rather than sharp interface. We believe that future studies must pay close attention to this question, even considering the inhomogeneous charge density at the surface associated with atomic structure on crystalline targets. Indeed it may well be that reasonably detailed characterization of such interfaces will result from such a project.

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Initiation of Superfluorescence in Coherently Pumped Three-Level Systems

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The fully quantum mechanical treatment of superfluorescence buildup in the linearized regime of superfluorescence is presented for coherently, optically pumped three-level systems. It is shown that to order τ_p/τ_R, where τ_p is the pump time duration and τ_R the characteristic superfluorescence time, fluorescence buildup during the dynamical pumping process contributes significantly to quantum initiation of superfluorescence.

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Superfluorescence (SF)[1] is the phenomenon whereby a collection of atoms or molecules is prepared initially in a state of complete inversion, and then allowed to undergo relaxation by collective, spontaneous decay. Since Dicke's initial work,[2] there has been a large amount of theoretical and experimental work dealing with this process.[3]

With the exception of the more recent work of Bowden and Sung[4] and Bowden and Mattar,[5] all theoretical treatments have dealt exclusively with the relaxation process from a prepared state of complete inversion in a two-level manifold of atomic energy levels, and thus do not consider the dynamical effects of the pumping process. Yet, all reported experimental work[3] has utilized optical pumping on a minimum manifold of three atomic or molecular energy levels by laser pulse injection into the nonlinear medium, which subsequently superfluoresces.

It was pointed out by Bowden and Sung[4] that for a system otherwise satisfying the conditions for superfluorescent emission, unless the characteristic superradiance time,[1] τ_R, is much greater than the pump-pulse temporal duration, τ_p, i.e., $\tau_R \gg \tau_p$, the process of coherent optical pumping on a three-level system can have dramatic effects on the SF. This is a condition which has not been realized over the full range of experimental data.[3]

It has not only been shown that the dynamics of the pumping process can have dramatic effects upon the SF pulse evolution,[4,5] but also that the

statistics of the quantum mechanical initiation of the SF has profound effects in terms of macroscopic temporal fluctuations.[6,7] So far, amplified-quantum-initiation statistics in SF emission have been discussed only with regard to two-level systems with the initial condition of complete inversion.[6,7] However, it is noted that Raymer and Mostowski have treated spontaneous initiation in a three-level system in the context of stimulated Raman scattering.[8]

It is the purpose of this paper to provide a more comprehensive treatment of SF by combining coherent pump dynamics on the three-level system and quantum mechanical initiation of SF emission. In what follows, we present our model and discuss the results in terms of the effects of amplified quantum initiation and coherent pump dynamics on SF pulse evolution.

The model upon which the calculation is based is comprised of a collection of identical three-level atoms, each having the energy-level scheme such that the $1 \leftrightarrow 3$ transition is induced by a coherent electromagnetic field pulse of frequency ω_0 approximately tuned to the transition. The transition $3 \leftrightarrow 2$ evolves by spontaneous emission at a much lower frequency ω. It is assumed that the energy-level spacing is such that $\epsilon_3 > \epsilon_2 \gg \epsilon_1$, and we neglect spontaneous relaxation in the pump transition $1 \leftrightarrow 3$. The energy levels ϵ_2 and ϵ_1 are not coupled radiatively because of parity considerations. The pump field is treated classically, i.e., as a coherent state,[9] and longitudinally uniform in the medium, which has been shown[4a] to be justified provided that the pump-pulse time duration τ_p is much larger than the longitudinal transit time in the medium, τ_E, i.e., $\tau_p \gg \tau_E$.

