Quasi-resonant van der Waals interaction between non-identical atoms

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We present a time-dependent quantum calculation of the van der Waals interaction between a pair of dissimilar atoms, one of which is initially excited while the other one is in its ground state. For small detuning, the interaction is predominantly mediated at all distances by the exchange of doubly resonant photons between the two atoms. We find that it presents, generally, both temporal and spatial oscillations. Spatially oscillating terms depend on the resonant frequencies of both atoms, while the frequency of the time oscillations is given by their detuning. We analyse the physical content of our findings and discuss to what extent previous conflicting stationary approaches provide compatible results.

Dispersion forces between neutral atoms are often interpreted as a result of the quantum fluctuation of both the electromagnetic (EM) field and the atomic charges. At zero temperature, in the electric dipole approximation and for atoms in their ground state, the atoms undergo a series of virtual E1 transitions to upper levels. It is the coupling of the charges of each atom to the quantum EM field that induces the correlation between their transient dipole moments, giving rise to a non-vanishing interaction. The lifetime of the virtual atomic transitions is very short in comparison to ordinary observation times and thus, the use of stationary quantum perturbation theory is well justified for the calculation of this interaction. Alternatively, same results are obtained using linear response theory. For short interatomic distances \( r \) in comparison to the relevant transition wavelengths, in the so-called non-retarded regime, the interaction scales as \( \sim 1/r^6 \). For large distances, in the so-called retarded regime, it goes like \( \sim 1/r^7 \).

The situation is different for excited atoms. In the first place, excited states are unstable and present finite lifetimes. This implies that, generically, the interaction between excited atoms must depend on time. Second, if any of the transitions from the excited to lower atomic levels is relevant to the interaction, the exchange of resonant photons between the atoms must be considered. It is the energy of the interaction mediated by resonant photons which is usually referred to as resonant van der Waals potential in the literature. In the retarded regime the resonant potential overtakes by far the non-resonant one. It is in this regime that different approaches yield conflicting results concerning the spatial oscillations of the interaction. This long-standing problem is the main motivation of the present Letter.

In the following, we address the time-dependent quantum computation of the interaction between two dissimilar atomic dipoles, one of which is excited. The excited atom is taken of type \( A \) while the atom in its ground state is considered of a different type \( B \). Without loss of generality we approximate the atoms by two-level systems of resonant frequencies \( \omega_A \) and \( \omega_B \) respectively, with respective linewidths \( \Gamma_A \) and \( \Gamma_B \). Further, in order to ensure the perturbative nature of the calculation and to avoid resonant energy transfer we set the detuning \( \Delta_{AB} = \omega_A - \omega_B \) such that \( |\Delta_{AB}| > (\Gamma_A + \Gamma_B)/2 \) and \( |\Delta_{AB}| \gg (W(T))/\hbar \), with \( W(T) \) being the interaction Hamiltonian at the time of observation, \( T \). Since the observation is made for atom \( A \) excited, we must have \( T \lesssim 2\pi\Gamma_A^{-1} \). Lastly, we assume without much loss of generality \( \Gamma_{A,B} < |\Delta_{AB}| \ll \omega_{A,B} \), which is easily met by pairs of alkali atoms. We will refer to this condition as quasi-resonant. We will see that it allows for a great reduction in the number of calculations and makes the resonant potential dominant at all distances. We will show that the interaction energy oscillates both in time and in space. It contains time-independent terms which oscillate in space with frequency \( 2\omega_A/c \), and time-dependent terms which oscillate in time with frequency \( \Delta_{AB} \) and in space with frequency \( 2\omega_B/c \). We compare our results to previous conflicting approaches and discuss in detail to which extent they provide compatible results.

