PHYSICAL REVIEW A VOLUME 45, NUMBER 9 1 MAY 1992

Two-photon resonance induced by the dipole-dipole interaction

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(Received 5 August 1991)

The two-photon absorption by a pair of two-level atoms interacting with an incident field is examined. If the laser field is weak, then the existence of the dipole-dipole interaction-induced two-photon resonance follows from the higher-order Fermi golden rule. The effects of spontaneous emission and intense fields on the dipole-dipole interaction-induced two-photon resonance is studied using density-matrix methods. The presence of an intense field is shown to result in a significant enhancement of the two-photon resonance. Numerical results for the probability of simultaneous excitation of two atoms, for the atomic correlations, and for the radiated field intensity are presented.

PACS number(s): 42.50.Fx, 42.50.Hz

I. INTRODUCTION

The interaction of a laser field with an atomic system consisting of a pair of atoms has been a subject of continuous interest mainly because it is a prototype model for studying the dynamics of dense atomic systems. Recently, however, there has been a renewal of interest in this system due to the experimental progress made in confining a few atoms or ions at small interatomic separations in ion and neutral-atom traps [1]. This setup facilitates the isolation of the single-atom effects from those arising due to the correlations between the atoms. In view of the above it is useful to consider the modifications in the dynamics of the two-atom system arising due to cooperative decay as well as the dipole-dipole interaction. Pairs of two-level atoms have been extensively studied in the past [2–17]. Correlations between a pair of *identical atoms* arising from the dipole-dipole (*d-d*) interaction have been known to give rise to new resonances [6–10] in the fluorescence spectrum. A correct interpretation for the origin [8] of these resonances is possible by considering the dressed levels of the system consisting of two atoms interacting with a laser field. The role of the *d-d* interaction has also been examined in the quantum beats produced in the intensity [11] and the intensity correlations [12,13] of the spontaneous emission from a pair of atoms. Recently the squeezing properties of the emitted fluorescence [14] have also been shown to yield information about the dynamical correlations between atoms. Note that the system of two identical two-level atoms is equivalent to a four-level atomic system. The four levels can be described in terms of the eigenstates associated with the collective spin operators $\mathbf{S}=\mathbf{S}_1+\mathbf{S}_2$. One has three symmetric states corresponding to $S=1$ and one antisymmetric state corresponding to $S=0$. At small but finite distances, the metastability of the antisymmetric state gives rise to the phenomenon of macroscopic quantum jumps whenever the system makes a transition from the symmetric state to the antisymmetric state via the cooperative decay process. At precisely the interatomic distances where the antisymmetric state is metastable, the *d-d* interaction also becomes important since it shifts the states away from resonance with the laser frequency and thus affects the jump rates [15] and the photon statistics [16] of the fluorescence produced by the system. The role of the *d-d* interaction has also been studied in the context of quantum jumps from a pair of three-level atoms [17].

Experimental investigations in the optical emission [18–20] and absorption [21] process in dense media have reported the observation of the *d-d* interaction-induced novel resonances at the sum frequency of the atomic transitions. Cooperative single-photon absorption by a pair of coupled atoms was first demonstrated by Varsanyi and Dieke [22]. They observed that weak absorption occurs at the sum of the transition frequencies of the coupled atoms. This effect was qualitatively explained by Dexter [23] using first-order perturbation theory. He showed that *d-d* interaction between the atoms leads to the simultaneous excitation of atoms, which can be utilized for pumping lasers [24]. A strong-field theory for calculating the pair-emission and absorption transition probability for weakly coupled atoms has been developed [25]. Simultaneous excitation of a pair of nonidentical atoms of frequencies ω_1 and ω_2 interacting with a laser field of frequency ω_l by a two-photon absorption process exhibits [26] a new *d-d* interaction-induced resonance at $2\omega_l=\omega_1+\omega_2$ which is in addition to the usual peaks at $\omega_l=\omega_1$ and ω_2 for noninteracting atoms. For a pair of independent atoms the transition probability for simultaneous excitation is just the product of the transition probabilities for finding each atom in the excited state. Therefore, as the laser frequency is scanned, the peaks occur whenever the laser is on resonance with the atomic frequencies. Simultaneous excitation by absorption of two photons occurs because one atom is resonantly excited by one photon and the other atom is off resonantly excited by the other photon. The emergence of an additional *d-d* interaction-induced two-photon resonance is a clear indication of a totally different channel of excitation where each of the atoms is off resonantly excited by absorption of a photon, but an energy compensation occurs to overcome this detuning. In the presence of the *d-d* interaction the atom for which the laser is tuned above its transition frequency can transfer its excess energy to the second atom for which the laser is tuned below its transition frequency. This process thus shows the two-photon resonance when the energy transfer exactly compensates for the detuning, i.e., at $\omega_1-\omega_l=-(\omega_2-\omega_l)$. Nayfeh and Hillard [27] have shown that when the atoms are not restricted to being two-level systems, the line shapes and the resonance positions for the two-photon-absorption process can change depending on whether only one atom is radiatively active or both the atoms are. It is thus interesting to study how this two-photon resonance gets modified when the strength of the *d-d* interaction increases (i.e., as the atoms come closer to each other) in the presence of an intense exciting field. It is also necessary to incorporate the effects of spontaneous emission from the atoms. Thus the purpose of this paper is to extend the previous studies on the two-photon resonance to get exact results for arbitrary strengths of the atom-atom and atom-field interaction including the spontaneous-decay effects. This has been achieved using the master-equation techniques [28]. For simplicity we consider only two two-level atoms. The density-matrix equation is solved numerically. We show that the two-photon resonance becomes more and more dominant as the atoms come closer to each other. In addition, the presence of a saturating field can enhance this resonance considerably. The origin of this enhancement is attributed to the increasing correlations between the atoms. In addition it is also found that the radiated power reflects the behavior of the transition probabilities.

In Sec. II we give the Hamiltonian for the system of two two-level atoms interacting with each other via the dipole-dipole interaction and interacting with an incident laser field. In Sec. III we give the perturbative result for the probability for two photon absorption using the Fermi golden rule. We study the effect of a saturating field on the two photon resonance using the master equation approach in Sec. IV. We give the numerical results for the transition probability for the simultaneous excitation of atoms, atomic correlations and the far-zone radiated-field intensity.

