
**UNIVERSIDAD DE VALLADOLID**
FACULTAD DE CIENCIAS

# TRABAJO DE FIN DE MÁSTER

Máster en Física

## Resonant van-der-Waals Interaction: Net Harmonic Force on a Two-Atom System

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## ABSTRACT

We perform a time-dependent quantum computation of the van der Waals interaction between two free identical atoms, one of which is initially excited. Making use of the perturbative approach developed in Refs.[1–3] it is shown that, in addition to an off-resonant interaction force between the two atoms, the two-atom system is subjected to a net force which oscillates periodically in time. We prove that the apparently *missing* momentum is carried by the resonant photons which mediate the periodic transfer of the excitation between the atoms, guaranteeing this way the satisfaction of the action-reaction principle in the quantum realm. Remarkably, since mediating photons remain virtual throughout the process, their momentum is virtual and thus unaccessible experimentally. Ultimately, as the de-excitation of the system takes place, that momentum turns into actual when carried off by the real photon which is emitted spontaneously. Hence, the directionality of such an emission constitutes a vestige of the net force acting upon the two-atom system while excited.

En este trabajo hemos llevado a cabo un cálculo cuántico, dependiente del tiempo, de la interacción de van der Waals entre dos átomos libres idénticos, uno de los cuales se encuentra inicialmente en un estado excitado. Haciendo uso del formalismo desarrollado en Refs.[1–3] mostramos que, además de una fuerza de interacción no-resonante entre los dos átomos, el sistema completo se encuentra sujeto a una fuerza neta que oscila periódicamente en el tiempo. Asímismo, probamos que el momento aparentemente *desaparecido* es acarreado por los fotones resonantes que median la transferencia periódica de la excitación entre los dos átomos, garantizando de esta manera el cumplimiento del principio de acción-reacción a nivel cuántico. El hecho de que los fotones mediadores de la fuerza permanezcan virtuales a lo largo del proceso hace que su momento sea virtual, siendo indetectable de forma experimental. En último término, ese momento se convierte en actual cuando es acarreado por el fotón real emitido espontáneamente cuando la desexcitación del sistema atómico tiene lugar. De hecho, la direccionalidad de dicha emisión constituye un vestigio de la fuerza neta que actúa sobre el sistema atómico mientras permanece excitado.


## I. INTRODUCTION

Long-range interactions between neutral atoms result from the coupling of the electromagnetic (EM) quantum field fluctuations to the current fluctuations of the atomic charges [4, 5]. In the electric dipole approximation they are known as van der Waals (vdW) interactions. In early quantum mechanical studies, London [6] found out that the interaction between close neutral atoms in the ground state decreases as $R^{-6}$ with the interatomic distance. Later, the quantum electrodynamical (QED) approach of Casimir and Polder [7] showed that, when retardation effects are introduced, the vdW interaction energy goes as $R^{-7}$ for large separations.

Further investigations on vdW-like interactions involving excited atoms –cf., Wylie and Sipe [8, 9] and others [10–12]– revealed that the resultant forces contain two separable components, namely resonant and off-resonant forces. The former are mediated by resonant *on-shell* photons whose frequency equals the transition frequency between intermediate atomic states. On the contrary, the latter are mediated by photons of any frequency.

On the other hand, Dicke's works [13] on the resonant interaction between two identical atoms showed that nearby identical atoms may exchange a quantum excitation in a coherent manner by means of the periodic exchange of a resonant photon. As a result, the two-atom system behaves as a single entangled quantum system whose evolution in time is coherent [14–16]. Also as a result of coherence, for the particular arrangement of atomic phases in the so-called symmetric and antisymmetric states, enhancement and inhibition of the spontaneous emission rate are obtained, respectively.

In the last decade, a renewal interest has flourished in the vdW interaction of excited atoms, motivated by the ever-increasing improvement in the techniques of atomic manipulation as well as by the possibility to build q-bits from binary atomic systems [17]. In this respect, different approaches to the computation of the resonant vdW interaction between two dissimilar atoms, one of which being excited, have led to conflicting results [1, 18–21]. The apparent discrepancy has been recently explained by Donaire [3], who has pointed out that the discrepancy lies in the different physical quantities being computed in each approach. That is, whereas Ref.[20] deals with the energy shift of the two-atom wavefunction, Ref.[21] computes the energy shifts of the wavefunctions of each atom, Ref.[1] deals with the vdW potential of the excited atom, and Ref.[18] computes the vdW potential of the ground state atom. Further, the latter two results imply an apparent lack of reciprocity between the vdW forces acting upon each atom. The explanation was given in Ref.[2] in terms of the virtual momentum carried off by the resonant photons which mediate the resonant vdW forces. In turn, the difference between the strengths of the resonant forces on each atom leads to a net force upon the two-atom system.

Following up with the latter findings, it is the purpose of this work to extend the investigation of the vdW forces into atomic systems made of two identical atoms, one of which is initially excited. The well-known degenerate character of this system –cf., Refs.[13, 14, 17]– is expected

to amplify the strength of the net vdW force found out in Ref.[2], making this way our system a suitable setup to probe experimentally this phenomenon.

This work is organized as follows. The fundamentals of the approach are explained in Sec II together with the computation of the time-evolution propagator of the atomic system. The off-resonant vdW interactions are discussed in Sec. III. In Sec.IV we compute in full detail the net resonant vdW force. In Section V we derive the expressions for the net force acting upong the system, as well as, the internal force between the atoms. Sec VI is devoted to prove the satisfaction of the action-reaction principle in our system, explaining the relationship between the net vdW force and directional spontaneous emission. We conclude with the Discussion in Sec. VI.

## II. TIME EVOLUTION OF TWO ATOM STATES

We consider the interaction of two two-level identical atoms $A$ and $B$ with excitation levels $\omega_A = \omega_B = \omega_0$. We assume that the atoms are placed at a stationary distance $\mathbf{R} = \mathbf{R}_B - \mathbf{R}_A$ from each other and their wave functions do not overlap. As initial conditon we consider that only the atom $A$ is excited, i.e. $|\psi(t_0)\rangle = |A_+, B_-\rangle$ at time $t_0 = 0$. Although in our calculations we are considering $t_0 \to -\infty$ this condition do not pretend to describe the initial instant time but an adiabatic excitation of atoms. This means that the Rabi frecuency $\Omega_{Rabi}$, which describes the time it takes to produce an atomic transition in a given light field, satisfies $\Omega_{Rabi}^{-1} \ll T - t_0$, where $T$ is the time of observation, see Appendix A. Then, dynamical description of atom states begins when the atom $A$ is already excited at time $t_0 = 0$. Lastly, off resonant van der Waals interaction which are previous to the initial time $t_0$ are neglected. The interaction Hamiltonian considered is the electric dipole approximation interaction of each atom with the quantum EM field where $W_{A,B} = -\mathbf{d}_{A,B} \cdot \mathbf{E}(\mathbf{R}_{\mathbf{A},\mathbf{B}})$, and the the total hamiltonian reads $H = H_A + H_B + H_{EM} + W_A + W_B$. As the initial state $|A_+, B_-\rangle$ is not a eigenstate of the hamiltonian, it is necessary to study the time evolution of the atomic states. Firstly, we are going to study the interaction of an isolated atom with the quantum EM field and then the interaction with a nearby atom.

The probability of decay of an atom $A$ per unit of time is known as decay rate $\Gamma_A(T)$

$$\Gamma_A(T) = \frac{d}{dT} P_{(A_+ \to A_-, \gamma_\mathbf{k})} = \frac{d}{dT} |\langle A_-, \gamma_\mathbf{k}| \, U(T) \, |A_+\rangle|^2, \tag{1}$$

which is represented diagrammaticaly in Fig. 1

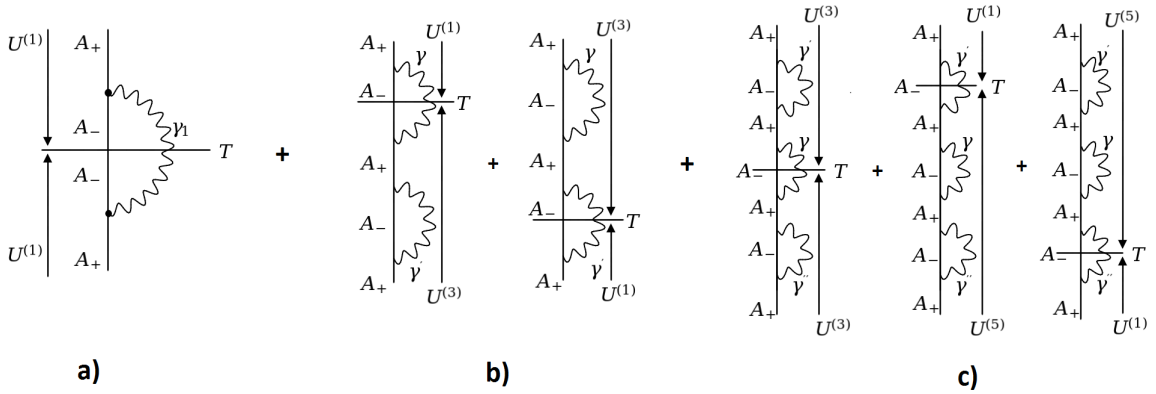


FIG. 1. Diagrammatic Representation of $\Gamma_A(T)$. Diagrams a), b) and c) represent the probability of spontaneous emission of an atom at order $W_A^4$, $W_A^4$ and $W_A^6$, respectively.

The contribution of the first diagram, which we denote by $\Gamma_0$ reads

$$\Gamma_0 = \frac{\omega_A^3 |\boldsymbol{\mu}_A|^2}{3\pi} \tag{2}$$

which coincides with the Einstein A coefficient [22], where $\boldsymbol{\mu}_A = \langle A_-| \, \mathbf{d}_A \, |A_+\rangle$ is the electric dipole transition element. Making que computation of the diagrams of several orders, see Appendix B, we find

$$\Gamma_A(T) = \Gamma_0 - \Gamma_0^2 T + \Gamma_0^3 \frac{T^2}{2!} + ... = \Gamma_0 e^{-\Gamma_0 T}, \tag{3}$$

impliying that the probability of emission is

$$P_{(A_+ \to A_-, \gamma_{\mathbf{k_0}})} = 1 - e^{-\Gamma_0 T}. \tag{4}$$

This implies that the time evolution of an atomic state

taking into account the decay processes is

$$U(T)\left|A_{+}\right\rangle = e^{-i\left(\omega_A - i\frac{\Gamma_0}{2}\right)T}\left|A_{+}\right\rangle . \tag{5}$$

This approach describes in an effective manner the evolution of an atom as an open system. Nevertheless the presence of a second atom $B$ produce the transmission of the excitation from atom $A$ to atom $B$. This process is given by the matrix element $\langle A_{-}, B_{+}|\, U(T)\,|A_{+}, B_{-}\rangle$, which is denoted by $\mathcal{U}_{(A_{+},B_{-})\to(A_{-},B_{+})}$. Furthermore, despite the presence of atom $B$ the excitation may remains in the atom $A$. This process is given by the matrix element $\langle A_{+}, B_{-}|\, U(T)\,|A_{+}, B_{-}\rangle$, which is denoted by $\mathcal{U}_{(A_{+},B_{-})\to(A_{+},B_{-})}$. The perturbative expansion of both processes is represented in Fig.2 diagrammatically.