We aim at computing the EM energy of atom \( A \) due to the presence of atom \( B \). To this end we apply standard time-dependent quantum perturbative techniques in the electric dipole approximation. At any given time \( T \) the state of the two-atom-vacuum system can be written as \( |\Psi(T)\rangle = U(T)|\Psi(0)\rangle \), where the state of the system at time 0 is \( |\Psi(0)\rangle = |A_+\rangle \otimes |B\rangle \otimes |0_s\rangle \). In this expression \( |A_+\rangle \otimes |B\rangle \otimes |0_s\rangle \) label the upper/lower internal states of the atoms \( A \) and \( B \) respectively and \( |0_s\rangle \) is the EM vacuum state. \( U(T) \) denotes the time evolution operator in the Schrödinger representation,

\[
U(T) = T - \exp \left\{ -i\hbar^{-1} \int_0^T dt \left[ H_A + H_B + H_{EM} + W(t) \right] \right\}.
\]

In this equation \( H_A + H_B \) is the free Hamiltonian of the internal atomic states,

\[
H_A + H_B = \hbar \omega_A |A_+\rangle \langle A_+ | + \hbar \omega_B |B_+\rangle \langle B_+ |,
\]

while the Hamiltonian of the free EM field is

\[
H_{EM} = \sum_{\mathbf{k},\epsilon} \hbar \omega (a_{\mathbf{k},\epsilon} a_{\mathbf{k},\epsilon}^\dagger + 1/2),
\]

where \( a_{\mathbf{k},\epsilon} \) and \( a_{\mathbf{k},\epsilon}^\dagger \) are the bosonic annihilation and creation operators of the EM field.
where $\omega = c k$ is the photon frequency, and the operators $a_{k,\epsilon}$ and $a_{k,\epsilon}^\dagger$ are the creation and annihilation operators of photons with momentum $\hbar k$ and polarization $\epsilon$ respectively. Finally, the interaction Hamiltonian reads $W = W_A + W_B$, with $W_{A,B} = -\mathbf{d}_{A,B} \cdot \mathbf{E}(\mathbf{R}_{A,B})$. In this expression $\mathbf{d}_{A,B}$ are the electric dipole operators of each atom and $\mathbf{E}(\mathbf{R}_{A,B})$ is the electric field operator evaluated at the position of each atom, which can be written in the usual manner as a sum over normal modes as

$$E(\mathbf{R}_{A,B}) = \sum_k E_k^-(\mathbf{R}_{A,B}) + E_k^+(\mathbf{R}_{A,B})$$

$$= i \sum_{k,\epsilon} \sqrt{\frac{\hbar c k}{2Vc_0}} [\epsilon a_k e^{i k \mathbf{R}_{A,B}} - \epsilon^* a_k^\dagger e^{-i k \mathbf{R}_{A,B}}],$$

where $V$ is a generic volume and $E_k^{(\pm)}$ denote the annihilation/creation electric field operators of photons of momentum $\hbar k$, respectively. While the internal atomic and EM degrees of freedom are quantum variables, the position vectors $\mathbf{R}_{A,B}$ are classical variables. We emphasize here that we do not make further simplifications to these potentials. In particular, we do not replace the EM response of any of the atoms by its ordinary polarizability, as it is the case in Ref. [21].

Next, considering $W$ as a perturbation to the free Hamiltonians, the unperturbed time-evolution diagonal operator of the atomic states is

$$\mathbb{U}^{\text{at}}(t) = [A_-] \langle A_- | + e^{-i \omega_A t} | A_+ \rangle \langle A_+ |$$

$$\otimes [B_-] \langle B_- | + e^{-i \omega_B t} | B_+ \rangle \langle B_+ |,$$

while that for free photons reads

$$\mathbb{U}^\gamma(t) = \sum_{\gamma_{k,\epsilon}} e^{-i \omega_{k,\epsilon} t} | \gamma_{k,\epsilon} \rangle \langle \gamma_{k,\epsilon} |.$$

In order to make contact with a realistic setup, we imagine that atom $A$ starts being excited at time $-\tau$ by a laser pulse of duration $\tau \ll \Gamma_A^{-1}$. This fixes our temporal resolution and implies that at time $\approx 0$ the initial state $|\Psi(0)\rangle$ is well-defined within a time interval of the order of $\tau$. We are now ready to compute the EM energy of atom $A$ due to the presence of atom $B$ at any time $T$ such that $0 \leq T \leq 2\pi \Gamma_A^{-1}$,

$$\langle W_A(T) \rangle = \langle \Psi(T) | W_A | \Psi(T) \rangle = -\langle \Psi(0) | \mathbb{U}^{\text{at}}(T) \mathbf{d}_A \cdot \mathbf{E}(\mathbf{R}_A) \mathbb{U}^{\gamma}(T) | \Psi(0) \rangle.$$  \hspace{1cm} (1)

