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Superradiance from an ultrathin film of three-level V-type atoms: interplay between splitting, quantum coherence and local-field effects

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Abstract

We carry out a theoretical study of the collective spontaneous emission (superradiance) from an ultrathin film comprised of three-level atoms with V configuration of the operating transitions. As the thickness of the system is small compared to the emission wavelength inside the film, the local-field correction to the averaged Maxwell field is relevant. We show that the interplay between the low-frequency quantum coherence within the subspace of the upper doublet states and the local-field correction may drastically affect the branching ratio of the operating transitions. This effect may be used for controlling the emission process by varying the doublet splitting and the amount of low-frequency coherence.

Keywords: Superradiance, quantum coherence, local field, three-level atoms

1. Introduction

After the pioneering works by Kocharovskaya and Khanin [1, 2], Harris [3] and Scully *et al* [4], the effects of the light–matter interactions in the presence of low-frequency quantum coherence between sublevels in the ground or excited states have received much attention. There appeared new phenomena such as *amplification without inversion* and *lasing without inversion* (AWI and LWI), *electromagnetically induced transparency* (EIT), etc (see [5, 6] for a review as well as the topical issue of quantum optics [7]). Yet, optical bistability in systems of V-type atoms has been considered in [8, 9]. The interplay between coherence and interference effects in dense V-type systems has been addressed in [10].

Recently, it was shown that, in a closed system of Λ-type atoms, the low-frequency coherence may give rise to superradiance without inversion (SRWI) [11–16]. In [17], the SRWI of an open (i.e. in the presence of a driven field) system of V-type atoms was discussed. We might stress that the standard superradiance (SR) effect, predicted by Dicke for a collection of two-level atoms [18], requires an initial inversion of level populations (see the review by Gross and Haroche [19] and the book by Benedict *et al* [20] for further details).

The goal of the present paper is to analyse theoretically the features of the SR of a closed system of V-type atoms. This problem was already discussed in the 1980s by Crubellier *et al* [21, 22], Molander and Stroud [23] and Keitel *et al* [24] within the context of subradiance—inhibited spontaneous emission. This effect has been observed experimentally by Pavolini *et al* [25] in a low-density gas of gallium atoms (the density of gallium atoms in the interaction region was about 10^{12} atoms cm^{-3}). Here, we consider a dense system comprised of V-type radiators and take into account the local-field correction (LFC) to the averaged Maxwell

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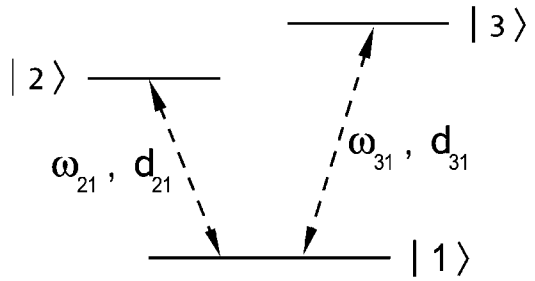

Figure 1. Scheme of the energy levels and transitions in a V-type atom.

field. This correction was neglected in previous theoretical studies [21–23] and this approach is then the novelty of this paper. As is well known from the studies of high-density two-level assemblies [26–39], the LFC significantly affects the resonant optical response of such systems, especially the response of ultrathin films [40–43]. In particular, the effects of the LFC on the SR of a Λ-type dense system have been discussed in [14–16, 44]. We show that, in the case of a dense V-type system, the interplay between the LFC, doublet splitting and low-frequency coherence dramatically influences the branching ratio of the operating transitions. This effect can be used for controlling the SR by varying the doublet splitting and the degree of coherence between the doublet states.

The outline of the paper is as follows. In section 2 we present the model we will be dealing with. We derive the truncated equations for the density matrix elements and electric field taking into account the LFC to the Maxwell field, within the rotating wave approximation (RWA). Section 3 is devoted to the particular case of degenerate doublets in the upper state, allowing for an analytical solution of the problem. Results of numerical simulations are described in section 4, where we provide interpretations as well of the peculiarities found numerically. We conclude the paper in section 5 with a brief summary of the results and how these can be related to actual measurements to infer the main characteristics of the LFC.

2. Model and truncated equations

We consider a system of three-level atoms with a doublet in the upper state (the so-called V configuration, see figure 1), forming an ultrathin film of thickness L smaller than the emission wavelength inside the film. This constraint is often met in experiments and simplifies the mathematical description of the model by taking away the propagation effect along the film normal. All the vectors, the SR electric field and transition dipole moments of the operating transitions, d_{21} and d_{31}, are considered to be parallel to each other as well as to the film plane (the transition between the levels of the doublet is not included). Under these assumptions, the vector nature of the above quantities is not important and the problem is reduced to its scalar form. It is also assumed, without loss of generality, that the transition dipole moments are real and positive: $d_{21} = d_{12} > 0$ and $d_{31} = d_{13} > 0$.

We use the semiclassical description of the optical dynamics of the system, treating the atom dynamics by means of the density matrix $\rho_{\alpha\beta}$ $(\alpha, \beta = 1, 2, 3)$, i.e. quantum mechanically, while the field evolution is assumed to obey the classical Maxwell equation. Under the limitations adopted above, the joint set of equations for the density matrix and Maxwell field are

$$\dot\rho_{31} = -\mathrm{i}\omega_{31}\rho_{31} - \mathrm{i}\frac{d_{31}\mathcal{E}'}{\hbar}(\rho_{33} - \rho_{11}) - \mathrm{i}\frac{d_{21}\mathcal{E}'}{\hbar}\rho_{32} \tag{1}$$

$$\dot\rho_{21} = -\mathrm{i}\omega_{21}\rho_{21} - \mathrm{i}\frac{d_{21}\mathcal{E}'}{\hbar}(\rho_{22} - \rho_{11}) - \mathrm{i}\frac{d_{31}\mathcal{E}'}{\hbar}\rho_{23} \tag{2}$$

$$\dot\rho_{32} = -\mathrm{i}\omega_{32}\rho_{32} - \mathrm{i}\frac{d_{21}\mathcal{E}'}{\hbar}\rho_{31} + \mathrm{i}\frac{d_{31}\mathcal{E}'}{\hbar}\rho_{12}, \tag{3}$$

$$\dot\rho_{33} = \mathrm{i}\frac{d_{31}\mathcal{E}'}{\hbar}(\rho_{13} - \rho_{31}), \tag{4}$$

$$\dot\rho_{22} = \mathrm{i}\frac{d_{21}\mathcal{E}'}{\hbar}(\rho_{12} - \rho_{21}), \tag{5}$$

$$\dot\rho_{11} = -\mathrm{i}\frac{d_{21}\mathcal{E}'}{\hbar}(\rho_{12} - \rho_{21}) - \mathrm{i}\frac{d_{31}\mathcal{E}'}{\hbar}(\rho_{13} - \rho_{31}). \tag{6}$$