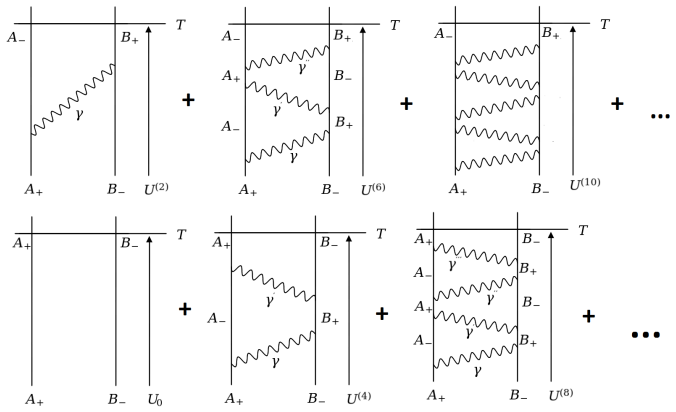


FIG. 2. Diagrammatic representation of the photon exchange. The matrix element $\mathcal{U}_{(A_{+},B_{-})\to(A_{-},B_{+})}$ is represented in the first row and there is an odd number of photons exchanged in each diagram. The matrix element $\mathcal{U}_{(A_{+},B_{-})\to(A_{+},B_{-})}$ is represented in the second row and there is a even number of photons exchanged in each diagram.

The computation of these diagrams, developed in Appendix B, yields

$$\begin{aligned}\mathcal{U}_{(A_{+},B_{-})\to(A_{-},B_{+})} &= ie^{-i\omega_A T}\sin\left[\tilde{\Omega}(R)\;\;T\right],\\ \mathcal{U}_{(A_{+},B_{-})\to(A_{+},B_{-})} &= e^{-i\omega_A T}\cos\left[\tilde{\Omega}(R)\;\;T\right],\end{aligned} \tag{6}$$

where

$$\tilde{\Omega}(R) = \mu^i\mu^j\omega_A^2 G_{ij}(\omega_A, R). \tag{7}$$

The Green's function, $\mathbb{G}(\omega_A, R)$, has a real part, $\Omega(R) = \mu^i\mu^j\omega_A^2\,\mathrm{Re}\left\{G_{ij}(\omega_A, R)\right\}$, which corresponds to the oscillation frecuency, and an imaginary part which implies a decay of the atomic states due the spontaneous emission of the photon that they exchange. We refer to this damping component as decay by influence, $\Gamma(R) = 2\mu^i\mu^j\omega_A^2\,\mathrm{Im}\left\{G_{ij}(\omega_A, R)\right\}$, which is taken out of the trigonometric functions as a negative exponential.

The frecuency of the excitation transmission must be $\Omega(R) \gg \Gamma$ for ensuring the photon exchange between atom $A$ and $B$. It follows therefore that $\Omega(R) \gg \Gamma(R) + \Gamma_A$ which establishes a condition on the interatomic distance $R$. Finally, $\Omega_{Rabi} \ll \Omega(R)$ [3].

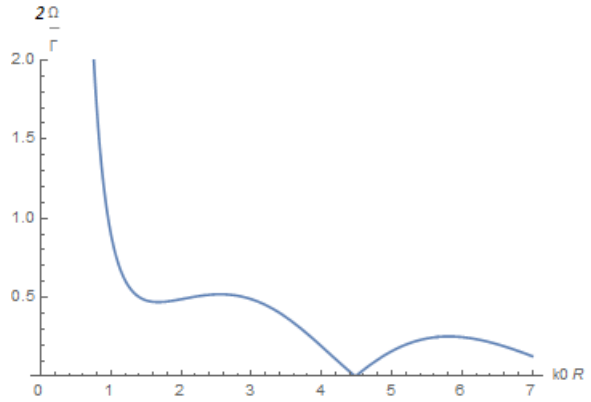


FIG. 3. Plot representation of $\frac{2\Omega}{\Gamma}$. This shows that the ratio starts to increase at closer distances than a wavelength $\lambda_0$

The time evolution of atomic states is given by the exchange of a resonant single-photon at a time. The sum of the terms of the perturbative expansion converges to trigonometric functions which shows the coherence time evolution of the atomic states [13, 15]. This confirms the degeneracy of the states $|A_{+}, B_{-}\rangle$ and $|A_{-}, B_{+}\rangle$.

Summing up, total time evolution of atomic states is given by

$$\begin{aligned}\mathcal{U}_{(A_{+},B_{-})\to(A_{-},B_{+})} &= ie^{-\frac{\Gamma_0+\Gamma(R)}{2}T}e^{-i\omega_A T}\sin\left(\Omega T\right),\\ \mathcal{U}_{(A_{+},B_{-})\to(A_{-},B_{+})} &= e^{-\frac{\Gamma_0+\Gamma(R)}{2}T}e^{-i\omega_A T}\cos\left(\Omega T\right).\end{aligned} \tag{8}$$

This implies that the population levels, neglecting decay rate terms, evolves coherently

$$\begin{aligned}P_{(A_{+},B_{-})\to(A_{-},B_{+})} &= \sin^2\left(\Omega T\right),\\ P_{(A_{+},B_{-})\to(A_{-},B_{+})} &= \cos^2\left(\Omega T\right),\\ P_{(A_{+},B_{-})\to(A_{-},B_{+})} + P_{(A_{+},B_{-})\to(A_{-},B_{+})} &= 1.\end{aligned} \tag{9}$$

This result takes into account the infinite number of shared photons between the atoms $A$ and $B$. From now on, we will denote the total decay rate of the two atom system as $\Gamma = \Gamma_0 + \Gamma(R)$, and the matrix elements $\mathcal{U}_{(A_{+},B_{-})\to(A_{-},B_{+})}$ and $\mathcal{U}_{(A_{+},B_{-})\to(A_{-},B_{+})}$ as $U_{cos}(T)$ and $U_{sin}(T)$ on diagramatic representation, respectively.

On the other hand, the initial excitation may also be spontaneously emitted. This process is given by the matrix element $\langle A_{-}, B_{-}, \gamma_{\mathbf{k}}|\, U(T)\,|A_{+}, B_{-}\rangle$ which is denoted by $\mathcal{U}_{(A_{+},B_{-})\to(A_{-},B_{-},\gamma_{\mathbf{k}})}$. This is represented diagramatically in Fig. 4

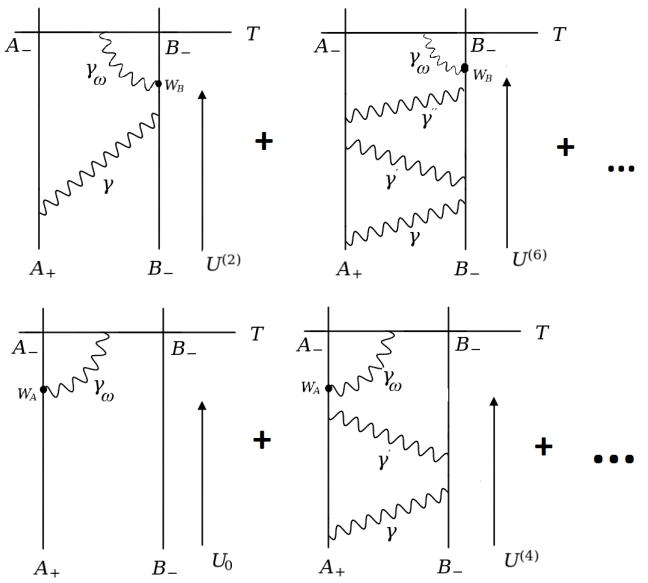


FIG. 4. Diagrammatic representation of the spontaneous photon emission preceded by the exchange of a resonant photon. The emitted photon has a generic momentum $\mathbf{k}$ .

The calculation of the diagrams if Fig. 4, see Appendix B, yields

$$\begin{aligned}&\mathcal{U}_{(A_+,B_-)\to(A_-,B_-,\gamma_\mathbf{k})}\\&=\frac{\langle A_-,\gamma_\mathbf{k}|\,W_A\,|A_+\rangle\,e^{\eta T}}{[\omega-(\omega_A+i\eta)]}e^{-\frac{\Gamma}{2}T}e^{-i\omega_A T}\cos(\Omega T)\\&-i\frac{\langle B_-,\gamma_\mathbf{k}|\,W_B\,|B_+\rangle\,e^{\eta T}}{[\omega-(\omega_A+i\eta)]}e^{-\frac{\Gamma}{2}T}e^{-i\omega_A T}\sin(\Omega T)\end{aligned}\tag{10}$$

This result implies that the photon spontaneous emission is preceded by the exchange of the initial excitation between both atoms. This photon possesses a generic momentum $\mathbf{k}$ and can be emitted from any atom. The first term on the right hand side of equation (10) is the amplitude of probability of photon emission of generic frecuency $\omega$, which will be denoted by $U^{\omega}_{cos}$. Likewise, the second term is the amplitud of probability of photon emission of generic frecuency $\omega$, which will be denoted by $U^{\omega}_{sin}$.

## III. OFF-RESONANT VAN DER WAALS INTERACTION

Generically, the coupling of an atom to the quantum EM field causes its EM self-interaction, which is attributed to an energy shift in the atomic levels. In free space this shift is known as the Lamb-shift [23]. In the presence of another atom, it has been proved that the off-resonant component of the interaction potential $W$ on the atom under study is equivalent to an energy shift of its wave function [3].

The vdW interaction is represented by the diagrams of Fig. 5.

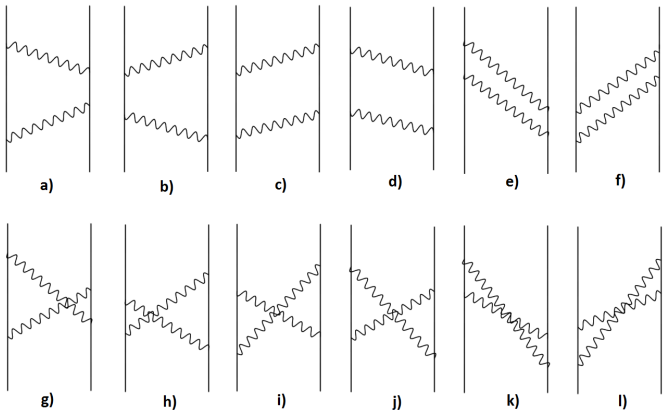


FIG. 5. Diagramatic representation of resonant and off-resonant vdW interaction diagrams

The twelve diagrams shown in Fig. 5 contribute to the fourth order interaction energy. The sum of all the expectation values of the interaction vertices $W_A$ and $W_B$ yields the vdW interaction energy [3]. The off-resonant vdW interaction between two atoms reads [8, 9, 19], see Appendix B,

$$\begin{aligned}&\langle W^{off}\rangle_{AB}\equiv\langle W^{off}_A\rangle_{AB}=\langle W^{off}_B\rangle_{AB}\\&=-\frac{2\mu^i_A\mu^j_B\mu^p_A\mu^q_B}{\pi}\\&\times\int_0^\infty du\;\frac{u^2\omega_A\omega_B}{[\omega_A^2+u^2]\,[\omega_B^2+u^2]}G_{ij}(iu,R)G_{pq}(iu,R).\end{aligned}\tag{11}$$

We are considering a system with an initially excitated atom. This implies the vdW is equal up to a sign to the vdW interaction between ground state atoms. The off resonant interaction is produced for al values of $\omega$ with the exception of $\omega_A$ where the system becomes degenerate. The initial atomic states evolve exchanging a single resonant photon at a time up to the off-resonant vdW interaction is meadures. This implies there are four processes, which are depicted diagrammaticaly in Fig.6 and Fig. 7, where the time evolution of the system is taken into account.

