(k_{A}R) \cdot \boldsymbol{\mu}_{B}] \sin\left(\Delta_{AB}T\right) \right],$$

$$(12)$$

where the first non-oscillating term coincides with the net force for the case of an adiabatic excitation [23].

In what follows we study the directionality of onephoton 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{\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}} \left[\cos(\Delta_{AB}T) \left[\cos(k_B R \cos\theta) \boldsymbol{\mu}_A \cdot \operatorname{Re} \mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_B - \sin(k_B R \cos\theta) \right] \\ \times \boldsymbol{\mu}_A \cdot \operatorname{Im} \mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_B \right] - \sin(\Delta_{AB}T) \left[\cos(k_B R \cos\theta) \boldsymbol{\mu}_A \cdot \operatorname{Im} \mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_B \right] \\ + \sin(k_B R \cos\theta) \boldsymbol{\mu}_A \cdot \operatorname{Re} \mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_B \right] \right] + \frac{e^{-\Gamma_A T k_A^5}}{\Delta_{AB}} \sin(k_A R \cos\theta) \boldsymbol{\mu}_A \cdot \operatorname{Im} \mathbb{G}(k_A R) \cdot \boldsymbol{\mu}_B \\ - e^{-(\Gamma_A + \Gamma_B)T/2} k_B^3 \left[\cos(\Delta_{AB}T) \cos(k_B R \cos\theta) - \sin(\Delta_{AB}T) \sin(k_B R \cos\theta) \right] \\ \times \int_0^\infty c \frac{dq}{\pi} \frac{q^2 (q^2 - k_A k_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^2 k_A^3}{\Delta_{AB}} \left[\cos(\Delta_{AB}T) \left[\cos(k_A R \cos\theta) \boldsymbol{\mu}_A \cdot \operatorname{Re} \mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_B - \sin(k_A R \cos\theta) \right] \\ \times \mu_A \cdot \operatorname{Im} \mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_B \right] - \sin(\Delta_{AB}T) \left[\cos(k_A R \cos\theta) \boldsymbol{\mu}_A \cdot \operatorname{Im} \mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_B \right] \\ + \sin(k_A R \cos\theta) \boldsymbol{\mu}_A \cdot \operatorname{Re} \mathbb{G}(k_B R) \cdot \boldsymbol{\mu}_B \right] \right] + \frac{e^{-\Gamma_A T k_A^5}}{\Delta_{AB}} \sin(k_A R \cos\theta) \boldsymbol{\mu}_A \cdot \operatorname{Im} \mathbb{G}(k_A R) \cdot \boldsymbol{\mu}_B \\ - e^{-(\Gamma_A + \Gamma_B)T/2} k_B^3 \left[\cos(\Delta_{AB}T) \cos(k_A R \cos\theta) - \sin(\Delta_{AB}T) \sin(k_A R \cos\theta) \right] \\ \times \int_0^\infty c \frac{dq}{\pi} \frac{q^2 (q^2 - k_A k_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}$$
(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, $d\Gamma_{\rm dir}/d\Theta$.

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,

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

Straight integration of this equation leads to Eq.(12) –up to two-photon emission terms– but 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 **K** defined in Eq.(4).

III. THE IDENTICAL ATOMS LIMIT IN THE WEAK-INTERACTION REGIME

We proceed to take the identical atoms 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.II 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., $k_0^2 \mu_A \cdot \text{ReG}(k_0 R) \cdot \mu_B \lesssim \hbar \epsilon_0 / T$ [2, 34]. This is the weak-interaction regime, which implies that the original atomic states are quasi-stationary despite the degeneracy of the system. In this limit, the vdW forces read

$$\langle \mathbf{F}_{A} \rangle_{T} = -\frac{2\omega_{0}^{4}e^{-\Gamma_{0}T}}{c^{4}\epsilon_{0}^{2}\hbar} T \left[\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{B} \boldsymbol{\nabla}_{\mathbf{R}} \left[\boldsymbol{\mu}_{B} \cdot \operatorname{Im}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{A} \right] + \boldsymbol{\mu}_{A} \cdot \operatorname{Im}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{B} \boldsymbol{\nabla}_{\mathbf{R}} \left[\boldsymbol{\mu}_{B} \cdot \operatorname{Re}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{A} \right] \right]$$

