PHYSICAL REVIEW A VOLUME 18, NUMBER 4 OCTOBER 1978

Cooperative behavior among three-level systems: Transient effects of coherent optical pumping

C. M. Bowden
Missile Research Directorate, High Energy Laser and Research Laboratory, US Army Missile Research and Development Command, Redstone Arsenal, Alabama 35809

C. C. Sung
Physics Department, University of Alabama in Huntsville, Huntsville, Alabama 35807
(Received 18 May 1978)

The dynamical evolution of an excited-state population in a macroscopic volume of three-level molecules is considered where the population is driven optically between the initial ground state and the excited states by an externally applied (c-number) coherent pump. The intermediate and ground states are taken as nonradiatively coupled whereas the excited-to-intermediate-state transition is coupled to the radiation field which is treated quantum mechanically. It is found that stimulated Raman processes can significantly influence the dynamical evolution of population inversion and macroscopic polarization by producing coherence effects among the populations. We examine the evolution of collective relaxation between the excited and intermediate levels in the time regime of the pump pulse duration. It is shown, using the mean-field approximation, that for uniform pumping and conditions such that the pump Rabi rate ω_R, the characteristic collective radiation time τ_R, and the pump pulse duration τ_P satisfy the inequalities, $\omega_R > 1/\tau_R > \gamma$ and $\tau_P \gtrsim \tau_R$ (γ is a characteristic internal dephasing rate for the molecules), the system is left in a state with classical transverse polarization when the pump pulse terminates. The system can evolve collectively from such a state only as superradiant (classical) evolution. For superfluorescent evolution which requires a state of preparation of complete inversion (zero transverse polarization), it is shown that the pump pulse must be effectively of area π and that $\tau_P < \tau_R$. Our results show that when the former conditions are satisfied the delay time τ_D for collective free pulse evolution is a function of both τ_P and τ_R. It is shown further that the pump pulse shape as well as temporal duration significantly affect the final state of preparation. The results of this work are interpreted in connection with recent reported results of experiments in superfluorescence and superradiance.

I. INTRODUCTION

Since Dicke's initial work[1] in which he showed that under certain conditions, a collection of two-level atoms, coupled only via their mutual radiation field, can evolve collectively from an initial state of inversion, there have been many theoretical treatments of various aspects and ramifications of the process which has come to be commonly known as Dicke superradiance.[2-6] Of particular note is the more recent work of Bonifacio and Lugiato[5] in which they draw the distinction between two kinds of cooperative relaxation processes in a macroscopic volume of two-level atoms. These two processes derive their distinction from the different initial conditions from which a system of two-level atoms can evolve collectively. In one case the system evolves from an initial state of complete inversion, i.e., zero initial macroscopic transverse dipole moment; in the other case the initial state is one of nonzero, though possibly small, macroscopic dipole moment. The first case exhibits the distinctly quantum-mechanical aspects of collective spontaneous relaxation and is predicted to manifest large quantum temporal fluctuations[3] and collective as well as individual-atom radiation reaction and frequency shifts.[3,7] This process has come to be known as superfluorescence. The second case can be adequately described in semiclassical terms and quantum fluctuations in the collective evolution are entirely negligible. The latter process is widely termed superradiance as distinct from superfluorescence.

The details of the dynamics of the free relaxation of a collection of excited atoms obviously strongly depend upon the initial state from which it evolves. Therefore, the preparation of the initial state, i.e., the pumping process itself, is an important aspect in any physical situation. An incoherent pump can invert the Boltzmann distribution in a collection of two-level atoms, but due to extremely small relative dispersion in the inversion, leave the system effectively completely inverted as far as subsequent collective evolution is concerned.[3] On the other hand, a π pulse may be applied to also leave the system of atoms in a state of complete inversion from which it undergoes free evolution. The first case is an example of incoherent pumping and the second is an example of coherent pumping on a collection of two-level atoms to induce an effective state of complete inversion. In either case, the system can undergo individual-atom spontaneous relaxation (fluorescence) stimulated spontaneous decay, or collective spontaneous relaxation (superfluorescence), depending upon other conditions.[5,6,8-10]

If the pumping is with respect to three levels rather than two, the effects can be quite different. For instance, an incoherent pump between the ground and third level of sufficient intensity can produce an effective state of complete inversion if the ambient temperature is sufficiently low and if the pump duration is not too long.[6] A coherent pump on the other hand can induce a coherent superposition of the original eigenstates of the three-level system and thus produce correlation in the population among the various levels. In particular, when a coherent pumping field is tuned to resonance between the ground and third levels, Raman transitions can transfer population coherently back and forth between the intermediate and ground states, even if they are not radiatively coupled. The time-dependent gain will in this case show oscillatory behavior at the Rabi frequency of the pump and coherence effects will be evident in the preparation of the system. One such effect of coherent pumping is to leave the system in a state of macroscopic, albeit small, transverse polarization, from which it evolves freely in a purely classical manner. It can be anticipated that coherence effects in the evolution of gain imparted by a coherent pumping field will be important when the pump Rabi frequency ω_R is large compared to some internal atomic decay rate γ, i.e., when $\omega_R/\gamma \gg 1$, and when the pump pulse duration τ_P satisfies the condition $\tau_P > \omega_R^{-1}$.

Predominantly, the experiments which have been reported to date which have been interpreted on the basis of superradiance of superfluorescence have used coherent pumping schemes.[11-17] In particular, with regard to the experiments of Gibbs[15] and Vrehen,[16] a dye laser was used to excite Cs vapor and the results have been interpreted on the basis of single-pulse and oscillatory superfluorescence[5,8] using analysis[8] based upon the two-level mean-field model for superfluorescence of Bonifacio and Lugiato.[5] In these cases an initial state of incoherent preparation, i.e., complete inversion, is assumed. This assumption leads to the determination of an initial value of the tipping angle of the Bloch vector in the mean-field model which is absolutely crucial to the analysis and comparison with experimental results. This assumption is commensurate with that of an incoherent pump preparation. However, there is ample evidence of coherence effects induced by coherent pumping in the same system[14,15] in the form of quantum beats[18] which indicate correlation in the population distribution among the various energy levels of the atoms.

This leaves open the question as to whether the assumption of incoherent preparation within an equivalent two-level manifold is valid at all in analysis of such experiments. It is the purpose of this paper to examine in detail some of the aspects of coherent optical pumping on multilevel systems and the effects on subsequent collective atomic relaxation and radiation pulse generation.

In Sec. II, we develop the Hamiltonian formulation of the problem for coherent optical pumping of a collection of three-level atoms in a macroscopic volume. Section III will be devoted to a discussion of collective- and individual-atom renormalizations, including Raman contributions which arise under a perturbative analysis of the dynamical evolution of the system. Then, in Sec. IV, we develop mean-field results based upon the model of Sec. II and compare our results with the two-level-atom model of Bonifacio and Lugiato[5] and with results of recent experiments.[13-17] The last section will be concerned with a summary of the work reported in this paper and the implications to previous theoretical work and recent experiments.

II. MODEL HAMILTONIAN

The model we use is that of a collection of N three-level atoms uniformly distributed in a macroscopic volume $\mathcal{V}$. The energy-level scheme is shown in Fig. 1. The frequency ω_0 is that of an externally applied coherent (classical) pumping field which we shall treat as a c-number and specify *apriori*. The transition from the third to the intermediate level is coupled to the radiation field treated quantum mechanically, whereas

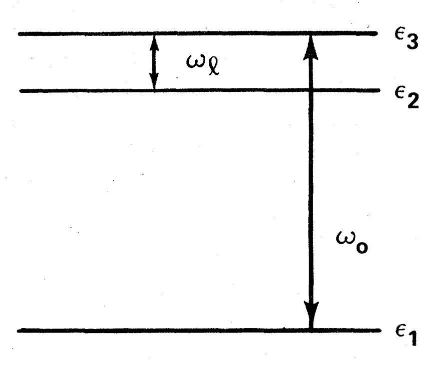

FIG. 1. Energy-level scheme for the three-level system. Here, ω_0 is the carrier frequency of the laser which couples levels 1 and 3. The transitions between levels 3 and 2 are coupled to the radiation field in the dipole approximation, whereas the transition between levels 2 and 1 is not directly radiatively coupled. Spontaneous relaxation between levels 3 and 1 is neglected.

levels 2 and 1 are not radiatively coupled. This level scheme is consistent, for instance, with requirements for rotational selection rules for pump induced *V-V* transitions with subsequent radiative transitions between rotational levels in the excited vibrational band for an optically pumped molecular species.[11,13,17] For the treatment here we ignore spontaneous relaxation between the third level and the ground state.