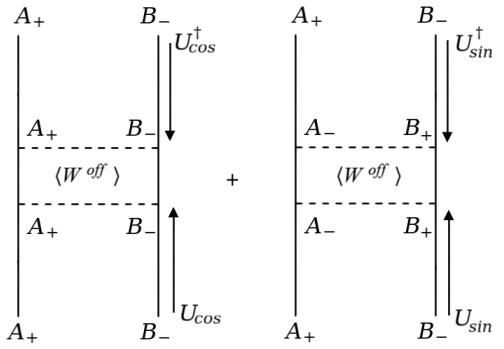


FIG. 6. Diagramatic representation of the vdW interaction between two identical atoms. The atomic states evolve in time from the initial state $|A_+, B_-\rangle$ exchanging a resonant photon until vdW off-resonant interaction operator is applied. Resonant vdW contributions are not taking into account in this diagram.

On one hand, the atoms evolve in time exchanging the initial excitation until the vdW off-resonant interaction takes place because the time evolution only shares one resonant photon at a time. The photon exchange is described by the matrix elements $\mathcal{U}_{(A_+,B_-)\to(A_-,B_+)}$ and $\mathcal{U}_{(A_+,B_-)\to(A_+,B_-)}$. Each diagramms of Fig. 6 oscillates in time but the coherence behaviour yields a non-oscillating vdW interaction which reads

$$\langle W^{off}\rangle_{(A_+,B_-)} = -e^{-\Gamma T}\langle W^{off}\rangle_{(A_-,B_-)}. \tag{12}$$

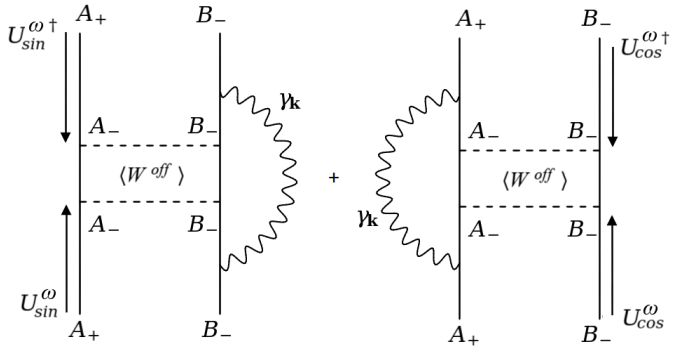


FIG. 7. Diagramatic representation of the vdW interaction between two identical atoms with a resonant photon emitted. Resonant photon does not intefere in the vdW interction.

On the other hand, the initial excitation is spontaneously emitted. The time evoultion is given by the diagrams of Fig. 7. The time evolution of the atomic staes is given by the matrix element $\mathcal{U}_{(A_+,B_-)\to(A_-,B_-,\gamma_{\mathbf{k}})}$. The coherent time evolution of the atom also yields a non-oscillating vdW interacton which yields

$$\langle W^{off}\rangle_{(A_-,B_-,\gamma_{\mathbf{k}})} = \left(1-e^{-\Gamma T}\right)\langle W^{off}\rangle_{(A_-,B_-)}. \tag{13}$$

We obtain that the total off-resonant vdW interaction of a two identical atom system is

$$\langle W^{off}\rangle(R,T) = \left(1-2e^{-\Gamma T}\right)\langle W^{off}\rangle_{(A_-,B_-)}. \tag{14}$$

## IV. RESONANT VAN DER WAALS INTERACTION

The resonant vdW interaction involves the exchange of a resonant photons. The time evolution of the atoms allows us to compute resonant vdW interaction at second order (Fig. 8). This is because an infinite number of resonant enchanges of photons are included on the time evolution of atomic states.

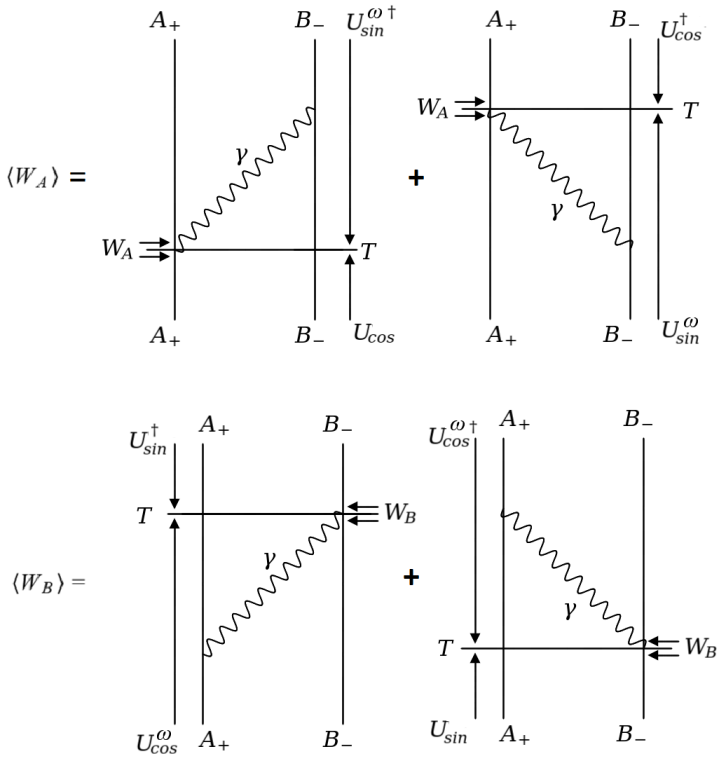


FIG. 8. Diagrammatic representation of resonant vdW interaction.

The calculations of the diagrammatic representation in Fig.8, see Appendix B, have been performed using the matrix element $\mathcal{U}_{(A_+,B_-)\to(A_-,B_-,\gamma_{\mathbf{k}})}$ for avoiding double counting of photon exchange.

Computing the calculations of the diagramatic representation we get

$$\begin{aligned}\langle W_A\rangle &= \langle A_+, B_-|\, U^{\omega\dagger}_{sin}(T) W_A U_{cos}(T)\,|A_+, B-\rangle \\ &+ \langle A_+, B_-|\, U^{\dagger}_{cos}(T) W_A U_{sin}(T)\,|A_+, B-\rangle \\ &= \mu^i_A \mu^j_A \omega^2_A \;\; e^{-\Gamma T} \;\; \mathrm{Im}\left\{G_{ij}(\omega_A, R)\right\}\sin(2\Omega T)\end{aligned} \tag{15}$$

$$\begin{aligned}\langle W_B\rangle &= \langle A_+,B_-|\,U^\dagger_{sin}(T)W_BU^\omega_{cos}(T)\,|A_+,B-\rangle \\ &+ \langle A_+,B_-|\,U^{\omega\dagger}_{cos}(T)W_BU_{sin}(T)\,|A_+,B-\rangle \\ &= -\mu^i_A\mu^j_A\omega^2_A\ e^{-\Gamma T}\ \ \mathrm{Im}\left\{G_{ij}(\omega_A,R)\right\}\sin(2\Omega T)\end{aligned} \tag{16}$$

We obtain that the interaction energy in each atom is equal up to the sign. Thus $\langle W_A+W_B\rangle = 0, \forall T$. This result is consistent with the fact that our initial state is a linear combination of the eigenstates symmetric and antisymetric states $|A_+,B_-\rangle = \frac{1}{\sqrt{2}}(|+\rangle+|-\rangle)$ [13].

## V. VAN DER WAALS FORCES

On the basis of interaction energy results, vdW force between atoms can be calculated. Straight application of the definition of force on each atom gives

$$\begin{aligned}\langle \mathbf{F}_A+\mathbf{F}_B\rangle &= \langle \dot{\mathbf{Q}}_A+\dot{\mathbf{Q}}_B\rangle \\ &= -i\partial_T\,\langle\psi(0)|\,U^\dagger(T)(\nabla_{\mathbf{R}_A}+\nabla_{\mathbf{R}_B})U(T)\,|\psi(0)\rangle \\ &= -\langle\nabla_{\mathbf{R}_A}W_A\rangle - \langle\nabla_{\mathbf{R}_B}W_B\rangle,\end{aligned} \tag{17}$$

where $\mathbf{Q}_{A,B}$ denote the kinetic momentum of the center of mass of the atoms $A$ and $B$ [2].

Straight application of the negative gradients upon the vdW potentials computed in the previous sections results in

$$\begin{aligned}\langle \mathbf{F}_A\rangle = \hat{\mathbf{R}}\Big[&2\mu^i_A\mu^j_A\omega^2_A\ \ e^{-\Gamma T}\ \ \nabla_{\mathbf{R}}\,\mathrm{Im}\left\{G_{ij}(\omega_A,R)\right\}\sin(2\Omega T) \\ &+\left(1-2e^{-\Gamma T}\right)\nabla_{\mathbf{R}}\langle W^{off}\rangle_{(A_-,B_-)}\Big].\end{aligned} \tag{18}$$

$$\begin{aligned}\langle \mathbf{F}_B\rangle = \hat{\mathbf{R}}\Big[&2\mu^i_A\mu^j_A\omega^2_A\ \ e^{-\Gamma T}\ \ \nabla_{\mathbf{R}}\,\mathrm{Im}\left\{G_{ij}(\omega_A,R)\right\}\sin(2\Omega T) \\ &-\left(1-2e^{-\Gamma T}\right)\nabla_{\mathbf{R}}\langle W^{off}\rangle_{(A_-,B_-)}\Big].\end{aligned} \tag{19}$$

These result yield

$$\begin{aligned}\mathbf{F}_{net}\equiv\langle \mathbf{F}_A+\mathbf{F}_B\rangle = 4\hat{\mathbf{R}}\ \ &\mu^i_A\mu^j_A\omega^2_A\ \ e^{-\Gamma T} \\ &\times\nabla_{\mathbf{R}}\,\mathrm{Im}\left\{G_{ij}(\omega_A,R)\right\}\sin\left(2\Omega T\right)\end{aligned} \tag{20}$$

$$\mathbf{F}_{int}\equiv\langle \mathbf{F}_A-\mathbf{F}_B\rangle = 2\hat{\mathbf{R}}\left(1-2e^{-\Gamma T}\right)\nabla_{\mathbf{R}}\langle W^{off}\rangle_{(A_-,B_-)} \tag{21}$$

There is a net force exerted on both atoms which makes the system to oscillate jointly. This net force is a consequence of the resonant vdW interaction. On the other hand, there is a force of interaction $\mathbf{F}_{int}$ as a cibseqyebce if the off-resonant vdW interaction.