The above expression admits an expansion in powers of $W$ which can be developed out of the time-ordered exponential equation for $\mathbb{U}(T)$,

$$\mathbb{U}(T) = [\mathbb{U}^{\text{at}} \otimes \mathbb{U}^\gamma](T)$$

$$= T \text{-exp} \int_0^T dt [\mathbb{U}^{\text{at}} \otimes \mathbb{U}^\gamma]^\dagger(t) \cdot W(t) \cdot [\mathbb{U}^{\text{at}} \otimes \mathbb{U}^\gamma](t).$$  \hspace{1cm} (2)

At leading order, Eq. (1) contains a series of terms of fourth order in $W$ where an electric field operator creates/annihilates a photon at time $T$ at the position of atom $A$. They correspond to the twelve well-known time-ordered diagrams of Fig. 1 [3, 13]. In the time-dependent approach, each diagram contributes to $\langle W_A(T) \rangle$ with two terms in which $W_A$ is flanked by two $\mathbb{U}$-matrices which make the system evolve, in opposite time directions, from the initial state to two different states at time $T$, which differ from one another in the state of atom $A$ and in the number of photons by one unit. In quasi-resonant condi-

FIG. 1: Diagrammatic representation of the twelve time-ordered processes which contribute to the van der Waals interaction between atoms $A$ and $B$ at the lowest order in $W$. $R$ denotes the distance between the atoms. The time variable runs along the vertical.
integrals, we find at leading order,
\[
\langle W_A(T) \rangle \approx \frac{1}{2} \int_{-\infty}^{\infty} \frac{dk}{(2\pi)^3} \int_{-\infty}^{\infty} \frac{dk'}{(2\pi)^3} \int_{0}^{4\pi} d\Omega \int_{0}^{4\pi} d\Omega' \times \\
\left[ \langle \Psi(0) \rangle | \mathcal{U}^T \otimes \mathcal{U} \rangle | (-T) \rangle \langle \Psi(0) \rangle \right] \left( -i \right)^3 \\
\times \int_{T}^{T} dt \int_{0}^{t} dt' \int_{0}^{t'} d\omega' \langle \Psi(0) \rangle | d_A \cdot E_{k'}(R_A) \rangle \\
\times \mathcal{U}^T(t) d_B \cdot E_k(R_B) \\
\times \mathcal{U}^T(t-t') d_A \cdot E_{k'}(R_A) \\
\times \mathcal{U}^T(t' + t'') \langle \Psi(0) \rangle + [k \leftrightarrow k'].
\]

The time integrals of the time-evolution operators in Eq. (3) yield a series of terms with poles along the real axis,
\[
\frac{1}{\Delta_{AB}(k - k_A)(k' - k_A)} - \frac{\cos(\Delta_{AB}T)}{\Delta_{AB}(k - k_B)(k' - k_B)} + \frac{\cos(\omega_A T)}{(k - k_A)(k' - k_B)(k' - k)}
\]

