# Recovery of superfluorescence in inhomogeneously broadened systems through rapid relaxation

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This paper shows that time-dependent hyperfine interactions of the nucleus with electrons can enhance the superfluorescence (SF) intensity by countering the destructive effect of inhomogeneous broadening on SF. [S0163-1829(97)05818-9]

## I. INTRODUCTION

The emission of a superfluorescence (SF) pulse from the ends of a thin, cylindrically shaped active medium is highly directional. The process is characterized by the SF time defined by[1]

$$\tau_{\mathrm{SF}}=\frac{8\pi\tau_n}{3\rho\lambda^2 l}, \tag{1}$$

where $\tau_n$ is the natural lifetime of an excited level,[2] $\rho$ is the density of excited atoms or nuclei, $\lambda$ is the emission wavelength, and $l$ is the length of the cylindrical inverted region. Following a fast inversion, a pulse is emitted after a time delay.[3] The time delay $\tau_D$ is characteristically much shorter than $\tau_n$ but several times longer than $\tau_{\mathrm{SF}}$. During $\tau_D$, the $N$ individual radiators become correlated so that they can radiate as a single dipole with a peak intensity proportional to $N^2$.

The presence of a dephasing mechanism, characterized by a dephasing time $\tau_\varphi$, may slow down or completely inhibit this process so that SF cannot occur. If the dephasing time is sufficiently short, correlations between radiators do not develop, and each radiator responds individually to the instantaneous electromagnetic field, resulting in natural spontaneous emission.

Our interest in SF stems from the following expectation: if a $\gamma$-ray laser[4] is developed, it will probably emit in a SF mode because characteristic features of SF decay may provide ways of overcoming specific nuclear problems that prevent more conventional continuous wave or pulsed-laser action. These specific nuclear problems are discussed in Ref. 5, where we investigated the possibility of experimentally realizing nuclear SF and provided several reasons why obtaining atomic (or molecular) SF is easier. One of these reasons, perhaps the most important impediment to SF in long-lived nuclear states, is the high level of inhomogeneous broadening,[6] which can destroy the resonant condition when narrow nuclear lines are involved. Several authors have addressed the problem of inhomogeneous broadening as it pertains to Mössbauer experiments and $\gamma$-ray laser development, and have proposed techniques for dealing with it.[7,8]

In this paper we concentrate on the effect of inhomogeneous broadening in SF emission. We show how inhomogeneous broadening produces dephasing and inhibits cooperative emission, thus reducing the intensity of the SF pulse. We also show how electronic relaxation or time-dependent hyperfine interactions can mitigate the effect of inhomogeneous broadening so that SF can be recovered.

## II. EMISSION FROM A SYSTEM OF IDENTICAL NUCLEI IN DIFFERENT ENVIRONMENTS

For $N$ atoms that are identical except for small differences in energy caused by inhomogeneous broadening,[6] the emission probability is given by[9]

$$P(\omega)=\int_0^\infty dt'\int_0^\infty dt''\langle V^{(-)}(t'')V^{(+)}(t')\rangle e^{i\omega(t'-t'')-\Gamma/2(t'+t'')}\times\sum_{j,k}^{N} e^{i(\Delta\omega_j t'-\Delta\omega_k t'')+i(\varphi_j-\varphi_k)}, \tag{2}$$

where $V^{(\pm)}(t)=e^{iHt}V^{(\pm)}e^{-iHt}$ and $V^{(\pm)}$ is the interaction term of the Hamiltonian responsible for the emission and absorption of a photon. $\Gamma$ is the reciprocal of the lifetime of the excited state, $H$ is the Hamiltonian for the total system, including all nuclear and solid-state energy terms, $\Delta\omega_j$ is the transition energy between the two nuclear levels of the $j$th nucleus divided by $\hbar$, and $\varphi_j$ is a spatial phase factor which depends on the relative position of the nucleus in the lattice. Inhomogeneous broadening occurs when, in general, $\Delta\omega_j \neq \Delta\omega_k$.