The Hamiltonian $\mathcal{H}$ which describes this system is the following:

$$\mathcal{H}=\sum_{k=1}^{3}\sum_{j=1}^{N}\epsilon_{kj}E_{k,k}^{(j)}+\sum_{l=1}^{\infty}\omega_l b_l^{+}b_l +\sum_{j=1}^{N}\sum_{l=1}^{\infty}g_l^{(j)}\left(E_{2,3}^{(j)}+E_{3,2}^{(j)}\right)\left(b_l e^{i\vec{k}_l\cdot\vec{x}_j}+b_l^{+}e^{-i\vec{k}_l\cdot\vec{x}_j}\right) +\frac{1}{2}\sum_{j=1}^{N}\left[\omega_R^{(j)}E_{3,1}^{(j)}e^{-i(\omega_0 t-\vec{k}_0\cdot\vec{x}_j)} +\omega_R^{(j)*}E_{1,3}^{(j)}e^{i(\omega_0 t-\vec{k}_0\cdot\vec{x}_j)}\right], \tag{2.1}$$

where we have used units where $\hbar=1$, $c=1$.

The first and second terms in Eq. (1) describe the collection of free atoms and the free electromagnetic field, respectively. The third term gives the interaction between the atoms and the electromagnetic field, in the dipole approximation, coupled to the transition between the upper pair of levels, Fig. 1. The last term gives the interaction between the externally applied classical electromagnetic field and the atoms. Here $\omega_R^{(j)}$ represents the electromagnetic field envelope of carrier frequency and wave vector ω_0 and k_0, respectively, at the *j*th atomic site [see (2.5)]. For convenience, we take each atom as located at a lattice site of a cubic lattice. This restriction will be relaxed later on and our results do not depend upon this requirement. The field operators obey Bose commutation relations,

$$[b_l, b_k^{\dagger}]=\delta_{l,k}\,. \tag{2.2}$$

The complete set of atomic operators describing transitions between states j and k, the $E_{jk}^{(m)}$ $(j,k=1,2,3)(m=1,2,\ldots,N)$ obey commutation relations isomorphic to those of the Lie algebra $u(3)$,[19-21]

$$[E_{ij}^{(m)},E_{lk}^{(n)}]=E_{ik}^{(m)}\delta_{lj}\delta_{m,n}-E_{lj}^{(m)}\delta_{k}\delta_{m,n}\,. \tag{2.3}$$

The coupling coefficients g_l are given by

$$g_l^{(j)}=u_{32}^{(j)}\omega_{32}(2\pi/\omega_l V)^{1/2}, \tag{2.4}$$

where V is the volume of quantization for the electromagnetic field and $u_{32}^{(j)}$ is the dipole-moment matrix element of the transition. We shall take V much larger than the volume occupied by the atoms and eventually take this volume to infinity. The c-number factor $\omega_R^{(j)}$ is the Rabi frequency for the externally applied pump field envelope $E^{(j)}(t)$ at the atomic site located at x_j, where

$$\omega_R^{(j)}=u_{13}^{(j)}E^{(j)}(t)\,. \tag{2.5}$$

For simplicity of discussion, we have limited the model to only one degree of polarization.

It is very simple to show that the Lie algebra $u(3)$, Eq. (2.3) has the realization in terms of bilinear combinations of Bose creation and annihilation operators[20,21]

$$E_{ij}^{(m)}=a_i^{(m)\dagger}a_j^{(m)}, \tag{2.6}$$

$$[a_i^{(m)},a_j^{(n)\dagger}]=\delta_{ij}\delta_{m,n}\,. \tag{2.7}$$

With the substitution of (2.6) into (2.1), the Hamiltonian $\mathcal{H}$ takes the form

$$\mathcal{H}=\sum_{k=1}^{3}\sum_{j=1}^{N}\epsilon_{kj}a_k^{(j)\dagger}a_k^{(j)}+\sum_{l=1}^{\infty}\omega_l b_l^{\dagger}b_l+\sum_{j=1}^{N}\sum_{l=1}^{\infty}g_l^{(j)}\left(a_2^{(j)\dagger}a_3^{(j)}+a_3^{(j)\dagger}a_2^{(j)}\right)\left(b_l e^{i\vec{k}_l\cdot\vec{x}_j}+b_l^{+}e^{-i\vec{k}_l\cdot\vec{x}_j}\right) +\frac{1}{2}\sum_{j=1}^{N}\left[\omega_R^{(j)}a_3^{(j)\dagger}a_1^{(j)}e^{-i(\omega_0 t-\vec{k}_0\cdot\vec{x}_j)}+\omega_R^{(j)*}a_1^{(j)\dagger}a_3^{(j)}e^{i(\omega_0 t-\vec{k}_0\cdot\vec{x}_j)}\right]. \tag{2.8}$$

For simplicity, we take the energy-level structure of the individual atoms as identical in the absence of any fields; and, without loss of generality, we take $\epsilon_{1j}=0$ $(j=1,2,\ldots,N)$. Furthermore, it is useful to define collective operators,

$$R_{32}(\vec{\alpha})=\sum_{j=1}^{N}a_3^{(j)\dagger}a_2^{(j)}e^{i\vec{\alpha}\cdot\vec{x}_j}, \tag{2.9a}$$

$$R_{31}(\vec{\alpha})=\sum_{j=1}^{N}a_3^{(j)\dagger}a_1^{(j)}e^{i\vec{\alpha}\cdot\vec{x}_j}, \tag{2.9b}$$

$$R_{21}(\vec{\alpha})=\sum_{j=1}^{N}a_2^{(j)\dagger}a_1^{(j)}e^{i\vec{\alpha}\cdot\vec{x}_j}, \tag{2.9c}$$

$$R_{33}=\sum_{j=1}^{N}a_3^{(j)\dagger}a_3^{(j)}, \tag{2.9d}$$

$$R_{22}=\sum_{j=1}^{N} a_2^{(j)\dagger} a_2^{(j)}, \tag{2.9e}$$

$$R_{11}=\sum_{j=1}^{N} a_1^{(j)\dagger} a_1^{(j)}. \tag{2.9f}$$

where $\alpha_i=(2\pi/L_i)\eta_i, \eta_i=0,1,2,\ldots,N-1,\ i=x,y,z,$ and $V=\Pi_i L_i$. Making use of the orthogonality relations,

$$\frac{1}{\sqrt{N}}\sum_{j=1}^{N} e^{i(\vec{\alpha}-\vec{\alpha}')\cdot\vec{x}_j}=\delta_{\vec{\alpha},\vec{\alpha}'}, \tag{2.10a}$$

$$\frac{1}{N}\sum_{\vec{\alpha}} e^{i\vec{\alpha}\cdot(\vec{x}_i-\vec{x}_j)}=\delta_{ij} \tag{2.10b}$$

the reciprocal relations are obtained,

$$a_p^{(j)\dagger} a_q^{(j)}=\frac{1}{N}\sum_{\vec{\alpha}} R_{pq}(\vec{\alpha}) e^{i\vec{\alpha}\cdot\vec{x}_j}, \quad p>q. \tag{2.11}$$

If the substitutions indicated by (2.9) and (2.11) are made in (2.8), the result is

$$\mathcal{H}=\omega_{31}R_{33}+\omega_{21}R_{22}+\sum_l \omega_l b_l^\dagger b_l+\sum_{l=1}^{\infty}\sum_{\vec{\alpha}} g_l\left[R_{32}(\vec{\alpha}) b_l f(\vec{k}_l-\vec{\alpha})+R_{23}(\vec{\alpha}) b_l^\dagger f^*(\vec{k}_l-\vec{\alpha})\right]$$
$$+\sum_{l=1}^{\infty}\sum_{\vec{\alpha}} g_l\left[R_{32}(\vec{\alpha}) b_l^\dagger f(\vec{k}_l+\vec{\alpha})+R_{23}(\vec{\alpha}) b_l f^*(\vec{k}_l+\vec{\alpha})\right]+\tfrac{1}{2}\sum_{\vec{\alpha}}\left[R_{31}(\vec{\alpha}) F(\vec{k}_0-\vec{\alpha},t) e^{-i\omega_0 t}+R_{13}(\vec{\alpha}) F^*(\vec{k}_0-\vec{\alpha},t) e^{i\omega_0 t}\right]. \tag{2.12}$$

Here,

$$f(\vec{\eta})=\frac{1}{N}\sum_{j=1}^{N} e^{i\vec{\eta}\cdot\vec{x}_j}=\langle e^{i\vec{\eta}\cdot\vec{x}_j}\rangle \tag{2.13}$$

$$F(\eta,t)=\frac{1}{N}\sum_{j=1}^{N}\omega_R^{(j)}(t) e^{i\vec{\eta}\cdot\vec{x}_j}=\langle\omega_R^{(j)}(t) e^{i\vec{\eta}\cdot\vec{x}_j}\rangle. \tag{2.14}$$

If the pump field is uniform throughout the volume containing the molecules, i.e., for uniform transverse pumping on a pencil-like medium, then $\omega_R^{(j)}(t)$ is independent of location of the jth molecule. In such case

$$F(\eta,t)\approx\omega_R(t) f(\eta), \tag{2.15}$$

where $f(\eta)$ is the product of diffraction functions in each of the three dimensions.[5] If, on the other hand, the pumping pulse propagates along the major axis of symmetry of a long rectangular medium, to be precise, we would have to couple the present model to Maxwell's equations for the excitation pulse. However, if we assume that the pumping pulse is either sustained externally or is saturating in the medium such that negligible percentage of the pump pulse energy is actually absorbed by the molecules, then as far as the dynamics of the system are concerned, the pump is specified *apriori.*

If we take the origin of coordinates at the center of a long rectangular volume containing the active material and if we assume a plane wave for the pump pulse, (2.14) becomes[5]

$$F(\eta,t)=\frac{\sin(\frac{1}{2}\eta_x L_x)}{\frac{1}{2}\eta_x L_x}\,\frac{\sin(\frac{1}{2}\eta_y L_y)}{\frac{1}{2}\eta_y L_y}\times\langle\omega_R^{(j)}(t) e^{i\eta_z x_z}\rangle, \tag{2.16}$$

where L_x, L_y, and L_z are the linear dimensions of material volume.