Further, the development of this expression contains terms in which both photons resonate either with the transition of atom A or with the transition of atom B only. This is a direct consequence of energy conservation. Upon integration in frequencies, the former terms are time-independent while the latter oscillate in time as \( \sim \cos \Delta_{AB}T \). Important is the fact that only the first term in Eq. (4) arises in the stationary perturbative approach [3, 7, 13]. However, the integration in frequencies of the third and fourth terms provides additional time-frequency-independent contributions which are missing in the stationary approach. Assuming that the causality condition \( T > 2R/c \) holds, with \( R = R_B - R_A \), replacing the time integrals in Eq. (3) with the result (4) and integrating in orientations and frequencies, we obtain
\[
\langle W_A(T) \rangle \approx \frac{U_{ijpq}}{R^6} \left[ \delta^{ij} \beta^{pq} - k_A^2 R^2 (\delta^{ij} \beta^{pq} + 2\alpha^{ij} \beta^{pq}) \right. \\
\left. + k_A^2 R^2 \alpha^{ij} \alpha^{pq} \cos(2k_A R) + \frac{2U_{ijpq}}{R^3} k_A [\delta^{ij} \beta^{pq}] \cos(2k_A R) \right. \\
\left. - k_A^2 R^2 \delta^{ij} \beta^{pq} \sin(2k_A R) \right. \\
\left. - \frac{U_{ijpq}}{R^6} (\delta^{ij} \beta^{pq} - k_B^2 R^2 (\delta^{ij} \beta^{pq} + 2\alpha^{ij} \beta^{pq}) \right. \\
\left. + k_B^2 R^2 \alpha^{ij} \alpha^{pq} \cos(2k_B R + \Delta_{AB} T) \right. - \frac{2U_{ijpq}}{R^3} k_B [\delta^{ij} \beta^{pq}] \cos(2k_B R + \Delta_{AB} T) \right. \\
\left. \times [\delta^{ij} \beta^{pq} - k_B^2 R^2 \delta^{ij} \beta^{pq} \sin(2k_B R + \Delta_{AB} T) \right. \\
\left. + \frac{U_{ijpq}}{R^6} (1 + ... + (k_{A,B} R)^4) \mathcal{O}(\Delta_{AB}/\omega_A) \right. + \ldots ,
\]

where \( T > 2R/c \) and \( U_{ijpq} = \mu_A^{ij} \mu_A^{pq} / (4\pi \epsilon_0)^2 \hbar \Delta_{AB} \), with \( \mu_A = \langle A^- | d_A | A^+ \rangle \), \( \mu_B = \langle B^- | d_B | B^+ \rangle \) and \( \beta^{ij} = \beta^{i1} - 3R^2 R^2/4 \). The last term in Eq. (4) indicates the order of the leading corrections to the dominant doubly resonant photon exchange terms of Eq. (3) [18]. As anticipated, the time-independent terms of Eq. (5) oscillate only in space with frequency \( 2k_A \). On the contrary, the time-dependent terms oscillate in time with frequency \( \Delta_{AB} \) and in space with frequency \( 2k_B \). Only for large integration times, \( \Delta T > \Delta_{AB} \), the time average of the latter vanishes. A necessary condition for this is \( \Delta_{AB} > \Gamma_A, \) since the lifetime of the excited state. In the short time limit, \( T \rightarrow 2R^+ c, \langle W_A(T) \rangle \) vanishes identically at our order of approximation. This is a consequence of the fact that, in order to establish the interaction, it is necessary that the excitation be transferred actually to atom B. Upon causality, \( T > R/c, \) the probability of excitation of atom B oscillates in time as \( | \langle \Psi(T) | A^- \rangle \otimes | B^+ \rangle \rangle \sim \sin^2 (\Delta_{AB}(R/c - T)/2), \) being maximum for the first time at \( T = R/c + \pi/\Delta_{AB} \). Correspondingly, \( \langle W_A(T) \rangle \) becomes maximum for the first time at \( T = 2R/c + \pi/\Delta_{AB} \). The lapse \( R/c \) between these two times is the time needed for a photon to travel back from \( R_B \) to \( R_A \) after the excitation of atom B.

A long-standing debate exists in the literature concerning the spatial oscillations of the two-atom interaction in the retarded regime when one of the atoms is excited [12–17]. The existence of spatial oscillations is indeed supported by experiments [22, 23]. According to our findings, for \( k_{A,B} R > 1 \) and \( T > 2R/c, \) the interaction oscillates both in time and in space as
\[
\langle W_A(T) \rangle \approx \frac{U_{ijpq}}{R^6} \alpha^{ij} \alpha^{pq} | k_A \cos(2k_A R) \right. \left. - k_B^2 \cos(2k_B R + \Delta_{AB} T) \right) \\
\times (\Delta_{AB}(R/c - T)/2) \sin(\Delta_{AB}(R/c + \pi/\Delta_{AB})).
\]

From the last expression we read that, at fixed time, the interaction is modulated by long-range oscillations of frequency \( \Delta_{AB}/c \), while short-range oscillations take place at frequency \( k_A + k_B \). Also as a function of time the interaction is modulated by oscillations of frequency \( \Delta_{AB} \). In Fig. 2 we plot the energy of the interaction between two alkali atoms, one of \( 87 \text{Rb} \) which is excited to the state \( 5P \) and another one of \( 40 \text{K} \) which is in its ground state, in the retarded regime.