$$- \frac{2e^{-\Gamma_{0}T}}{c^{4}\epsilon_{0}^{2}\hbar} \frac{\partial}{\partial\omega} \left[\omega^{4}\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(kR) \cdot \boldsymbol{\mu}_{B} \boldsymbol{\nabla}_{\mathbf{R}} \left[\boldsymbol{\mu}_{B} \cdot \operatorname{Re}\mathbb{G}(kR) \cdot \boldsymbol{\mu}_{A} \right] - \omega^{4}\boldsymbol{\mu}_{A} \cdot \operatorname{Im}\mathbb{G}(kR) \cdot \boldsymbol{\mu}_{B} \boldsymbol{\nabla}_{\mathbf{R}} \left[\boldsymbol{\mu}_{B} \cdot \operatorname{Im}\mathbb{G}(kR) \cdot \boldsymbol{\mu}_{A} \right] \right] \right]$$

$$+ \frac{\omega_{0}^{3}e^{-\Gamma_{0}T}}{c^{4}\epsilon_{0}^{2}\hbar} \left[\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{B} \boldsymbol{\nabla}_{\mathbf{R}} \left[\boldsymbol{\mu}_{B} \cdot \operatorname{Re}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{A} \right] - \boldsymbol{\mu}_{A} \cdot \operatorname{Im}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{B} \boldsymbol{\nabla}_{\mathbf{R}} \left[\boldsymbol{\mu}_{B} \cdot \operatorname{Im}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{A} \right] \right]$$

$$- \frac{2\omega_{0}^{2}e^{-\Gamma_{0}T}}{c^{3}\epsilon_{0}^{2}\hbar} \boldsymbol{\nabla}_{\mathbf{R}} \left[\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{B} \right] \int_{0}^{\infty} \frac{dq}{\pi} \frac{(q^{2} - k_{0}^{2})q^{2}\boldsymbol{\mu}_{A} \cdot \mathbb{G}(iqR) \cdot \boldsymbol{\mu}_{B}}{(q^{2} + k_{0}^{2})^{2}}$$

$$+ \frac{4\omega_{0}^{2}\left(1 - 2e^{-\Gamma_{0}T}\right)}{c^{3}\epsilon_{0}^{2}\hbar} \int_{0}^{\infty} \frac{dq}{\pi} \frac{q^{4}\boldsymbol{\mu}_{A} \cdot \mathbb{G}(iqR) \cdot \boldsymbol{\mu}_{B}}{(q^{2} + k_{0}^{2})^{2}} \boldsymbol{\nabla}_{\mathbf{R}} \left[\boldsymbol{\mu}_{B} \cdot \mathbb{G}(iqR) \cdot \boldsymbol{\mu}_{A} \right],$$

$$(15)$$

$$\langle \mathbf{F}_{B} \rangle_{T} = \frac{2\omega_{0}^{4}e^{-\Gamma_{0}T}}{c^{4}\epsilon_{0}^{2}\hbar} T \left[\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{B} \nabla_{\mathbf{R}} \left[\boldsymbol{\mu}_{B} \cdot \operatorname{Im}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{A} \right] - \boldsymbol{\mu}_{A} \cdot \operatorname{Im}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{B} \nabla_{\mathbf{R}} \left[\boldsymbol{\mu}_{B} \cdot \operatorname{Re}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{A} \right] \right]$$

$$+ \frac{2e^{-\Gamma_{0}T}\omega_{0}^{2}}{c^{4}\epsilon_{0}^{2}\hbar} \left[\frac{\partial}{\partial\omega} \left[\omega^{2}\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(kR) \cdot \boldsymbol{\mu}_{B} \right]_{\omega=\omega_{0}} \nabla_{\mathbf{R}} \left[\boldsymbol{\mu}_{B} \cdot \operatorname{Re}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{A} \right]$$