Qualitative results can be obtained if we assume a step-function envelope for the pumping pulse and neglect transient effects arising from the pulse entry and exit from the active volume. This confines the analysis to pulses much shorter than the length of the medium and ones that are much longer. If we take the direction of propagation $x_3=z$, and a plane-wave pump pulse specified by

$$\begin{aligned}&\omega_R(t)=0, \quad z<z_i\\&\omega_R(t)=\overline{\omega}_R, \quad z_i\leq z\leq z_f\\&\omega_R(t)=0, \quad z>z_f\\&c\tau_p'\equiv z_f-z_i,\end{aligned} \tag{2.17}$$

where $c\tau_p'$ is the length of the pulse of amplitude given by $\overline{\omega}_R$ in the medium; then the last factor in (2.16) becomes

$$\langle\omega_R^{(j)}(t) e^{i\eta_z x_z}\rangle=\frac{\overline{\omega}_R}{L_z}\int_{z_i}^{z_f} dz'\, e^{i\eta_z z'}. \tag{2.18}$$

This factor when used in (2.12) and (2.14) has the argument $\eta_z=k_0-\alpha_z$. The terms of the form (2.16) with (2.18) give maximum contribution to the summation in the last term in (2.12) for η_x, η_y, $\eta_z \to 0$. Then, neglecting transient effects, (2.18) can be evaluated explicitly to give

$$F(\vec{k}_0 - \vec{\alpha}, t) \to \overline{\omega}_R \tau_p' c/L_z . \qquad (2.19)$$

If the pump pulse is longer than the medium, and remembering that $c\tau_p'$ is the length of the pulse in the medium, (2.19) becomes

$$F(\vec{k}_0 - \vec{\alpha}, t) \approx \overline{\omega}_R, \quad \tau_p' > L_z/c . \qquad (2.20)$$

As expected, this is equivalent to the result for uniform (transverse) pumping, (2.15) (for $f(\vec{k}_0 - \vec{\alpha}) = 1$, $\vec{\alpha} = k_0$, and zero otherwise).

If, instead, the pump pulse duration is short compared to the length of the medium, and again, neglecting transient effects, (2.19) can be written as

$$F(\vec{k}_0 - \vec{\alpha}, t) \approx \theta_p \gamma_E , \quad \tau_p' < L_z/c \qquad (2.21)$$

where

$$\theta_p \equiv \overline{\omega}_R \tau_p' \qquad (2.22)$$

is the pump pulse area, and

$$\gamma_E^{-1} \equiv L_z/c \qquad (2.23)$$

is the transit time for the pump field in the medium. Equation (2.21) is quite different in appearance than (2.20). However, both have the same physical interpretation as the rate at which energy is exchanged back and forth between the collective medium and the pumping field.

III. DRESSED REPRESENTATION: COLLECTIVE- AND SINGLE-ATOM RENORMALIZATIONS

It is useful to transform the Hamiltonian (2.12) by a unitary transformation which eliminates the explicit rapidly time-varying part which appears in the exponential factors in the last term. The transformation T which does this is

$$T = e^{-i\omega_0(R_{33}+R_{22})t} . \qquad (3.1)$$

Using the commutation relations, derived from (2.7) and (2.9),

$$[R_{ij}(\alpha), R_{km}(\alpha')] = R_{im}(\Lambda)\delta_{kj} - R_{kj}(\Lambda)\delta_{im}, \qquad (3.2)$$

where Λ is given by

$$\Lambda = \alpha \mathcal{S}(i,j) + \alpha' \mathcal{S}(k,m) , \qquad (3.3)$$

$$\mathcal{S}(p,q) = \begin{cases} 1 & p \geq q \\ -1 & p < q , \end{cases} \qquad (3.4)$$

the transformed Hamiltonian $\mathcal{H}' = T^{-1}\mathcal{H}T$ is

$$\mathcal{H}' = \Omega R_{33} + \omega R_{22} + \sum_l \omega_l b_l^\dagger b_l + \sum_{l=1}^{\infty}\sum_{\vec{\alpha}} g_l [R_{32}(\vec{\alpha}) b_l f(\vec{k}_l - \vec{\alpha}) + R_{23}(\vec{\alpha}) b_l^\dagger f^*(\vec{k}_l - \vec{\alpha})]$$
$$+ \sum_{l=1}^{\infty}\sum_{\vec{\alpha}} g_l [R_{23}(\vec{\alpha}) b_l f(\vec{k}_l + \vec{\alpha}) + R_{32}(\vec{\alpha}) b_l^\dagger f^*(\vec{k}_l + \vec{\alpha})] + \tfrac{1}{2}\sum_{\vec{\alpha}}[R_{31}(\vec{\alpha}) F(\vec{k}_0 - \vec{\alpha}, t) + R_{13}(\vec{\alpha}) F^*(\vec{k}_0 - \vec{\alpha}, t)] . \qquad (3.5)$$

Here,

$$\Omega = \omega_{31} - \omega_0 , \quad \omega = \omega_{21} - \omega_0 . \qquad (3.6)$$

The set of states corresponding to the basis in which

$$\mathcal{H}_0 = \Omega R_{33} + \omega R_{22} + \sum_l \omega_l b_l^\dagger b_l \qquad (3.7)$$

is diagonal, we call the bare states of (3.5). The renormalizations which come about through the dynamical evolution of the system described by (3.5) can be obtained by transforming $\mathcal{H}'$ to the unitary representation for which the original bare states are the eigenstates of the time-independent transformed Hamiltonian.[22] For the case here, in connection with (3.5), the Hamiltonian is explicitly time dependent through the factors $F(t)$. The perturbation procedure developed by Coulter[22] for the transformation to the dressed representation is for a Hamiltonian explicitly independent of the time. In order to apply Coulter's method to our case here we have modified his procedure to account for the explicit time dependence of the Hamiltonian to be transformed, and the development is given in the Appendix.

As shown in the Appendix, the dressed Hamiltonian $\mathcal{H}''$ which is unitarily equivalent to $\mathcal{H}'$ through second order in the atom-field coupling is given by the sum of five terms,

$$\mathcal{H}''(t) = \mathcal{H}_0 + D_1(t) + D_2(t) - [\dot{k}_1(t) + \dot{k}_2(t)] - \tfrac{1}{2} i [k_1(t), \dot{k}_1(t)] . \qquad (3.8)$$

Some simpliciation in the calculation is obtained by choosing $\Omega = 0$ in (3.5) and (3.7), i.e., the pumping field is taken to be precisely tuned to the pump transition.

From the Appendix, and for $\Omega = 0$, we have

$$D_1(t)=\sum_{\substack{\vec{\alpha}\\ \bar{l}\in\Delta}} g_{\bar{l}}\, f(\vec{k}_{\bar{l}}-\vec{\alpha})[R_{32}(\vec{\alpha})b_{\bar{l}}+R_{23}(\vec{\alpha})b_{\bar{l}}^{\dagger}]+\tfrac{1}{2}\sum_{\vec{\alpha}}[R_{31}(\vec{\alpha})F(\vec{k}_0-\vec{\alpha},t)+R_{13}(\vec{\alpha})F^*(\vec{k}_0-\vec{\alpha},t)], \tag{3.9}$$

$$D_2(t)=-\sum_{\substack{l\\ l\neq\bar{l}}}\sum_{\alpha,\alpha'}\frac{g_l^2 f(\vec{k}_l-\vec{\alpha})f(\vec{k}_l-\vec{\alpha}')}{\omega_l-\omega_{32}}\{b_l^{\dagger}b_l[R_{33}(\vec{\alpha}-\vec{\alpha}')-R_{22}(\vec{\alpha}-\vec{\alpha}')]+R_{32}(\vec{\alpha})R_{23}(\vec{\alpha}')\}$$