II. THE HAMILTONIAN FOR THE MODEL

Consider a system of two two-level atoms of frequencies ω_1 and ω_2, which interact with a single laser field of frequency ω_l. The incident monochromatic field is written as

$$\mathbf{E}(\mathbf{r},t)=\boldsymbol{\epsilon}(\mathbf{r})e^{-i\omega_l t}+\text{c.c.}\ , \tag{2.1}$$

where $\boldsymbol{\epsilon}(\mathbf{r})$ is the amplitude and ω_l is the frequency of the field. Figure 1 shows the energy levels of the free atoms. The levels $|j\rangle$ and $|i\rangle$ represent, respectively, the ground state and the excited state of the atom of frequency ω_1. Similarly, the states $|\beta\rangle$ and $|\alpha\rangle$ are, respectively, the ground state and excited state of the second atom of frequency ω_2. The two two-level-atom system is equivalent to a four-level atomic system obtained as a direct product of the one-atom states. These states will be represented as $|k,\eta\rangle$ with $k=i$ or j and $\eta=\alpha$ or β having energy $(\omega_k+\omega_\eta)$ $(\hbar=1)$. We denote by $|j\beta\rangle$ the state when both the atoms are in the ground state. The state when both the atoms are excited is denoted by $|i\alpha\rangle$. The states corresponding to the case when either one of the atoms is excited is denoted as $|i\beta\rangle$ and $|j\alpha\rangle$. The dipole moment of each atom is given by

$$\mathbf{d}_i=\mathbf{d}_i(S_i^+ + S_i^-),\quad i=1,2. \tag{2.2}$$

Here $S_i^\pm$, S_i^z are the spin-$\frac{1}{2}$ operators for each atom. In what follows we work in a frame rotating with the frequency of the external field. The Hamiltonian for the system of two two-level atoms interacting with a laser of frequency ω_l is given by

$$H=H_0+H_{dd}+H_{\text{ext}}\ , \tag{2.3}$$

where

$$H_0=\sum_{i=1}^{2}\Delta_i S_i^z,\quad \Delta_i=\omega_i-\omega_l,\quad i=1,2 \tag{2.4}$$

is the unperturbed Hamiltonian of the two atoms and Δ_i

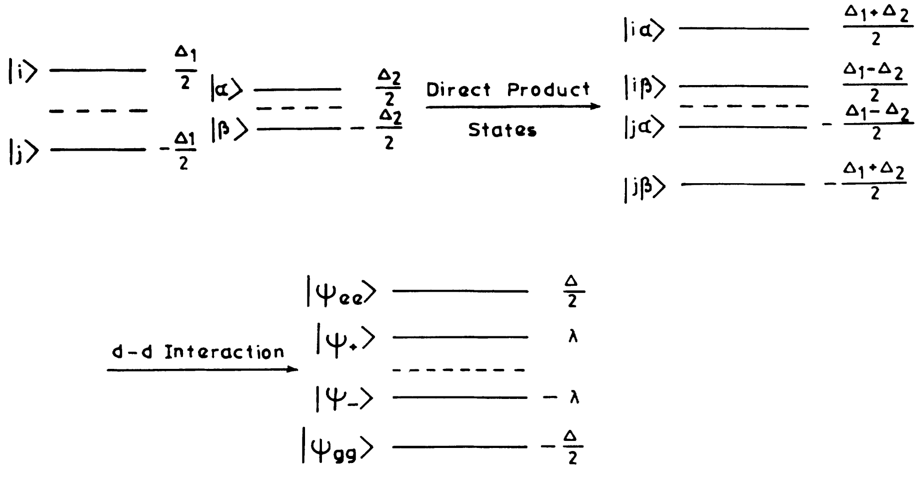

FIG. 1. Schematic diagram of the energy levels of the two two-level atoms of frequencies ω_1 and ω_2 interacting with the incident laser field of frequency ω_l. These are shown in the frame rotating with the frequency ω_l of the field. The diagram also shows the energies of the direct-product states and the states obtained after the dipole-dipole interaction has been diagonalized.

gives the detuning of the field. The second term in Eq. (2.3) describes the dipole-dipole interaction between the atoms and can be approximated by

$$H_{dd}=\sum_{\substack{i,j\\ i\neq j}}\left[VS_i^+S_j^-+\text{H.c.}\right],\quad i,j=1,2. \tag{2.5}$$

In writing Eq. (2.5) non-energy-conserving terms like $S_1^+S_2^+$ and $S_1^-S_2^-$ have been dropped. The dipole-dipole interaction cannot simultaneously excite or deexcite both the atoms. The static form of the *d*-*d* interaction is given by

$$V=\frac{1}{|\mathbf{r}_{12}|^3}\left[\mathbf{d}_1\cdot\mathbf{d}_2-3\left[\mathbf{d}_1\cdot\frac{\mathbf{r}_{12}}{|\mathbf{r}_{12}|}\right]\left[\mathbf{d}_2\cdot\frac{\mathbf{r}_{12}}{|\mathbf{r}_{12}|}\right]\right], \tag{2.6}$$

where $\mathbf{r}_{12}$ is the separation between the two atoms. The last term in Eq. (2.3) describes the interaction of each atom with the laser field, which in rotating-wave approximation is given by

$$H_{\text{ext}}=\sum_{i=1}^{2}\left[g_iS_i^++\text{H.c.}\right],\quad g_i=-\mathbf{d}_i\cdot\epsilon_i\ , \tag{2.7}$$

where g_i is the coupling constant of each atom to the field and ϵ_i is the field at the position of the ith atom. The unperturbed energy levels for the four atomic states obtained as a direct product of the single-atom states are shown in Fig. 1. Various detunings shown in the figure are defined as

$$\begin{aligned}&\Delta_1=\omega_1-\omega_l,\quad \Delta_2=\omega_2-\omega_l\ ,\\&\Delta=\Delta_1+\Delta_2=\omega_1+\omega_2-2\omega_l\ ,\\&\delta=\Delta_1-\Delta_2=\omega_1-\omega_2\ .\end{aligned} \tag{2.8}$$

In the basis of the four direct-product states $|i,\alpha\rangle,|i,\beta\rangle,|j,\alpha\rangle,|j,\beta\rangle$, the matrix representation of the Hamiltonian is

$$H=\begin{bmatrix}\frac{\Delta}{2} & 0 & g_2 & g_1\\ 0 & -\frac{\Delta}{2} & g_1^* & g_2^*\\ g_2^* & g_1 & \frac{\delta}{2} & V\\ g_1^* & g_2 & V^* & -\frac{\delta}{2}\end{bmatrix}. \tag{2.9}$$

We first diagonalize Eq. (2.9) in the absence of the laser field. The new eigenstates and the corresponding eigenvalues are given by

$$\begin{aligned}&|\psi_{ee}\rangle=|i\alpha\rangle\ ,\quad \Delta/2\\&|\psi_+\rangle=S_{11}^{-1}|i\beta\rangle+S_{12}^{-1}|j\alpha\rangle\ ,\quad \lambda\\&|\psi_-\rangle=S_{21}^{-1}|i\beta\rangle+S_{22}^{-1}|j\alpha\rangle\ ,\quad -\lambda\\&|\psi_{gg}\rangle=|j\beta\rangle\ ,\quad -\Delta/2\end{aligned} \tag{2.10}$$

where

$$\lambda=\left[\frac{\delta^2}{4}+|V|^2\right]^{1/2}, \tag{2.11}$$

and the diagonalization matrix S is given by

$$S=\begin{bmatrix}\dfrac{1}{\left[1+\dfrac{\left[\frac{\delta}{2}-\lambda\right]^2}{V^2}\right]^{1/2}} & \dfrac{-1}{\left[1+\dfrac{\left[\frac{\delta}{2}+\lambda\right]^2}{V^2}\right]^{1/2}}\\[3ex] -\dfrac{\left[\frac{\delta}{2}-\lambda\right]/V}{\left[1+\dfrac{\left[\frac{\delta}{2}-\lambda\right]^2}{V^2}\right]^{1/2}} & \dfrac{\left[\frac{\delta}{2}+\lambda\right]/V}{\left[1+\dfrac{\left[\frac{\delta}{2}+\lambda\right]^2}{V^2}\right]^{1/2}}\end{bmatrix}. \tag{2.12}$$