## VI. CONSERVATION OF TOTAL MOMENTUM AND DIRECTIONAL EMISSION OF PHOTONS

No external source is exerciting the net force, $\mathbf{F}_{net}$. The only entity which con compensate this apparent lose of momentum is the shared excitation, i.e. the photon exchanged by the atoms. The momentum of the photon $\mathbf{P}$, following the interaction diagrams, has associated time derivative of its momentum

$$\langle\dot{\mathbf{P}}\rangle = \sum_{\mathbf{k}}\partial_T\,\langle A_+,B_-|\,U^{(\omega\dagger}_{sin}(T)\ \mathbf{k}\ U^\omega_{cos}(T)\,|A_+,B-\rangle + (c.c.). \tag{22}$$

where the first term corresponds to the first column of diagrams of the interaction and its complex conjugated (c.c.) to the second column. Performing the time derivative to the time evolution operators we arrive at

$$\begin{aligned}\langle\dot{\mathbf{P}}\rangle &= \sum_{\mathbf{k}}\Big[\langle A_+,B_-|\,U^\dagger_{sin}(T)\ \ i\mathbf{k}W_B\ \ U_{cos}(T)\,|A_+,B-\rangle \\ &+\langle A_+,B_-|\,U^{\omega\dagger}_{sin}(T)\ \ (-i\mathbf{k})W_A\ \ U_{cos}(T)\,|A_+,B-\rangle\Big] + (c.c.) \\ &= \langle\dot{\mathbf{P}}\rangle_1+\langle\dot{\mathbf{P}}\rangle_2 = \langle\nabla_{\mathbf{R}_A}W_A\rangle+\langle\nabla_{\mathbf{R}_B}W_B\rangle = -\langle\mathbf{F}_A+\mathbf{F}_B\rangle\end{aligned} \tag{23}$$

This result can be depicted diagrammaticaly as

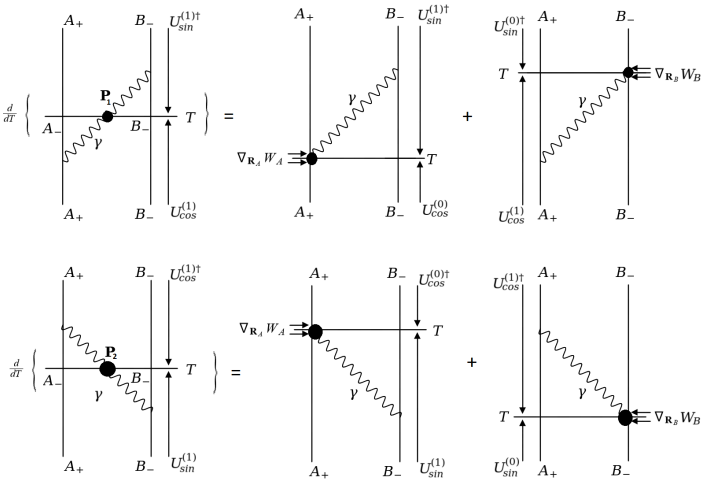


FIG. 9. Diagrammatic representation of the time derivative of the momentum of the photon.

The first two terms correspond to the first photon diagram while the complex conjugated corresponds to the second one. Each photon pressure coincides with the sum of the gradient of the both vertices where the photon is emitted and absorbed.

$$\begin{aligned}\langle\dot{\mathbf{P}}\rangle_{1,2} = &\mu^i_A\mu^j_A\omega^2_A\ \ e^{-(\Gamma_A+2\Gamma(R))T} \\ &\times\nabla_{\mathbf{R}}\,\mathrm{Im}\left\{G_{ij}(\omega_A,R)\right\}\sin(2\Omega T)\end{aligned} \tag{24}$$

Finally, the sum of each diagram coincides with $\langle \nabla_{\mathbf{R}_A} W_A \rangle + \langle \nabla_{\mathbf{R}_B} W_B \rangle$.

The influence of atom $B$ on atom $A$ allow us to discuss the directionality of photon emission. Spontaneous decay of atom $A$ (1) has an uniform angular distribution

$$\frac{d\Gamma_A}{d\Theta} = \left(\frac{\omega_A}{2\pi}\right)^2 \mu_A^i \mu_A^j (\delta_{ij} - \hat{k}_i \hat{k}_j), \tag{25}$$

where $\Theta$ is the solid angle. Nevertheless, the angular distribution of the decay rate by influece, where $\theta$ denotes the angle between $\mathbf{R}$ and $\mathbf{k}$, is

$$\frac{d\Gamma(\mathbf{R})}{d\Theta} = \left(\frac{\omega_A}{2\pi}\right)^2 \mu_A^i \mu_A^j (\delta_{ij} - \hat{k}_i \hat{k}_j) \sin(\omega_A R cos\theta). \tag{26}$$

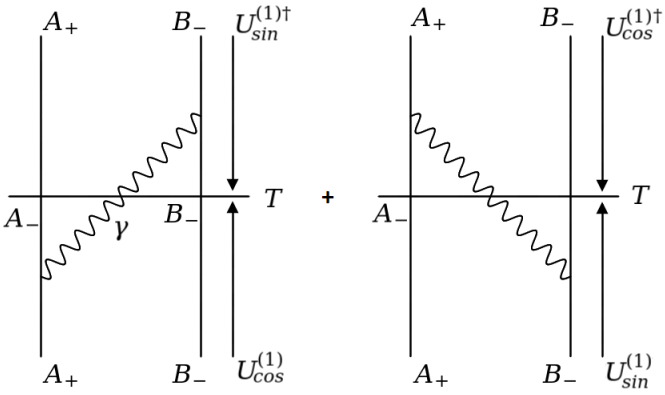


FIG. 10. Diagrammatic representation of probability of spontaneous emission

Then the total emission distribution of atom $A$ is influenced by the presence of atom $B$ which makes possible to distinct a forward and a backward emission.

$$\frac{d\left(\Gamma_A + \Gamma(\mathbf{R})\right)}{d\Theta} = \left(\frac{\omega_A}{2\pi}\right)^2 \mu_A^i \mu_A^j (\delta_{ij} - \hat{k}_i \hat{k}_j) \times \left[1 + \sin(\omega_A R cos\theta)\right]. \tag{27}$$

The maximal differential probability between forward and backward emission occurs at $R = \frac{\pi}{2\omega_A}$.

## VII. CONCLUSIONS AND DISCUSSION

The performed calculations yield that the two atom system is subjected to a net force, $\mathbf{F}_{net}$, which makes the whole system to oscillate jointly. In physical grounds, the net oscillating force is a consequence of the alternating breaking and restoration of Parity symmetry in the atomic system. This force is balance by the time derivative of the photon momentum which is greater when the photon has not been emitted yet. This means that the action-reaction principle is satisfied. Furthermore, a directional emission of photons is observed. We may give two possible interpretations of these results. Firstly, we can afirm that the force exercised on the system vanishes when the photon is being emitted. Nevertheless, this interpretation require the photon to exert a time dependent force on the atoms. Secondly, as time exponential decaying comes from the spontaneous decay processes, this exponential factor indicates the probability of making a measurement of the oscillating force on the atoms. As the initial time of the system is well known, several measurements at different times would be necessary to discern the oscillating behaviour of the force from its damped behaviour, which would correspond to a force dissipation, or a spontaneous emission of a real photon. Although this interpretations could be valid, an experimental testing can give us the final answer.

### Appendix A: Time Dependent Perturbative Quantum Electrodynamics (QED)

The fundamentals of the quantum mechanical approach of matter-radiation interaction based on [24, 25] are explained in this section. We also introduce a standard time-dependent perturbation theory [26] and a diagrammatic representation of them based on the Refs [1–3].

### 1. Radiation Field Quantization

Classical electromagnetism is described by the electric and magnetic fields, $\mathbf{E}(\mathbf{r},t)$ and $\mathbf{B}(\mathbf{r},t)$ respectively, which obey Maxwell's equations

$$\begin{aligned} \nabla\cdot\mathbf{E} &= \rho, \\ \nabla\cdot\mathbf{B} &= 0, \\ \nabla\wedge\mathbf{E} &= -\frac{\partial\mathbf{B}}{\partial t}, \\ \nabla\wedge\mathbf{B} &= \mathbf{J}+\frac{\partial\mathbf{E}}{\partial t}. \end{aligned} \tag{A1}$$

These fields can be expressed in terms of a scalar potential $\phi(\mathbf{r},t)$ and a vector potential $\mathbf{A}(\mathbf{r},t)$,

$$\begin{aligned} \mathbf{E} &= -\nabla\phi-\frac{\partial\mathbf{A}}{\partial t}, \\ \mathbf{B} &= \nabla\wedge\mathbf{A}. \end{aligned} \tag{A2}$$

Both potentials are not uniquely determinated, as they are defined up to a gauge transformation of the form

$$\mathbf{A}'\to\mathbf{A}+\nabla\chi\,,\ \phi'\to\phi-\frac{\partial\chi}{\partial t}, \tag{A3}$$

which leaves $\mathbf{E}$ and $\mathbf{B}$ invariant.

By Maxwell's equations (A1) the dynamical equations of the potentials are

$$\begin{aligned} \Box^2\mathbf{A} &= \mathbf{J}-\nabla\left(\nabla\cdot\mathbf{A}-\frac{\partial\phi}{\partial t}\right), \\ \nabla^2\phi+\frac{\partial}{\partial t}\nabla\cdot\mathbf{A} &= 0, \end{aligned} \tag{A4}$$

whose final form will depend on the gauge fixing.

In order to quantize the electromagnetic field, canonical formalism is convenient for defining dynamical variables. Firstly, we associate to the free electromagnetic field a Lagrangian density of the form

$$\mathcal{L}=-\frac{1}{4}F^{\mu\nu}F_{\mu\nu}=\frac{1}{2}\left(\mathbf{E}^2-\mathbf{B}^2\right), \tag{A5}$$

where $A^\mu=(\phi,\mathbf{A})$ and $F_{\mu\nu}=\partial_\mu A_\nu-\partial_\nu A_\mu$. The canonical momentum asociated to $A_i$ is

$$\Pi^i(\mathbf{r})=\frac{\partial\mathcal{L}}{\partial\left(\partial_0 A_i\right)}=-F^{0i}, \tag{A6}$$

which vanishes identically for $\nu=0$, indicating that $A^0$ is not a dynamical variable. Then the canonical variables are the $A_i$ with a cojugated canonical momentum $-F^{0i}=E^i$, $i=1,2,3$. This implies that the Hamiltonian of the free electromagnetic field is

$$H=\frac{1}{2}\int d^3x\left(\mathbf{E}^2+\mathbf{B}^2\right). \tag{A7}$$

First of all we must fix the gauge. In our particular case we are using Coulomb Gauge $\nabla\cdot\mathbf{A}=0$ since we are going to study radiative processes. This implies that the vector potential $\mathbf{A}$ can be separated in a longitudinal $\mathbf{A}_\parallel$ and a transverse component $\mathbf{A}_\perp$ such as

$$\begin{aligned} \mathbf{A} &= \mathbf{A}_\parallel+\mathbf{A}_\perp\,,\ A_0=0, \\ \nabla\cdot\mathbf{A}_\perp &= 0\,,\ \nabla\wedge\mathbf{A}_\parallel=0. \end{aligned} \tag{A8}$$

and the equation of motion of the vector potential (A4) takes the form

$$\Box^2\mathbf{A}_\perp=0. \tag{A9}$$

Finally, we must replace the electromagnetic canonical variables by operators and impose commutation rules which are compatible with the fixed gauge. Coulomb Gauge commutator rules are

$$\begin{aligned}[A_i(\mathbf{r}), E_j(\mathbf{r}')] &= i\hbar\delta_{ij}\delta^T_{ij}(\mathbf{r}-\mathbf{r}'),\\ [A_i(\mathbf{r}), A_j(\mathbf{r}')] &= [E_i(\mathbf{r}), E_j(\mathbf{r}')] = 0,\end{aligned} \tag{A10}$$

with

$$\delta^T_{ij}(\mathbf{r}-\mathbf{r}') = \int \frac{d^3k}{(2\pi)^3}\left(\delta_{ij} - \frac{k_i k_j}{\mathbf{k}^2}\right) e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}. \tag{A11}$$