It is convenient to write Eq. (2) in the form that separates cooperative (SF) and noncooperative emissions, so that

$$P(\omega)=I_{N^1}(\omega)+I_{N^2}(\omega), \tag{3}$$

where

$$I_{N^1}(\omega)=N\int_0^\infty dt'\int_0^\infty dt''\langle V^{(-)}(t'')V^{(+)}(t')\rangle\times e^{i\omega(t'-t'')-\Gamma/2(t'+t'')} \tag{4}$$

 

and

$$I_{N^2}(\omega)=\int_0^\infty dt'\int_0^\infty dt''\langle V^{(-)}(t'')V^{(+)}\times(t')\rangle e^{i\omega(t'-t'')-\Gamma/2(t'+t'')}\times\sum_{j,k\neq j}^{N} e^{i(\Delta\omega_j t'-\Delta\omega_k t'')+i(\varphi_j-\varphi_k)}. \quad (5)$$

The first term on the right-hand side of Eq. (3) as defined by Eq. (4) represents the total single nucleus uncooperative emission. The second term on the right side of Eq. (3) as defined by Eq. (5) represents the SF cooperative emission with the characteristic narrow angular distribution and shortened emission time following an initial delay. This double sum exhibits the inhomogeneous broadening effect. Assuming that the nuclei are identical except for the different positions in the solid lattice and different amounts of detuning, the expectations in Eq. (3) are the same for any two atoms, and, therefore, the double sum involves only the exponential terms.

Since our primary interest is the total intensity as a function of time and not as a function of the line shape or photon energy distribution, we can integrate $I_{N^2}(\omega)$ over $\omega$, to get

$$\frac{1}{2\pi}\int_{-\infty}^{\infty} I_{N^2}(\omega)d\omega=\int_{-\infty}^{\infty}\hat{I}_{N^2}(t')dt',$$

where

$$\hat{I}_{N^2}(t')=\langle V^{(-)}(t')V^{(+)}(t')\rangle\times\sum_{j,k\neq j}^{N} e^{i(\Delta\omega_j-\Delta\omega_k)t'-\Gamma t'+(\varphi_j-\varphi_k)i}. \quad (6)$$

The exponential on the right-hand side of Eq. (6) exhibits phase differences in the contributions of the different radiators to the SF emission term. This effect of inhomogeneous broadening reduces the intensity of the emitted SF pulse.

Assuming that the inhomogeneously broadened line shape can be modeled by a Lorentzian distribution, Eberly[10] showed that the SF intensity given by Eq. (6) after the summation is performed takes the form[11]

$$I_{N^2}(t)=N^2|\langle V\rangle|^2 e^{-2\Gamma^* t}, \quad (7)$$

where $\Gamma^*$ is the inhomogeneously broadened linewidth and $\langle V\rangle$ is the product of the matrix elements in Eq. (6). For fast pumping, essentially completed near $t=0$ (pumping time $t_{\rm pump}\ll\tau_{\rm SF}\ll 1/\Gamma^*<1/\Gamma$), inhomogeneous broadening has little effect on the SF emission because the nuclei do not have time to dephase. An examination of the exponential factor on the right-hand side of Eq. (7) shows that only after a relaxation time of the order of $T_2^*=1/\Gamma^*$ do the phase differences caused by the energy differences between different nuclei $[i,j,(\Delta\omega_i-\Delta\omega_j)t]$ become large enough to affect the emission significantly or prevent the SF pulse from occurring.

## III. TIME-DEPENDENT PERTURBATIONS

### A. In absorption measurements

Inhomogeneous broadening in samples used in a resonance experiment can reduce the resonance effect observed. However, for fluctuations that are fast enough, if the inhomogeneous broadening is caused by resonances that are coupled by fluctuating fields, the resonance effect often can be recovered.

For example, assume that the field at the nucleus is jumping between two values at a particular site in the source or absorber, so that the resonance energy correspondingly jumps between $\pm\delta$. Blume[9] has shown how to calculate the emission and absorption line shapes for resonators acting independently when the system under consideration is subjected to time-dependent perturbing fields. We follow his analysis, assuming a time-dependent field acting on the nucleus, and write

$$H(t)=H_0+\delta_i f(t), \quad (8)$$

where $H_0$ is the unperturbed Hamiltonian, $f(t)$ is a random function of $t$, $\delta_i$ is the perturbation energy with $i=0$ for the ground state, and $i=1$ for the excited state.