It is evident from Eq. (2.10) that dipole-dipole interaction mixes up the intermediate levels $|i\beta\rangle$ and $|j\alpha\rangle$ only. The new energy levels lead to a resonant cooperative absorption of light which results in a two-photon resonance, which cannot occur for free atoms. This is because in the absence of dipole-dipole interaction the destructive interference between the two channels of two photon absorption $|j\beta\rangle\rightarrow|j\alpha\rangle\rightarrow|i\alpha\rangle$ and $|j\beta\rangle\rightarrow|i\beta\rangle\rightarrow|i\alpha\rangle$ is complete.

III. THE DIPOLE-DIPOLE INTERACTION-INDUCED TWO-PHOTON RESONANCE—A PERTURBATIVE ANALYSIS

We now calculate the transition probability for simultaneous excitation of atoms by a two-photon absorption process when both the atoms are initially in the ground state. We work in the basis [Eq. (2.10)] that diagonalizes the unperturbed Hamiltonian and the dipole-dipole interaction. For weak fields, the probability for the two-photon absorption process can be calculated using second-order time-dependent perturbation theory. The Fermi golden rule for the rate R_{12} for transition from $|\psi_{gg}\rangle\rightarrow|\psi_{ee}\rangle$ gives

$$R_{12}=2\pi\left|\frac{\langle i\alpha|H_{\text{ext}}|\psi_+\rangle\langle\psi_+|H_{\text{ext}}|j\beta\rangle}{i\left[\frac{\Delta}{2}+\lambda\right]}+\frac{\langle i\alpha|H_{\text{ext}}|\psi_-\rangle\langle\psi_-|H_{\text{ext}}|j\beta\rangle}{i\left[\frac{\Delta}{2}-\lambda\right]}\right|^2\delta(\omega_1+\omega_2-2\omega_l)\ , \tag{3.1}$$

where the intermediate states $|\psi_+\rangle$ and $|\psi_-\rangle$ are the new states given in Eq. (2.10), and H_{ext} is the atom-field interaction defined by Eq. (2.7). The paths $|j\beta\rangle \to |\psi_+\rangle \to |i\alpha\rangle$ and $|j\beta\rangle \to |\psi_-\rangle \to |i\alpha\rangle$ are the two excitation channels. Simultaneous excitation of atoms occurs along either of these two channels. As long as the matrix elements in Eq. (3.1) are nonzero, R_{12} shows a two-photon resonance at $2\omega_l=\omega_1+\omega_2$. In terms of the exact eigenstates given by Eq. (2.10), R_{12} is found to be

$$R_{12}=\frac{2\pi g^4}{16\lambda^4}\left|\frac{\left[1+\left(\frac{\delta}{2}-\lambda\right)^2/V^2\right]\left(V+\frac{\delta}{2}+\lambda\right)^2}{\left(\frac{\Delta}{2}+\lambda\right)}+\frac{\left[1+\left(\frac{\delta}{2}+\lambda\right)^2/V^2\right]\left(V+\frac{\delta}{2}-\lambda\right)^2}{\left(\frac{\Delta}{2}-\lambda\right)}\right|^2 \times\delta(\omega_1+\omega_2-2\omega_l)\ , \tag{3.2}$$

where for simplicity we have set $g_1=g_2=g$.

For the case when the dipole-dipole coupling is also weak, the corrections to the states can be obtained by using first-order perturbation theory, which implies that there are no corrections to the energy eigenvalues. Then

$$|\psi_+\rangle=|i\beta\rangle-\frac{|j\alpha\rangle\langle j\alpha|H_{dd}|i\beta\rangle}{E_{j\alpha}-E_{i\beta}}\ ,$$
$$|\psi_-\rangle=|j\alpha\rangle-\frac{|i\beta\rangle\langle i\beta|H_{dd}|j\alpha\rangle}{E_{i\beta}-E_{j\alpha}}\ , \tag{3.3}$$

with $E_{j\alpha}=E_j+E_\alpha$, $E_{i\beta}=E_i+E_\beta$. Putting in the exact form of H_{dd} from Eq. (2.5)

$$|\psi_+\rangle=|i\beta\rangle+\frac{V}{\delta}|j\alpha\rangle\ ,$$
$$|\psi_-\rangle=|j\alpha\rangle-\frac{V}{\delta}|i\beta\rangle\ , \tag{3.4}$$

and

$$R_{12}=2\pi\left|\frac{g_1g_2(\omega_1+\omega_2-2\omega_l)}{(\omega_1-\omega_l)(\omega_2-\omega_l)}-\frac{V(g_1^2+g_2^2)}{(\omega_1-\omega_l)(\omega_2-\omega_l)}\right|^2 \times\delta(\omega_1+\omega_2-2\omega_l)\ . \tag{3.5}$$

Note that Eq. (3.5) is a sixth-order perturbative result for R_{12}. It is obtained to second order in dipole-dipole coupling and to fourth order in external field amplitudes. It is clear that in the absence of dipole-dipole coupling ($V=0$), $R_{12}=0$, since $(\omega_1+\omega_2-2\omega_l)\delta(\omega_1+\omega_2-2\omega_l)=0$. The process in which two free nonidentical atoms are simultaneously excited by the absorption of two photons of frequency ω_l, such that $2\omega_l=\omega_1+\omega_2$ is not allowed because of exact destructive interference between the two excitation channels $|j\beta\rangle \to |j\alpha\rangle \to |i\alpha\rangle$ and $|j\beta\rangle \to |i\beta\rangle \to |i\alpha\rangle$. If the interaction with the complete radiation field (including the vacuum degrees of freedom) is included, however, the resulting shifts, widths, and mixing of the single-excitation states $|i\beta\rangle$ and $|j\alpha\rangle$ allows the transition to occur. In the presence of the coupling, R_{12} shows a resonance at $\omega_1+\omega_2=2\omega_l$. Thus this two-photon resonance is a dipole-dipole interaction-induced extra resonance. We also note that it would not be possible to take the limit of Eq. (3.5) for the case of identical atoms as the two-photon resonance condition would reduce to the single-photon resonance condition $\omega_1=\omega_l$, whereas the validity of the second-order perturbation theory demands that the intermediate resonances should be avoided. Thus one has to use Eq. (3.2) which holds to all orders in *d-d* interaction. In fact Eq. (3.2) for identical atoms reduces to

$$R_{12}=\frac{8\pi g^4}{V^2}\delta(\Delta)\ . \tag{3.6}$$

Note, however, that Eq. (3.6) ignores the width of the intermediate states $\psi_\pm$ which should, in principle, be included. Thus Eq. (3.6) holds in the limit that the strength of the dipole-dipole interaction is much bigger than the width of the intermediate states $\psi_\pm$. Note further that the widths of the intermediate states $\psi_\pm$ can be obtained from equations in the Appendix.