Then by means of a Fourier Transform in (A9) we can expand the quantized fields in terms of transverse plane waves

$$\begin{aligned}\mathbf{A}_\perp(\mathbf{r}) &= \sum_{\alpha=1}^{2}\int \frac{d^3k}{(2\pi)^3}\frac{1}{\sqrt{2\omega_\mathbf{k}}}\ \left[\epsilon a_{\mathbf{k},\alpha}e^{i\mathbf{k}\cdot\mathbf{r}} + \epsilon^* a^\dagger_{\mathbf{k},\alpha}e^{-i\mathbf{k}\cdot\mathbf{r}}\right],\\ \mathbf{E}_\perp(\mathbf{r}) &= i\sum_{\alpha=1}^{2}\int \frac{d^3k}{(2\pi)^3}\sqrt{\frac{\omega_\mathbf{k}}{2}}\ \epsilon_{\mathbf{k},\alpha}\left[\epsilon a_{\mathbf{k},\alpha}e^{i\mathbf{k}\cdot\mathbf{r}} - \epsilon^* a^\dagger_{\mathbf{k},\alpha}e^{-i\mathbf{k}\cdot\mathbf{r}}\right],\\ \mathbf{B}_\perp(\mathbf{r}) &= i\sum_{\alpha=1}^{2}\int \frac{d^3k}{(2\pi)^3}\frac{1}{\sqrt{2\omega_\mathbf{k}}}\ \mathbf{k}\wedge\left[\epsilon a_{\mathbf{k},\alpha}e^{i\mathbf{k}\cdot\mathbf{r}} - \epsilon^* a^\dagger_{\mathbf{k},\alpha}e^{-i\mathbf{k}\cdot\mathbf{r}}\right],\end{aligned} \tag{A12}$$

with the following properties,

$$\begin{aligned}\epsilon_{\mathbf{k},\alpha}\cdot\epsilon_{\mathbf{k},\alpha'} &= \delta_{\alpha\alpha'}\ ,\ \mathbf{k}\cdot\epsilon_{\mathbf{k},\alpha} = 0,\\ I^{ij} = \sum_{\alpha=1}^{2}\epsilon^i_{\mathbf{k},\alpha}\epsilon^j_{\mathbf{k},\alpha} &= \left(\delta^{ij} - \frac{k^i k^j}{\mathbf{k}^2}\right)\ ,\ \omega_\mathbf{k} = |\mathbf{k}|,\\ \left[a_{\mathbf{k},r}, a^\dagger_{\mathbf{k}',s}\right] &= \delta_{rs}\delta(\mathbf{k}-\mathbf{k}'),\\ [a_{\mathbf{k},r}, a_{\mathbf{k}',s}] = \left[a^\dagger_{\mathbf{k},r}, a^\dagger_{\mathbf{k}',s}\right] &= 0.\end{aligned} \tag{A13}$$

Finally, substituting the quantized fields (A12) in the electromagnetic Hamiltonian (A7) and momentum operator yields,

$$\begin{aligned}H_{EM} &= \sum_\alpha\int\frac{d^3\mathbf{k}}{(2\pi)^3}\ \omega_\mathbf{k}\ a^\dagger_{\mathbf{k},\alpha}a_{\mathbf{k},\alpha},\\ \mathbf{P} &= \sum_\alpha\int\frac{d^3\mathbf{k}}{(2\pi)^3}\ \mathbf{k}\ a^\dagger_{\mathbf{k},\alpha}a_{\mathbf{k},\alpha}.\end{aligned} \tag{A14}$$

Quantum electromagnetic field in Coulomb gauge can be described as a set of vibration modes whose field amplitude is perpendicular to the direction of propagation. This modes are interpreted as particles called photons. The space state $|\gamma\rangle$ where operators $a_{\mathbf{k},\alpha}$, $a^\dagger_{\mathbf{k},\alpha}$ act, annihilating and creating photons respectively, represents the physical situation where there are many types of photons with different sets of $(\mathbf{k}_j, \alpha_j)$,

$$|n^1_{\mathbf{k_1},\alpha_1}, n^2_{\mathbf{k_2},\alpha_2}, ...n^j_{\mathbf{k_j},\alpha_j}, ...\rangle = |n^1_{\mathbf{k_1}}\rangle\otimes|n^2_{\mathbf{k_2},\alpha_2}\rangle\otimes...\otimes|n^j_{\mathbf{k_j},\alpha_j}\rangle\otimes... \tag{A15}$$

In this equation $n^j_{\mathbf{k_j},\alpha_j}$ is called the occupation number for the state $(\mathbf{k}_j, \alpha_j)$. The state of zero photons, $n^j = 0\ \forall j$, is called vacuum state and is represented as $|0_\gamma\rangle$ which satisfies

$$a_{\mathbf{k}_j,\alpha_j}|0_\gamma\rangle = 0\ ,\ \forall(\mathbf{k}_j,\alpha_j), \tag{A16}$$

and each $|n^j_{\mathbf{k_j},\alpha_j}\rangle$ is defined as

$$|n^j_{\mathbf{k_j},\alpha_j}\rangle = \sqrt{\frac{2\left(\omega_{k_i}\right)^{n^j}}{n^j!}}\left(a^\dagger_{\mathbf{k},\alpha}\right)^{n^j}|0_\gamma\rangle\,. \tag{A17}$$

Atomic interactions mediated by radiation are carried out by these transverse photons. In the following sections atom-radiation interactions are discussed.

### 2. Time-Dependent Perturbation Theory

Based on [26], we introduce a Time-Dependent Perturbation approach which is used all along this paper. Quantum mechanical systems are described by Schrödinger Equation

$$i\frac{\partial}{\partial t}\left|\psi(t)\right\rangle = H(t)\left|\psi(t)\right\rangle, \tag{A18}$$

which has associated a time evolution operator $U(t,t_0)$ such as

$$\left|\psi(t)\right\rangle = U(t,t_0)\left|\psi(t_0)\right\rangle. \tag{A19}$$

Few problems can be exactly solved in quantum mechanics approximate solutions are obtained by perturbation methods. The most usual way to tackle this problem is to separate the total hamiltonian of a quantum system in two parts

$$H(t) = H_0(t) + W(t), \tag{A20}$$

one that can be analytically solved $H_0(t)$, known as free hamiltonian, and $W(t)$, called perturbation, which represents the processes that need a perturbative analysis. When the $H_0(t)$ solutions are known, with a time evolution operator associated $U_0(t)$ is convenient to define the total time evolution operator $U(t)$ as

$$U(t) = U_0(t)U_I(t), \tag{A21}$$

where $U_I(t)$ is the time evolution operator associated with the perturbation $W(t)$. Substituting in the Schrödinger equation (A18) we obtain

$$i\frac{\partial}{\partial t}U_I = W_I U_I, \tag{A22}$$

or equivalently

$$U_I(t) = 1 - i\int_0^t d\tau\ \ W_I(\tau)U_I(\tau), \tag{A23}$$

where $W_I = U_0^\dagger W U_0$. The time evolution of the system due to the perturbation $W$ has been separated from the free part associated with $H_0$. We are adopting, throughout the discussion, this representation known as "Interaction Picture". Kets and observables will be subscripted by an "I" under this representation. Finally, the equation of motion results

$$i\frac{\partial}{\partial t}\left|\psi_I(t)\right\rangle = W_I\left|\psi_I(t)\right\rangle, \tag{A24}$$

where kets and observables are related with the previous representation or "Schödinger Picture" as

$$\begin{aligned}\left|\psi_I(t)\right\rangle &= U_0^\dagger\left|\psi_S(t)\right\rangle \\ A_I(t) &= U_0^\dagger A_S(t)U_0.\end{aligned} \tag{A25}$$

If we solve the integral equation (A23) by successive interation we obtain

$$U_I(t) = 1 + \sum_{n=1}^{\infty} U_I^{(n)}(t) \tag{A26}$$

where

$$U_I^{(n)}(t,t_0) = (-i)^n \int_{t_0}^{t} d\tau_1 \int_{t_0}^{\tau_1} d\tau_2 ... \int_{t_0}^{\tau_{n-1}} d\tau_n\ \ W_I(\tau_1)W_I(\tau_2)...W_I(\tau_n). \tag{A27}$$

for $t > \tau_1 > \tau_2 > ... > \tau_n > 0$.

Due to the time order condition we can write the time evolution operator in the interaction picture as

$$U_I(t,t_0) = \mathcal{T}\exp\left\{\left[-i\int_{t_0}^{t} d\tau W_I(\tau)\right]\right\}, \tag{A28}$$

where $\mathcal{T}$ is the time order operator action on the exponential, giving the same result as (A26).

We are interested in coming back to Schrödinger picture since we want to separate the proper time dependence of operators from the time variation of the total system. Then, using equations (A25) we finally get

$$U(t,t_0) = U_0(t,t_0)\ \mathcal{T}\exp\left\{\left[-i\int_{t_0}^{t} d\tau\ U_0^{\dagger}(\tau,t_0)\ W(\tau)\ U_0(\tau,t_0)\right]\right\}. \tag{A29}$$

As an example, first order and second order terms result

$$\begin{aligned} U^{(1)}(t,t_0) &= -i\int_{t_0}^{t} d\tau_1 U_0(t-\tau)W(\tau_1)U_0(\tau_1,t_0) \\ U^{(2)}(t,t_0) &= -\int_{t_0}^{t} d\tau_1 \int_{t_0}^{\tau_2} d\tau_2\ U_0(t-\tau)W(\tau_1)U_0(\tau_1,\tau_2)W(\tau_2)U_0(\tau_2,t_0) \end{aligned} \tag{A30}$$

Ultimately, all matter-radiation interactions considered in this paper will be evolve in time adiabatically. This means that the interactions will be switched on "very slowly" up to the time of interaction

$$W_\eta(t) = e^{-\eta t}W(t)\,,\ \eta \to 0^+. \tag{A31}$$

In all calculations is assumed that $t_0 \to -\infty$. This initial condition means that the Rabi frecuency $\Omega_{Rabi}$, which describes the time it takes to produce an atomic transition in a given light field, satisfies $\Omega_{Rabi} \ll T - t_0$, i.e. the atom is being excitated slowly enough to perform an atomic excitation transition. Furthermore, this approximation allows us to neglect spurious terms arisen from the sudden approximation which does not describe properly the finite time excitation of atoms [20].

This time-dependent approach will be used all along this paper for the description of matter-radiation interaction.

### 3. Atom-Radiation Interaction

The interaction model used in this paper, based on [25], consists on a non relativistic quantum mechanical description of spinless charged particles coupled with the quantum radiation field already described.