For the equilibrium case, Blume derives the emission probability

$$P(\omega)=\frac{2}{\Gamma}\,\mathrm{Re}\int_0^\infty dt e^{i\omega t-(\Gamma/2)t}\overline{\langle V^{(-)}V^{(+)}(t)\rangle}, \quad (9)$$

where the bar indicates an average with respect to all functions $f(t)$, so that

$$\overline{\langle V^{(-)}V^{(+)}(t)\rangle}=e^{i(E_0-E_1)t}\langle V^{(-)}V^{(+)}\rangle\langle e^{i(\delta_0-\delta_1)\int_0^t f(t')dt'}\rangle_{\rm av}. \quad (10)$$

For convenience, we have selected units for which $\hbar=1$. Blume[9] also shows that the following identity holds for any value of $\alpha=\delta_0-\delta_1$:

$$\langle e^{i\alpha\int_0^t f(t')dt'}\rangle_{\rm av}=\left(\cos x\Omega t+\frac{1}{x}\sin x\Omega t\right)e^{-\Omega t}=G(\Omega,\alpha,t), \quad (11)$$

where

$$x=\left[\frac{\alpha^2}{\Omega^2}-1\right]^{1/2}. \quad (12)$$

$\Omega$ is the relaxation rate, or rate of jumping between values of $f(t)=+1$ and $-1$, with equal probabilities of finding the system in either $+1$ or $-1$. The identity given in Eq. (11) can be used to calculate the average on the right side of Eq. (10) and the emission probability (line shape) given by Eq. (9).

To apply Eq. (11) to inhomogeneous broadening, we further assume that $\alpha$ is a random variable with a probability density $\gamma$. In this paper, we consider both Gaussian and Lorentzian distributions as examples. For a Gaussian distribution, we characterize the broadening by the standard deviation $\sigma$. For a Lorentzian distribution, the broadening is characterized by the inhomogeneous broadening parameter[12] $a$. Thus, for a Gaussian distribution

$$\gamma(\alpha,\sigma)=\frac{1}{\sigma\sqrt{2\pi}}\,e^{-\alpha^2/2\sigma^2}.$$

The inhomogeneous broadening factor is given by

$$\overline{G(\Omega,\sigma,t)}=\int_{-\infty}^{\infty}\gamma(\alpha,\sigma)G(\Omega,\alpha,t)\,d\alpha, \tag{13}$$

which can be used with Eq. (9) to calculate the inhomogeneously broadened line.

We can now examine two special cases: $\Omega/\alpha\gg 1$ and $\Omega/\alpha\ll 1$. From Eq. (11), for $\Omega\gg\alpha$, $G(t)\approx 1$ and the inhomogeneous broadening effect disappears. For $\Omega\ll\alpha$, $G(t)\approx 1/2(e^{i\alpha t}+e^{-i\alpha t})$ and with $\alpha$ as a random variable distributed so that it represents the appropriate inhomogeneous broadening in Eq. (13), the maximum broadening effect occurs.

For values of $\Omega/\alpha$ between these two extremes, we do not get such simple results, and a complete calculation using Eqs. (9), (10), (11), and (13) is required to obtain the line shape of an inhomogeneously broadened system in the presence of relaxation. Figure 1 depicts the effect of such fluctuations in the fields causing relaxation between resonances. The first column shows the effect of fluctuations on the absorption cross section at a particular site. The second column shows the effect on the inhomogeneously broadened system of nuclei with a Gaussian distribution of $\sigma$ (the positions of the resonances shown in the first column indicated by arrows). The third column shows the observed line shape in a Mössbauer experiment when both the source and absorber have the same amount of inhomogeneous broadening. In row (a), $\sigma=0$, implying no inhomogeneous broadening. The maximum effect is observed. For rows (b)–(f), the inhomogeneous broadening is assumed constant at $\sigma=20$, and the relaxation rate, $\Omega$, varies from 0.001 to 500, in units of the natural lifetime. At high relaxation rates [Fig. 1(f)], the inhomogeneous broadening is essentially wiped out and the line shape approaches the result obtained when no broadening is present [Fig. 1(a)]. For arbitrary $\sigma$, the relaxation rate has to be much greater than the inhomogeneous broadening ($\Omega\gg\sigma$) for the broadening effects to be wiped out.