IV. NONPERTURBATIVE ANALYSIS OF THE EXTRA TWO-PHOTON RESONANCE—EFFECTS OF INTENSE FIELDS

The perturbative analysis [Eq. (3.5)] shows the existence of a new resonance at $2\omega_l=\omega_1+\omega_2$ for coupled atoms, the strength of which depends on the dipole-dipole interaction V. The width of the resonance can be obtained from considerations involving the spontaneous emission of photons. For treating spontaneous emission we need to consider the interaction of the atoms with a large number of vacuum modes of the radiation field. We work in the transverse gauge. It is also desirable to examine the effects of an intense electromagnetic field on the two-photon resonance. One thus needs a formulation which can account for both spontaneous-emission effects as well as intense-field effects. This can be done within the framework of master equations [28]. This has the advantage that even the dipole-dipole interaction can be handled to all orders. The master equation for the evolution of the atomic density-matrix operator obtained by tracing over the radiation field variables is given by [28]

$$\frac{\partial\rho}{\partial t}=-i[H,\rho]-\sum_{i,j=1}^{2}\gamma_{ij}(S_i^+S_j^-\rho-2S_j^-\rho S_i^++\rho S_i^+S_j^-)\ , \tag{4.1}$$

where H is the total Hamiltonian defined by Eq. (2.3) with the static dipole-dipole interaction now replaced by the retarded dipole-dipole interaction

$$\Omega_{12}=\tfrac{3}{2}\left[[\mathbf{d}_1\cdot\mathbf{d}_2-3(\mathbf{d}_1\cdot\mathbf{r}_{12})(\mathbf{d}_2\cdot\mathbf{r}_{12})]\left[\frac{\sin x}{x^2}+\frac{\cos x}{x^3}\right]-[\mathbf{d}_1\cdot\mathbf{d}_2-(\mathbf{d}_1\cdot\mathbf{r}_{12})(\mathbf{d}_2\cdot\mathbf{r}_{12})]\frac{\cos x}{x}\right],\quad x=k_0r_{12},\quad k_0\approx\frac{\omega_1}{c}\approx\frac{\omega_2}{c}\ . \tag{4.2}$$

In Eq. (4.1) γ_{ij} is the collective decay constant of the atomic system given by

$$\gamma_{ij}=\tfrac{3}{2}\gamma\left[[(\hat{\mathbf{d}}_1\cdot\hat{\mathbf{d}}_2)-(\hat{\mathbf{d}}_1\cdot\mathbf{r}_{12})(\hat{\mathbf{d}}_2\cdot\mathbf{r}_{12})]\frac{\sin x}{x}+[(\hat{\mathbf{d}}_1\cdot\hat{\mathbf{d}}_2)-3(\hat{\mathbf{d}}_1\cdot\mathbf{r}_{12})(\hat{\mathbf{d}}_2\cdot\mathbf{r}_{12})]\left[\frac{\cos x}{x^2}-\frac{\sin x}{x^3}\right]\right],\quad x=k_0r_{12} \tag{4.3}$$

where $\hat{\mathbf{d}}_i$, $i=1,2$ are unit vectors along the electric dipole moments. In the limit when $\mathbf{r}_{12}\to 0$ we can write

$$\gamma_{12}\approx\gamma,\quad \Omega_{12}\approx V\ , \tag{4.4}$$

where V is given by Eq. (2.6). We will work at distances such that Eq. (4.4) is indeed a good approximation. Note that the master equation (4.1) treats the external field and the dipole-dipole interaction to all orders and thus multiphoton processes induced by the external field are automatically included. Only the spontaneous-emission events are considered to be single-photon processes. Note further that if the retardation effects are completely ignored, i.e., if one formally sets the velocity of light equal to infinity, then $\Omega_{ij}\to V$ and $\gamma_{ij}\to 0$.

The two two-level-atom system is equivalent to a four-level atomic system. On using the normalization condition $\mathrm{Tr}\rho=1$, these reduce to the set of 15 coupled equations given in the Appendix. In the matrix notation these equations are given as

$$\frac{d\psi}{dt}=M\psi+I\ , \tag{4.5}$$

where ψ is the 15×1 column matrix of the mean atomic operators defined in Eq. (A1), and M is the 15×15 matrix of the coefficients which can be easily constructed from Eq. (A1). The only nonzero matrix elements of 15×1 column matrix I are

$$I_3=-\gamma_1,\quad I_6=-\gamma_2\ . \tag{4.6}$$

Equation (4.5) allows us to easily study the long-time behavior of the atomic system. Of particular interest is the behavior of the transition probability for simultaneous excitation of atoms, the characteristics of the radiated power, and the correlations between the atoms.

We first examine the transition probability for simultaneous excitation of atoms by a two-photon absorption process. This is given by

$$\begin{aligned}P_{12}=\rho_{i\alpha,i\alpha}&=\langle S_1^+S_1^-S_2^+S_2^-\rangle\\&=\langle(\tfrac{1}{2}+S_1^z)(\tfrac{1}{2}+S_2^z)\rangle\\&=\tfrac{1}{4}+\tfrac{1}{2}\langle S_1^z\rangle+\tfrac{1}{2}\langle S_2^z\rangle+\langle S_1^zS_2^z\rangle\ .\end{aligned} \tag{4.7}$$

The first three terms in Eq. (4.7) are the contributions to P_{12} arising from each atom independently. If one ignores the dipole-dipole interaction and if one ignores the cooperative decay, i.e., if one sets $\gamma_{12}=0$ in Eq. (4.1), then the atoms are not correlated and P_{12} is the product of the probabilities of finding each atom in the excited state. This is given by

$$P_{12}=\langle S_1^+S_1^-\rangle\langle S_2^+S_2^-\rangle=\frac{|g_1|^2}{\gamma_1^2+\Delta_1^2}\frac{|g_2|^2}{\gamma_2^2+\Delta_2^2}\ . \tag{4.8}$$

Each term of the product in the above equation is the transition probability for the excitation of a single atom by a one-photon absorption process. P_{12} thus shows resonance at $\Delta_1=0$ and $\Delta_2=0$, i.e., at $\omega_l=\omega_1$ and $\omega_l=\omega_2$. The last term in Eq. (4.7) gives the cooperative contribution, which is nonzero only in the presence of the *d-d* interaction. This term is responsible for the origin of the two-photon resonance at $\omega_1+\omega_2=2\omega_l$. A perturbative solution for the steady-state transition probability P_{12} for simultaneous excitation of the two atoms can be obtained using Eq. (4.5). The perturbative solution to first order in the dipole-dipole coupling and fourth order in the field coupling, in the limit when the detunings Δ_1 and Δ_2 are large, is given by

$$P_{12}\approx\frac{|g|^4}{\Delta_1^2\Delta_2^2}-\frac{4|g|^4\Delta V}{\Delta_1^2\Delta_2^2(4\gamma^2+\Delta^2)}>0\ , \tag{4.9}$$

where we have assumed that $g_1=g_2=g$ and $\gamma_1=\gamma_2=\gamma$. In this expression we have retained only those terms which contribute to the two-photon resonance. Note that since Eq. (4.9) is a perturbative result, the second term is smaller than the first term, so that P_{12} remains positive. This shows the presence of the dipole-dipole coupling-induced resonance at $\Delta=\Delta_1+\Delta_2=0$. It is evident that the extra resonance vanishes in the absence of the dipole-dipole interaction ($V=0$). Equation (4.9) predicts a dispersionlike line shape proportional to the dipole-dipole interaction strength V. The situation would be different if the V^2 term dominates (cf. Fig. 2). Note that the sixth-order perturbative result given in Eq. (3.5) gives the rate for exciting both atoms, whereas the perturbative result given in Eq. (4.9) gives the actual steady-state probability for exciting both atoms by a two-photon absorption process.