$$+\sum_{\substack{l\\ l\neq\bar{l}}}\frac{g_l^2 f(\vec{k}_l+\vec{\alpha})f(\vec{k}_l+\vec{\alpha}')}{(\omega_l+\omega_{32})}\{b_l^{\dagger}b_l[R_{33}(\vec{\alpha}-\vec{\alpha}')-R_{22}(\vec{\alpha}-\vec{\alpha}')]-R_{23}(\vec{\alpha})R_{32}(\vec{\alpha}')\}, \tag{3.10}$$

$$\dot{k}_1=0, \tag{3.11}$$

and

$$\dot{k}_2=i\sum_{\substack{l\\ l\neq\bar{l}}}\sum_{\alpha,\alpha'}\frac{g_l f(\vec{k}_l-\vec{\alpha})\dot{F}^*(\vec{k}_0-\vec{\alpha}',t)}{(\omega_l-\omega_{32})^2}b_l R_{12}(\vec{\alpha}-\vec{\alpha}')-i\sum_{\substack{l\\ l\neq\bar{l}}}\sum_{\vec{\alpha},\vec{\alpha}'}\frac{g_l f(\vec{k}_l-\vec{\alpha})\dot{F}(\vec{k}_0-\vec{\alpha}',t)}{(\omega_l-\omega_{32})^2}b_l^{\dagger}R_{21}(\vec{\alpha}'-\vec{\alpha})$$

$$-i\sum_{\substack{l\\ l\neq\bar{l}}}\sum_{\vec{\alpha},\vec{\alpha}'}\frac{g_l f(\vec{k}_l+\vec{\alpha})\dot{F}(\vec{k}_0-\vec{\alpha}',t)}{(\omega_l+\omega_{32})^2}b_l R_{21}(\vec{\alpha}'-\vec{\alpha})+i\sum_{\substack{l\\ l\neq\bar{l}}}\sum_{\vec{\alpha},\vec{\alpha}'}\frac{g_l f(\vec{k}_l+\vec{\alpha})\dot{F}^*(\vec{k}_0-\vec{\alpha}',t)}{(\omega_l+\omega_{32})^2}b_l^{\dagger}R_{12}(\vec{\alpha}-\vec{\alpha}'). \tag{3.12}$$

Here $\bar{l}\in\Delta$ restricts the first summation in (3.9) within a thin shell in k space centered at $|k| \approx\omega_{32}/c$ and within $\Delta=\delta\omega_{32}/c$.

The first term in (3.9) describes the exchange of energy back and forth between the atoms and the field. The second term is the pump driving term and drives the population between the first and third levels. It is to be noted that the effect of the pumping pulse in developing gain in the medium is limited by defraction through the function F, i.e., the material geometry, as well as the propagation and electromagnetic properties of the pump pulse in the atomic medium. A simple manifestation of this is given by comparing (2.20) and (2.21). These are the first-order contributions. The second-order contributions are given by D_2 and $\dot{k}_2$.

The first terms within the two curley brackets in (3.10) are easily recognized as ac Stark contributions with associated frequency renormalizations The second terms in these brackets do not depend upon photon number and together give rise to single-atom and collective-frequency renormalization contributions.[5,7] These have already been discussed elsewhere [see Eqs. (4)–(8) of Ref. 7]. If F is set equal to zero, D_1 and D_2 are identical with the terms obtained earlier for the system comprised of a collection of two-level atoms evolving from an excited state.[7]

The last term of the Hamiltonian, $\dot{k}_2$ given in (3.12), contains only pump-induced Raman terms which transfer population directly between the ground state and the intermediate level. These terms depend upon the time derivative of the slowly varying envelope of the electromagnetic field of the pump, i.e., $\dot{F}$ and $\dot{F}^*$. There exists also rapidly varying contributions to the Raman transitions at the rate ω_0, but the contributions do not appear here because of the transformation to the slowly varying operator representation, (3.1). The first two terms in (3.12) represent resonant, stimulated Raman transitions with associated frequency renormalization. Due to the difference denominators of these terms, their contributions can be at least as important as the collective frequency shifts which arise from (3.10). The appearance of the time derivative of the pump "diffraction function" F in these terms indicates that the shape of the pump pulse envelope can cause significant contributions to the Raman transitions, and hence, to population transfer from the ground to the intermediate level, even though they are not radiatively coupled. It is to be noted further that since the functions F carry the coherence of the pumping field, the process described by the terms in (3.12) are coherent, stimulated Raman transitions. The last two terms in (3.12) have nonresonant denominators and are therefore less significant than the first two terms. These terms describe nonenergy conserving Raman processes.

IV. MEAN-FIELD EQUATIONS: DYNAMICAL EVOLUTION OF GAIN

This section will be used to develop some important qualitative results of the dynamical evolution of gain induced by coherent pumping. We shall use the mean-field approach and assume uniform pumping or its equivalent, i.e., conditions (2.15) or (2.20).

To obtain the appropriate set of Heisenberg's equations of motion, we consider again the Hamiltonian (2.12) and transform to the interaction representation,

$$\mathcal{H}_I(t) = \sum_{l=1}^{\infty}\sum_{\vec{\alpha}} g_l[R_{32}(\vec{\alpha})b_l e^{i\sigma_l^{(-)}t} f(\vec{k}_l - \vec{\alpha}) + R_{23}(\vec{\alpha})b_l^\dagger e^{-i\sigma_l^{(-)}t} f^*(\vec{k}_l - \vec{\alpha})]$$
$$+ \sum_{l=1}^{\infty}\sum_{\vec{\alpha}} g_l[R_{32}(\vec{\alpha})b_l^\dagger e^{i\sigma_l^{(+)}t} f(\vec{k}_l + \vec{\alpha}) + R_{23}(\vec{\alpha})b_l e^{-i\sigma_l^{(+)}t} f^*(\vec{k}_l + \vec{\alpha})]$$
$$+ \tfrac{1}{2}\sum_{\vec{\alpha}}[R_{31}(\vec{\alpha})F(\vec{k}_0 - \vec{\alpha}, t)e^{i\Omega t} + R_{13}(\vec{\alpha})F^*(\vec{k}_0 - \vec{\alpha}, t)e^{-i\Omega t}]. \quad (4.1)$$

Here

$$\sigma_l^{(\pm)} = \omega_{32} \pm \omega_l, \quad \Omega = \omega_{31} - \omega_0. \quad (4.2)$$

In a manner similar to that of Ref. 5, it is convenient to define collective operators for the quantized internal radiation field,

$$A_0(\vec{\alpha}) \equiv \sum_l b_l e^{i\sigma_l^{(-)}t} f(\vec{k}_l - \vec{\alpha}),$$
$$A_0^\dagger(\vec{\alpha}) = \sum_l b_l^\dagger e^{-i\sigma_l^{(-)}t} f^*(\vec{k}_l - \vec{\alpha}). \quad (4.3)$$

The collective operators (4.3) satisfy the commutation relations,

$$[A_0(\vec{\alpha}), A_0^\dagger(\vec{\alpha}')] = \delta_{\vec{\alpha},\vec{\alpha}'}, \quad (4.4)$$

where we have used (2.2) and

$$f(\vec{k}_l - \vec{\alpha}) = \prod_{i=1}^{3} \operatorname{sinc}[\tfrac{1}{2}(\vec{k}_l - \vec{\alpha})_i L_i], \quad (4.5)$$

$$\sum_l f(\vec{k}_l - \vec{\alpha}) f^*(\vec{k}_l - \vec{\alpha}') = \delta_{\vec{\alpha},\vec{\alpha}'}. \quad (4.6)$$

If we drop the counterrotating terms in (4.1) and note that $g_l \to g_{\vec{\alpha}}$ in the first summation since $f(\vec{k}_l - \vec{\alpha})$, (4.5), becomes δ-function-like in the neighborhood of $\vec{k}_l \approx \vec{\alpha}$, then (4.1) becomes, using (4.3),

$$\mathcal{H}_I(t) = \sum_{\vec{\alpha}} g_{\vec{\alpha}}[R_{32}(\vec{\alpha})A_0(\vec{\alpha}) + R_{23}(\vec{\alpha})A_0^\dagger(\vec{\alpha})]$$
$$+ \tfrac{1}{2}\sum_{\vec{\alpha}}[R_{31}(\vec{\alpha})\mathfrak{F}(\vec{k}_0 - \vec{\alpha}, t) + R_{13}(\vec{\alpha})\mathfrak{F}^*(\vec{k}_0 - \vec{\alpha}, t)], \quad (4.7)$$

where we have used

$$\mathfrak{F}(\vec{k}_0 - \vec{\alpha}, t) \equiv F(k_0 - \vec{\alpha}, t)e^{i\Omega t}$$

in (4.7).