### B. Effect on the SF intensity

To illustrate the effect of time-dependent perturbations on the SF pulse, we generalize Eq. (10) by introducing the random functions $f_i(t)$, which represent mutually independent random processes that are otherwise identical with $f(t)$, and by replacing $\Delta\omega_i$, for all $i$, with $\alpha f_i(t)$. To simplify the calculation we assume in addition $\phi_j=\phi_k$, thus ignoring spatial phase variations. We then get from Eq. (6)

$$\overline{\hat{I}_{N^2}(t)}=\langle V^{(-)}V^{(+)}\rangle e^{-\Gamma t}\sum_{j\neq k}\langle e^{i\alpha\int_0^t f_j(t')dt'-i\alpha\int_0^t f_k(t')dt'}\rangle_{\rm av}. \tag{14}$$

Since the exponential factors on the right-hand side of Eq. (14) are statistically independent, the average of the product is equal to the product of the averages; therefore,

$$\overline{\hat{I}_{N^2}(t)}=(N^2-N)\langle V^{(-)}V^{(+)}\rangle e^{-\Gamma t}[\langle e^{i\alpha\int_0^t f(t')dt'}\rangle_{\rm av}]^2. \tag{15}$$

The average can then be calculated as in Eq. (13). After approximating $N^2-N$ by $N^2$, the result is

$$\overline{\hat{I}_{N^2}(t)}=N^2\langle V^{(-)}V^{(+)}\rangle e^{-\Gamma t}\overline{G(\Omega,\alpha,t)}^2, \tag{16}$$

where $G(\Omega,\alpha,t)$ is given by Eq. (11).

As expected from the previous emission results based on Eq. (11), for $\Omega\gg\alpha$, the inhomogeneous broadening effect that destroys the coherence disappears, and a maximum SF pulse is obtained. For $\Omega\ll\alpha$, the maximum phase destruction occurs and the SF pulse disappears.

We now examine the dependence of the reduction factor $\overline{G(\Omega,\sigma,t)}^2$ on the inhomogeneous broadening that produces dephasing (characterized by $\sigma$) and the relaxation that reduces the dephasing (characterized by the relaxation rate $\Omega$).

In Fig. 2, we show $\overline{G(\Omega,\sigma,t)}^2$ with parameters $\Omega=0$ and $\sigma=0$, 1, 10, or 100, plotted as a function of time normalized to the natural lifetime $\tau_n$. The horizontal line labeled $\sigma=0$ shows no reduction in the SF pulse intensity because no inhomogeneous broadening exists. The other curves show the decrease in SF pulse intensity when different dephasing rates caused by different amounts of inhomogeneous broadening are present.

Figure 3 shows the effect of relaxation on the reduction factor when $\sigma=10^3$ for different values of the relaxation rate, $\Omega$. The curves decay rapidly for small values of $\Omega$ and less rapidly as $\Omega$ increases, approaching the limit of 1.0 as $\Omega$ approaches infinity. The results shown in Fig. 3 indicate that time-dependent hyperfine interactions can reduce the effect of inhomogeneous broadening. As the relaxation rate, $\Omega$, increases, the intensity factor also increases. In the limit of $\Omega\gg\sigma$, the SF pulse intensity approaches unity, which is the value it would have all the time if no inhomogeneous broadening ($\sigma=0$) were present.

### C. Effect on the SF pulse shape

In Ref. 5, we showed that inhomogeneous broadening degrades the SF pulse shape and that this effect is more pronounced than the degradation resulting from homogeneous broadening. In the previous sections, we also showed how this effect of inhomogeneous broadening can be mitigated through time-dependent hyperfine interactions.

In this section, we employ the Maxwell-Bloch equations developed in Ref. 5 and summarized in the Appendix here to show how the time-dependent effects alter the total SF pulse shape. With this approach, we can study the effect of time-dependent interactions in inhomogeneously broadened systems on the complete SF pulse shape, i.e., time delay, multiplicity of peaks, and the peak intensities. In this formalism, the effect of both the inhomogeneous broadening and time-dependent hyperfine interactions is contained in what was originally a ''coupling constant'' (in the Appendix) and is now a time-dependent function $g'(t)$, as discussed in Ref. 5. We use Eqs. (11) and (13) to calculate $g'(t)=\overline{G(\Omega,\sigma,t)}$ which replaces the constant $g$ in Eq. (1) of Ref. 5. The SF pulse shape is calculated using the resulting equation.

