Dipole-Dipole Coupling in the Presence of Dispersing and Absorbing Bodies

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Abstract:

An effective Hamiltonian and equations of motion for treating both the resonant dipole-dipole interaction between two-level atoms and the resonant atom-field interaction are derived, which can suitably be used for studying the influence of arbitrary dispersing and absorbing material surroundings on these interactions. It is shown that the dipole-dipole interaction and the atom-field interaction, respectively, are closely related to the real part and the imaginary part of the (classical) Green tensor. The theory is applied to the study of the transient behavior of two atoms that initially share a single excitation, with special emphasis on the role of the two competing processes of virtual and real photon exchange in the energy transfer between the atoms. To illustrate the powerfulness of the theory, specific results for the case of the atoms being near a dispersing and absorbing microsphere are presented. In particular, the regimes of weak and strong atom-field coupling are addressed.
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1 Introduction

The progress in trapping single atoms in microresonator-type equipments has offered novel possibilities of studying fundamental quantum processes on the level of a very few atoms. Let us consider two two-level atoms and assume that one of them is initially prepared in the upper state. Various processes are possible. (i) The excited atom can spontaneously emit a real photon that leaves the resonator or is reabsorbed by the atom. (ii) The emitted photon is lost because of material absorption. (iii) The excited atom undergoes radiationless decay because of material absorption. (iv) The emitted photon is absorbed by the second atom that is initially in the lower state, i.e., the excitation energy is exchanged between the two atoms. (v) The excitation energy is exchanged between the atoms via virtual photon emission and absorption, which is commonly termed resonant dipole-dipole interaction. All

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these processes sensitively depend on the electromagnetic-field structure which is controlled by the material surroundings forming the microresonator.

In the regime of weak atom-field coupling, the mutual interaction of atoms has typically been described by an effective two-body potential involving atomic variables only. By analyzing a one-dimensional cavity model, it has been shown that this concept may fail to give a correct description of the interaction at least in the strong-coupling regime, where the electromagnetic field degrees of freedom can no longer be eliminated. In order to describe both the resonant dipole-dipole interaction and the resonant atom-field interaction of two two-level atoms in a high-$Q$ cavity, an effective Hamiltonian has been proposed. Apart from the fact that the introduced coupling parameters are not specified and their relation to each other thus remains unclear (also see [3, 4]), material absorption cannot be taken into account, because of the underlying concept of mode decomposition.

2 Hamiltonian

In order to treat the problem more rigorously, let us start from the multipolar-coupling Hamiltonian for $N$ two-level atoms [positions $\mathbf{r}_A$, transition frequencies $\omega_A$, transition dipole moments $\mathbf{d}_A$ ($A = 1, 2, ..., N$)] that interact with the electromagnetic field via electric-dipole transitions in the presence of dispersing and absorbing bodies:

$$\hat{H} = \int d^3r \int_0^\infty d\omega \hbar \omega \hat{f}^\dagger(\mathbf{r}, \omega)\hat{f}(\mathbf{r}, \omega) + \sum_A \frac{\hbar}{2} \omega_A \hat{\sigma}_A^z - \sum_A \int_0^\infty d\omega \left[ \mathbf{d}_A \mathbf{E}(\mathbf{r}_A, \omega) + \text{H.c.} \right]$$

(1)

Here, $\mathbf{d}_A = \mathbf{d}_A \hat{\sigma}_A + \mathbf{d}_A^* \hat{\sigma}_A^\dagger$.

$$\mathbf{E}(\mathbf{r}, \omega) = i \sqrt{\frac{\hbar}{\pi \varepsilon_0} \frac{\omega^2}{c^2}} \int d^3r' \sqrt{\varepsilon_1(\mathbf{r}', \omega)} \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)\hat{f}(\mathbf{r}', \omega).$$

(2)

Here, $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$ is the classical Green tensor satisfying the inhomogeneous Helmholtz equation

$$\left[ \frac{\omega^2}{c^2} \varepsilon(\mathbf{r}, \omega) - \nabla \times \nabla \times \right] \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = -\delta(\mathbf{r} - \mathbf{r}')$$