If we use (4.7) the Heisenberg equations of motion for the collective atomic and field operators are the following:

$$\dot{R}_{33} = -i\sum_{\vec{\alpha}} g_{\vec{\alpha}}[A_0(\vec{\alpha})R_{32}(\vec{\alpha}) - A_0^\dagger(\vec{\alpha})R_{23}(\vec{\alpha})]$$
$$- \tfrac{i}{2}\sum_{\vec{\alpha}}[R_{31}(\vec{\alpha})\mathfrak{F}(\vec{k}_0 - \vec{\alpha}, t) - R_{13}(\vec{\alpha})\mathfrak{F}^*(\vec{k}_0 - \vec{\alpha}, t)], \quad (4.8a)$$

$$\dot{R}_{22} = i\sum_{\vec{\alpha}} g_{\vec{\alpha}}[A_0(\vec{\alpha})R_{32}(\vec{\alpha}) - A_0^\dagger(\vec{\alpha})R_{23}(\vec{\alpha})], \quad (4.8b)$$

$$\dot{R}_{11} = \tfrac{i}{2}\sum_{\vec{\alpha}}[R_{31}(\vec{\alpha})\mathfrak{F}(\vec{k}_0 - \vec{\alpha}, t) - R_{13}(\vec{\alpha})\mathfrak{F}^*(\vec{k}_0 - \vec{\alpha}, t)], \quad (4.8c)$$

$$\dot{R}_{32}(\vec{\alpha}') = i\sum_{\vec{\alpha}} g_{\vec{\alpha}}A_0^\dagger(\vec{\alpha})[R_{22}(\vec{\alpha}' - \vec{\alpha}) - R_{33}(\vec{\alpha}' - \vec{\alpha})]$$
$$+ \tfrac{i}{2}\sum_{\vec{\alpha}} R_{12}(\vec{\alpha}' - \vec{\alpha})\mathfrak{F}^*(\vec{k}_0 - \vec{\alpha}, t), \quad (4.8d)$$

$$\dot{R}_{13}(\vec{\alpha}') = -i\sum_{\vec{\alpha}} g_{\vec{\alpha}}A_0(\vec{\alpha})R_{12}(\vec{\alpha}' - \vec{\alpha})$$
$$+ \tfrac{i}{2}\sum_{\vec{\alpha}}\mathfrak{F}(\vec{k}_0 - \vec{\alpha}, t)[R_{33}(\vec{\alpha} - \vec{\alpha}') - R_{11}(\vec{\alpha} - \vec{\alpha}')], \quad (4.8e)$$

$$\dot{R}_{12}(\vec{\alpha}') = -i\sum_{\vec{\alpha}} g_{\vec{\alpha}}A_0^\dagger(\vec{\alpha})R_{13}(\vec{\alpha} + \vec{\alpha}')$$
$$+ \tfrac{i}{2}\sum_{\vec{\alpha}} R_{32}(\vec{\alpha} - \vec{\alpha}')\mathfrak{F}(\vec{k}_0 - \vec{\alpha}, t), \quad (4.8f)$$

and

$$\dot{A}_0(\vec{\alpha}') = -ig_{\vec{\alpha}'}R_{23}(\vec{\alpha}') - \kappa A(\alpha'). \quad (4.8g)$$

In the last equation, (4.8g), we have added a phenomenological damping term to account for linear losses from the radiation field, and also we have taken [see (4.3) and (4.2)],

$$\frac{\partial A_0(\alpha')}{\partial t} = 0.$$

It is to be noted from (4.8) that the conservation relation

$$\dot{R}_{33}+\dot{R}_{22}+\dot{R}_{11}=0 \tag{4.9}$$

is satisfied identically.

If now we restrict to just two modes for the emission field, $\vec{\alpha}$ and $-\vec{\alpha}$, and the pump carrier mode k_0, and let $\Omega=0$ in (4.2) for simplicity, then for either case (2.15) or (2.20), the equations of motion (4.8) reduce to

$$\dot{R}_{33}=-\dot{R}_{22}-\dot{R}_{11}, \tag{4.10a}$$

$$\dot{R}_{22}=ig[A_0(\vec{\alpha})R_{32}(\vec{\alpha})+A_0(-\vec{\alpha})R_{32}(-\vec{\alpha}) -A_0^\dagger(\vec{\alpha})R_{23}(\vec{\alpha})-A^\dagger(-\vec{\alpha})R_{23}(-\vec{\alpha})], \tag{4.10b}$$

$$\dot{R}_{11}=i(\omega_R/2)[R_{31}(\vec{k}_0)-R_{13}(\vec{k}_0)], \tag{4.10c}$$

$$\dot{R}_{32}(\pm\vec{\alpha})=igA_0^\dagger(\pm\vec{\alpha})[R_{33}(\pm\vec{\alpha}-\vec{\alpha})-R_{22}(\pm\vec{\alpha}-\vec{\alpha})] -igA_0^\dagger(\mp\vec{\alpha})[R_{33}(\pm\vec{\alpha}+\vec{\alpha})-R_{22}(\pm\vec{\alpha}-\vec{\alpha})] +i(\omega_R/2)R_{12}(\vec{k}_0\mp\vec{\alpha}), \tag{4.10d}$$

$$\dot{R}_{12}(\vec{k}_0\mp\vec{\alpha})=-igA_0^\dagger(\vec{\alpha})R_{13}(\vec{k}_0+\vec{\alpha}\mp\vec{\alpha}) -igA_0^\dagger(-\vec{\alpha})R_{13}(\vec{k}_0-\vec{\alpha}\mp\vec{\alpha}) +i(\omega_R/2)R_{32}(\pm\vec{\alpha}), \tag{4.10e}$$

$$\dot{R}_{13}(\vec{k}_0)=igA_0(\vec{\alpha})R_{12}(\vec{k}_0-\vec{\alpha})-igA_0(-\vec{\alpha})R_{12}(\vec{k}_0+\vec{\alpha}) +i(\omega_R/2)[R_{33}-R_{11}], \tag{4.10f}$$

and

$$\dot{A}_0(\pm\vec{\alpha})=-igR_{23}(\pm\vec{\alpha})-\kappa A_0(\pm\vec{\alpha}), \tag{4.10g}$$

where we have set

$$g_\alpha=g_{-\alpha}=g.$$

The Eqs. (4.10a)–(4.10g) are not closed because of the mode dependence of the arguments of the operators.

It is of interest to examine the dynamical buildup of population inversion between the upper two levels, Fig. 1, within a time for which negligible radiation, due to transitions between levels three and two, has evolved. We shall describe the evolution of transverse polarization between the upper pair of energy levels in terms of the usual tipping angle Θ, and consider the small-tipping-angle regime. We are interested in the determination of the state of the system for short, intense pump pulses immediately after the pump pulse has terminated in the medium.

The time duration of the pump pulse in the medium τ_p is assumed to be small compared to T_2 for the radiative transition of interest, i.e.,

$$\tau_p \ll T_2 .$$

In this case, the atom-field coupling g in the above equations is independent of the time and we may neglect dephasing in the population buildup. Furthermore, consistent with the assumption that τ_p is short compared to the time it takes for appreciable radiation to evolve, the average population in the intermediate level remains negligible on the average, compared to that in the third level, i.e.,

$$\langle R_{33}\rangle \gg \langle R_{22}\rangle .$$

Thus we set

$$R_{22}=\dot{R}_{22}=0 \tag{4.11}$$

in (4.10).

If, in addition, we assume that

$$|\alpha/k_0|\ll 1, \tag{4.12}$$

neglect double-frequency terms compared to similar terms of single-frequency variation, and make the substitutions,

$$A_T=A_0(\vec{\alpha})+A_0(-\vec{\alpha}), \quad R_{32\,T}=R_{32}(\vec{\alpha})+R_{32}(-\vec{\alpha}), \tag{4.13}$$

and

$$R_{12\,T}=R_{12}(\vec{k}_0+\vec{\alpha})+R_{12}(\vec{k}_0-\vec{\alpha}),$$

we arrive at the following set of equations:

$$\dot{R}_{33}=-\dot{R}_{11}, \tag{4.14a}$$

$$\dot{R}_{11}=i(\omega_R/2)[R_{31}(k_0)-R_{13}(k_0)], \tag{4.14b}$$

$$\dot{R}_{13}(k_0)=-i(g/2)A_TR_{12\,T}+i(\omega_R/2)(R_{33}-R_{11}), \tag{4.14c}$$

$$\dot{R}_{32\,T}=-2igA_T^\dagger R_3+i(\omega_R/2)R_{12\,T}, \tag{4.14d}$$

$$\dot{R}_{12\,T}=-2igA_T^\dagger R_{13}(k_0)+i(\omega_R/2)R_{32\,T}, \tag{4.14e}$$

and

$$\dot{A}_T=-igR_{23\,T}-\kappa A_T. \tag{4.14f}$$

Here, we have made the substitution

$$R_3\equiv\tfrac{1}{2}(R_{33}-R_{22}). \tag{4.15}$$

All variables in (4.14) are taken as expectation values, and we imply the decoupling

$$\langle A_TR_3\rangle=\langle A_T\rangle\langle R_3\rangle,$$

$$\langle A_TR_{12}\rangle=\langle A_T\rangle\langle R_{12}\rangle,$$

and

$$\langle A_TR_{13}\rangle=\langle A_T\rangle\langle R_{13}\rangle.$$

The equations (4.14) now comprise a closed set.