We will next show how the two results, Eqs. (3.5) and (4.9), can be connected. For this purpose we need to introduce the width of the state $|i\alpha\rangle$. Equations (4.7) and (A1) show that the state $|i\alpha\rangle$ *decays at the rate* 4γ. The rate R_{12} [Eq. (3.5)] does not account for the width of the final state. The width of the intermediate state is not important as ω_l is greatly detuned. The result Eq. (3.5) upon accounting for the width (4γ) of the final state, is modified to $[\delta(x)=(2\gamma/\pi)/(4\gamma^2+x^2)]$

$$R_{12}=\left|\frac{g^2\Delta}{\Delta_1\Delta_2}-\frac{2Vg^2}{\Delta_1\Delta_2}\right|^2\frac{4\gamma}{4\gamma^2+\Delta^2}\ , \tag{4.10}$$

where we have set $g_1=g_2=g$. The pumping of the state $|i\alpha\rangle$ at the rate R_{12} and its decay at the rate 4γ would result in the steady-state population of the state $|i\alpha\rangle$ being given by

$$\rho_{i\alpha,i\alpha}=\frac{R_{12}}{4\gamma}\ , \tag{4.11}$$

which on combining with Eq. (4.10) reduces to

$$\rho_{i\alpha,i\alpha}=\left[\frac{g^2\Delta}{\Delta_1\Delta_2}-\frac{2Vg^2}{\Delta_1\Delta_2}\right]^2\frac{1}{4\gamma^2+\Delta^2}\ . \tag{4.12}$$

Note that here the term to lowest order in V is the same as Eq. (4.9), assuming that the terms of order V^2 are still smaller. Note further that the V-independent term in Eq. (4.12) is the same as the corresponding term in Eq. (4.9). To see this explicitly we recall that the denominators $\omega_1-\omega_l$ and $\omega_2-\omega_l$ in Eq. (3.5) should in principle have the damping of the corresponding state, i.e., $\omega_1-\omega_l\rightarrow\omega_1-\omega_l-i\gamma$ and $\omega_2-\omega_l\rightarrow\omega_2-\omega_l-i\gamma$, and thus the terms

$$\left|g^2\left[\frac{1}{(\omega_1-\omega_l)}+\frac{1}{(\omega_2-\omega_l)}\right]\right|^2$$

should be replaced by

$$\begin{aligned}\left|g^2\left[\frac{1}{(\omega_1-\omega_l-i\gamma)}+\frac{1}{(\omega_2-\omega_l-i\gamma)}\right]\right|^2&=g^2\left|\frac{(\Delta-2i\gamma)}{(\omega_1-\omega_l-i\gamma)(\omega_2-\omega_l-i\gamma)}\right|^2\\&\approx g^4\frac{(\Delta^2+4\gamma^2)}{\Delta_1^2\Delta_2^2}\ .\end{aligned}$$

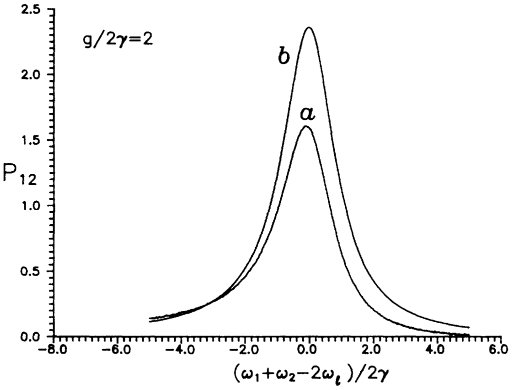

FIG. 2. Transition probability P_{12} for simultaneous excitation of a pair of nonidentical atoms ($\omega_1\neq\omega_2$) as a function of the detuning $(\omega_1+\omega_2-2\omega_l)/2\gamma$, for two different values of the dipole-dipole coupling $V/2\gamma$. The laser field is assumed to be weak. The atom-field coupling parameter is chosen to be the same for each of the two atoms, $g/2\gamma=2$. The difference in atomic frequencies is chosen as $(\omega_1-\omega_2)/2\gamma=200$, and the collective decay parameter is chosen to be $\gamma_{12}/2\gamma=0.5$. The curves are labeled by the dipole-dipole coupling parameter: curve a, $V/2\gamma=5$, and curve b, $V/2\gamma=20$. The actual values of P_{12} for a and b are, respectively, 10^{-5} and 10^{-4} times smaller than those shown in the figure.

To study the behavior of the two-photon resonance for arbitrary strengths of the d-d interaction and the atom-field interaction it is necessary to obtain the exact solutions of Eq. (4.5). The steady-state solution is

$$\psi=(-M)^{-1}I\ . \tag{4.13}$$

The exact solution requires the inversion of the 15×15 M matrix. We do this numerically. We present the numerical results for the transition probability P_{12} for simultaneous excitation of atoms as a function of the detuning $\Delta=\omega_1+\omega_2-2\omega_l$. For simplicity we choose the coupling constants g_1 and g_2 and the decay constants γ_1 and γ_2 for each atom to be equal, so that $g_1=g_2=g$ and $\gamma_1=\gamma_2=\gamma$. 2γ is the rate at which excited state of a single atom decays. We normalize all frequency parameters with respect to 2γ. Figure 2 shows the behavior of P_{12} when the driving field is weak ($g/2\gamma=2$), for two different values of the dipole-dipole coupling strength. It shows the presence of the dipole-dipole interaction-induced two-photon resonance at $2\omega_l=\omega_1+\omega_2$. As compared to the peak value for $V/2\gamma=5$, the two-photon resonance increases by one order of magnitude for $V/2\gamma=20$. The dipole-dipole interaction between the atoms determines the relative size of the two-photon resonance. Thus as the atoms come nearer to each other, the two-photon resonance becomes more and more prominent. At optical frequencies $\omega\approx3\times10^{15}$ Hz and dipole moments of ≈1 D, dipole-dipole coupling of $V/2\gamma=5$ corresponds to interatomic distance ≈292 nm and $V/2\gamma=20$ to a distance ≈184 nm between the atoms. The resonance line shapes have an asymmetric shape. This arises because of the partial interference between the two excitation channels, via the $|\psi_+\rangle$ and $|\psi_-\rangle$ intermediate states [Eq. (3.3)]. Figure 2 does not show the dispersive feature of Eq. (4.9) as the dipole-dipole interaction is large enough so that the V^2 term dominates. We next consider two-photon excitation of two atoms by intense fields ($g/2\gamma=20$). This result is shown in Fig. 3. For large dipole-dipole coupling of $V/2\gamma=20$, the strong field leads to an enhancement of the two-photon resonance by three orders of magnitude as compared to the weak-field case shown in Fig. 2.