$$+ \frac{\partial}{\partial\omega} \left[\omega^{2}\boldsymbol{\mu}_{A} \cdot \operatorname{Im}\mathbb{G}(kR) \cdot \boldsymbol{\mu}_{B} \right]_{\omega=\omega_{0}} \nabla_{\mathbf{R}} \left[\boldsymbol{\mu}_{B} \cdot \operatorname{Im}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{A} \right]$$

$$- \frac{\omega_{0}^{3}e^{-\Gamma_{0}T}}{c^{4}\epsilon_{0}^{2}\hbar} \left[\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{B} \nabla_{\mathbf{R}} \left[\boldsymbol{\mu}_{B} \cdot \operatorname{Re}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{A} \right] + \boldsymbol{\mu}_{A} \cdot \operatorname{Im}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{B} \nabla_{\mathbf{R}} \left[\boldsymbol{\mu}_{B} \cdot \operatorname{Im}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{A} \right]$$

$$+ \frac{2\omega_{0}^{2}e^{-\Gamma_{0}T}}{c^{4}\epsilon_{0}^{2}\hbar} \left[\boldsymbol{\mu}_{A} \cdot \operatorname{Re}\mathbb{G}(k_{0}R) \cdot \boldsymbol{\mu}_{B} \right] \int_{0}^{\infty} \frac{dq}{\pi} \frac{(q^{2} - k_{0}^{2})q^{2}\boldsymbol{\mu}_{A} \cdot \mathbb{G}(iqR) \cdot \boldsymbol{\mu}_{B}}{(q^{2} + k_{0}^{2})^{2}}$$

$$- \frac{4\omega_{0}^{2}\left(1 - 2e^{-\Gamma_{0}T}\right)}{c^{3}\epsilon_{0}^{2}\hbar} \int_{0}^{\infty} \frac{dq}{\pi} \frac{q^{4}\boldsymbol{\mu}_{A} \cdot \mathbb{G}(iqR) \cdot \boldsymbol{\mu}_{B}}{(q^{2} + k_{0}^{2})^{2}} \nabla_{\mathbf{R}} \left[\boldsymbol{\mu}_{B} \cdot \mathbb{G}(iqR) \cdot \boldsymbol{\mu}_{A} \right],$$

$$(16)$$



FIG. 6: Graphical representation of the net force on a binary system of identical atoms according to Eq.(17) as a function of $k_0 R$, $\langle \mathbf{F}_A + \mathbf{F}_B \rangle_{\text{idem}}$ -solid curve, normalized to $\mathcal{N}_1 = \frac{|\boldsymbol{\mu}_A|^2 |\boldsymbol{\mu}_B|^2 \omega_0^7 T}{10^2 c^7 \hbar \epsilon_0^2}$; and net force on a binary system of dissimilar atoms according to Ref.[32], $\langle \mathbf{F}_A + \mathbf{F}_B \rangle_{\text{diss}}$ -dashed curve, normalized to $\mathcal{N}_2 = \frac{|\boldsymbol{\mu}_A|^2 |\boldsymbol{\mu}_B|^2 \omega_A^7}{10^2 c^7 \hbar \epsilon_0^2 \Delta_{AB}}$, with $\omega_A \approx \omega_0$.

and the net force upon the atomic system is

$$\langle \mathbf{F}_A + \mathbf{F}_B \rangle_T = -\frac{4e^{-\Gamma_0 T}}{c^4 \epsilon_0^2 \hbar} \tag{17}$$