Here, the dots denote the time derivative and $\mathcal{E}'$ stands for the acting field:

$$\mathcal{E}' = -\frac{2\pi L}{c}\dot{\mathcal{P}} + \frac{4\pi}{3}\mathcal{P} \tag{7}$$

L and c being the film thickness and the speed of light, respectively; $\mathcal{P} = N_0(d_{31}\rho_{31} + d_{21}\rho_{21} + \text{c.c.})$ is the electric polarization of the unit volume, with N_0 being the atom number concentration. The first term in equation (7) represents the Maxwellian emission field, while the second term is the LFC.

In order to further specify the model we are dealing with, we first stress that it is applicable to the description of SR of a thin dielectric crystalline film rather than a dense gas system. In the latter case, the pressure broadening terms turn out to be important. Having the same nature and order of magnitude as the LFC [30], these terms have to be added to the equations for the off-diagonal density matrix elements. In solid crystalline media, the levels are usually broadened due to crystal imperfections as well as coupling to phonons. Under specific conditions, the width of the levels is smaller than the LFC (in frequency units). The existence of Frenkel exciton states in dielectric solids, which are due to the interatomic dipolar coupling [45, 46] or, in other words, due to the LFC, represents an unambiguous confirmation of this fact. Because of that, we do not take into account either relaxation of populations or dephasing of the electric polarization of a single atom, assuming that the SR process is faster (the estimates of the corresponding constants are presented in section 5).

We seek a solution of equations (1)–(6) in the form: $\mathcal{E}' = E'\exp(-\mathrm{i}\omega_c t) + \text{c.c.}$, $\rho_{31} = R_{31}\exp(-\mathrm{i}\omega_c t)$, $\rho_{21} = R_{21}\exp(-\mathrm{i}\omega_c t)$, where $\omega_c = (\omega_{31} + \omega_{21})/2$; E' and R_{31}, R_{21} are the complex slowly varying (in the scale of $2\pi/\omega_c$) amplitudes of the field and of the off-diagonal density matrix elements, respectively. Hereafter the latter will be referred to as optical coherences. Within the RWA, the equations for the amplitudes are

$$\begin{aligned}\dot R_{31} = {} & -\mathrm{i}\frac{\omega_{32}}{2}R_{31} + \left(\frac{1}{\tau_R} - \mathrm{i}\Delta_L\right)[\mu_{31}(\rho_{33} - \rho_{11}) \\ & + \mu_{21}\rho_{32}](\mu_{21}R_{21} + \mu_{31}R_{31}),\end{aligned} \tag{8}$$

$$\dot R_{21} = \mathrm{i}\frac{\omega_{32}}{2}R_{21} + \left(\frac{1}{\tau_R} - \mathrm{i}\Delta_L\right)[\mu_{21}(\rho_{22} - \rho_{11})$$

$$+\mu_{31}\rho_{23}](\mu_{21}R_{21}+\mu_{31}R_{31}), \tag{9}$$

$$\dot\rho_{32}=-\mathrm{i}\omega_{32}\rho_{32}-\left[\left(\frac{1}{\tau_R}+\mathrm{i}\Delta_L\right)\mu_{21}R_{31}(\mu_{21}R_{21}^*+\mu_{31}R_{31}^*)+\left(\frac{1}{\tau_R}-\mathrm{i}\Delta_L\right)\mu_{31}R_{21}^*(\mu_{21}R_{21}+\mu_{31}R_{31})\right], \tag{10}$$

$$\dot\rho_{33}=\mu_{31}\left[\left(-\frac{1}{\tau_R}+\mathrm{i}\Delta_L\right)(\mu_{21}R_{21}+\mu_{31}R_{31})R_{31}^*+\mathrm{c.c.}\right], \tag{11}$$

$$\dot\rho_{22}=\mu_{21}\left[\left(-\frac{1}{\tau_R}+\mathrm{i}\Delta_L\right)(\mu_{21}R_{21}+\mu_{31}R_{31})R_{21}^*+\mathrm{c.c.}\right], \tag{12}$$

$$\dot\rho_{11}=\frac{2}{\tau_R}|\mu_{21}R_{21}+\mu_{31}R_{31}|^2. \tag{13}$$

Here we have defined $\mu_{31}=d_{31}/d$ and $\mu_{21}=d_{21}/d$, where $d=\sqrt{(d_{31}^2+d_{21}^2)/2}$, $\Delta_L=4\pi d^2N_0/3\hbar$, $\tau_R^{-1}=2\pi k_cLd^2N_0/\hbar$, $k_c=\omega_c/c$. When deriving equations (8)–(13), we exploited the fact that the equation for the slowly varying field amplitude E' can be cast in the form

$$\frac{\mathrm{d}E'}{\hbar}=\left(\frac{\mathrm{i}}{\tau_R}+\Delta_L\right)(\mu_{21}R_{21}+\mu_{31}R_{31}), \tag{14}$$

and we introduced this expression directly into the density matrix equation. The quantities τ_R^{-1} and Δ_L represent the magnitudes of the SR field and of the LFC (in frequency units), respectively [42]. Recall that $\Delta_L>\tau_R^{-1}$ since the relationship $k_cL<1$ holds for an ultrathin film.

It is to be noted that equations (8)–(13) have the following integrals of motion:

$$\rho_{11}+\rho_{22}+\rho_{33}=1, \tag{15}$$

$$\rho_{11}^2+\rho_{22}^2+\rho_{33}^2+2(|\rho_{32}|^2+|R_{31}|^2+|R_{21}|^2)=\text{constant}, \tag{16}$$

where the first equation establishes the normalization condition for the total level population, while the second one represents the conservation of the trace of the atomic density matrix squared. Physically, it express the conservation of the symmetry under atom permutations [21, 24].