These equations can be reduced to an even simpler set by neglecting the reaction on the pump transition of the coupled emission field Raman transitions, i.e., the first term on the right-hand side of (4.14c). In the small-tipping-angle limit, the absolute value of this term is expected to be small on the average compared to that of the second term. With this additional approximation, and in the mean-field limit, qualitative analytical results are easily obtained.

Thus, neglecting the first term on the right-hand side of (4.14c), the pump equations are decoupled and are given by,

$$\dot{R}_3 = -(\omega_R/4)\left(R_{31}(k_0) - R_{13}(k_0)\right) \quad (4.16a)$$

$$\dot{R}_{13}(k_0) = i(\omega_R/2)(2R_3 - R_{11}) \quad (4.16b)$$

$$\dot{R}_{11} = -2\dot{R}_3 . \quad (4.16c)$$

With the initial condition

$$R_{11}(0) = N , \quad (4.17)$$

where N is the effective total population, Eqs. (4.16) are easily solved to give

$$R_3 = \tfrac{1}{2}N \sin^2 \tfrac{1}{2}\phi , \quad (4.18a)$$

$$R_{13} = -\tfrac{1}{2}iN \sin\phi , \quad (4.18b)$$

where

$$\phi(t) = \int_0^t \omega_R(t')dt' \quad (4.19)$$

is the pump pulse area, $\omega_R(t)$ is the Rabi frequency of the pumping pulse, and N is the effective total number of atoms being pumped. We have dropped the mode-dependent notation in these equations and in those which follow. Equations (4.14a)–(4.14f) are now reduced to (4.18), together with

$$\dot{R}_{32\,T} = -2igA_T^\dagger R_3 + \tfrac{1}{2}i\omega_R R_{12\,T} , \quad (4.20a)$$

$$\dot{R}_{12\,T} = \tfrac{1}{2}i\omega_R R_{32\,T} - 2igA_T^\dagger R_{13} , \quad (4.20b)$$

$$\dot{A}_T^\dagger = igR_{32\,T} - \kappa A_T^\dagger . \quad (4.20c)$$

to form a closed set of equations. It is useful to write the derivative of (4.20a) in the form

$$\frac{d}{dt}\left[\frac{\dot{R}_{32\,T} + 2igR_3 A_T^\dagger}{\tfrac{1}{2}i\omega_R}\right] = \dot{R}_{12\,T} . \quad (4.21)$$

We make the mean-field approximation,

$$\dot{A}_T^\dagger \ll \kappa A^\dagger , \quad (4.22)$$

then using (4.20b) and (4.20c) in (4.21), and using (4.19), we have

$$\ddot{R}_{32} - \left[\frac{\sin^2\frac{1}{2}\phi}{\tau_R} + \frac{\dot{\omega}_R}{\omega_R}\right]\dot{R}_{32} - \left[\frac{\omega_R \sin\phi}{2\tau_R} - \frac{\dot{\omega}_R \sin^2\frac{1}{2}\phi}{\omega_R\tau_R} + \frac{\dot{\omega}_R \sin\phi}{2\tau_R} - \frac{\omega_R^2}{4}\right]R_{32} = 0 , \quad (4.23)$$

where

$$\tau_R = \kappa/g^2 N \quad (4.24)$$

is the characteristic "superradiance" time.[5,8]

Equation (4.23) describes the temporal evolution of the tipping angle θ, in the small-angle regime, through the identification

$$R_{32} = -\tfrac{1}{2}iN\sin\theta \approx -\tfrac{1}{2}iN\theta . \quad (4.25)$$

The appearance of the time derivative of the pulse envelope in (4.23) is a direct result of the presence of the Raman terms in (4.20a) and (4.20b). This is, in fact, to be anticipated from the appearance of the slowly varying renormalized Raman contributions in the "dressed" Hamiltonian (3.12).

With the identification (4.25), Eq. (4.23) becomes

$$\ddot{\theta} - \left[\frac{\sin^2\frac{1}{2}\phi}{\tau_R} + \frac{\dot{\omega}_R}{\omega_R}\right]\dot{\theta} - \left[\frac{\omega_R \sin\phi}{\tau_R} - \frac{\dot{\omega}_R}{\omega_R}\frac{\sin^2\frac{1}{2}\phi}{\tau_R} - \frac{\omega_R^2}{4}\right]\theta = 0 . \quad (4.26)$$

For

$$\dot{\omega}_R = 0 , \quad (4.27)$$

i.e., a rectangular-pump-pulse envelope, (4.26) becomes

$$\ddot{\theta} - \left[\frac{\sin^2\frac{1}{2}\phi}{\tau_R}\right]\dot{\theta} + \frac{\omega_R^2}{4}\left[1 - \frac{4\sin\phi}{\omega_R\tau_R}\right]\theta = 0 . \quad (4.28)$$

To be consistent with the small-tipping-angle approximation (4.25), we require the pump pulse intensity to be such that

$$\omega_R\tau_R \gg 1 . \quad (4.29)$$

This allows us to neglect the last term in the coefficient of θ in (4.28) relative to unity. Thus, using (4.29), (4.28) has the solution

$$\theta_p(t=\tau_p) = \frac{1}{(N/2)^{1/2}} \exp\left[\frac{\tau_p}{4\tau_R} - \frac{\sin(\omega_R\tau_p)}{4\omega_R\tau_R}\right] \times \sin\tfrac{1}{2}(\omega_R\tau_p) , \quad (4.30)$$

where τ_p is the pump-pulse time duration. The initial conditions have been chosen so that $\theta_p = 0$ at $t=0$ [see the initial condition (4.17)] and reduces to the value $\theta_p = 1/(N/2)^{1/2}$ for impulse excitation in the limit of vanishingly small pump pulse width τ_p and fixed pump-pulse area, i.e.,

$$\theta_p \xrightarrow[\tau_p/\tau_R \to 0]{} \frac{1}{(N/2)^{1/2}} , \quad \omega_R\tau_p = \pi , \quad (4.31)$$

where N is the effective population inversion. Equation (4.30) is the most important result of this section.

Expression (4.30) shows that the envelope of the transverse polarization grows as an exponential in the pump-pulse duration τ_p. However, the envelope is also modulated at the pump Rabi rate which is a direct manifestation of the presence of the Raman terms in (4.20). Furthermore, the scaling of the pump-pulse duration τ_p with τ_R in the argument of the exponential is also affected significantly by the Raman contributions. If, for instance, we neglect the Raman terms in (4.20), i.e., for arbitrarily setting $R_{12}=0$, (4.30) becomes

$$\theta_p = \frac{1}{(N/2)^{1/2}} \exp\left[\frac{\tau_p}{2\tau_R} - \frac{\sin(\omega_R \tau_p)}{2\omega_R \tau_R}\right], \quad R_{12}=0\,, \tag{4.32}$$

which, when compared with (4.30), is seen to be far from a correct result. Therefore, the Raman contribution must be included.

The modulation which appears in (4.30) is a direct result of the fact that the coherent, external pump drives the system into a coherent linear superposition of its original eigenstates. A consequence of (4.30) for a pump pulse containing many Rabi cycles is that after a time τ_p, the system is left in a state of macroscopic (classical) dipole moment and will evolve collectively as superradiant[8] evolution as opposed to superfluorescent[5,8] evolution which demands that the initial state of free pulse evolution be a state of zero (quantum-mechanical) macroscopic transverse dipole moment. It is to be noted further that from (4.30), the effect of the pump in preparing the initial state of free pulse evolution scales as τ_R rather than τ_D,[24] the free pulse delay time.

If

$$\dot{\omega}_R \neq 0\,, \tag{4.33}$$

we can still draw some qualitative conclusions. If we replace (4.27) by

$$|\dot{\omega}_R/\omega_R| \gg 1/\tau_R\,, \tag{4.34}$$

and (4.29) by

$$|\dot{\omega}_R/\omega^2{}_R| \ll \omega_R \tau_R \tag{4.35}$$

then (4.26) becomes

$$\ddot{\theta} - (\dot{\omega}_R/\omega_R)\dot{\theta} + (\omega^2{}_R/4)\theta = 0\,. \tag{4.36}$$

This is just the form for an oscillator equation with positive or negative damping depending upon the sign of $\dot{\omega}_R$. Thus, a positive slope in the pump-pulse envelope tends to drive the tipping angle to larger values, whereas a negative slope tends to retard the increase of the angle. The effects on the evolution of the tipping angle θ of the time derivative of the pump-pulse envelope is another manifestation of the Raman contributions in (4.20), and is not surprising in the light of (3.12).