In the electric-dipole and rotating-wave approximations, the Hamiltonian $\mathcal{H}$ which describes this system of N dynamically pumped three-level atoms is

$$\mathcal{H} = \sum_{r=1}^{3}\sum_{j=1}^{N} \epsilon_{rj} R_{rr}^{(j)} + \hbar \sum_{l=0}^{\infty} \omega_l a_l^{\dagger} a_l - i \sum_{j=1}^{N}\sum_{l=0}^{\infty} [g_l^{(j)} a_l R_{32}^{(j)} \exp(i\vec{k}_l \cdot \vec{r}_j) - g_l^{*(j)} a_l^{\dagger} R_{23}^{(j)} \exp(-i\vec{k}_l \cdot \vec{r}_j)] - \frac{i\hbar}{2} \sum_{j=1}^{N} \{\omega_R^{(j)} R_{31}^{(j)} \exp[-i(\omega_0 t - \vec{k}_0 \cdot \vec{r}_j)] - \omega_R^{*(j)} R_{13}^{(j)} \exp[i(\omega_0 t - \vec{k}_0 \cdot \vec{r}_j)]\}, \quad (1)$$

where the canonical atomic operators $R_{kl}^{(j)}$ obey the Lie algebra defined by the commutation rules,[4]

$$[R_{ij}^{(m)}, R_{lk}^{(n)}] = R_{ik}^{(m)} \delta_{lj} \delta_{mn} - R_{lj}^{(m)} \delta_{ik} \delta_{mn}. \quad (2)$$

The field operators $a_l, a_l^{\dagger}$ are the usual Bose operators and ω_R is the pump field Rabi rate.

We assume plane-wave propagation and take the initial condition with all the atoms in the ground state ϵ_1. The Heisenberg equations of motion for the slowly varying rightward propagating collective atomic variables,[10] $\tilde{R}_{kl}$, and field variables, $A_R^{(-)}$, in the linearized regime of small SF signal (negligible population in level 2) and strong pump,[4]

$$\langle \tilde{R}_{22} \rangle \approx d\langle \tilde{R}_{22} \rangle / dt \approx 0; \quad \omega_R \tau_R \gg 1, \quad (3)$$

lead to the following set of coupled equations in retarded time $\tau = t - z/c$:

$$d\tilde{R}_{33}/d\tau = -\omega_R \tilde{R}_{31}, \quad (4a)$$

$$d\tilde{R}_{31}/d\tau = \omega_R (\tilde{R}_{33} - \tfrac{1}{2}), \quad (4b)$$

$$d\tilde{R}_{32}/d\tau = -\tfrac{1}{2}\omega_R \tilde{R}_{12} + A_R^{(-)} \tilde{R}_{33} + f_R^{(-)}, \quad (4c)$$

$$d\tilde{R}_{12}/d\tau = \tfrac{1}{2}\omega_R \tilde{R}_{32} + A_R^{(-)} \tilde{R}_{13} + h_R^{(-)}, \quad (4d)$$

$$\partial A_R^{(-)}/\partial z = (1/\tau_R L) \tilde{R}_{32}. \quad (5)$$

In deriving these equations, we have assumed normal ordering of the field relative to the atomic variables.[6] In the above equations, L is the longitudinal length of the medium and τ_R, the SF characteristic time, is given by $\tau_R = (L/c)[(2\pi|g|^2/\hbar)\rho]^{-1}$, where ρ is the atomic density taken as uniform and g is the coupling in the neighborhood of resonance for the fluorescence transition. We have taken the pump as perfectly tuned to the atomic transition $1 \leftrightarrow 3$ and consider times τ short compared with the natural relaxation time τ_0 of the $3 \leftrightarrow 2$ transition. The origin of the Langevin terms $f_R^{(-)}$ and $h_R^{(-)}$ is discussed below.

The Maxwell equation (5) for the field $A_R^{(-)}$ is derived from the expression

$$A_R^{(-)}(z,\tau) = \frac{n_s}{\hbar^2} \sum_i |g_l|^2 \sum_{z_m} \int_{-z_m/c}^{\tau + (z - z_m)/c} \exp[ic(k_l - k)(\tau - \tau')] \tilde{R}_{32}(z_m, \tau') d\tau' \quad (6)$$