The $(\mathbf{x}_i, \mathbf{p}_i)$ variables associated with each particle of charge $q_j$ are operators in the Hilbert Space unless where they are arguments of the electromagnetic field where they specify the point of evaluation of the fields. Under the minimal coupling scheme the total Hamiltonian of a system of charged particles is

$$H = H_{EM} + \sum_j \frac{\left(\mathbf{p}_j - e_j\mathbf{A}_\perp(\mathbf{x}_j)\right)^2}{2m_i} + \sum_{i>j}\frac{e_i e_j}{4\pi|\mathbf{x}_i - \mathbf{x}_j|}. \tag{A32}$$

The coulombian interaction is not included in the minimal coupling so it is added with the last term of the hamiltonian above. With this interaction approach we may define the state system as the tensor product of the atomic states and photonic states $|\psi\rangle = |s\rangle \otimes |\gamma\rangle$. Atomic states can describe bounded states or free states of electrons. On the other hand, photonic states gives the number of photons which are interacting with atoms. We may separate the free hamiltonian $H_0 = H_{EM} + H_{mat}$ from the interaction terms $W$ and make some approximations. The matter hamiltonian $H_{mat}$ is the sum of the kinetic energy of each particle and the coulombian interaction between them

$$H_{mat} = \sum_j \frac{\mathbf{p}_j^2}{2m_i} + \sum_{i>j}\frac{e_i e_j}{4\pi|\mathbf{x}_i - \mathbf{x}_j|}. \tag{A33}$$

In our calculations, hydrogen atoms with one valence atom are involved so we set that the atom can be described as a system of energy levels

$$H_{mat} = \sum_s \omega_s\, |s\rangle\, \langle s|\,. \tag{A34}$$

Furthermore, we may neglect, in first approximation, the interaction of the nucleus with electromagnetic field as well as its dynamics. This implies we only have to describe electrons whose vector position, assuming a classical two body dynamic of the atom, is given by

$$\mathbf{x}_j = \mathbf{R}_j^{cm} + \frac{M_n}{m_e + M_n}\mathbf{r}_j \approx \mathbf{R}_j^{cm} + \mathbf{r}_j, \tag{A35}$$

as nucleous is much massive than electrons $M_n \gg m_e$. Furthermore the centre of mass position is much larger than the radius of the hydrogen type atom $\mathbf{R}_j^{cm} \gg \mathbf{r}_j$, $\mathbf{R}_j^{cm}$ will be consider a classical variable meanwhile $\mathbf{r}_j$ a quantum operator.

With all this considerations the interaction hamiltonian $W$, eliminating $\mathbf{A}_\perp^2$ terms,in the Coulomb gauge, remains

$$W = -\sum_j \frac{e_j}{m_j} \mathbf{p}_j \cdot \mathbf{A}_\perp(\mathbf{x}_j). \tag{A36}$$

where the sum runs over for every electron of the system. Computing this interaction vertex between in a excitation process we obtain

$$\langle A^+| W |A^-, \gamma_{\mathbf{k}}^\alpha\rangle = -\frac{e}{m} \langle A^+| \mathbf{p} \cdot \epsilon_{\mathbf{k},\alpha}\ e^{i\mathbf{k}\cdot\mathbf{r}} |A^-\rangle e^{i\mathbf{k}\cdot\mathbf{R^{cm}}}. \tag{A37}$$

As in a typical atomic transition in the optical region the wavelength of the emitted photon is much greater than the radius of the atom, $\lambda = \frac{2\pi}{|\mathbf{k}|} \gg |\mathbf{r}|$ we can approximate $e^{i\mathbf{k}\cdot\mathbf{r}} \approx 1$ by its leading term so the interaction has only a $(\mathbf{p} \cdot \epsilon_{\mathbf{k},\alpha})$ term. This approach is known as the electric dipole $E1$ approximation. Using the relation $[H_{mat}, \mathbf{r}] = -i\frac{\mathbf{p}}{m}$ we obtain that the Hamiltonian interaction reads

$$W_{E1} = -\mathbf{d}_A \cdot \mathbf{E}_\perp(\mathbf{R}^{cm}) \tag{A38}$$

where $\mathbf{d}_A = e\mathbf{r}$ is the electric dipolar operator. Unlike the minimal coupling approach electric dipole approximation includes coulombian interaction between atoms. Henceforce, we assume that all fields are transverse and the symbol $\perp$ will be omitted.

### 4. Diagrammatic Representation of Interactions

Based on the previous interaction model we are going to present the diagrammatic representation of interaction processes of Ref. [1–3]. These diagrams contain temporal and spatial axis,vertical and horizontal respectively, where the photon and atomic dynamics is depicted. Photons' propagators are represented by wavy lines and atoms states' propagators by straight lines, with a specific atomic state, showing the position of their centre of mass. The coupling between photons and atoms is represented by a vertex of interaction.

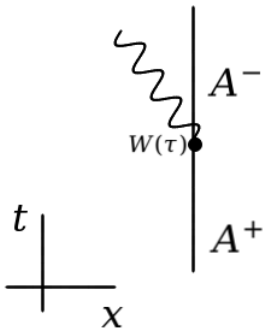


FIG. 11. Interaction Vertex

This diagrams represents the probability amplitude between two arbitrary states $|a\rangle$ and $|b\rangle$ at a time $T$

$$\mathcal{A}_{a\to b} = \langle b| U(T) |a\rangle . \tag{A39}$$

Using the series expansion (A27) of the time evolution operator $U(T)$ yields that the probability amplitude is an infinite sum of amplitudes of order n in the perubation $W$

$$\mathcal{A}_{a\to b} = \sum_{n=0}^{\infty} \mathcal{A}_{a\to b}^{(n)} = \sum_{n=0}^{\infty} \langle b| U^{(n)}(T) |a\rangle . \tag{A40}$$

Each term of the sum below corresponds to a diagram where the number of interaction vertex is equal to the order of the expansion term. With this approach we can describe self-interaction and interatomic interactions.

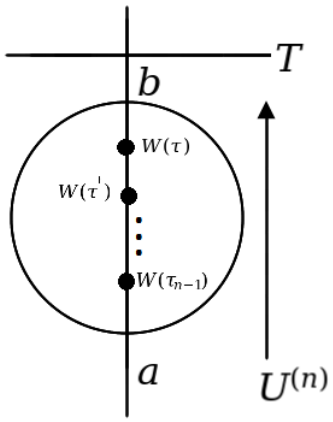


FIG. 12. Interactions $W$ take place until the time of observation $T$. There is no time location but a time order of the perturbations. Spatial distribution of perturbations is allowed provided that causality is preserved.

A diagramatic representation of probability,

$$P = |\mathcal{A}_{a\to b}|^2 = \sum_{n=0}^{\infty} \mathcal{A}^{(n)\dagger}_{a\to b} \sum_{m=0}^{\infty} \mathcal{A}^{(m)}_{a\to b} \tag{A41}$$

consists of the norm of $\mathcal{A}$. As probability amplitudes are expanded as a sum of partial amplitudes $\mathcal{A}^n$, probability is expresed as the sum of products of two partial probability amplitudes.

$$P \approx \mathcal{A}^{(0)\dagger}_{a\to b}\mathcal{A}^{(0)}_{a\to b} + \mathcal{A}^{(1)\dagger}_{a\to b}\mathcal{A}^{(0)}_{a\to b} + \mathcal{A}^{(0)\dagger}_{a\to b}\mathcal{A}^{(1)}_{a\to b} + \mathcal{A}^{(1)\dagger}_{a\to b}\mathcal{A}^{(1)}_{a\to b} + ... \tag{A42}$$

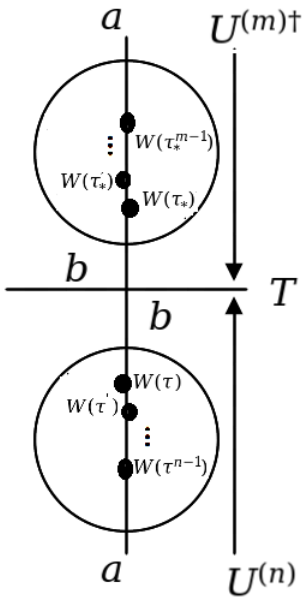


FIG. 13. Probability term of $(n + m)$ order. Both partial amplitudes evolve from the initial state $|a\rangle$ up to the time of observation $T$ whose final state is $|b\rangle$.

Terms with $n \neq m$ stand for interference terms. The order of each product is the sum of the orders of the amplitudes involved.

Finally, diagrammatic representation is also useful in the calculation of expectation values of operators.

$$\langle \mathcal{O}(t)\rangle = \langle \psi(t)|\, \mathcal{O}\, |\psi(t)\rangle = \langle \psi(0)|\, U^\dagger(t)\mathcal{O}U(t)\, |\psi(0)\rangle\,. \tag{A43}$$

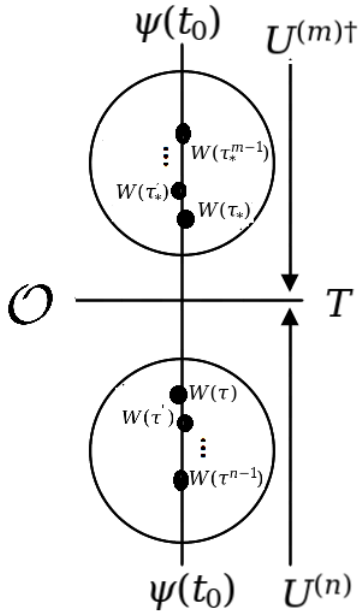


FIG. 14. Digramatic representation of the expectation value of the operator $\mathcal{O}$

Each time evolution operator is power expanded, up to the desired order, and the system evolves from the initial state $|\psi(t_0)\rangle$ to the time T at which the operator $\mathcal{O}$ is applied.

### 5. Interactions Between Atoms in Electric Dipole Approximation: Diagrammatic Rules

Let $A$ and $B$ two two levels atoms separated a distance $\mathbf{R} = \mathbf{R}_B - \mathbf{R}_A$. Under the $E1$ approximation the coulombian interaction is neglected and the interaction hamiltonian of the system results $W = -\mathbf{d}_A \cdot \mathbf{E}_\perp(\mathbf{R}_A) - \mathbf{d}_B \cdot \mathbf{E}_\perp(\mathbf{R}_B)$. All the interactions are retarded taking account of the finite speed of propagation of electromagnetic radiation. This implies that the interactions between atoms are at least of second order in $W$. Suppose the initial state of the system is $|A_+, B_-\rangle$. The transmision of the initial excitation from $A$ to $B$ leading to a final state $|A_-, B_+\rangle$ is given by the matrix element $\mathcal{A}^{(2)} = \langle A_-, B_+| U^{(2)} |A_+, B_-\rangle$ which is represented by the time ordered diagrams

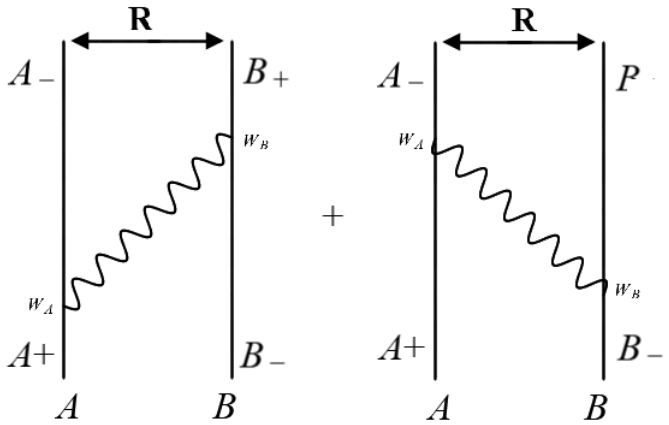


FIG. 15. Diagramatic representation of the interaction between two atoms by the exchange of a photon.