(3)

with $\varepsilon(\mathbf{r}, \omega) = \varepsilon_R(\mathbf{r}, \omega) + i \varepsilon_i(\mathbf{r}, \omega)$ being the complex (Kramers-Kronig consistent) permittivity of the material surroundings. The fields $\hat{f}(\mathbf{r}, \omega)$ and $\hat{f}^\dagger(\mathbf{r}, \omega)$ are bosonic ones which play the role of the fundamental variables of the electromagnetic field and the medium, including a reservoir necessarily associated with the losses in the medium. It is not difficult to see that $\hat{f}(\mathbf{r}, \omega)$ obeys the Heisenberg equation of motion

$$\dot{\hat{f}}(\mathbf{r}, \omega) = -i\omega \hat{f}(\mathbf{r}, \omega) + \frac{\omega^2}{c^2} \sqrt{\frac{\varepsilon_1(\mathbf{r}, \omega)}{\hbar \pi \varepsilon_0}} \sum_A \mathbf{d}_A \mathbf{G}^*(\mathbf{r}_A, \mathbf{r}, \omega).$$

(4)

To treat the off-resonant atom-field interaction, we proceed as follows. (i) The $\omega$-integrals in Eq. (1) are decomposed into on-resonant parts (denoted by $\int_0^\omega d\omega ...$) and off-resonant parts (denoted by $\int_0^\omega d\omega ...$). (ii) For $\hat{f}(\omega)$ [and $\hat{f}^\dagger(\omega)$] in the off-resonant part of the third term on the right-hand side in Eq. (1), the formal solution of Eq. (1) is substituted, i.e.,

$$\hat{f}(\mathbf{r}, \omega, t) = \hat{f}_\text{free}(\mathbf{r}, \omega, t) + \frac{\omega^2}{c^2} \sqrt{\frac{\varepsilon_1(\mathbf{r}, \omega)}{\hbar \pi \varepsilon_0}} \sum_A \int_0^t dt' \hat{d}_A(t') \mathbf{G}^*(\mathbf{r}_A, \mathbf{r}, \omega)e^{-i\omega(t-t')},$$

(5)
where \( \hat{f}_{\text{free}}(r, \omega, t) \) evolves freely. (iii) In the resulting expression, slowly varying atomic operators are put in front of the \( t' \)-integrals, so that the \( t' \)-integrals can be performed to (approximately) yield \( \zeta \)-functions which are (approximately) replaced by their principal-value parts (because of the off-resonance condition). (iv) The expectation value with respect to the off-resonant medium-assisted field is taken, by assuming that it is in the vacuum state.

If the differences between the atomic transition frequencies are small compared to the frequency scale of variation of the Green tensor, then the procedure outlined changes the Hamiltonian (1) into the effective Hamiltonian

\[
\hat{H}_{\text{eff}} = \int d^3r \int_0^\infty d\omega \bar{h} \omega \hat{f}^\dagger(r, \omega) \hat{f}(r, \omega) + \sum_A \frac{\bar{h} \bar{\omega}_A \hat{\sigma}_A}{\bar{\omega}_A} - \sum_{A, A'} h_{\Delta A, A'} \hat{\sigma}_A \hat{\sigma}_{A'} - \sum_A \int_0^\infty d\omega \left[ \hat{d}_A \hat{E}(r_A, \omega) + \text{H.c.} \right],
\]

where the notation \( \sum'_{A, A'} \) indicates that \( A \neq A' \). The \( \bar{\omega}_A \) are the shifted transition frequencies, and the \( \delta_{A, A'} \) (\( A \neq A' \)) are the resonant dipole-dipole coupling strengths,

\[
\bar{\omega}_A = \omega_A - \delta_{A, A},
\]

\[
\delta_{A, A} = \delta_{A, A}^+ - \delta_{A, A}^-,
\]

\[
\delta_{A, A'} = \delta_{A, A'}^+ + \delta_{A, A'}^-,
\]

\[
\delta_{A, A'}^{(+)} = \frac{\mathcal{P}}{\pi \hbar c_0} \int_0^\infty d\omega \frac{\omega^2}{\omega - (+)\omega_{A'}} \frac{d_A \text{Im} G(r_A, r_{A'}, \omega) d_{A'}}{\omega_{A'}},
\]