in a manner similar to that leading to Eq. (36) of Ref. 6. Here n_s is a normalization due to the introduction of collective atomic operators.[10] The above expression is obtained by formally integrating the Heisenberg equations of motion for the fluorescence field and then separating out the self-field of the atom and the vacuum contribution. The former separated field leads to natural atomic relaxation τ_0 in the normally ordered Heisenberg equation of motion, and the latter to the Langevin terms $f_R^{(-)}$ and $h_R^{(-)}$ in (4c) and (4d), which give rise to Gaussian random quantum-initiation statistics in the SF evolution.[6,7] These are written explicitly as

$$f_R^{(-)}(z,\tau)=f(\tau)\tilde{R}_{33}(\tau,z), \tag{7}$$

$$h_R^{(-)}(z,\tau)=f(\tau)\tilde{R}_{31}(\tau,z), \tag{8}$$

where $f(\tau)$ satisfies the ensemble averages over the vacuum fluctuations,

$$\langle f(\tau)f^{\dagger}(\tau')\rangle=(1/N\tau_R)\delta(\tau-\tau'),$$
$$\langle f^{\dagger}(\tau)f(\tau')\rangle=0, \tag{9}$$

and N is the total number of atoms. It should be noted that we have ignored any contributions of natural atomic relaxation in the dynamics under conditions of SF initiation.[1]

The Heisenberg equations of motion for $\tilde{R}_{33}$ and $\tilde{R}_{31}$ in the approximation (3) exhibit no operator character and form a closed set in themselves. We have thus taken them as expectation values, and ω_R and $\tilde{R}_{31}$ are taken as real without loss of generality. The result is (4a) and (4b). Because of the strong-pump approximation, $\omega_R\tau_R\gg1$, the collective pump transition variables $\tilde{R}_{33}$ and $\tilde{R}_{31}$ are dynamically determined by the pumping process entirely and can therefore be replaced by their factorized expectation values in (4c) and (4d). Then the only operator character in these equations are the Langevin force terms $f_R^{(-)}$ and $h_R^{(-)}$. Thus, Eqs. (4c)–(5) can be regarded in terms of expectation values with respect to the atomic system, but not the field reservoir, i.e., the ordering of products of the Langevin force terms in expectation values is essential, as exhibited in (9). From this point on, Eqs. (4)–(5) are regarded in terms of expectation values with respect to the atomic system.

It is reemphasized that the strong-pump approximation (3) has resulted in a decoupling of the pump-term equations, (4a) and (4b), from the remainder. Thus, for all the atoms initially in the ground state at $\tau=0$, we have

$$\tilde{R}_{33}=\sin^2\tfrac{1}{2}\omega_R\tau;\quad \tilde{R}_{31}=-\tfrac{1}{2}\sin\omega_R\tau. \tag{10}$$

The remainder of the equations of motion (4c)–(5) can be solved by Laplace transform. These combine in the Laplace regime, in the limit $\omega_R\tau_R\gg1$, and to first order in τ_p/τ_R, to give the solution in terms of $\bar{\tilde{R}}_{32}^{(0)}(s,\tau)$,

$$\frac{\partial^2\bar{\tilde{R}}_{32}^{(0)}(s,\tau)}{\partial\tau^2}+\frac{1}{4}\omega_R{}^2\bar{\tilde{R}}_{32}^{(0)}(s,\tau)=\frac{u(\tau)}{s}, \tag{11}$$

where

$$u(\tau)\equiv\partial f_R^{(-)}/\partial\tau-\tfrac{1}{2}\omega_R h_R^{(-)} \tag{12}$$

and

$$\bar{\tilde{R}}_{32}^{(0)}(s,\tau)=\frac{2}{\omega_R}\int_0^{\tau}d\tau'\frac{u(\tau')}{s}\sin\tfrac{1}{2}\omega_R(\tau-\tau'). \tag{13}$$