To complete the mathematical formalism we should specify the initial conditions for equations (8)–(13). We assume that the doublet states are initially populated, i.e. there exist nonzero $\rho_{33}(0)$ and $\rho_{22}(0)$. We also allow an initial low-frequency coherence $\rho_{32}(0)$. In order to trigger the emission process, we set a fixed (not-fluctuating) value for the initial electric polarization in the operating channels, $R_{31}(0)=R_{21}(0)=R_0$. This corresponds to triggering the SR by an ultrashort external pulse of a small area, with a duration $T_p<\min\{2\pi/\omega_{32},\tau_R^{-1}\}$ [20, 48, 49].

Equations (8)–(13) are written within the original basis of states $|1\rangle$, $|2\rangle$ and $|3\rangle$. From physical reasons, especially in the case of a degenerated doublet (see below), another set of states turns out to be very useful: $|1\rangle$, $|+\rangle=(1/\sqrt{2})(\mu_{21}|2\rangle+\mu_{31}|3\rangle)$ and $|-\rangle=(1/\sqrt{2})(\mu_{21}|3\rangle-\mu_{31}|2\rangle)$. The convenience of this set is clear from the fact that only the superposition $|+\rangle$ is coupled to the ground state $|1\rangle$ (it will be referred to as the bright state hereafter), while the remainder one is decoupled (the dark state). The dipole moments of the transitions $|1\rangle\to|+\rangle$ and $|1\rangle\to|-\rangle$ are $\langle1|\hat d|+\rangle=\sqrt{2}d$ and $\langle1|\hat d|-\rangle=0$. In this regards, the dark channel does not contribute to the SR.

Within the new basis, $|1\rangle$, $|+\rangle$, $|-\rangle$, the density matrix elements can be expressed as follows:

$$R_{+1}=\frac{1}{\sqrt{2}}(\mu_{21}R_{21}+\mu_{31}R_{31}), \tag{17}$$

$$\rho_{++}=\tfrac{1}{2}(\mu_{21}^2\rho_{22}+\mu_{31}^2\rho_{33}+2\mu_{21}\mu_{31}\Re\rho_{32}), \tag{18}$$

$$R_{-1}=\frac{1}{\sqrt{2}}(\mu_{21}R_{31}-\mu_{31}R_{21}), \tag{19}$$

$$\rho_{--}=\tfrac{1}{2}(\mu_{21}^2\rho_{33}+\mu_{31}^2\rho_{22}-2\mu_{21}\mu_{31}\Re\rho_{32}), \tag{20}$$

$$\rho_{+-}=\tfrac{1}{2}[\mu_{21}\mu_{31}(\rho_{33}-\rho_{22})+\mu_{21}^2\rho_{23}-\mu_{31}^2\rho_{32}], \tag{21}$$

where now ρ_{++} and ρ_{--} stand for populations of the bright and dark states, respectively, ρ_{+-} represents the low-frequency coherence, while R_{+1} and R_{-1} describe the coherence of the bright and dark channels, respectively. We stress that R_{+1} determines the field, as seen from equation (7). The equations for these matrix elements are

$$\dot R_{+1}=-\mathrm{i}\frac{\omega_{32}}{4}[(\mu_{31}^2-\mu_{21}^2)R_{+1}+2\mu_{21}\mu_{31}R_{-1}]+2\left(\frac{1}{\tau_R}-\mathrm{i}\Delta_L\right)(\rho_{++}-\rho_{11})R_{+1}, \tag{22}$$

$$\dot\rho_{++}=\mathrm{i}\frac{\omega_{32}}{2}\mu_{21}\mu_{31}(\rho_{+-}-\rho_{-+})-\frac{4}{\tau_R}|R_{+1}|^2, \tag{23}$$

$$\dot\rho_{11}=\frac{4}{\tau_R}|R_{+1}|^2, \tag{24}$$

$$\dot R_{-1}=-\mathrm{i}\frac{\omega_{32}}{4}[(\mu_{21}^2-\mu_{31}^2)R_{-1}+2\mu_{21}\mu_{31}R_{+1}]+2\left(\frac{1}{\tau_R}-\mathrm{i}\Delta_L\right)R_{+1}\rho_{-+}, \tag{25}$$

$$\dot\rho_{+-}=\mathrm{i}\frac{\omega_{32}}{2}[(\mu_{21}^2-\mu_{31}^2)\rho_{+-}+\mu_{21}\mu_{31}(\rho_{++}-\rho_{--})]+2\left(-\frac{1}{\tau_R}+\mathrm{i}\Delta_L\right)R_{+1}R_{-1}^*, \tag{26}$$

$$\dot\rho_{--}=\mathrm{i}\frac{\omega_{32}}{2}\mu_{21}\mu_{31}(\rho_{-+}-\rho_{+-}). \tag{27}$$

As can be seen from equations (22)–(27), the bright channel ($|+\rangle\to|1\rangle$) is coupled to the dark one ($|-\rangle\to|1\rangle$) through ω_{32} terms, and thus, at $\omega_{32}=0$, the former turns out to be independent of the latter (see section 3 for more details). At the same time, the behaviour of the dark channel is driven by the bright one even in the presence of degeneracy of the doublet states: the field terms proportional to τ_R^{-1} and Δ_L play this role.

3. Degenerated doublet ($\omega_{32}=0$)

We first analyse the degenerated case. Then equations (22)–(27), describing the bright channel and which we are interesting in, reduce to

$$\frac{\mathrm{d}}{\mathrm{d}t}|R_{+1}|=\frac{4}{\tau_R}Z|R_{+1}|, \tag{28}$$

$$\dot Z=-\frac{4}{\tau_R}|R_{+1}|^2, \tag{29}$$

$$\dot\phi=-4\Delta_LZ, \tag{30}$$

where the new variables are $Z\equiv(\rho_{++}-\rho_{11})/2$ and ϕ is the phase of R_{+1}. We stress that equations (28)–(30) are

similar to the SR equations of an ultrathin film of two-level atoms [20, 50–52], but replacing τ_R by $\tau_R/2$ and Δ_L by $2\Delta_L$, which is reasonable due to the presence of two emission channels.