$$\times \left\{ \omega_0^4 T \left[\boldsymbol{\mu}_A \cdot \operatorname{Re}\mathbb{G}(k_0 R) \cdot \boldsymbol{\mu}_B \boldsymbol{\nabla}_{\mathbf{R}} \left[\boldsymbol{\mu}_B \cdot \operatorname{Im}\mathbb{G}(k_0 R) \cdot \boldsymbol{\mu}_A \right] \right. \\ \left. - \omega_0^4 \frac{\partial}{\partial \omega} \left[\boldsymbol{\mu}_A \cdot \operatorname{Im}\mathbb{G}(k R) \cdot \boldsymbol{\mu}_B \boldsymbol{\nabla}_{\mathbf{R}} \left[\boldsymbol{\mu}_B \cdot \operatorname{Im}\mathbb{G}(k R) \cdot \boldsymbol{\mu}_A \right] \right]_{\omega = \omega_0} \right. \\ \left. - \frac{5\omega_0^3}{2} \boldsymbol{\mu}_A \cdot \operatorname{Im}\mathbb{G}(k_0 R) \cdot \boldsymbol{\mu}_B \boldsymbol{\nabla}_{\mathbf{R}} \left[\boldsymbol{\mu}_B \cdot \operatorname{Im}\mathbb{G}(k_0 R) \cdot \boldsymbol{\mu}_A \right] \right. \\ \left. + \omega_0^3 \boldsymbol{\mu}_A \cdot \operatorname{Re}\mathbb{G}(k_0 R) \cdot \boldsymbol{\mu}_B \boldsymbol{\nabla}_{\mathbf{R}} \left[\boldsymbol{\mu}_B \cdot \operatorname{Re}\mathbb{G}(k_0 R) \cdot \boldsymbol{\mu}_A \right] \right\},$$

which contains fully resonant terms only.

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

$$\frac{d\Gamma_{\text{dir}}}{d\Theta} = \frac{-\boldsymbol{\mu}_A \cdot (\mathbb{I} - \hat{\mathbf{k}} \otimes \hat{\mathbf{k}}) \cdot \boldsymbol{\mu}_B}{2(\pi\epsilon_0 \hbar)^2} e^{-\Gamma_0 T} \Big\{ Tk_0^5 \sin(k_0 R \cos \theta) \boldsymbol{\mu}_A \cdot \text{Re}\,\mathbb{G}(k_0 R) \cdot \boldsymbol{\mu}_B \\
- k_0^5 \sin(k_0 R \cos \theta) \frac{\partial}{\partial \omega} [\boldsymbol{\mu}_A \cdot \text{Im}\,\mathbb{G}(kR) \cdot \boldsymbol{\mu}_B]_{\omega=\omega_0} - 2c^{-1}k_0^4 \sin(k_0 R \cos \theta) \boldsymbol{\mu}_A \cdot \text{Im}\,\mathbb{G}(k_0 R) \cdot \boldsymbol{\mu}_B \\
+ H(\cos \theta) \Big[3c^{-1}k_0^4 [\cos(k_0 R \cos \theta) \boldsymbol{\mu}_A \cdot \text{Re}\,\mathbb{G}(k_0 R) \cdot \boldsymbol{\mu}_B - \sin(k_0 R \cos \theta) \boldsymbol{\mu}_A \cdot \text{Im}\,\mathbb{G}(k_0 R) \cdot \boldsymbol{\mu}_B] \\
- c^{-1}Rk_0^5 \cos \theta [\sin(k_0 R \cos \theta) \boldsymbol{\mu}_A \cdot \text{Re}\,\mathbb{G}(k_0 R) \cdot \boldsymbol{\mu}_B + \cos(k_0 R \cos \theta) \boldsymbol{\mu}_A \cdot \text{Im}\,\mathbb{G}(k_0 R) \cdot \boldsymbol{\mu}_B] \Big] \Big\},$$
(18)

where H is the Heaviside function. The terms of Eq.(18) are in correspondence with those in Eq.(17) –but for two-photon emission terms, such that