[\( \mathcal{P} \) – principal value]. The notation \( A^* (A'') \) means that \( d_A (d_{A'}) \) in Eq. (11) has to be replaced with its complex conjugate \( d_A^* (d_{A'}^*) \). Recalling the Kramers-Kronig relation for the Green tensor, we may approximately rewrite Eq. (11) as

\[
\delta_{A, A'} = \frac{\bar{\omega}_{A'}^2}{\hbar c_0} d_A^* \text{Re} G(r_A, r_{A'}, \bar{\omega}_{A'}) d_{A'},
\]

which reveals that the resonant dipole-dipole interaction is closely related to the real part of the Green tensor. Accordingly, from Eqs. (10) and (8) it (approximately) follows that

\[
\delta_{A, A} = \frac{\omega_A^2}{\hbar c_0} d_A^* \text{Re} G(r_A, r_A, \bar{\omega}_A) d_A - 2\delta_{A, A}^+.
\]

Note that the Green tensor can typically written as a sum of the vacuum Green tensor \( G_V \) and a reflection part \( G_R \). Due to the singularity of \( \text{Re} G_V \) at equal space points, Eq. (12) actually applies to the reflection part only. The vacuum part can be thought of as being already included in \( \omega_A \).

3 Equations of motion

Let us assume that the atoms initially share a single excitation while the medium-assisted electromagnetic field is in the vacuum state. In the Schrödinger picture, we then may write,
on omitting off-resonant terms, the state vector of the system in the form of

$$|\psi(t)\rangle = \sum_A C_A(t) e^{-i(\tilde{\omega}_A - \tilde{\omega})t}|U_A\rangle\{0\} + \int d^3r \int_0^{\infty} d\omega C_L(r, \omega, t)e^{-i(\omega - \tilde{\omega})t}|L\rangle \hat{\Phi}(r, \omega)|\{0\}\rangle$$  \hspace{1cm} (13)

($\tilde{\omega} = \frac{1}{2} \sum_A \tilde{\omega}_A$). Here, $|U_A\rangle$ is the atomic state with the $A$th atom in the upper state and all the other atoms in the lower state, and $|L\rangle$ is the atomic state with all atoms in the lower state. Accordingly, $|\{0\}\rangle$ is the vacuum state of the rest of the system, and $\int d^3r (r, \omega)|\{0\}\rangle$ is the state, where a single quantum is excited.

Basing on the Hamiltonian [6], it is straightforward to derive the equations of motion for the slowly varying probability amplitudes $C_A(t)$ and $C_L(t)$. By formally integrating the equation for $C_L(t)$ under the initial condition that $C_L(t = 0) = 0$, and substituting the formal solution into the equation for $C_A(t)$, we obtain the following system of coupled integrodifferential equations for the $C_A(t)$:

$$\dot{C}_A(t) = \sum_{A' \neq A} i\delta_{A, A'} e^{i(\tilde{\omega}_A - \tilde{\omega}_{A'})t} C_{A'}(t) + \sum_{A'} \int_0^t dt' \int_0^{\infty} d\omega K_{A, A'}(t, t'; \omega) C_{A'}(t'),$$  \hspace{1cm} (14)

where

$$K_{AA'}(t, t'; \omega) = -\frac{1}{\hbar \epsilon_B} \left[ \frac{\omega^2 \epsilon}{c^2} e^{-i(\omega - \tilde{\omega}_A)t} e^{i(\omega - \tilde{\omega}_{A'})t'} d_A \text{Im} G(r_A, r_{A'}, \omega) d_{A'} \right].$$  \hspace{1cm} (15)

Equations (14) and (15) reveal that the resonant atom-field interaction is closely related to the imaginary part of the Green tensor. Since the real part and the imaginary part of the Green tensor are related to each other, it is clear that the resonant dipole-dipole coupling strengths and the resonant atom-field coupling strengths cannot be chosen independently from each other in general.