The "tipping angle" $\theta(z,\tau)$ is defined as[6,7]

$$\theta^2(z,\tau)\equiv\langle\tilde{R}_{23}(z,\tau)\tilde{R}_{32}(z,\tau)\rangle \tag{14}$$

and we are interested in its value in the SF regime $\tau>\tau_p$, $\omega_R=0$. In the time frame after the pump is turned off, i.e., for $\tau>\tau_p$,

$$f_R^{(-)}(\tau)=f(\tau)\tilde{R}_{33}(\tau_p), \tag{15}$$

$$h_R^{(-)}(\tau)=f(\tau)\tilde{R}_{31}(\tau_p), \tag{16}$$

and Eqs. (4a)–(5) are solved for $\omega_R=0$ with the initial condition being the value of each variable at $\tau=\tau_p$. The procedure already outlined leads in a straightforward way to the result, for $\tau'=\tau-\tau_p$, $\tau'>0$,

$$\theta^2(\tau')=\{I_0(2[(1/L\tau_R)\tilde{R}_{33}(\tau_p)z\tau']^{1/2})\}^2\langle\tilde{R}_{23}^{(0)}(\tau_p)\tilde{R}_{32}^{(0)}(\tau_p)\rangle$$
$$+\frac{\tilde{R}_{33}{}^2(\tau_p)}{N\tau_R}\int_0^{\tau'}d\tau''\{I_0(2[(1/L\tau_R)\tilde{R}_{33}(\tau_p)z(\tau'-\tau'')]^{1/2})\}^2, \tag{17}$$

which is the main result of this paper.

It is interesting to compare the result (17) with the corresponding result from the two-level model with an initial state of complete inversion (i.e., impulse excitation) of Ref. 6, Eq. (64). We see that the two cases differ by the first term in (17) as well as the factor $\tilde{R}_{33}{}^2(\tau_p)$ of the square of the pump-induced inversion onto the second term and the appearance of $\tilde{R}_{33}(\tau_p)$ in the argument of the Bessel's functions I_0. In the linearized regime of Refs. 6 and 7, $R_{33}=1$; in our case this need not be so. But, if $\omega_R\tau_p=\pi$, i.e., a π pump pulse, then the second term in (17) is exactly equivalent to

Eq. (64) of Ref. 6. However, the first term in (17) still remains, which arises from spontaneous relaxation of the $3 \rightarrow 2$ transition during the dynamics of the pumping process. The first term in (17) therefore, uniquely characterizes the effects of dynamical pumping in a three-level system on SF quantum initiation.

The evolution of the expectation value in the first term of (17) gives

$$\langle R_{23}^{(0)}(\tau_p) R_{32}^{(0)}(\tau_p)\rangle \approx \left(\frac{4}{N}\sin^2\frac{\omega_R \tau_p}{2}\right)\frac{\tau_p}{\tau_R}$$

to first order in τ_p/τ_R. Thus, for a π-pulse, impulse excitation, i.e., $\omega_R \tau_p = \pi$, $\tau_p \rightarrow 0$, all effects of the pump dynamics vanish, and (17) reduces to the results of Ref. 6, Eq. (64).

However, it is apparent that $\tau_p/\tau_R \ll 1$ is a condition not satisfied in SF experiments over the full range of atomic densities, and therefore, the effects of the dynamical pumping process on amplified quantum initiation could be quite important.

The opposite regime, i.e., for $\tau_p/\tau_R \gg 1$ has been treated in a numerical calculation for a three-level system including propagation and transverse effects,[5] and the results show that the injected pump-pulse initial characteristics can have a significant deterministic effect on the SF pulse evolution. That simulation was done semiclassically, i.e., without the effects of quantum initiation. The procedure outlined here actually leads to a formulation of the statistics of quantum initiation in that regime as well and will be the subject of a future publication.[11]

Further details of our development as well as explicit quantum statistical ramifications of temporal fluctuations in SF emission will be presented in a future publication.[12]

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$$R_{kl}(z,t) = \frac{1}{n_s}\sum_{j\in\{j\}_z} R_{kl}^{(j)}(t),$$

where n_s is the mean number of atoms in a slice and $\{j\}_z$ denotes the collection of atoms in $z - d/2 < z_j < z + d/2$, $d/2\pi\lambda \ll 1$. The procedure is presented explicitly in Ref. 6. $\check{R}_{kl}$ is then the rightward propagating collective atomic variable.

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