The power emitted by the two-atom system into a unit solid angle around a point $\mathbf{R}$, per unit time, is obtained from the normal component of the Poynting vector

$$\begin{aligned}I(\mathbf{R},t)&=R^2\hat{\mathbf{R}}\cdot\hat{S}\\&=\frac{cR^2}{4\pi}\langle:\mathbf{E}\times\mathbf{B}:\rangle\cdot\hat{\mathbf{R}}\\&=\frac{cR^2}{4\pi}\langle E^{(-)}(\mathbf{R},t)E^{(+)}(\mathbf{R},t)\rangle\ ,\end{aligned} \tag{4.14}$$

where :: are the normal ordered products. The operators $E^{(+)}$ ($E^{(-)}$) denote the positive-(negative-) frequency

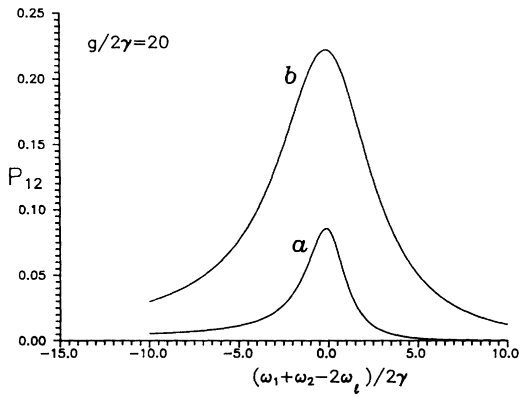

FIG. 3. Transition probability P_{12} when the incident field is assumed to be strong, $g/2\gamma=20$. All other parameters are same as in Fig. 2. The two curves are labeled by the dipole-dipole coupling parameter: curve a, $V/2\gamma=5$ and curve b, $V/2\gamma=20$.

components of the electromagnetic field. In the far zone $k_1R \gg 1$ and $k_2R \gg 1$, where k_1 and k_2 are given by $k_i=\omega_i/c$, $i=1,2$. The positive-frequency part E^+ of the fluorescence-field operator is given by

$$\mathbf{E}^{(+)}(\mathbf{R},t)=\mathbf{E}_0^{(+)}(\mathbf{R},t) - \sum_{i=1}^{2} k_i^2 \frac{\mathbf{R}_i\times(\mathbf{R}_i\times\mathbf{d})}{|\mathbf{R}_i|} S_i^- \left[t-\frac{|\mathbf{R}|}{c}\right] \times\exp(-ik_i\hat{\mathbf{R}}\cdot\mathbf{r}_i)\ . \tag{4.15}$$

Here $\mathbf{r}_i$ is the position vector of each atom and $\mathbf{R}_i=\mathbf{R}-\mathbf{r}_i\cdot E_0^+(\mathbf{R},t)$ denotes the free field. The second term in Eq. (4.15) depends directly on the atomic operators and represents the radiation field emitted by the atom. Substituting (4.15) in (4.14), $I(\mathbf{R},t)$ is evaluated under the assumption that in the far zone $k_1R\sim k_2R$ leading to

$$I(\mathbf{R},t)=\frac{3\gamma\omega_0}{\pi}\sin^2\theta \sum_{i,j=1}^{2} e^{ik_0\hat{\mathbf{R}}\cdot\mathbf{r}_{ij}}\langle S_i^+(t)S_j^-(t)\rangle\ , \tag{4.16}$$

where θ is the angle between $\mathbf{d}$ and $\mathbf{R}$, and we have ignored retardation effects. Integrating over the solid angle around $\mathbf{R}$, the total radiation rate is

$$I(t)=\int I(\mathbf{R},t)d\Omega_{\mathbf{R}}\approx 2\omega_0\gamma\sum_{i,j}\langle S_i^+(t)S_j^-(t)\rangle\ , \tag{4.17}$$

where it is assumed that interatomic distance $\mathbf{r}_{12}$ is much smaller than the optical wavelengths so that $\exp(ik_0\hat{\mathbf{R}}\cdot\mathbf{r}_{ij})\approx 1$.

The nature of the intensity of the radiation emitted by the two-atom system in the far-zone region is shown in Figs. 4 and 5. The radiated power reflects the behavior of P_{12}. For weak fields (Fig. 4) the two-photon resonance becomes more and more prominent as the atoms come closer. This effect is enhanced in the presence of the intense driving field (Fig. 5). Thus the two-photon resonance induced by the dipole-dipole interaction can be probed by fluorescent-intensity measurements.

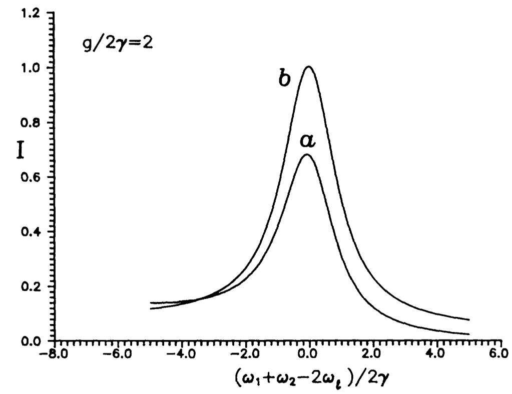

FIG. 4. Radiated field intensity I as a function of the detuning $(\omega_1+\omega_2-2\omega_l)/2\gamma$, for two different values of the dipole-dipole coupling constant $V/2\gamma$ when the incident field is weak $(g/2\gamma=2)$. All other parameters are the same as in Fig. 2. The curves are labeled by the dipole-dipole coupling parameter: curve a, $V/2\gamma=5$, and curve b, $V/2\gamma=20$. The actual values of I for a and b are, respectively, 10^{-4} and 10^{-3} times smaller than those shown in the figure.

The spontaneous emission in general leads to correlations among atoms and thus the absorption and emission characteristics of the two-atom system differ from those of independent atoms. An idea of atomic correlations induced by spontaneous emission can be obtained by studying the quantity

$$G=\langle S_1^+S_2^-\rangle-\langle S_1^+\rangle\langle S_2^-\rangle\ . \tag{4.18}$$

This represents correlations between the dipole moments of two atoms and becomes zero as the distance between the atoms becomes large, i.e., as $kR_{ij}\gg 1$. The behavior of ReG and ImG is plotted in Figs. 6 and 7, respectively. The real part of G shows a dispersive structure and the imaginary part an absorptionlike structure. The atoms are strongly correlated for large dipole-dipole coupling strength and strong fields.