4 Two atoms near a microsphere

To illustrate the theory, let us consider two identical atoms ($A$ and $B$) near a dispersing and absorbing microsphere and assume that they have equivalent positions and dipole orientations with respect to the sphere such that the relations $K_{A', A} = K_{B', B}$ and $K_{A', B} = K_{B', A}$ are valid, where

$$K_{A, A'} = \frac{i\tilde{\omega}_A^2}{\hbar \epsilon_0 c^2} d_A G(r_A, r_{A'}, \tilde{\omega}_{A'}) d_{A'} = -\frac{1}{2} \Gamma_{A, A'} + i\delta_{A, A'}.$$  \hspace{1cm} (16)

Examples of $\Gamma_{AB}$ and $\delta_{AB}$ as functions of frequency in the vicinity of a surface-guided field resonance are shown in Figs. [3] and [4], where a single-resonance Drude-Lorentz-type dielectric has been assumed (for details, see [3]). It is seen that near a microsphere resonance the dipole-dipole coupling strength sensitively change with frequency. In Fig. [3], the two atoms are located at diametrically opposite positions (outside the sphere), which is an example of the interatomic distances being much larger than the wavelengths. Using Eqs. [5] and (13) and approximating the microsphere resonance line by a Lorentzian, it is not difficult to prove that the absolute value of $\delta_{AB}$ vanishes exactly on resonance and peaks at half widths of half
Figure 1: $\Gamma_{AB}$, $\delta_{AB}$ (dashed line), and $\delta_{AA}$ (dotted line), which almost coincides with $\delta_{AB}$, in units of the free-space spontaneous decay rate $\Gamma_0$, are shown for two atoms situated near a dielectric microsphere of single-resonance Drude-Lorentz-type $\omega_T$, transverse frequency; $\omega_P = 0.5 \omega_T$, plasma frequency; $\gamma = 10^{-6} \omega_T$, absorption parameter; $d = 20 \lambda_T$, sphere diameter ($\lambda_T = 2\pi c / \omega_T$); $\Delta r_A \equiv r_A - 0.5 d = \Delta r_B \geq 10^{-3} \lambda_T$, distance of the atoms from the sphere surface; $d_A = d_B$, radially oriented real transition dipole moments; (a) $\Delta r_A = 0.02 \lambda_T$; $R = 20.04 \lambda_T$, interatomic distance; (b) $\omega = 1.05048621 \omega_T$. For comparison, the inset in (b) shows the free space case.

Figure 2: Same as in Fig. 1(a), but for a short interatomic distance $R = 0.01 \lambda_T$. maximum. Fig. 1(b) reveals that for interatomic distances that are large compared to the distances that would be required in free space strong dipole-dipole coupling can be realized, even for moderately small atom-sphere distances. For the example considered, the dipole-dipole coupling strength achieved, e.g., at an interatomic distance of about 20 wavelengths can be as strong as that one in free space at a distance of about 0.012 wavelengths. The
Figure 3: $P_A(t)$ (solid line) and $P_B(t)$ (dashed line) are shown for weak resonant atom-field coupling, with the data being taken from Fig. [A] curves 1: $\omega_A = 1.050485\omega_T$, $\Gamma_{AA} = \Gamma_{BB} = 640.848\Gamma_0$, $\delta_{AB} = 0$; curves 2: $\omega_A = 1.0504867\omega_T$, $\Gamma_{AA} = \Gamma_{BB} = 8372\Gamma_0$, $\delta_{AB} = 0$.

relation between the strengths of dipole-dipole and atom-field coupling sensitively depends on the relative positions and dipole orientations of the atoms to each other and to the microsphere. By moving the atoms close to each other, the contribution of the vacuum part to the Green tensor increases, which allows one to vary $\delta_{AB}$ while keeping $\Gamma_{AB}$ almost unchanged (see Fig. 2). Note that for other resonance lines, $\Gamma_{AB}$ and $\delta_{AB}$ can switch signs [7]. Clearly, the aforegiven discussion also holds for whispering gallery resonances.