Several qualitative conclusions about the system behaviour can be drawn from the direct analysis of equations (28)–(30). First of all, the derivative $\mathrm{d}|R_{+1}|/\mathrm{d}t$ is positive and thus $|R_{+1}|$ will give rise to the SR if $Z(0) > 0$ or, in other words, if there is an initial population inversion between the bright and ground states:

$$\rho_{++}(0) = \tfrac{1}{2}[\mu_{21}^2\rho_{22}(0) + \mu_{31}^2\rho_{33}(0) + 2\mu_{21}\mu_{31}\Re\rho_{32}(0)] > \rho_{11}(0). \tag{31}$$

From here it can be shown that, in order to meet this inequality, the total initial population of the doublet, $\rho_{22}(0) + \rho_{33}(0)$, must be larger than the population in the ground state, $\rho_{11}(0)$. In other words, in contrast to the case of a close Λ system, where the SRWI can be observed [11], the SR in a close V-type system requires the population inversion between operating levels. For the sake of simplicity, let us set $\rho_{22}(0) = \rho_{33}(0) = \rho_{32}(0) \equiv A = (1/2)[1 - \rho_{11}(0)]$. It corresponds to the excitation of the bright state $|+\rangle = (1/\sqrt{2})(\mu_{21}|2\rangle + \mu_{31}|3\rangle)$ with amplitude A. Then, the inequality (31) takes the form $A(1 + \mu_{21}\mu_{31}) > \rho_{11}(0)$. Bearing in mind that $\mu_{21}\mu_{31} < 1$, we get $2A > \rho_{11}(0)$, i.e. the total population of the doublet must indeed be larger than that in the ground state.

With the substitutions $Z = B\cos\Theta$, and $|R_{+1}| = B\sin\Theta$, where $B \approx Z(0)$, equations (28)–(30) can be solved analytically and the solution is

$$Z = -Z(0)\tanh\left(\frac{t - t_D}{\tau_R'}\right), \tag{32}$$

$$|R_{+1}| = Z(0)\mathrm{sech}\left(\frac{t - t_D}{\tau_R'}\right), \tag{33}$$

$$\phi = -4\Delta_L\int_0^t Z(\tau)\,\mathrm{d}\tau = 4Z(0)\Delta_L\tau_R' \times\left[\ln\cosh\left(\frac{t - t_D}{\tau_R'}\right) - \ln\cosh\left(\frac{t_D}{\tau_R'}\right)\right], \tag{34}$$

$$t_D = \tau_R'\ln\left[\frac{2Z(0)}{|R_{+1}(0)|}\right]. \tag{35}$$

As is seen, the SR pulse is characterized by a delay time t_D and a duration $\tau_R' = \tau_R/4Z(0)$. The unique effect of the LFC on the SR from a degenerated V system is the SR phase modulation which changes the SR frequency:

$$\Omega(t) = \dot{\phi} = 4Z(0)\ \Delta_L\tanh\left(\frac{t - t_D}{\tau_R'}\right), \tag{36}$$

from $-4Z(0)\Delta_L$ to $4Z(0)\Delta_L$. For this reason, it is similar to what is well known for two-level dense systems [20, 50–52], namely within the mean-field approximation we are in fact dealing with, the LFC does not affect the SR kinetics but determines the width of the SR spectrum.

4. Nondegenerated doublet

We show below that the scenario of the SR from a nondegenerated V system changes dramatically in the presence of the LFC. It is to be noticed that, concerning the SR from a Λ system, this fact has already been mentioned in [13–16, 44]. In order to investigate systematically the peculiarities of the SR in the case of a V system, we perform the numerical solution of equations (8)–(13). In all our calculations, the dipole moments of the operating transitions $|3\rangle \to |1\rangle$ and $|2\rangle \to |1\rangle$ are equal to each other, thus implying that $\mu_{21} = \mu_{31} = 1$. The initial values of amplitudes of the high-frequency coherences are set to $R_{31}(0) = R_{32}(0) = 10^{-8}$. Time is expressed in units of τ_R. The other initial magnitudes, such that the level populations $\rho_{11}(0)$, $\rho_{22}(0)$ and $\rho_{33}(0)$, the low-frequency coherence $\rho_{32}(0)$, the doublet splitting ω_{32} and the LFC Δ_L, will be regarded as variable parameters.

4.1. Effects of the low-frequency coherence neglecting the LFC

We first analyse the SR of a nondegenerated V system setting, as a first step, the LFC to zero (see also the discussions in [21, 22, 25]). In spite of the fact that this assumption might be unphysical for an ultrathin film (recall that $\Delta_L > \tau_R$), the analysis of this ideal case will help us in understanding more complicated situations with $\Delta_L \neq 0$. Nondegeneracy means that the magnitude of the splitting ω_{32} is larger than the spectrum width of the SR in the presence of degeneracy. The latter can be estimated on the basis of equations (32)–(35) as $\tau_R'^{-1} = 4Z(0)\tau_R^{-1}$ for $\Delta_L = 0$. Values of ω_{32} about several units of τ_R^{-1} suffice to model the outlined condition as $4Z(0) \leqslant 2$.