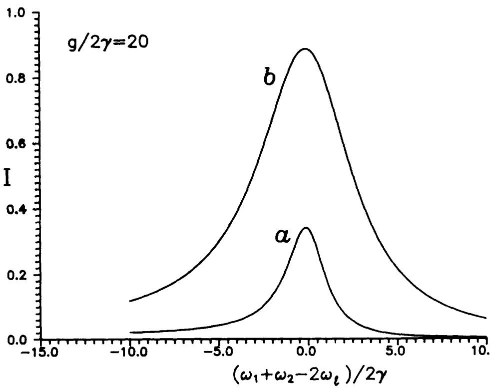

FIG. 5. Radiated field intensity I when the incident field is assumed to be strong, $(g/2\gamma=20)$. All other parameters are same as in Fig. 2. The curves are labeled by the dipole-dipole coupling parameter: curve a, $V/2\gamma=5$, and curve b, $V/2\gamma=20$.

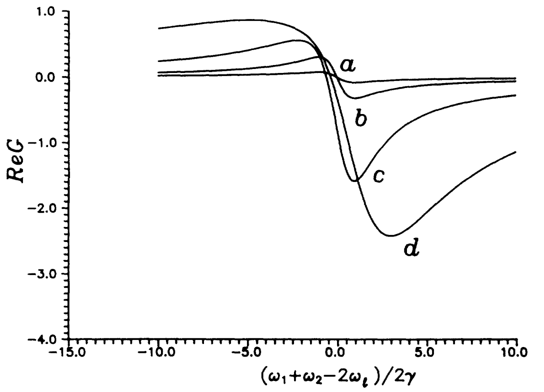

FIG. 6. Real part of the atomic-correlation function G as a function of the detuning $(\omega_1+\omega_2-2\omega_l)/2\gamma$. Different curves are labeled by the values of the atom-field coupling parameter $g/2\gamma$ and the dipole-dipole coupling parameter $V/2\gamma$. All other parameters are the same as in Fig. 2. Curves a and b correspond to the weak-field case: curve a, $g/2\gamma=2$, $V/2\gamma=5$, and curve b, $g/2\gamma=2$, $V/2\gamma=20$. Curves c and d, correspond to the strong-field case: curve c, $g/2\gamma=20$, $V/2\gamma=5$, and curve d, $g/2\gamma=20$, $V/2\gamma=20$. The actual values for a, b, c, and d are, respectively, 0.2×10^{-4}, 0.2×10^{-4}, 10^{-2}, and 10^{-2} times smaller than those shown in the figure.

In conclusion, we have examined the phenomenon of cooperative excitation of two two-level atoms by a two-photon absorption process. The transition probability for simultaneous excitation of atoms shows a dipole-dipole coupling-induced two-photon resonance. Master-equation formalism is adopted to include the effects of spontaneous emission and intense external fields. We find that the extra resonance is significantly enhanced in the presence of an intense driving field. The intensity of the emitted fluorescence reflects the behavior of the pairwise excitation of atoms. Measurement of the fluorescence intensity from a pair of trapped atoms can thus be a possible means for probing pair interactions. In addition, the dipole-dipole interaction introduces nonzero correlations between the atoms which increase significantly as the atoms come closer and closer to each other as expected. The intensity of the external field also affects the interatomic correlations significantly. Resonant enhancement of the correlations between the atoms and the presence of the extra two-photon resonance in the absorption spectrum suggests that the fluorescence spectrum of the radiation scattered from a pair of coupled atoms will be modified from the three-peaked Mollow spectrum [29] of isolated atoms. The case of identical atoms is discussed in Refs. [6–8] and the case of nonidentical atoms will be reported in a future work. The frequencies of emission are given by $\omega_l+(\lambda_i-\lambda_j)$, where λ_i's $(i=1,2,3,4)$ are the eigenvalues of Eq. (2.9). In general, these eigenvalues are to be obtained numerically. For example, for the case when the detunings are such that the two-photon resonance condition is satisfied, i.e., $\Delta_1+\Delta_2=0$, and $\Delta_1=20\gamma$, $V=40\gamma$, and $g=40\gamma$, the atomic system is found to emit radiation at the frequencies ω_l, $\omega_l+52.1$, $\omega_l-18.03$, and $\omega_l-34.07$. It is also expected that the dipole-dipole interaction will give rise to new effects in the nonlinear response of dense atomic systems which would also be the subject of a future study.

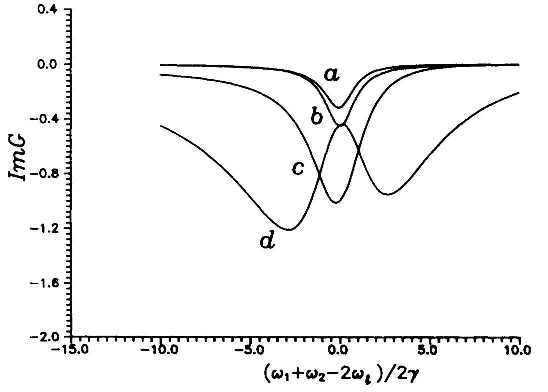

FIG. 7. Imaginary part of the atomic correlation function G as a function of $(\omega_1+\omega_2-2\omega_l)/2\gamma$ for the same parameters as in Fig. 6. Different curves are labeled by the coupling parameters: curve a, $g/2\gamma=2$, $V/2\gamma=5$; curve b, $g/2\gamma=2$, $V/2\gamma=20$; curve c, $g/2\gamma=20$, $V/2\gamma=5$; and curve d, $g/2\gamma=20$, $V/2\gamma=20$. The actual values for a, b, c, and d are, respectively, 10^{-6}, 10^{-5}, 10^{-3}, and 10^{-3} times smaller than those shown in the figure.

ACKNOWLEDGMENT

We would like to thank the Department of Science and Technology, Government of India, for financial support.

APPENDIX

In this appendix we present explicitly the coupled equations for various mean values involving the single-atom operators and atomic correlations. These follow from the master equation (4.1):

$$\langle \dot{S}_1^+\rangle=\dot{\psi}_1=(-\gamma_1+i\Delta_1)\langle S_1^+\rangle+2\gamma_{12}\langle S_1^zS_2^+\rangle$$
$$-2iV\langle S_1^zS_2^+\rangle-2ig_1^*\langle S_1^z\rangle\ ,$$

$$\langle \dot{S}_1^-\rangle=\dot{\psi}_2=(-\gamma_1-i\Delta_1)\langle S_1^-\rangle+2\gamma_{12}\langle S_1^zS_2^-\rangle$$
$$+2iV\langle S_1^zS_2^-\rangle+2ig_1\langle S_1^z\rangle\ ,$$

$$\langle \dot{S}_1^z\rangle=\dot{\psi}_3=-2\gamma_1\langle S_1^z\rangle-\gamma_1-\gamma_{12}\langle S_1^+S_2^-\rangle$$
$$-\gamma_{12}\langle S_1^-S_2^+\rangle-iV\langle S_1^+S_2^-\rangle$$
$$+iV\langle S_1^-S_2^+\rangle-ig_1\langle S_1^+\rangle+ig_1^*\langle S_1^-\rangle\ ,$$