Introducing the probability amplitudes

$$C_\pm(t) = 2^{-\frac{1}{2}} [C_A(t) \pm C_B(t)] e^{\mp i\delta_{AB} t}$$

of the superposition states $|\pm\rangle = 2^{-1/2} (|U_A\rangle \pm |U_B\rangle)$, from Eqs. [14] we find that the equations for $C_+(t)$ and $C_-(t)$ decouple,

$$\dot{C}_\pm(t) = \int_0^t dt' \int_0^\infty d\omega K_\pm(t, t'; \omega) e^{\mp i\delta_{AB}(t-t')} C_\pm(t'),$$

$$K_\pm(t, t'; \omega) = K_{A\pm A}(t, t'; \omega) \pm K_{A\pm B}(t, t'; \omega).$$

The integrodifferential equation (18) should be solved numerically in general. Closed solutions can be found in the limiting cases of weak and strong atom-field interaction.

In the weak-coupling regime the Markov approximation applies, thus

$$C_\pm(t) = 2^{-1/2} e^{-\Gamma_\pm t/2}, \quad \Gamma_\pm = \Gamma_{A\pm A} \pm \Gamma_{A\pm B}.\quad (20)$$

The upper-state occupation probabilities $P_{A(B)}(t) = |C_{A(B)}(t)|^2$ then read

$$P_{A(B)}(t) = \frac{1}{2} [\cosh (\Gamma_{A\pm B} t) + (-) \cos (2\delta_{A\pm B} t)] e^{-\Gamma_{B\pm B} t}.\quad (21)$$

From Eq. (21) it is seen that when $|\delta_{A\pm B}| \gg \Gamma_{B\pm B}$, then the excitation energy is exchanged back and forth between atoms $A$ and $B$ (Fig. 3, curves 1). In the opposite case
Figure 4: $P_A(t)$ (solid line) and $P_B(t)$ (dashed line) are shown for strong resonant atom-field coupling, with the data being taken from Figs. (a) and (b) $\Gamma_0 = 10^{-6} \omega_T$, $\Omega_+ \approx 128 \Gamma_0$, $\Delta \omega_m \approx 5 \times 10^{-7} \omega_T$; (i) $\tilde{\omega}_A = 1.04853747 \omega_T$, $R \approx 0.01 \lambda_T$, $\delta_{AB} \approx -2129 \Gamma_0$; (ii) $\tilde{\omega}_A = 1.05045444 \omega_T$, $R \approx 0.027 \lambda_T$, $\delta_{AB} \approx -32.2 \Gamma_0$; (iii) $\tilde{\omega}_A = \omega_m = 1.0504867 \omega_T$, $R = 20.04 \lambda_T$, $\delta_{AB} \approx 0$.

of $|\delta_{A\cdot B}| \ll \Gamma_{B\cdot B}$, $P_A$ and $P_B$ quickly approach equal values and then decay very slowly (curves 2). During this slow decay process, the two atoms are partially entangled [7]. Both types of temporal evolution can also appear in free space, but only for interatomic distances that are much smaller than the wavelengths.

In the strong-coupling limit, we restrict our attention to the case when the absolute value of the two-atom term $K_{A\cdot B}(t,t';\omega)$ is of the same order of magnitude as the absolute value of the single-atom term $K_{A\cdot A}(t,t';\omega)$, so that there is a strong contrast in the magnitude of $K_{+}(t,t';\omega)$ and $K_{-}(t,t';\omega)$. As a consequence, the strong-coupling regime can be realized for either the state $|+\rangle$ or the state $|-\rangle$, but not for both at the same time. Assuming that the field resonance strongly coupled to the atoms has a Lorentzian shape, with $\omega_m$ and $\Delta \omega_m$ being the central frequency and the half width at half maximum respectively, we can perform the frequency integral in Eq. (18) in a closed form, on extending it to $\pm \infty$. The further calculation can then be performed as described in [8]. In particular for exact resonance, i.e., $\omega_m = \tilde{\omega}_A \mp \delta_{A\cdot B}$, we derive