The photon propagation is given in the matrix element by the correlation function

$$\langle 0| \mathbf{E}(\mathbf{R}_A)\mathbf{E}(\mathbf{R}_B) |0\rangle = \int d\Theta \int dk k^3 \left(\mathbb{I} - \hat{\mathbf{k}}\hat{\mathbf{k}}\right) e^{i\mathbf{k}\cdot\mathbf{R}} \tag{A44}$$

Integration in the solid angle yields

$$\langle 0|\,\mathbf{E}(\mathbf{R}_A)\mathbf{E}(\mathbf{R}_B)\,|0\rangle = -\int_0^\infty \frac{dk}{\pi} k^2\,\mathrm{Im}\,\mathbb{G}^{(0)}(k,R) \tag{A45}$$

where $\mathbb{G}^{(0)}(k,R)$ is the dyadic Green's function of the electric field induced at $\mathbf{R}$ induced by an electric dipole of frecuency $\omega = ck$ in free space. It reads

$$\mathbb{G}^{(0)}(k,R) = \langle 0|\,[\mathbf{E}(\mathbf{R}_A)\mathbf{E}(\mathbf{R}_B))]\,|0\rangle = -\frac{ke^{ikR}}{4\pi}\left[\frac{\alpha}{kR} + \frac{i\beta}{(kR)^2} - \frac{\beta}{(kR)^3}\right] \tag{A46}$$

where $\alpha = \mathbb{I} - \hat{\mathbf{R}}\hat{\mathbf{R}}$, $\beta = \mathbb{I} - 3\hat{\mathbf{R}}\hat{\mathbf{R}}$. We denote the real and imaginary part of the the Green's function as $\mathrm{Re}\{\mathbb{G}^{(0)}(k,R)\}$ and $\mathrm{Im}\{\mathbb{G}^{(0)}(k,R)\}$, respectively. The real part corresponds to the coulombian potential between both atoms while the imaginary part corresponds to the radiation field interaction. Furthermore, the reciprocity in frecuency $\omega$ implies $\mathbb{G}^{(0)*}(\omega) = \mathbb{G}^{(0)}(-\omega)$. As Green's function do not depends on time we may give some rules for "reading" time dependent diagrams:

1. Compute the integration of the time evolution represented on the diagram. This integration typically yields a product of fractions with polynomials of order one in $\omega$ as denominators.

2. Each photon propagation corresponds to an electric correlation function $\langle 0|\,\mathbf{E}(\mathbf{R}_A)\mathbf{E}(\mathbf{R}_B)\,|0\rangle$. The integration in frecuencies must consider the time integrals computed of the previous step

3. Each vertex has associated a transition dipole element $\boldsymbol{\mu}_A = \langle A_-|\,d_A\,|A_+\rangle$

This rules has an implicity integration on every photon orientation.

## Appendix B: Calculations Details

### 1. Decay Rate $\Gamma(T)$ Eq.(3)

The time evolution of inferior probability amplitude at leading order of $\Gamma(T)$ which is represented in Fig. 1 a) reads

$$-i\int_{-\infty}^{T} d\tau\ e^{i\omega(T-\tau)}e^{\eta\tau}e^{-i\omega_A\tau} = -\frac{e^{\eta T}e^{-i\omega_A T}}{\omega - (\omega_A + i\eta)}. \tag{B1}$$

Then the probability of decay at leading order is

$$P_A = -\mu_A^i\mu_A^j\int_0^\infty \frac{d\omega}{\pi}\frac{e^{2\eta T}\,\mathrm{Im}\{G_{ij}(\omega,R)\}}{[\omega - (\omega_A + i\eta)]\,[\omega - (\omega_A - i\eta)]}, \eta \to 0^+. \tag{B2}$$

As $\mathrm{Im}\{G_{ij}\} = \frac{G_{ij} - G_{ij}^*}{2i}$, we integrate along the contour $C_+$ the Green's function $G_{ij}$ and its conjugated part $G_{ij}^*$ along the the contour $C_-$.

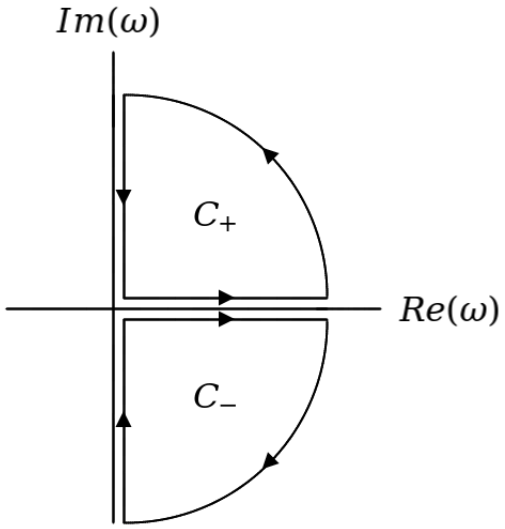


FIG. 16. Contours of integration $C_+$ and $C_-$

The integrals along the arcs tends to zero as $\omega \to \infty$ and we neglected the contributions of the vertical axis since are exponentially damped and they are small. Then the value of the integral on the real axis is the residue associated to the simple poles. The final result, as $R \to$, reads

$$\Gamma_A = \frac{d}{dT}\frac{e^{2\eta T}|\boldsymbol{\mu}_A|^2\omega_A^3}{6\pi} = \frac{\omega_A^3|\boldsymbol{\mu}_A|^2}{3\pi}. \tag{B3}$$

The poles of the second order decay rate, which corresponds to the diagram Fig. 1 b), are given by

$$(-i)^3\int_{-\infty}^{T}d\tau\int_{-\infty}^{\tau}d\tau'\int_{-\infty}^{\tau'}d\tau'' e^{-i\omega(T-\tau)}e^{\eta\tau}e^{-i\omega_A(\tau-\tau')}e^{\eta\tau'}e^{-i\omega(\tau'-\tau'')}e^{\eta\tau''}e^{-i\omega_A\tau''}\times -\frac{e^{\eta T}e^{-i\omega_A T}}{\omega-(\omega_A-i\eta)}. \tag{B4}$$

This lead to the integration in frecuencies

$$2Re\left\{i\mu_A^i\mu_A^j\mu_A^p\mu_A^q\int_0^\infty\frac{d\omega}{\pi}\int_0^\infty\frac{d\omega'}{\pi}\frac{\omega^2\omega'^2\,\mathrm{Im}\{G_{ij}(\omega,R)\}\,\mathrm{Im}\{G_{pq}(\omega',R)\}}{2\eta\left[\omega-(\omega_A-i\eta)\right]\left[\omega-(\omega_A+i3\eta)\right]\left[\omega'-(\omega_A+i\eta)\right]}\right\},\eta\to 0^+, \tag{B5}$$

which yields

$$\Gamma_A^{(2)} = -\mu_A^i\mu_A^j\mu_A^p\mu_A^q\frac{d^2}{dT^2}\frac{\omega_A^4 e^{4\eta T}}{2\eta^2}\,\mathrm{Im}\{G_{ij}(\omega,R)\}\,\mathrm{Im}\{G_{pq}(\omega,R)\} = -2\Gamma_0^2. \tag{B6}$$

Finally, the poles of the third order decay rate, which corresponds to the diagram Fig. 1 c), are separated in the contribution of the first diagram and the contribution of the conjugated diagrams. The first diagram has the poles

$$-\mu_A^i\mu_A^j\mu_A^p\mu_A^q\mu_A^r\mu_A^s\int_0^\infty\frac{d\omega}{\pi}\int_0^\infty\frac{d\omega'}{\pi}\int\frac{d\omega''}{\pi}\frac{e^{6\eta T}\omega^2\,\mathrm{Im}\{G_{ij}(\omega,R)\}\omega'^2\,\mathrm{Im}\{G_{pq}(\omega',R)\}\omega''^2\,\mathrm{Im}\{G_{rs}(\omega'',R)\}}{4\eta^2\left[\omega-(\omega_A-i\eta)\right]\left[\omega'-(\omega_A+i3\eta)\right]\left[\omega'-(\omega_A-i3\eta)\right]\left[\omega''-(\omega_A+i\eta)\right]},\eta\to 0^+, \tag{B7}$$

The time integration of conjugated diagrams results

$$(-i)^5\int_{-\infty}^{T}d\tau\int_{-\infty}^{\tau}d\tau'\int_{-\infty}^{\tau'}d\tau''\int_{-\infty}^{\tau''}d\tau'''\int_{-\infty}^{\tau'''}d\tau^{iv}e^{-i\omega(T-\tau)}e^{\eta\tau}e^{-i\omega_A(\tau-\tau')}e^{\eta\tau'}e^{-i\omega'(\tau'-\tau'')}e^{\eta\tau''}e^{-i\omega_A(\tau''-\tau''')}\times e^{\eta\tau'''}e^{-i\omega''(\tau'''-\tau^{iv})}e^{\eta\tau^{iv}}e^{-i\omega_A\tau^{iv}}. \tag{B8}$$

As result of time integration we obtain

$$-2\,\mathrm{Re}\left\{\mu_A^{ijpqrs}\int_0^\infty\frac{d\omega}{\pi}\int_0^\infty\frac{d\omega'}{\pi}\int_0^\infty\frac{d\omega''}{\pi}\frac{e^{6\eta T}\omega^2\,\mathrm{Im}\{G_{ij}(\omega,R)\}\omega'^2\,\mathrm{Im}\{G_{pq}(\omega',R)\}\omega''^2\,\mathrm{Im}\{G_{rs}(\omega'',R)\}}{8\eta^2\left[\omega''-(\omega_A-i\eta)\right]\left[\omega''-(\omega_A+i5\eta)\right]\left[\omega'-(\omega_A+i3\eta)\right]\left[\omega-(\omega_A+i\eta)\right]}\right\},\eta\to 0^+. \tag{B9}$$

The sum of both contributions yields

$$\Gamma^{(3)}=-\mu_A^i\mu_A^j\mu_A^p\mu_A^q\mu_A^r\mu_A^s\,\mathrm{Im}\{G_{ij}(\omega,R\to 0)\}\,\mathrm{Im}\{G_{pq}(\omega,R\to 0)\}\,\mathrm{Im}\{G_{rs}(\omega,R\to 0)\}\frac{d^3}{dT^3}\frac{e^{6\eta T}\omega_A^6}{6\eta^3}=\frac{9}{2}\Gamma_0^3. \tag{B10}$$

Summing the contributions of each diagram we obtain

$$\Gamma(T)=\Gamma_0-2\Gamma_0^2T+\frac{9}{2}\Gamma_0^3T^2. \tag{B11}$$

which do not coincide with Eq. (3). It is necessary to divide each term of the sum by $n^{n-1}$ where n is the order in $T$ of the calculated diagram. We adopt the criterion of dividing each term of the perturbative expansion of a probability amplitude by $n^n$ for the resumation of the perturbative expansion terms.