Figure 4 shows some typical results. The solid curves give the result obtained when a Lorentzian distribution[13] is assumed for the inhomogeneous broadening, and the dashed

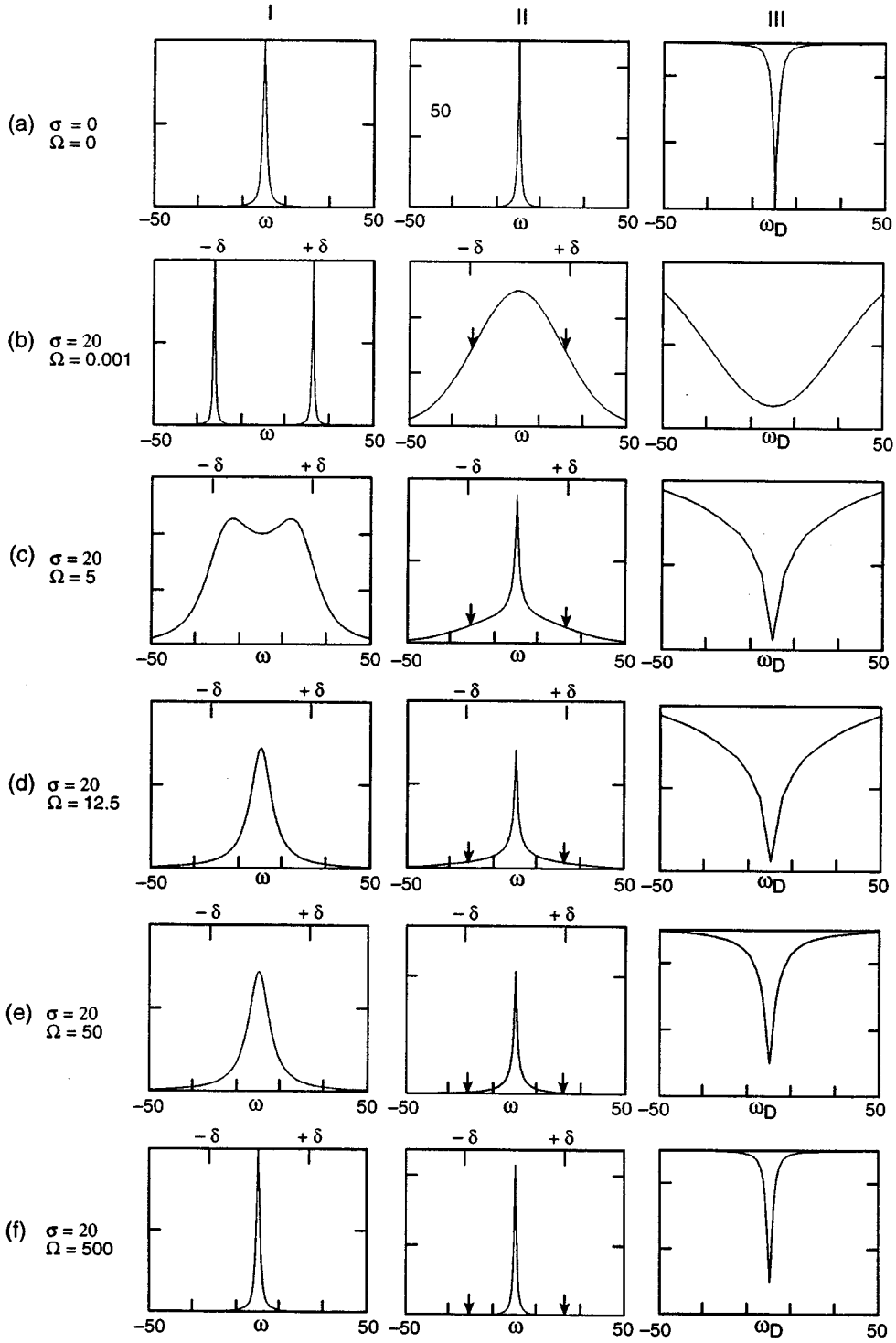


FIG. 1. Effect of relaxation on inhomogeneously broadened lines. Column I shows two resonances that contribute to the inhomogeneously broadened line (absorption cross section) shown in column II. Column III shows the Mössbauer line shape, assuming both the source and absorber are broadened as in column II. Rows (a)–(e) give the results for different values of broadening and relaxation. Row (a) shows the result when there is no broadening ($\sigma=0$) and no relaxation ($\Omega=0$). Rows (b)–(f) show the result when $\sigma=20$ and the relaxation increases ($\Omega=0.01-500$). For all these results, the natural linewidth is assumed to be $\Gamma=1$.

curves give the result when a Gaussian distribution is assumed. The curve labeled (a) is obtained assuming no inhomogeneous broadening and no relaxation ($a=0$, $\Omega=0$). The curves labeled (b)–(e) are obtained by increasing inhomogeneous broadening and exhibit the degradation of the SF pulse. We next consider the recovery of the SF pulse.