Figure 2 shows the kinetics of the SR pulse and the level populations calculated for $\omega_{32} = 5\tau_R^{-1}$ with the following initial conditions: all the population is in the doublet states, $\rho_{22}(0) = \rho_{33}(0) = 0.5$ and, additionally, there exists a low-frequency coherence, $\rho_{32}(0) = 0.5$. Within the subspace of states $|+\rangle$ and $|-\rangle$, this corresponds to the excitation of only the pure bright state $|+\rangle(\rho_{++}(0) = 1)$, while all other populations and coherences are equal to zero, $\rho_{--}(0) = \rho_{11}(0) = \rho_{+-}(0) = 0$ (see equations (17)–(21)). As is seen from figure 2, the SR pulse deactivates *completely* the state $|+\rangle$. All the population is finally transferred to the ground state $|1\rangle$, as it takes place in the case of two-level SR under the condition of total inversion (see, for instance, [19, 20]). The modulation of the kinetics with frequency ω_{32} is explained by the fact that, at nonzero splitting, the bright state $|+\rangle$ is not a stationary state: it periodically (with frequency ω_{32}, i.e. rapidly in the scale of the SR) exchanges population with the dark state $|-\rangle$. Indeed, keeping in equations (23), (26) and (27) only the terms with the density matrix elements within the subspace of states $|+\rangle$ and $|-\rangle$ and introducing the notations $z = \rho_{++} - \rho_{--}$ and $y = \mathrm{i}(\rho_{+-} - \rho_{-+})$, we obtain

$$\dot{y} = -\omega_{32}\,z, \tag{37}$$

$$\dot{z} = \omega_{32}\,y. \tag{38}$$

These equations describe harmonic oscillations, with frequency ω_{32}, of a vector (y, z) in the YZ plane. Values $z = 1, -1$ correspond to the total population of bright and dark states, respectively. The magnitude $y = 2\Im\rho_{+-}$ reflects the low-frequency coherence. Note that $z^2 + y^2 =$ constant. For the initial conditions we are dealing with ($z(0) = 1$ and $y(0) = 0$), for approximately one-half of the period of these

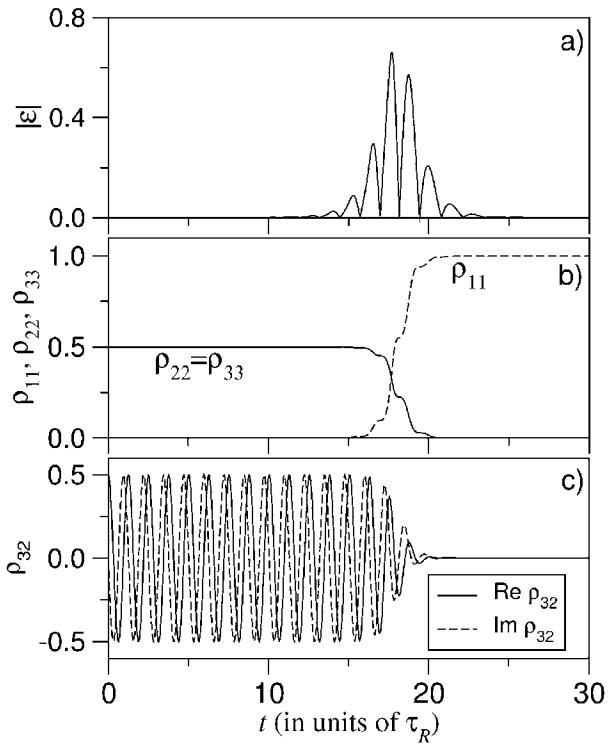

Figure 2. (a) The SR field $|\varepsilon| = d|E|\tau_R/\hbar$, (b) the level populations ρ_{11}, ρ_{22} and ρ_{33} and (c) the low-frequency coherence ρ_{32} calculated for zero LFC ($\Delta_L = 0$) at fixed magnitude of the doublet splitting $\omega_{32} = 5\tau_R^{-1}$. The initial conditions are: $\rho_{22}(0) = \rho_{33}(0) = \rho_{32}(0) = 0.5$ and $R_{21}(0) = R_{31}(0) = 10^{-8}$.

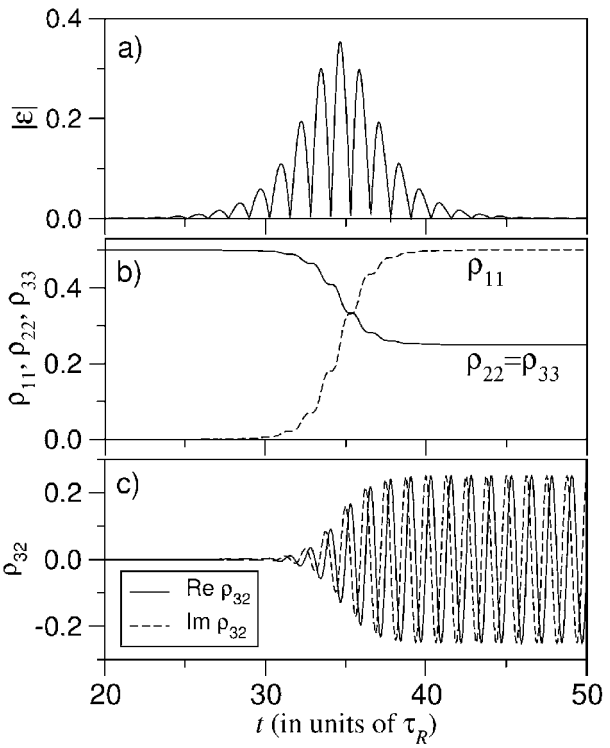

Figure 3. Same as in figure 2, except for $\rho_{32}(0) = 0$.

oscillations the system remains in the bright state while for the other half it does not. This also explains why the delay time of the SR in the present case ($t_D \approx 18\tau_R$) is twice as large compared to that time at $\omega_{32} = 0$. Indeed, using the above initial conditions in equation (35), one obtains $t_D \approx 9\tau_R$ for a degenerated doublet.

From the above discussion it is clear that changing the sign of the initial low-frequency coherence, i.e. setting $\rho_{32}(0) = -0.5$ or, in other words, $\rho_{--}(0) = 1$, will not affect the SR kinetics. After a half period of oscillations with frequency ω_{32}, the previous initial condition is restored.

In figure 3 we depicted the SR kinetics calculated for the same initial conditions as above, except for the low-frequency coherence $\rho_{32}(0)$, which was set to zero. Within the subspace of states $|+\rangle$ and $|-\rangle$, they now correspond to the excitation of an incoherent mixture of the bright and dark states: $\rho_{++}(0) = \rho_{--}(0) = 0.5$ and $\rho_{+-}(0) = 0$ (see equations (18), (20) and (21)). One can notice significant changes in the main features of the SR pulse as compared to the previous case: both the delay time and the pulse duration increased by approximately a factor of two and, in addition, the doublet states remained equally populated after the SR pulse has been emitted: $\rho_{22}(\infty) = \rho_{33}(\infty) = 0.25$. More specifically, the population of the bright state, i.e. only one-half of the total population accumulated in the upper states, is transferred to the ground state during the SR, while the other half remains trapped in the dark state. The increase of the delay time and the duration of the SR pulse by a factor of two is simply explained by the fact that, in the present case, the initial value $Z(0) = \rho_{++}(0) - \rho_{11}(0) = 0.5$ is twice as small as compared to the previous situation. Recall that both t_D and τ_R are inversely proportional to $Z(0)$.