$$\langle \dot{S}_2^+\rangle=\dot{\psi}_4=(-\gamma_2+i\Delta_2)\langle S_2^+\rangle+2\gamma_{12}\langle S_2^zS_1^+\rangle$$
$$-2iV\langle S_2^zS_1^+\rangle-2ig_2^*\langle S_2^z\rangle\ ,$$

$$\langle \dot{S}_2^-\rangle=\dot{\psi}_5=(-\gamma_2-i\Delta_2)\langle S_2^-\rangle+2\gamma_{12}\langle S_2^zS_1^-\rangle$$
$$+2iV\langle S_2^zS_1^-\rangle+2ig_2\langle S_2^z\rangle\ ,$$

$$\langle \dot{S}_2^z\rangle=\dot{\psi}_6=-2\gamma_2\langle S_2^z\rangle-\gamma_2-\gamma_{12}\langle S_1^+S_2^-\rangle$$
$$-\gamma_{12}\langle S_1^-S_2^+\rangle+iV\langle S_1^+S_2^-\rangle$$
$$-iV\langle S_1^-S_2^+\rangle-ig_2\langle S_2^+\rangle+ig_2^*\langle S_2^-\rangle\ ,$$

$$
\begin{aligned}
\langle S_1^+\overset{\cdot}{}S_2^+\rangle=\dot{\psi}_7={}&[-(\gamma_1+\gamma_2)+i\Delta]\langle S_1^+S_2^+\rangle\\
&-2ig_1^*\langle S_1^zS_2^+\rangle-2ig_2^*\langle S_2^zS_1^+\rangle\ ,\\
\langle S_1^-\overset{\cdot}{}S_2^-\rangle=\dot{\psi}_8={}&[-(\gamma_1+\gamma_2)-i\Delta]\langle S_1^-S_2^-\rangle\\
&+2ig_1\langle S_1^zS_2^-\rangle+2ig_2\langle S_2^zS_1^-\rangle\ ,\\
\langle S_1^z\overset{\cdot}{}S_2^z\rangle=\dot{\psi}_9={}&-2(\gamma_1+\gamma_2)\langle S_1^zS_2^z\rangle-\gamma_1\langle S_2^z\rangle\\
&-\gamma_2\langle S_1^z\rangle+\gamma_{12}\langle S_1^+S_2^-\rangle\\
&+\gamma_{12}\langle S_1^-S_2^+\rangle-ig_1\langle S_2^zS_1^+\rangle-ig_2\langle S_1^zS_2^+\rangle\\
&+ig_1^*\langle S_2^zS_1^-\rangle+ig_2^*\langle S_1^zS_2^-\rangle\ ,\\
\langle S_1^+\overset{\cdot}{}S_2^-\rangle=\dot{\psi}_{10}={}&[-(\gamma_1+\gamma_2)+i\delta]\langle S_1^+S_2^-\rangle+\gamma_{12}\langle S_1^z\rangle\\
&+\gamma_{12}\langle S_2^z\rangle+4\gamma_{12}\langle S_1^zS_2^z\rangle\\
&-iV(\langle S_1^z\rangle-\langle S_2^z\rangle)+2ig_2\langle S_2^zS_1^+\rangle\\
&-2ig_1^*\langle S_1^zS_2^-\rangle\ ,\\
\langle S_1^-\overset{\cdot}{}S_2^+\rangle=\dot{\psi}_{11}={}&[-(\gamma_1+\gamma_2)-i\delta]\langle S_1^-S_2^+\rangle+\gamma_{12}\langle S_1^z\rangle\\
&+\gamma_{12}\langle S_2^z\rangle+4\gamma_{12}\langle S_1^zS_2^z\rangle\\
&+iV(\langle S_1^z\rangle-\langle S_2^z\rangle)-2ig_2^*\langle S_2^zS_1^-\rangle\\
&+2ig_1\langle S_1^zS_2^+\rangle\ ,\\
\langle S_1^z\overset{\cdot}{}S_2^+\rangle=\dot{\psi}_{12}={}&-\gamma_1\langle S_2^+\rangle+[-(2\gamma_1+\gamma_2)+i\Delta_2]\langle S_1^zS_2^+\rangle\\
&-\frac{\gamma_{12}}{2}\langle S_1^+\rangle-2\gamma_{12}\langle S_2^zS_1^+\rangle-i\frac{V}{2}\langle S_1^+\rangle\\
&-ig_1\langle S_1^+S_2^+\rangle+ig_1^*\langle S_1^-S_2^+\rangle\\
&-2ig_2^*\langle S_1^zS_2^z\rangle\ ,\\
\langle S_1^z\overset{\cdot}{}S_2^-\rangle=\dot{\psi}_{13}={}&-\gamma_1\langle S_2^-\rangle-[2\gamma_1+\gamma_2+i\Delta_2]\langle S_1^zS_2^-\rangle\\
&-\frac{\gamma_{12}}{2}\langle S_1^-\rangle-2\gamma_{12}\langle S_2^zS_1^-\rangle+i\frac{V}{2}\langle S_1^-\rangle\\
&+ig_1^*\langle S_1^-S_2^-\rangle-ig_1\langle S_1^+S_2^-\rangle\\
&+2ig_2\langle S_1^zS_2^z\rangle\ ,\\
\langle S_2^z\overset{\cdot}{}S_1^+\rangle=\dot{\psi}_{14}={}&-\gamma_2\langle S_1^+\rangle+[-(2\gamma_2+\gamma_1)+i\Delta_1]\langle S_2^zS_1^+\rangle\\
&-\frac{\gamma_{12}}{2}\langle S_2^+\rangle-2\gamma_{12}\langle S_1^zS_2^+\rangle-i\frac{V}{2}\langle S_2^+\rangle\\
&-ig_2\langle S_1^+S_2^+\rangle+ig_2^*\langle S_1^+S_2^-\rangle\\
&-2ig_1^*\langle S_1^zS_2^z\rangle\ ,\\
\langle S_2^z\overset{\cdot}{}S_1^-\rangle=\dot{\psi}_{15}={}&-\gamma_2\langle S_1^-\rangle-[2\gamma_2+\gamma_1+i\Delta_1]\langle S_2^zS_1^-\rangle\\
&-\frac{\gamma_{12}}{2}\langle S_2^-\rangle-2\gamma_{12}\langle S_1^zS_2^-\rangle+i\frac{V}{2}\langle S_2^-\rangle\\
&+ig_2^*\langle S_1^-S_2^-\rangle-ig_2\langle S_1^-S_2^+\rangle\\
&+2ig_1\langle S_1^zS_2^z\rangle\ .
\end{aligned}
\tag{A1}
$$

The interdependence of the dynamical evolution of one atom on the other through the dipole-dipole coupling is evident in these equations. When $V=0$, these equations reduce to the optical Bloch equations for each single atom.

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