In contrast to our result, the stationary quantum approach of Power and Thirunamachandran in Ref. [12] predicts no oscillations for \( \langle W_A \rangle \) in the far field. The key point in their calculation is the addition of small imaginary parts to the resonant frequency of atom A in such a way that poles get shifted off the real axis. They used the prescription that a positive/negative imaginary part must be added for emitted/absorbed photons in order to account for the finite linewidth of the excited atom.
In particular, for $\Delta_{AB} < \omega_{A,B}$ the dominant term in their stationary calculation is the first one in Eq. [4], but with the real poles shifted as $\left[\Delta_{AB}(k - k_A - i\eta/c)(k' - k_A + i\eta/c)\right]^{-1}$, $\eta \to 0^+$. After integrating in orientations an analogous equation to Eq. [3], they must have obtained for the energy in the far field limit, $k_A R \gg 1$,

$$\frac{\mathcal{U}_{ijpq}}{4\pi^2 R^2} \alpha_{ij}^{\alpha pq} \int_{-\infty}^{+\infty} dk \int_{-\infty}^{+\infty} dk' k'^2 k^2 e^{i(k-k')R} \left(\frac{e^{i(k+k')R} + e^{-(k+k')R} - e^{i(k-k')R} - e^{-(k-k')R}}{(k-k_A - i\eta/c)(k' - k_A + i\eta/c)}\right) \eta \to 0^+. \tag{7}$$

Since the pole in $k$ lies on the upper half of the complex plane and the pole in $k'$ lies on the lower half, the only nonvanishing contribution to the above integral comes from the term proportional to $e^{i(k-k')R}$. Since the real part of the poles is in both cases $k_A$, taking the limit $\eta \to 0^+$ the exponent vanishes after evaluating the residues and the integral yields the non-oscillating result $\frac{\mathcal{U}_{ijpq}}{R^2} k_A^4 \alpha_{ij}^{\alpha pq}$.

![FIG. 2: Graphical representation of the EM interaction between a $^{87}$Rb atom in state 5P ($k_A = 2\pi 12737$ cm$^{-1}$) and a $^{40}$K atom in its ground state ($k_B = 2\pi 13023$ cm$^{-1}$), in the retarded regime, $k_A R \gg 1$, and averaging over dipole orientations and fine structure levels. (i) Fixed time snapshot at $T_0 > 2R/c$. (ii) Fixed time snapshot at $T_0 + \pi/\Delta_{AB}$. In red dashed line, the envelope curve of the interaction at time $T_0$.](image-url)

In the previous stationary calculation of McLone and Power [13] and Gomberoff et al. [14] the poles in Eq. [7] were not shifted. As a result, when taking the principal value of the integrals in Eq. [4] with $\eta = 0$, the four exponentials in the numerator contribute as $\sim 2 \cos 2k_A R + 2 = 4 \cos^2 k_A R$ after adding up the residues, yielding the oscillating result $\frac{\mathcal{U}_{ijpq}}{R^2} k_A^4 \alpha_{ij}^{\alpha pq} \cos^2 k_A R$. As mentioned after Eq. [4], some time-independent terms are missing in the stationary calculation, which explains the discrepancy of this result with the time-independent component of ours in Eq. [4].