4.2. *Effects of the LFC* ($\Delta_L \neq 0$)

We turn now to studying the LFC effects on the SR kinetics. Therefore, in what follows the magnitude of LFC, Δ_L, is regarded as a variable parameter while the doublet splitting, ω_{32}, is set to a fixed value. More specifically, we present the results of numerical calculations for $\omega_{32} = 5\tau_R^{-1}$, meaning that the splitting is larger than the full width of the SR spectrum which is $\approx 2\tau_R^{-1}$. For small magnitudes of ω_{32} compared to τ_R^{-1}, the SR kinetics is well described by equations (32)–(35). We also assume that the total initial population of the system, $\rho_{22}(0) = \rho_{33}(0) = 0.5$ and $\rho_{11}(0) = 0$, with equal populations of the doublet states, $\rho_{22}(0) = \rho_{33}(0) = 0.5$. As we will show below, the output depends on the initial value of the low-frequency coherence, $\rho_{32}(0)$, as well. We restrict ourselves to two limiting cases: $\rho_{32}(0) = 0$ and $\rho_{32}(0) = \sqrt{\rho_{22}(0)\rho_{33}(0)} = 0.5$. The former corresponds to the initial excitation of incoherent mixture of the doublet states $|2\rangle$ and $|3\rangle$, while the latter implies the initial excitation of the pure state $|+\rangle$. The initial values of the high-frequency coherences are set to $R_{21}(0) = R_{31}(0) = 10^{-8}$ as before.

4.2.1. SR from an incoherent mixture ($\rho_{32}(0) = 0$). Figure 4 shows the SR kinetics calculated for $\rho_{22}(0) = \rho_{33}(0) = 0.5$ (the total initial inversion) in the absence of the initial low-frequency coherence, $\rho_{32}(0) = 0$, varying the LFC magnitude, Δ_L. As is seen from this figure, increasing Δ_L affects drastically the SR kinetics. While Δ_L is smaller than some 'critical' value, the scenario of the SR is similar to that described

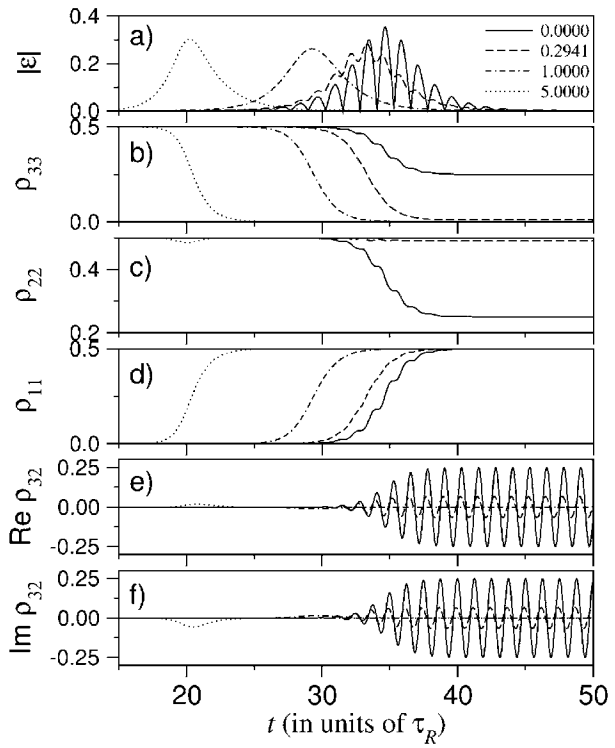

Figure 4. Effects of the LFC on (a) the SR field $|\varepsilon| = d|E|\tau_R/\hbar$, the level populations (b) ρ_{33}, (c) ρ_{22}, (d) ρ_{11} and the low-frequency coherence ρ_{32} (panels (e) and (f)) calculated at a fixed doublet splitting $\omega_{32} = 5\tau_R^{-1}$. The values of the LFC (in units of τ_R^{-1}) are given in (a). The initial conditions are $\rho_{22}(0) = \rho_{33}(0) = 0.5$ (total initial inversion), $\rho_{32}(0) = 0$ (no initial low-frequency coherence) and $R_{21}(0) = R_{31}(0) = 10^{-8}$.

in the previous subsection, i.e. the transitions $|2\rangle \to |1\rangle$ and $|3\rangle \to |1\rangle$ evolve synchronously and the doublet states remain equally populated after the SR pulse has been emitted: $\rho_{22}(\infty) = \rho_{33}(\infty) = 0.25$. For larger Δ_L, the transition $|2\rangle \to |1\rangle$ in fact does not evolve, conserving almost all the initial population in the state $|2\rangle$, while the population of the state $|3\rangle$ is entirely transferred to the ground state $|1\rangle$. This explains the changes which occur in the SR kinetics on increasing Δ_L: disappearance of the oscillatory structure and shortening both the SR pulse duration and delay time by approximately a factor of two. Indeed, as $|3\rangle \to |1\rangle$ is the only transition contributing to the emission, the problem is reduced to the two-level scheme with parameters $\tau_R' = 4Z(0)\tau_R = \tau_R$ and $t_D = -\tau_R' \ln R_{31}(0) \approx 17\tau_R$, which are characteristic for the two-level SR. It is worth mentioning that the suppression of the transition $|2\rangle \to |1\rangle$ occurs at $d_{21} = d_{31}$, i.e. under the condition of equivalent coupling of the individual transitions to the field.