$$C_{\pm}(t) = 2^{-\frac{1}{2}} e^{-\Delta \omega_m t/2} \cos(\Omega_{\pm} t/2), \quad \Omega_{\pm} = \sqrt{2 \Gamma_{\pm} \Delta \omega_m}.$$  \hspace{1cm} (22)
For the probability amplitudes of the remaining states $|∓\rangle$, which are weakly coupled to the field, we have $C_∓(t) = 2^{-1/2}e^{-\Gamma_{∓}t/2}$. It then follows that

$$P_{A(B)}(t) = \frac{1}{4} \left[ e^{-\Gamma_{∓}t} + e^{-\Delta\omega_m t} \cos^2(\Omega_∓t/2) + (-) 2e^{-(\Delta\omega_m + \Gamma_{∓})t/2} \cos(\Omega_±t/2) \cos(2\delta_{A,B}t) \right].$$

(23)

The upper (lower) signs refer to the case where the state $|+\rangle (|−\rangle)$ is strongly coupled to the medium-assisted field. Typical examples are shown in Fig. 3. When the atoms are sufficiently close to each other $(4|\delta_{A,B}| ≫ \Omega_±)$, then a beating-type behavior of $P_A(t)$ and $P_B(t)$ as shown in Figs. 3(iA) and (iB), respectively, may be observed. The beat is between the oscillation of the frequency $2\delta_{A,B}$, which arises from the two-atom dipole-dipole coupling, and the Rabi oscillation of frequency $\Omega_±$. Recall that the collapses and revivals of the atomic level populations in the Jaynes-Cummings model are caused by the presence of oscillations with noncommensurate single-atom Rabi frequencies. Partial trapping of the excitation energy in atom $A$ as shown in Fig. 3(ii) may be observed if the distance between the atoms is slightly increased $(4|\delta_{A,B}| \simeq \Omega_±)$. This trapping can be understood as resulting from a destructive interference between the two channels of energy transfer, one via virtual and the other via real medium-assisted field excitation. Finally, Fig. 3(iii) shows that for larger distances the motion becomes governed by the Rabi oscillations $(4|\delta_{A,B}| \simeq 0 ≪ \Omega_±)$.

5 outlook

The present analysis has left a number of open questions, on which future work will concentrate. Depending upon the strengths of the resonant dipole-dipole coupling and the resonant atom-field coupling and their relations to each other, multiplet spectra of the emitted light can be expected. Further, the problem of mutual interaction of atoms whose transition frequencies must be regarded as being different with regard to the variation of the medium-assisted Green tensor needs special emphasis, because in such a case the effective Hamiltonian $H$ does not apply.

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References

[1] E. V. Goldstein and P. Meystre, Phys. Rev. A 56 (1997) 5135.
[2] G. Kurizki, A. G. Kofman, and V. Yudson, Phys. Rev. A 53 (1996) R35.
[3] G. Kurizki and A. Z. Genack, Phys. Rev. Lett. 61 (1988) 2269.
[4] Q. Zheng, T. Kobayashi, and T. Sekiguchi, Phys. Rev. Lett. 77 (1996) 406; G. Kurizki, A. G. Kofman, and A. Z. Genack, *ibid.* 77 (1996) 407.
[5] L. Knöll, S. Scheel, and D.-G. Welsch, in *Coherence and Statistics of Photons and Atoms*, edited by J. Peřina (John Wiley & Son, New York, 2001), p. 1.
[6] Ho Trung Dung, L. Knöll, and D.-G. Welsch, Phys. Rev. A 65 (2002) 043813.
[7] Ho Trung Dung, L. Knöll, and D.-G. Welsch, Phys. Rev. A 64 (2001) 0013804; Ho Trung Dung, S. Scheel, L. Knöll, and D.-G. Welsch, J. Opt. B: Quant. Semiclass. Opt. 4 (2002) S169.