Recently, Safari and Karimpour have published a letter [17] where they claim to obtain for $k_A R \gg 1$ the same oscillating behaviour as Gomberoff et al. [14]. However, a straightforward comparison of Eq. (19) of Ref. [17] and Eqs. (14,26) of Ref. [14] reveals that this is indeed not the case. Whereas the result of the latter is the one outlined above, $\sim \cos^2 k_A R$, the authors of the former have found $\sim \cos 2k_A R$, despite the fact that both approaches are based on fourth order stationary perturbation theory. The origin of the discrepancy is in the algebraic manipulation inherited by the authors of Ref. [17] from Ref. [7]. In the Appendix B of Ref. [7] the authors have tried to express the total contribution of the twelve diagrams of Fig. 1 as a single frequency integral whose integrand is a function of the ordinary polarizabilities of the two atoms. In doing so by means of Eq. (B2) of Ref. [7], the authors have replaced effectively the denominator of Eq. [7], which is a symmetric and separable function of $k$ and $k'$ for $\eta = 0$, by the expression $\left[\Delta_{AB}(k - k_A)\right]^{-1}[1/(k' - k) + 1/(k' + k)]$, which is neither symmetric nor separable. As a consequence, that replacement makes the frequency integrals depend arbitrarily on the order of integration. Next, integrating in $k'$ first and in $k$ later, one obtains $\frac{\mathcal{U}_{ijpq}}{R^2} k_A^4 \alpha_{ij}^{\alpha pq} \cos 2k_A R$, which agrees with Eq. (19) of Ref. [17] for $k_A R \gg 1$, $\Delta_{AB} \ll \omega_{A,B}$, upon averaging in atomic orientations. Interestingly, this result equals the time-independent term found in Eq. (4). However, this coincidence can only be accidental, since the above replacement and the subsequent prescription on the order of integration are not connected to the time-dependent terms of Eq. (4) which cause the actual discrepancy with respect to the result of Refs. [13,14].

It is worth noting that while we have invoked the existence of finite lifetimes $\sim \Gamma_1^{-1}$ in order to impose physical constraints on the detuning $\Delta_{AB}$ and on the observation time $T$, no explicit reference to these quantities appear in our expression for $\langle W_A(T) \rangle$. As a matter of fact, only the emission through the exchange of resonant photons between the two atoms has been implicitly accounted for in our calculation of $\langle W_A(T) \rangle$. However, our calculation lacks the inclusion of the spontaneous emission of each atom into free space, whose rates are $k_A^3 \mu_{A,B}^2/3\pi\epsilon_0 \hbar$, respectively. The processes corresponding to the latter phenomenon are generally subdominant in comparison to those depicted in Fig. 1 since their leading contribution to $\langle W_A(T) \rangle$ is of order $\mathcal{O}(W^6) \sim \mu_1^2 \mu_3^3$, $\mu_2^2 \mu_3^2$—see Fig. 3. They might only be relevant for the case that the lifetimes are of the order of the temporal frequency of the interaction, $\Gamma_{A,B} \sim \Delta_{AB}$, but they cannot affect in any case the oscillatory behaviour found here for the terms of order $\mu_2^2 \mu_3^2$. This
argument opposes to the reason given in Ref. [12] to add an imaginary shift to the real poles at $O(W^4)$, which in turn causes the cancelation of the spatial oscillations.

In this Letter we have shown that the van der Waals interaction between two dissimilar atoms, one of which is initially prepared in an excited state, presents generically oscillations both in time and in space. In quasi-resonant conditions the interaction is dominated at all distances by the exchange of doubly resonant photons between the two atoms. It is modulated in space by long-range oscillations both in time and in space. In quasi-resonant conditions the interaction is transferred to atom $B$. In the retarded regime the interaction takes the form of Eq. (6). Only for large integration times, $\delta T \gg \Delta_{AB}^{-1}$, that expression reduces to a time-independent term which oscillates in space with frequency $2k_A$. The latter, however, does not agree with the result of stationary perturbation theory [13, 14].

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[18] Some of the diagrams of Fig. 1 discarded at our level of approximation provide also terms with poles in both $k$ and $k'$. However, after integrated in frequencies, they yield contributions of the order of $\Delta_{AB}/(\omega_A + \omega_B)$ times smaller than the ones found here, and hence negligible. For instance, in diagram (g) this is due to its lack of resonance with the transition of atom $B$, since both photons are simply resonant. Nonetheless, should we allowed for $\Delta_{AB} \sim \omega_{AB}$, those contributions together with the ones found here would have become the dominant ones only in the retarded regime.
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