The physics of such a behaviour is as follows. A V atom represents two transitions coupled to each other by the common field which includes the emission term ($\sim\tau_R^{-1}$) and the LFC ($\sim\Delta_L$) (see equation (14)). In the presence of population inversion, the amplitudes of optical oscillations, R_{21} and R_{31}, grow in time. Their increments are equal to each other in the absence of the LFC and under the condition $\mu_{13} = \mu_{12}$ and $\rho_{33}(0) = \rho_{22}(0) = 0.5$. However, they become different after appearing in the LFC (see below). This makes the transition $|3\rangle \to |1\rangle$ to evolve faster than $|2\rangle \to |1\rangle$. The final state of the system depends on the relationship between R_{31} and R_{21} at those times when the SR pulse is already well developed, i.e. at $t \approx t_D$. If $|R_{21}(t_D)| \approx |R_{31}(t_D)|$ then both transitions still evolve synchronously, while at $|R_{21}(t_D)| \ll |R_{31}(t_D)|$ the initial population of level $|3\rangle$ (0.5 in our case) is transferred to the level $|1\rangle$ before the oscillations $|2\rangle \to |1\rangle$ begin to build up. It makes the population inversion between levels $|2\rangle$ and $|1\rangle$ equal to zero and thus prevents the superradiant evolution of this channel which explains the disappearance of the oscillations of the SR pulse.

An analysis of the linear stage of the emission, i.e. keeping all the quantities equal to their initial values, except for R_{21} and R_{31}, provides a solid support for the above arguments. Equations (8)–(13), linearized with respect to R_{21} and R_{31} and adapted to the conditions used in the numerical simulations ($\mu_{31} = \mu_{21} = 1$, $\rho_{33}(0) = \rho_{22}(0)$, $\rho_{11}(0) = \rho_{32}(0) = 0$), have the form

$$\dot{R}_{31} = \left[-\mathrm{i}\frac{\omega_{32}}{2} + \left(\frac{1}{\tau_R} - \mathrm{i}\Delta_L\right)W\right]R_{31} + \left(\frac{1}{\tau_R} - \mathrm{i}\Delta_L\right)WR_{21}, \tag{39}$$

$$\dot{R}_{21} = \left[\mathrm{i}\frac{\omega_{32}}{2} + \left(\frac{1}{\tau_R} - \mathrm{i}\Delta_L\right)W\right]R_{21} + \left(\frac{1}{\tau_R} - \mathrm{i}\Delta_L\right)WR_{31}, \tag{40}$$

where $W \equiv \rho_{33}(0) - \rho_{11}(0) = \rho_{22}(0) - \rho_{11}(0)$. Solving these coupled equations is straightforward. Below, we write down the solution in the limit $\tau_R^{-1}, \Delta_L \ll \omega_{32}$:

$$R_{21} \simeq R_0 \mathrm{e}^{\lambda_1 t}, \qquad R_{31} \simeq R_0 \mathrm{e}^{\lambda_2 t}, \tag{41}$$

where

$$\lambda_{1,2} = \mathrm{i}\left(\pm\frac{\omega_{32}}{2} - \Delta_L W\right) + \frac{W}{\tau_R}\left(1 \mp 2W\frac{\Delta_L}{\omega_{32}}\right). \tag{42}$$

As seen from equation (42), the increment of R_{31} is indeed larger than that for R_{21}, i.e. R_{31} grows faster than R_{21}. Thus, during the linear stage of the SR

$$\frac{|R_{31}|}{|R_{21}|} = \exp\left(4W^2\frac{\Delta_L}{\omega_{32}}\frac{t}{\tau_R}\right). \tag{43}$$

Recall that the linear solutions for R_{31} and R_{21} are valid almost up to the SR pulse maximum (see, for instance, [44]). Then, applying this formula for $t = t_D$ and equating the exponent to unity, one obtains an estimate for Δ_L^c:

$$\Delta_L^c = \frac{\omega_{32}}{4W^2}\frac{\tau_R}{t_D}, \tag{44}$$

which separate two regimes of the SR. At $\Delta_L < \Delta_L^c$, both transitions evolve synchronously, while for the opposite sign of the inequality, the transition $|2\rangle \to |1\rangle$ is blocked for the reasons discussed above.

Concerning numerical data ($\omega_{32} = 5\tau_R^{-1}$, $W = 0.5$ and $t_D \approx 35\tau_R$), equation (44) yields $\Delta_L^c = \omega_{32}/35 = (1/7)\tau_R^{-1}$. This estimate is in good agreement with the numerical data (see figure 4). We stress that the estimated value of Δ_L^c is smaller than the half-width of the SR spectrum given by τ_R^{-1}. In other words, even if the LFC (the dynamical resonance frequency

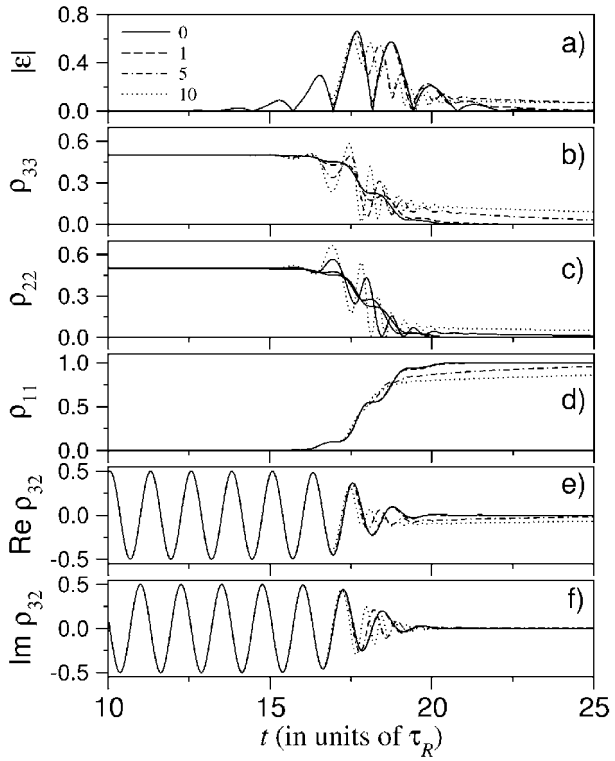

Figure 5. Same as in figure 4, except for $\rho_{32}(0) = 0.5$.

shift) are spectroscopically hidden due to the natural pulse broadening, it drastically affects the SR kinetics.