### 2. Matrix Elements $\mathcal{U}$ Eqs.(6)

In this section, I write the calculation of first two terms of the perturbative expansion of the matrix element $\mathcal{U}_{(A_+,B_-)\to(A_-,B_+)}$. The contour of integration of Fig. 16 is used. The time integral of the first order term of the matrix element $\mathcal{U}_{(A_+,B_-)\to(A_-,B_+)}$ of Fig. 2 is

$$-\int_{-\infty}^{T}d\tau\int_{-\infty}^{\tau}d\tau' e^{-i\omega_B(T-\tau)}e^{\eta\tau}e^{-\omega(\tau-\tau'}e^{\eta\tau'}e^{-i\omega_a\tau'},\eta\to 0^+. \tag{B12}$$

This lead to the integration in frecuencies

$$\mathcal{U}_{sin}^{(1)}=i\mu_A^i\mu_A^j\int_0^\infty\frac{d\omega}{\pi}\frac{e^{2\eta T}e^{-i\omega_A T}\omega^2\,\mathrm{Im}\left\{G_{ij}\left(\omega_A,R\right)\right\}}{2\eta\left[\omega-\left(\omega_A+i\eta\right)\right]}=i\mu_A^i\mu_A^j\omega_A^2e^{-i\omega_A T}G_{ij}\left(\omega_A,R\right)\equiv i\mu_A^2\omega_A^2e^{-i\omega_A T}G\left(\omega_A,R\right) \tag{B13}$$

The time integral of the third order term of the matrix element $\mathcal{U}_{(A_+,B_-)\to(A_-,B_+)}$ of Fig. 2 is

$$\begin{aligned}-\int_{-\infty}^{T}d\tau\int_{-\infty}^{\tau}d\tau'\int_{-\infty}^{\tau'}d\tau''\int_{-\infty}^{\tau''}d\tau'''\int_{-\infty}^{\tau'''}d\tau^{iv}\int_{-\infty}^{\tau^{iv}}d\tau^{v}e^{-i\omega_B(T-\tau)}e^{\eta\tau}e^{-i\omega(\tau-\tau')}e^{\eta\tau'}\\ \times e^{-i\omega_A(\tau'-\tau'')}e^{\eta\tau''}e^{-i\omega'(\tau''-\tau''')}e^{\eta\tau'''}e^{-i\omega_B(\tau'''-\tau^{iv})}e^{\eta\tau^{iv}}e^{-i\omega'''(\tau^{iv}-\tau^{v})}e^{\eta\tau^{v}}e^{-i\omega_A\tau^{v}}.\end{aligned} \tag{B14}$$

This time integral leads to

$$\begin{aligned}\mathcal{U}_{sin}^{(3)}=&i\mu_A^i\mu_A^j\mu_A^p\mu_A^q\mu_A^r\mu_A^s\int_0^\infty\frac{d\omega}{\pi}\int_0^\infty\frac{d\omega'}{\pi}\int_0^\infty\frac{d\omega''}{\pi}\frac{e^{6\eta T}e^{-i\omega_A T}\omega^2\,\mathrm{Im}\left\{G_{ij}\left(\omega,R\right)\right\}\omega'^2\,\mathrm{Im}\left\{G_{ij}\left(\omega',R\right)\right\}\omega''^2\,\mathrm{Im}\left\{G_{ij}\left(\omega'',R\right)\right\}}{48\eta^3\left[\omega-\left(\omega_A+i\eta\right)\right]\left[\omega'-\left(\omega_A+i3\eta\right)\right]\left[\omega''-\left(\omega_A+i5\eta\right)\right]}\\ =&i\frac{9}{2}\mu_A^i\mu_A^j\mu_A^p\mu_A^q\mu_A^r\mu_A^s\omega_A^6G_{ij}\left(\omega_A,R\right)G_{pq}\left(\omega_A,R\right)G_{rs}\left(\omega_A,R\right)\equiv i\frac{9}{2}\mu_A^6\omega_A^6G^6\left(\omega_A,R\right)\end{aligned} \tag{B15}$$

Dividing each term by $n^n$ we obtain the first two terms of the function the matrix element $\mathcal{U}_{(A_+,B_-)\to(A_-,B_+)}$ (6). Similar calculations are needed for the $\mathcal{U}_{(A_+,B_-)\to(A_+,B_-)}$ matrix element.

### 3. Matrix Elements $\mathcal{U}$ Eqs.(10)

In this section, I write the calculation of the diagrams of the second row of Fig. 4 corresponding to the matrix element $\mathcal{U}_{(A_+,B_-)\to(A_-,B_-,\gamma_{\mathbf{k}})}$. The time integration of the first diagram taking into account the vertex of interaction yields

$$\mathcal{U}_{cos}^{(1)\omega}=-i\int_{-\infty}^{T}d\tau\;\;e^{i\omega(T-\tau)}e^{\eta\tau}\left\langle A-,\mathbf{k}\right|W\left|A_+\right\rangle e^{-i\omega_A\tau}=-\frac{e^{\eta T}e^{-i\omega_A T}}{\omega-\left(\omega_A+i\eta\right)}=-\frac{\left\langle A-,\mathbf{k}\right|W\left|A_+\right\rangle e^{\eta T}e^{-i\omega_A T}}{\omega-\left(\omega_A+i\eta\right)}. \tag{B16}$$

The photon of the final state has a generic momentum $\mathbf{k}$.

The time integral of the second diagram results

$$(-i)^5 \int_{-\infty}^{T} d\tau \int_{-\infty}^{\tau} d\tau' \int_{-\infty}^{\tau'} d\tau'' \int_{-\infty}^{\tau''} d\tau''' \int_{-\infty}^{\tau'''} d\tau^{iv} e^{-i\omega(T-\tau)} e^{\eta\tau} e^{-i\omega_A(\tau-\tau')} e^{\eta\tau'} e^{-i\omega'(\tau'-\tau'')} e^{\eta\tau''} e^{-i\omega_B(\tau''-\tau''')} \times e^{\eta\tau'''} e^{-i\omega''(\tau'''-\tau^{iv})} e^{\eta\tau^{iv}} e^{-i\omega_A\tau^{iv}}. \tag{B17}$$

After lengthy similar calculation the time integration yields

$$\mathcal{U}_{cos}^{(3)\omega} = \frac{\langle A-, \gamma_{\mathbf{k}}| W |A_+\rangle e^{5\eta T} e^{-i\omega_A T}}{8\eta^2 [\omega - (\omega_A + 5i\eta)]} \mu_A^4 \omega_A^4 G^2(\omega_A, R). \tag{B18}$$

The remaining pole of $\mathcal{U}_{cos}^{(1)}\,\mathcal{U}_{cos}^{(3)}$ are equivalent by means of integration. This implies that

$$U_{cos}^{\omega} \approx -\frac{\langle A-, \gamma_{\mathbf{k}}| W |A_+\rangle e^{\eta T} e^{-i\omega_A T}}{\omega - (\omega_A + i\eta)} \left(1 - \mu_A^4 \omega_A^4 \frac{e^{4\eta T}}{8\eta^2} G^2(\omega_A, R)\right). \tag{B19}$$

Dividing each term by it $n^n$ this matrix element converges to

$$U_{cos}^{\omega} = -\frac{\langle A-, \gamma_{\mathbf{k}}| W |A_+\rangle e^{\eta T} e^{-i\omega_A T}}{\omega - (\omega_A + i\eta)} e^{-\frac{\Gamma(R)}{2}T} \cos(\Omega T). \tag{B20}$$

### 4. Off Resonant vdW Interaction

Since $G_{ij}$ is analytic in the upper half-plane, it is useful ot consider the integral

$$I_{\pm} = \int_C dz \frac{G_{ij}(z, R)}{\omega_a \pm z + i\eta} \tag{B21}$$

on the contour of Fig. 17 [9].

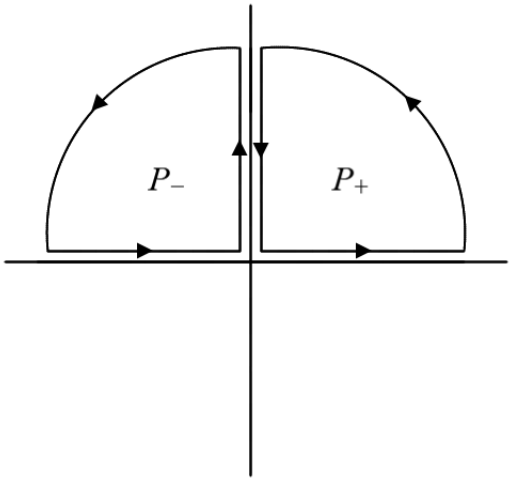


FIG. 17. Integration contours for Green's Function

For the $I_{\pm}$ we use the contour $P_{\pm}$ on te complex z plane. In the upper half-plane $G_{ij}(z, R)$ falls off at least as fast as $|z|$ so the integrand of (B21) vanishes at $|z| \to \infty$. Hence, using $I_+$ we find

$$\int_0^{\infty} d\omega \frac{G_{ij}(\omega, R)}{\omega_a + \omega + i\eta} = i \int_0^{\infty} d\omega \frac{G_{ij}(i\xi, R)}{\omega_a + i\xi + i\eta} \tag{B22}$$

whereas form $I_-$ we get

$$\int_C dz \frac{G_{ij}(z, R)}{\omega_a \pm z + i\eta} = i \int_0^{\infty} d\omega \frac{G_{ij}(i\xi, R)}{\omega_a - i\xi + i\eta} + 2\pi i G_{ij}(\omega_a, R). \tag{B23}$$

From this properties we are able to vdW non resonant interactions by terms of a Wick rotation $\omega = iu$. We obtain

$$\langle W^{off}\rangle_{AB} = -\frac{2\mu_A^i\mu_B^j\mu_A^p\mu_B^q}{\pi}\int_0^\infty du\ \frac{u^2\omega_A\omega_B}{[\omega_A^2+u^2]\,[\omega_B^2+u^2]}G_{ij}(iu,R)G_{pq}(iu,R). \tag{B24}$$

From this Wick rotation is clear that the vdW non resonant potential changes its sign when one of the atoms is initially excited.

### 5. Resonant vdW Interaction

In this section, I write the integral expressions of $\langle W_A\rangle$ and $\langle W_A\rangle$ of Eqs. (15) and (16) and Fig. 8. In all the calculation $\omega \gg \Omega$ is considered.

$$\begin{aligned}\langle W_A\rangle =& -2\,\mathrm{Re}\,\mu_A^i\mu_A^j\int_0^\infty \omega^2\,\mathrm{Im}\left\{G_{ij}(\omega,R)\right\}e^{-i\omega_A T}\cos(\Omega T)\times\int_{-\infty}^{T}d\tau e^{i\omega(T-\tau)}e^{\eta\tau}e^{i\omega_A\tau}\sin(\Omega\tau)\\ =& \,\mathrm{Re}\left\{i\int\frac{d\omega}{\pi}\frac{\omega^2\,\mathrm{Im}\left\{G_{ij}(\omega,R)\right\}}{[\omega-(\omega_A-i\eta)]}e^{2\eta T}\sin(2\Omega T)\right\},\eta\to 0^+.\end{aligned} \tag{B25}$$

$$\begin{aligned}\langle W_A\rangle =& -2\mu_A^i\mu_A^j\,\mathrm{Re}\int_0^\infty \omega^2\,\mathrm{Im}\left\{G_{ij}(\omega,R)\right\}e^{-i\omega_A T}\sin(\Omega T)\times\int_{-\infty}^{T}d\tau e^{i\omega(T-\tau)}e^{\eta\tau}e^{i\omega_A\tau}\cos(\Omega\tau)\\ =& \,\mathrm{Re}\left\{-i\mu_A^i\mu_A^j\int\frac{d\omega}{\pi}\frac{\omega^2\,\mathrm{Im}\left\{G_{ij}(\omega,R)\right\}}{[\omega-(\omega_A+i\eta)]}e^{2\eta T}\sin(2\Omega T)\right\},\eta\to 0^+.\end{aligned} \tag{B26}$$


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