V. CONCLUSIONS AND SUMMARY OF RESULTS

In this paper we have shown the essential qualitative aspects of coherent optical pumping on a three-level system in the small-tipping-angle limit for subsequent free-pulse evolution. The results presented have significance for general laser theory as well as for distinguishing characteristics between observed superradiance and superfluorescence. The Hamiltonian (2.12) is the working Hamiltonian for a collection of three-level atoms in terms of collective operators (2.9) and includes the radiation field modes, pump-pulse excitation, and propagation.

The essential characteristics of the effective coupling between a traveling plane-wave pump pulse and the collective active medium are compared for a pump pulse which is long compared to the excitation volume (2.20) and one which is much shorter than that volume (2.21). As far as the excitation is concerned, there is no difference between a long pulse traveling along the major axis of a pencil-like medium (2.20) and a transverse pump pulse of the same amplitude and duration, (2.15). If, however, the pulse is short compared to the longitudinal material axis, the pump-pulse area divided by the photon transit time in the medium, rather than the Rabi rate, determines the effective coupling between the pump and material medium. The Rabi frequency is effectively reduced by the ratio of the pump time duration τ_p to the photon transit time τ_E, (2.21)–(2.23).

The "dressed" Hamiltonian[22] in the slowly varying operator representation, (3.1), (3.8)–(3.12), exhibits all of the single-atom and collective renormalizations and frequency shifts present in the two-level model for superfluorescence[5,7] obtained earlier using the "dressed" representation.[7] However, slowly varying Raman terms appear with corresponding single-atom frequency shifts displayed in (3.12). These Raman terms, as well as having difference denominators, are proportional to the time derivative of the pump-pulse envelope and can be expected to make relatively strong contributions in the dynamical evolution of the system. This is borne out by the appearance of the time derivative of the pump-pulse envelope in the equation for the evolution of the tipping angle in the small-angle limit (4.26) and (4.36). So, the pump-pulse shape as well as pulse area and temporal duration can have significant effects on the evolution of the tipping angle.

The main results of this paper are presented in Sec. IV where the Heisenberg equations of motion (4.8) are developed and applied for the restriction to two counterpropagating modes of the radiation field for the radiated pulse, (4.10)–(4.15) in the mean-field limit (4.22). In the small-tipping-angle limit for the radiating pulse evolution, the equations of motion reduce to the set (4.18)–(4.20). These equations combine under the mean-field approximation (4.22) to give a second-order differential equation for the tipping angle θ, (4.26). For a step-function pump pulse, (4.27) and (4.28), the tipping angle at the termination of the pump pulse is given by (4.30). The initial conditions have been adjusted so that (4.30) gives the value consistent with the uncertainty principle for an impulse excitation and an excitation pulse area of π.

The modulation of the exponential envelope in (4.30) is a direct result of the Raman contributions and is an essential difference between the coherent preparation of a multilevel system as compared to a two-level system. The effect of "dropping" the Raman terms in (4.20) is given by (4.32). The main effect, on comparing with (4.30), other than the absence of the modulation, is a difference of a factor of $\frac{1}{2}$ in the argument of the exponent. This, in general, is an important discrepancy, and therefore it is a significant error to ignore the Raman contributions. Furthermore, the temporal evolution of the tipping-angle scales as τ_R rather than τ_D,[24] where τ_D is the delay time between an excitation "impulse" and the peak of the superradiant or superfluorescent pulse which evolves. These are coherent effects and are manifestations of the fact that a coherent pump drives the system into a coherent linear superposition of its original eigenstates. Other manifestations of effects produced by coherent pumping on multilevel systems are the observations of multiple wave mixing[14] and quantum beats.[15]

A conclusion from these results is that for coherent pumping on a multilevel system where the pump duration τ_p is less than the atomic dephasing time T_2 and either on the order of, or larger than, the characteristic superradiance time τ_R,[5,8,9] or such that the pump contains many Rabi cycles, or both, the system is always left in a state of macroscopic transverse polarization after the pump-pulse termination. Subsequent collective radiative pulse evolution is then purely classical with polarization determined by the pump polarization. In order to ensure evolution of the free radiated pulse as a quantum-mechanical dipole moment, the pump pulse must be much shorter than τ_R and it must be effectively a π pulse. Typically, τ_R is on the order of nanoseconds to tens of nanoseconds. Subnanosecond pump π pulses may be practically difficult to achieve.

Another observational effect of (4.30) is in connection with the free pulse delay time τ_D. If we take[8]

$$\tau_D = -2\tau_R \ln\theta_p , \tag{5.1}$$

for an active volume with large Fresnel number, then using (4.30) and suppressing the fluctuations suggested by the sine factor,

$$\tau_D = 2\tau_R \ln(N/2)^{1/2} - \tau_p/2 . \tag{5.2}$$

Equation (5.2) predicts an almost linear dependency of τ_D with inverse pressure, but with negative intercept. Such a linear dependency with pressure and with negative intercept has been observed.[25] A test of this model would be to adjust the pump-pulse duration τ_p while maintaining the pump-pulse area constant over the same range of data.

The appearance of the sine factor in (4.30) leads to the possibility for wide fluctuations in radiated pulse amplitude and width as well as delay time. For a many-π pump pulse, it may be experimentally difficult to ensure the same identical value for the total pulse area from one "shot" to the next. Then, from "shot" to "shot" temporal fluctuations can be anticipated which are purely of classical origin. Such large "fluctuations" have their origin entirely in coherent-pump-induced cycling, rather than quantum fluctuations which are anticipated from superfluorescent evolution.[5]

The effects of the shape of the pump-pulse envelope on the evolution of the tipping angle are suggested by (3.12), (4.26), and (4.36). Generally, a positive slope to the time derivative of the pump-pulse envelope causes an exponential increase in the evolution of the tipping angle in the small-angle regime, whereas a negative slope tends to inhibit the evolution to larger tipping angles. Due to the importance of the pump-pulse shape in preparing the system for collective free radiation pulse evolution, experiments on the same material and otherwise same experimental conditions could be quite different when different pump sources are used. These are matters to be considered in any experimental design.

It is concluded that when multilevel systems are pumped with coherent sources, the dynamics of radiative pulse evolution cannot be separated from the dynamics of the pumping process. Consequently, in the light of coherent pump effects, extreme care must be taken in the analysis and interpretation of experimental results which exhibit cooperative effects.

APPENDIX

We closely parallel the procedures developed by Coulter,[22] based upon Heitler's method,[23] to derive the unitary transformation to the "dressed representation" to the second order of the perturbation. Whereas Coulter's method treats an explicitly time-independent Hamiltonian, we treat the case here for a Hamiltonian explicitly dependent upon the time t.

It is assumed that the Hamiltonian $\mathcal{H}(t)$ can be written in the form

$$\mathcal{H}(t) = \mathcal{H}_0 + \lambda H_I(t), \tag{A1}$$

where $\mathcal{H}_0$ is explicitly time independent, λ is a perturbation parameter and the eigenstates $|n_0\rangle$ and associated eigenvalues ϵ_0 for $\mathcal{H}_0$ are known,

$$\mathcal{H}_0 |n_0\rangle = \epsilon_0 |n_0\rangle . \tag{A2}$$

We call the set of states $|n_0\rangle$ the "bare" states.

Let us assume that $\mathcal{H}(t)$ has eigenstates $|n(t)\rangle$ such that

$$\mathcal{H}(t)|n(t)\rangle = \epsilon(t)|n(t)\rangle , \tag{A3}$$

and suppose a unitary transformation $U(t)$ exists such that

$$U(t)|n(t)\rangle = |n_0\rangle . \tag{A4}$$

Then

$$\mathcal{H}(t)U^{-1}(t)|n_0\rangle = \epsilon(t)U^{-1}(t)|n_0\rangle ,$$

and

$$U(t)\mathcal{H}(t)U^{-1}(t)|n_0\rangle = \epsilon(t)|n_0\rangle . \tag{A5}$$

Let

$$\mathcal{H}'(t) \equiv U(t)\mathcal{H}(t)U^{-1}(t) \tag{A6}$$

then

$$\mathcal{H}'(t)|n_0\rangle = \epsilon(t)|n_0\rangle . \tag{A7}$$

Thus, if we can find $U(t)$, then $\mathcal{H}'(t)$ has eigenstates which are the original "bare" states. In other words, in the "dressed" state representation, the original, known "bare" states are the "dressed" states. In parallel with Coulter,[22] we let

$$U(t) = e^{ik(t)}, \quad k(t) = k^{\dagger}(t) \tag{A8}$$

and set about to calculate $U(t)$ through the desired order in perturbation. But first we determine the canonical form of the Hamiltonian in the transformed representation.