$$\langle \mathbf{F}_A + \mathbf{F}_B \rangle_T = -\langle \dot{\mathbf{P}}_{\perp}^{\gamma} \rangle_T \simeq -\hbar k_0 \int_0^{4\pi} d\Theta \hat{\mathbf{k}} \frac{d\Gamma_{\text{dir}}}{d\Theta}.$$
 (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, $\mu_{A,B}^x = \mu_{A,B}^{y^{-1}} = \mu_{A,B}^{z}$. Note that, in order to preserve the perturbative nature of our calculation, the following inequality must be satisfied, $24\pi \operatorname{Tr}\{\operatorname{Re} \mathbb{G}(k_0 R)\} \lesssim k_0/\Gamma_0 T$. Considering the lower bound value of this inequality, $\Gamma_0 T \sim 1$, it implies for isotropic dipoles $k_0 R \gtrsim 1.3$, as indicated with the vertical straight line in Fig.6. That implies that the actual maximum value of the net force on identical atoms is achieved at $k_0 R \approx 2.5$. For comparison, we represent the net force on a binary system of dissimilar atoms [32], which is of the order of $1/\Delta_{AB}T$ times smaller. The force on dissimilar atoms presents a maximum at $k_0 R \approx 1.3$, which coincides approximately with the value at which the force on the identical atoms vanishes for the first time.

IV. CONCLUSIONS

In the first place, starting with the perturbive timedependent 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. Namely, 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 quasi-stationary 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 non-conservative forces, they will be computed in a separate publication [39].

Second, we have taken the identical atoms limit upon the perturbative expressions for the vdW forces on dissimilar atoms. That compels us to constraint 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 $T = \log 2/\Gamma_0$, semi-resonant reciprocal forces arise -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 -Eqs.(18) and (19).