It is to be noticed that the above behaviour of the SR of a V system resembles the peculiarity of the SR for the Λ arrangement of nondegenerated levels [14–16, 44], namely in the presence of the LFC, all the population from the upper level of the Λ system is transferred to the lower level of the doublet, while the higher doublet level remains unpopulated after the SR pulse has gone.

4.2.2. SR from the pure state ($\rho_{32}(0) = 0.5$). Figure 5 shows the effects of the LFC on the SR kinetics obtained for $\rho_{22}(0) = \rho_{33}(0) = \rho_{32}(0) = 0.5$ or, in other words, when initially the pure bright state $|+\rangle$ is fully populated, $\rho_{++}(0) = 1$, while $\rho_{11}(0) = \rho_{--}(0) = \rho_{+-}(0) = 0$. As is seen from figure 5(a), the present case differs noticeably from the one where initially no low-frequency coherence is created ($\rho_{32}(0) = 0$, see the previous section). First of all, the LFC does not affect the SR delay time at all. Therefore, the linear stage of the SR is not useful here in predicting the changes in the SR kinetics, as they occur when the nonlinearity is already well developed.

The changes concern the second half and final stage of the pulse. The pulse shows an oscillatory structure which now cannot be associated with ω_{32} oscillations. The frequency of the oscillations grows upon increasing Δ_L and, in fact, reflects the magnitude of the latter. The populations of the doublet states also undergo antiphased (with respect to each other) oscillations at the same frequency as the pulse does. This indicates that the doublet states start to exchange the population when the SR pulse is developed. A qualitative interpretation of this effect is as follows. Recall that the LFC shifts the frequency of the transitions $|2\rangle \to |1\rangle$ and $|3\rangle \to |1\rangle$ by $\Delta_L(\rho_{22} - \rho_{11})$ and $\Delta_L(\rho_{33} - \rho_{11})$, respectively. Initially, these shifts are equal to each other. Figure 5(b) shows that the transition $|3\rangle \to |1\rangle$ starts to develop first. This reduces the initial detuning, ω_{32}, between the transitions. As a result, the radiation, which is emitted via the transition $|3\rangle \to |1\rangle$, is absorbed by the transition $|2\rangle \to |1\rangle$. It further reduces the detuning, stronger for larger Δ_L. After that, the transition $|2\rangle \to |1\rangle$ starts to emit while $|3\rangle \to |1\rangle$ starts to absorb, i.e. the transitions exchange their role.

The initial population is mostly transferred to the ground state, as it takes place for the same initial conditions at $\Delta_L = 0$. However, a small part of the population remains trapped in the dark state $|-\rangle$, unlike the case of $\Delta_L = 0$. This is the reason why the SR pulse has a long tail.

5. Summary and concluding remarks

We studied theoretically the SR from an ultrathin film of V-type atoms, taking into account the LFC to the average Maxwell field. We show that the interplay between the doublet splitting, low-frequency coherence (within the subspace of the doublet states) and LFC may significantly affect the scenario of SR. Several conclusion can be drawn from our results:

(i) Under the condition of degeneracy, the three-level problem is equivalent to that for a two-level system with a renormalized SR time. The role of the LFC is also similar to that for the two-level problem and manifests itself as a phase modulation of the SR pulse.

(ii) For a nondegenerated V system, the LFC correction affects drastically the SR scenario, allowing development of one of the transitions and blocking the other one under specific conditions. This effect may occur even if the LFC is small compared to the SR spectrum, i.e. when the LFC is spectroscopically hidden.

(iii) The SR scenario is sensitive to the amount of low-frequency coherence (within the subspace of the doublet states) as well as to the magnitude of the total inversion, thus providing a way to control the SR regimes.

To conclude, we discuss the conditions required to prove experimentally the predicted regimes of the V-type SR. First of all, one should look for a dense ensemble of radiators where the LFC is large compared to the linewidth. In [31], O_2^- ions in $KCl : O_2^-$ crystals and bound I_2 excitons at donor sites in CdS single crystals were considered suitable for observing the LFC effects. In relation to our model, one should bear in mind that, in disordered ensembles of dipole radiators, like the case of O_2^- centres and bound I_2 excitons, the LFC fluctuates. The average LFC drives the level shifts, while the fluctuating part contributes to the dipole–dipole line broadening. It turns out that both shift and linewidth are of the same order of magnitude [53], as it takes place in dense gas systems [30]. Because of this fact, these systems can hardly present the effects we are discussing.

Thin films of some organic compounds, such as naphthalene and anthracene, as well as materials containing unoccupied d or f orbitals, such as Cr_2O_3 or MnO_2, might be promising materials for this task. As they are crystalline, the intermolecular dipole–dipole interaction (and,

subsequently, the LFC) does not fluctuate. Furthermore, at low temperatures, the optical excitations in these materials are Frenkel excitons [45–47]. This fact implies that the intermolecular dipole–dipole interaction (the LFC, in other words) dominates over dephasing. As the density of the optically active units in crystals is generally high ($N_0 \sim 10^{21} - 10^{22}$ cm^{-3}), the SR time constant τ_R may be small compared to the dephasing time. Indeed, $\tau_R = \hbar\lambda_c/(2\pi)^2 d^2 N_0 L = (8\pi/3)(N_0\lambda_c^3)^{-1}(\lambda_c/L)\tau_0$, where $\lambda_c = 2\pi/k_c$ and $\tau_0 = 3\hbar/4d^2k_c^3$ is the spontaneous emission time of a single emitter. Let us take $\lambda_c = 5 \times 10^{-5}$ cm and assume that the transitions are dipole allowed ($\tau_0 \sim 10^{-8}$ s), which is typically the case for organic crystals. Then, for a film thickness $L = 0.1\lambda_c$ we estimate τ_R as being of the order of 10 fs. The exciton absorption linewidth is typically about a few hundreds of cm^{-1}. This gives 1 ps as an estimate for the dephasing time, that is, 100 times longer than τ_R. On the other hand, vibronic structure of aromatic crystals seems suitable for forming a V-configured system. The SR of high density Frenkel excitons was observed in single organic crystals of R-phycoerythrin molecules at room temperature [54]. The SR pulse was found to be phase-modulated, thus indicating the relevance of the LFC. Therefore, R-phycoerythrin single crystals are promising candidates to prove the effects predicted in this work.

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