We look for the Hamiltonian $\mathcal{H}''(t)$ in the transformed representation (A6) which is canonically equivalent to (A1). Thus consider the Schrödinger equation,

$$i\hbar\frac{\partial}{\partial t}|\psi(t)\rangle = \mathcal{H}(t)\,|\psi(t)\rangle . \tag{A9}$$

From (A3) and (A4) we have

$$\begin{aligned}|\psi(t)\rangle &= \sum_n c_n(t)|n(t)\rangle \\ &= \sum_n c_n(t)U^{-1}(t)|n_0\rangle \\ &= U^{-1}(t)\sum_n c_n(t)|n_0\rangle .\end{aligned} \tag{A10}$$

If we make the identification

$$|\psi_0(t)\rangle = \sum_n c_n(t)|n_0\rangle , \tag{A11}$$

then

$$|\psi(t)\rangle = U^{-1}(t)|\psi_0(t)\rangle . \tag{A12}$$

Using (A12) in (A9),

$$i\hbar\frac{\partial}{\partial t}U^{-1}(t)|\psi_0(t)\rangle = \mathcal{H}(t)U^{-1}(t)|\psi_0(t)\rangle . \tag{A13}$$

Multiplying both sides of (A13) by $U(t)$, and using (A6), leads to

$$i\hbar\frac{\partial}{\partial t}|\psi_0(t)\rangle = \left[\mathcal{H}'(t) - i\hbar U(t)\frac{\partial}{\partial t}U^{-1}(t)\right]|\psi_0(t)\rangle . \tag{A14}$$

Thus, the Hamiltonian $\mathcal{H}''(t)$ in the transformed representation canonically equivalent to (A1) is, from (A14)

$$\mathcal{H}''(t) = \mathcal{H}'(t) - i\hbar U(t)\frac{\partial}{\partial t}U^{-1}(t) . \tag{A15}$$

Furthermore, if $\mathcal{H}''(t)$ is to be Hermition, it is required that

$$\dot{U}(t)U^{-1}(t) = -U(t)\dot{U}^{-1}(t) , \tag{A16}$$

or, equivalently

$$\frac{\partial}{\partial t}[U(t)U^{-1}(t)] = 0 . \tag{A17}$$

So, in the transformed representation, we have for the canonical Hamiltonian, from (A15) and (A7),

$$\begin{aligned}\mathcal{H}''(t)|n_0\rangle &= \mathcal{H}'(t)|n_0\rangle + i\hbar\dot{U}^{-1}(t)|n_0\rangle \\ &= [\epsilon(t) + i\hbar\dot{U}(t)U^{-1}(t)]|n_0\rangle .\end{aligned} \tag{A18}$$

Now, returning to (A2) and (A7), we have that

$$[\mathcal{H}_0, \mathcal{H}'(t)] = 0 . \tag{A19}$$

Let

$$\mathcal{H}'(t) = \mathcal{H}_0 + D(t) \tag{A20}$$

and

$$D(t) = \sum_{n=1}^{\infty}\lambda^n D_n(t) \tag{A21}$$

and for (A8) let

$$\kappa(t) = \sum_{n=1}^{\infty}\lambda^n\kappa_n(t) = \kappa^{\dagger}(t) . \tag{A22}$$

It is to be noted that if the condition of unitarity in (A22) is satisfied, (A16) is automatically satisfied.

From (A20), (A19), and (A21) we have

$$[\mathcal{H}_0, D_n(t)] = 0 . \tag{A23}$$

We proceed to calculate $\mathcal{H}'(t)$ and $i\hbar\dot{U}(t)U^{-1}(t)$ through second order in the parameter λ. Thus, using (A1), (A2), (A21), (A6), (A8), and (A22), we arrive at the relations

$$D_1(t) = i[\kappa_1(t), \mathcal{H}_0] + \mathcal{H}_I(t) \quad \text{(A24)}$$

$$D_2(t) = i[\kappa_2(t), \mathcal{H}_0] + \frac{i^2}{2!}[\kappa_1(t), [\kappa_1(t), \mathcal{H}_0]] \quad \text{(A25)}$$

$$+ i[\kappa_1(t), \mathcal{H}_I(t)], \quad \text{(A26)}$$

and in addition,

$$\{\dot{U}(t)U^{-1}(t)\} = i(\dot{\kappa}_1(t) + \dot{\kappa}_2(t)) - \tfrac{1}{2}[\kappa_1(t), \dot{\kappa}_1(t)] \quad \text{(A27)}$$

through second order. Thus, the canonical Hamiltonian (A15) is, through second order, in the perturbation,

$$\mathcal{H}''(t) = \mathcal{H}_0 + D_1(t) + D_2(t) - \hbar(\dot{\kappa}_1(t) + \dot{\kappa}_2(t)) - \tfrac{1}{2}i\hbar[\kappa_1(t), \dot{\kappa}_1(t)]. \quad \text{(A28)}$$

[1]R. H. Dicke, Phys. Rev. 93, 99 (1954).
[2]N. E. Rehler and J. H. Eberly, Phys. Rev. A 3, 1735 (1971).
[3]R. Bonifacio, P. Schwendimann, and F. Haake, Phys. Rev. A 4, 302; 854 (1971), and references therein.
[4]R. Friedberg and S. R. Hartmann, Phys. Rev. A 11, 1728 (1974).
[5]R. Bonifacio and L. A. Lugiato, Phys. Rev. A 11, 1507 (1975); 12, 587 (1975).
[6]J. C. MacGillivray and M. S. Feld, Phys. Rev. A 14, 1169 (1976).
[7]L. M. Narducci and C. M. Bowden, J. Phys. A 9, L75 (1976).
[8]R. Bonifacio, M. Gronchi, L. A. Lugiato, and A. M. Ricca, in *Cooperative Effects in Matter and Radiation*, edited by C. M. Bowden, D. W. Howgate, and H. R. Robl (Plenum, New York, 1977), p. 193.
[9]R. Saunders and R. K. Bullough, in *Cooperative Effects in Matter and Radiation*, edited by C. M. Bowden, D. W. Howgate, and H. R. Robl (Plenum, New York, 1977), p. 209.
[10]J. C. MacGillivray and M. S. Feld, in *Cooperative Effects in Matter and Radiation*, edited by C. M. Bowden, D. W. Howgate, and H. R. Robl (Plenum, New York, 1977), p. 1.
[11]N. Skribanowitz, J. P. Herman, J. C. MacGillivray, and M. S. Feld, Phys. Rev. Lett. 30, 309 (1973).
[12]M. Gross, C. Fabre, P. Pillet, and S. Haroche, Phys. Rev. Lett. 36, 1035 (1976).
[13]A. T. Rosenberger, S. J. Petuchowski, and T. A. DeTemple, in *Cooperative Effects in Matter and Radiation*, edited by C. M. Bowden, D. W. Howgate, and H. R. Robl (Plenum, New York, 1977), p. 15.
[14]A. Flusberg, T. Mossberg, and S. R. Hartmann, in *Cooperative Effects in Matter and Radiation*, edited by C. M. Bowden, D. W. Howgate, and H. R. Robl (Plenum, New York, 1977), p. 37.
[15]H. M. Gibbs, in *Cooperative Effects in Matter and Radiation*, edited by C. M. Bowden, D. W. Howgate, and H. R. Robl (Plenum, New York, 1977), p. 79.
[16]Q. H. F. Vrehen, in *Cooperative Effects in Matter and Radiation*, edited by C. M. Bowden, D. W. Howgate, and H. R. Robl (Plenum, New York, 1977), p. 79.
[17]J. J. Ehrlich, C. M. Bowden, D. W. Howgate, S. H. Lehnigk, A. T. Rosenberger, and T. A. DeTemple, "Swept-gain Superradiance in CO_2 Pumped CH_3F" in *Coherence and Quantum Optics IV*, edited by L. Mandel and E. Wolf (Plenum, New York, 1978), p. 923.
[18]S. Haroche, J. A. Paisner, and A. L. Schowlow, Phys. Rev. Lett. 30, 948 (1973).
[19]R. Gilmore, *Lie Groups, Lie Algebras, and Some of their Applications* (Wiley, New York, 1974), Chap. 6, Sec. 2.
[20]R. Gilmore, C. M. Bowden, and L. M. Narducci, Phys. Rev. A 12, 1019 (1975).
[21]R. Gilmore, C. M. Bowden, and L. M. Narducci, "*c*-Number Representation for Multilevel Systems and the Quantum-Classical Correspondence", in *Quantum Statistics and the Many-Body Problem*, edited by S. B. Trickey, W. R. Kirk, and J. W. Dufty (Plenum, New York, 1975).
[22]C. A. Coulter, Phys. Rev. A 10, 1946 (1974).
[23]W. Heitler, *The Quantum Theory of Radiation* (Clarendon, Oxford, 1957), p. 146.
[24]J. C. MacGillivray and M. S. Feld, in *Cooperative Effects in Matter and Radiation*, edited by C. M. Bowden, D. W. Howgate, and H. R. Robl (Plenum, New York, 1977), p. 7.
[25]A. T. Rosenberger, S. J. Petuchowski, and T. A. DeTemple, in *Cooperative Effects in Matter and Radiation*, edited by C. M. Bowden, D. W. Howgate, and H. R. Robl (Plenum, New York, 1977), p. 24.