Beyond the weak-interaction regime the calculation of the vdW forces between identical atoms becomes nonperturbative as a result of degeneracy. That implies that non-perturbative time-evolution propagators are to be computed [40, 41]. 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 A: 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, but 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{split} \langle W_A \rangle_T &= \frac{2\omega_A^4 e^{-\Gamma_A T}}{e^4 \epsilon_0^2 \hbar \Delta_{AB}} \Big[[\mu_A \cdot \operatorname{ReG}(k_A R) \cdot \mu_B]^2 - [\mu_A \cdot \operatorname{ImG}(k_A R) \cdot \mu_B]^2 \Big] \\ &- \frac{2\omega_B^4 e^{-(\Gamma_A + \Gamma_B)T/2}}{e^4 \epsilon_0^2 \hbar \Delta_{AB}} \Big[[\mu_A \cdot \operatorname{ReG}(k_B R) \cdot \mu_B]^2 - [\mu_A \cdot \operatorname{ImG}(k_B R) \cdot \mu_B]^2 \Big] \cos(\Delta_{AB} T) \\ &+ \frac{4\omega_B^4 e^{-(\Gamma_A + \Gamma_B)T/2}}{e^4 \epsilon_0^2 \hbar \Delta_{AB}} \Big[[\mu_A \cdot \operatorname{ReG}(k_B R) \cdot \mu_B \mu_B \cdot \operatorname{ImG}(k_B R) \cdot \mu_A] \sin(\Delta_{AB} T) \\ &- \frac{2\omega_A^4 e^{-\Gamma_A T}}{e^4 \epsilon_0^2 \hbar (\mu_A + \omega_B)} \Big[[\mu_A \cdot \operatorname{ReG}(k_B R) \cdot \mu_B]^2 - [\mu_A \cdot \operatorname{ImG}(k_A R) \cdot \mu_B]^2 \Big] \\ &+ \frac{2\omega_B^2 e^{-(\Gamma_A + \Gamma_B)T/2}}{e^4 \epsilon_0^2 \hbar (\mu_A + \omega_B)} \Big[[\mu_A \cdot \operatorname{ReG}(k_B R) \cdot \mu_B \cos(\Delta_{AB} T) - \mu_A \cdot \operatorname{ImG}(k_B R) \cdot \mu_B \sin(\Delta_{AB} T)] \\ &\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)} \mu_B \cdot \mathbb{G}(iqR) \cdot \mu_A - \frac{4\omega_A \omega_B (2e^{-\Gamma_A T} - 1)}{e^3 \epsilon_0^2 \hbar} \int_0^\infty \frac{dq}{\pi} \frac{q^4 [\mu_A \cdot \mathbb{G}(iqR) \cdot \mu_B]^2}{(q^2 + k_A^2)(q^2 + k_B^2)}, \quad (A1) \\ \langle W_B \rangle_T &= \frac{2\omega_A^4 e^{-\Gamma_A T}}{e^4 \epsilon_0^2 \hbar \Delta_{AB}} \Big[[\mu_A \cdot \operatorname{ReG}(k_A R) \cdot \mu_B]^2 + [\mu_A \cdot \operatorname{ImG}(k_A R) \cdot \mu_B]^2 \Big] \\ &- \frac{2\omega_A^2 \omega_B^2 e^{-(\Gamma_A + \Gamma_B)T/2}}{e^4 \epsilon_0^2 \hbar \Delta_{AB}} \Big[\mu_A \cdot \operatorname{ReG}(k_A R) \cdot \mu_B \mu_A \cdot \operatorname{ReG}(k_B R) \cdot \mu_A - \operatorname{ImG}(k_B R) \cdot \mu_B + \mu_A \cdot \operatorname{ImG}(k_A R) \cdot \mu_B \mu_A \cdot \operatorname{ImG}(k_B R) \cdot \mu_B \Big] \\ &\times \cos(\Delta_{AB} T) \\ &- \frac{2\omega_A^2 \omega_B^2 e^{-(\Gamma_A + \Gamma_B)T/2}}{e^4 \epsilon_0^2 \hbar \Delta_{AB}} \Big[[\mu_A \cdot \operatorname{ReG}(k_A R) \cdot \mu_B \mu_B \cdot \operatorname{ImG}(k_B R) \cdot \mu_A - \operatorname{ImG}(k_A R) \cdot \mu_B \mu_B \cdot \operatorname{ReG}(k_B R) \cdot \mu_A \Big] \\ &\times \sin(\Delta_{AB} T) \\ &- \frac{2\omega_A^2 \omega_B^2 e^{-(\Gamma_A + \Gamma_B)T/2}}{e^4 \epsilon_0^2 \hbar \Delta_{AB}} \Big[[\mu_A \cdot \operatorname{ReG}(k_A R) \cdot \mu_B \mu_B \cdot \operatorname{ImG}(k_A R) \cdot \mu_B \Big]^2 \Big] \\ &+ \frac{2\omega_A^2 a^2 e^{-(\Gamma_A + \Gamma_B)T/2}}{e^4 \epsilon_0^2 \hbar \Delta_{AB}} \Big[[\mu_A \cdot \operatorname{ReG}(k_A R) \cdot \mu_B (\Delta (A R) \cdot \mu_B)]^2 \Big] \\ &+ \frac{2\omega_A^2 a^2 e^{-(\Gamma_A + \Gamma_B)T/2}}{e^4 \epsilon_0^2 \hbar \Delta_{AB}} \Big[\mu_A \cdot \operatorname{ReG}(k_A R) \cdot \mu_B \cos(\Delta_{AB} T) + \mu_A \cdot \operatorname{ImG}(k_A R) \cdot \mu_B \sin(\Delta_{AB} T) \Big] \\ &\times \sin(\Delta_{AB} T) \\ &- \frac{2\omega_A^2 a^2 e^{-(\Gamma_A + \Gamma_B)T/2}}{e^4 \epsilon_0^2 \hbar \Delta_{AB}} \Big[[\mu_A \cdot \operatorname{ReG}(k_A R) \cdot \mu_B \cos(\Delta_{AB} T) + \mu_A \cdot \operatorname{ImG}(k_A R) \cdot \mu_B \sin(\Delta_{AB} T) \Big] \\ &+ \frac{2\omega_A^2 a^2 e^{-(\Gamma_A + \Gamma_B)T/2}}{e^4 \epsilon_0^2 \hbar \Delta_{AB}} \Big[\mu_A \cdot \operatorname{ReG}(k_A R) \cdot \mu_B \cos(\Delta_{AB} T) + \mu_A \cdot \operatorname{ImG}(k_$$

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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