Figure 5 shows the effect of relaxation on the system with an inhomogeneous broadening parameter $a=10^3$. The curve labeled (a) is obtained with some relaxation present ($\Omega=0.001$). The curves labeled (b)–(e) are obtained with increasing relaxation. At $\Omega=10^4$ [curve (e)], the optimal SF [curve 4(a)] is almost fully recovered.

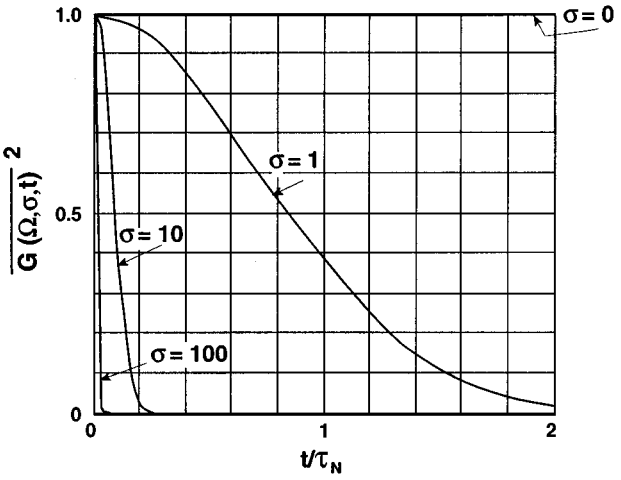


FIG. 2. SF pulse emission intensity reduction as a function of inhomogeneous broadening and time. For each curve shown, $\Omega = 0$ and $\sigma = 0$, 1, 10, or 100.

## IV. CONCLUSIONS

The number of cooperating nuclei and the inherent nuclear properties determine the SF pulse intensity. Our results show that if a candidate exists that, when inverted, would produce SF were it not for inhomogeneous broadening, it is still possible to get an SF emission if a fast enough relaxation of the levels producing the inhomogeneous broadening can be realized.

Our calculations show that inhomogeneous broadening affects an SF emission by causing a dephasing of the different nuclear dipoles that are forming correlations. This dephasing is time dependent and has a rate proportional to the inhomogeneous broadening. Immediately after inversion, SF emission is less affected by the broadening than later. Thus, for a strong SF pulse, the SF delay time should be much shorter than the dephasing time $\tau_{SF} \ll T_2^*$.

We also show that time-dependent hyperfine interactions of the nucleus with electrons can induce a reduction of the dephasing and an increase in the probability of SF emission

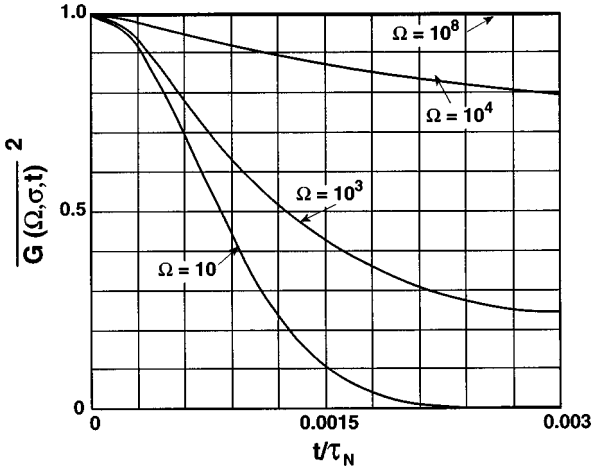


FIG. 3. SF pulse emission intensity reduction as a function of inhomogeneous broadening and relaxation. For each curve shown, $\sigma = 10^3$ and the relaxation rate $\Omega = 10$, $10^3$, $10^4$, or $10^8$.

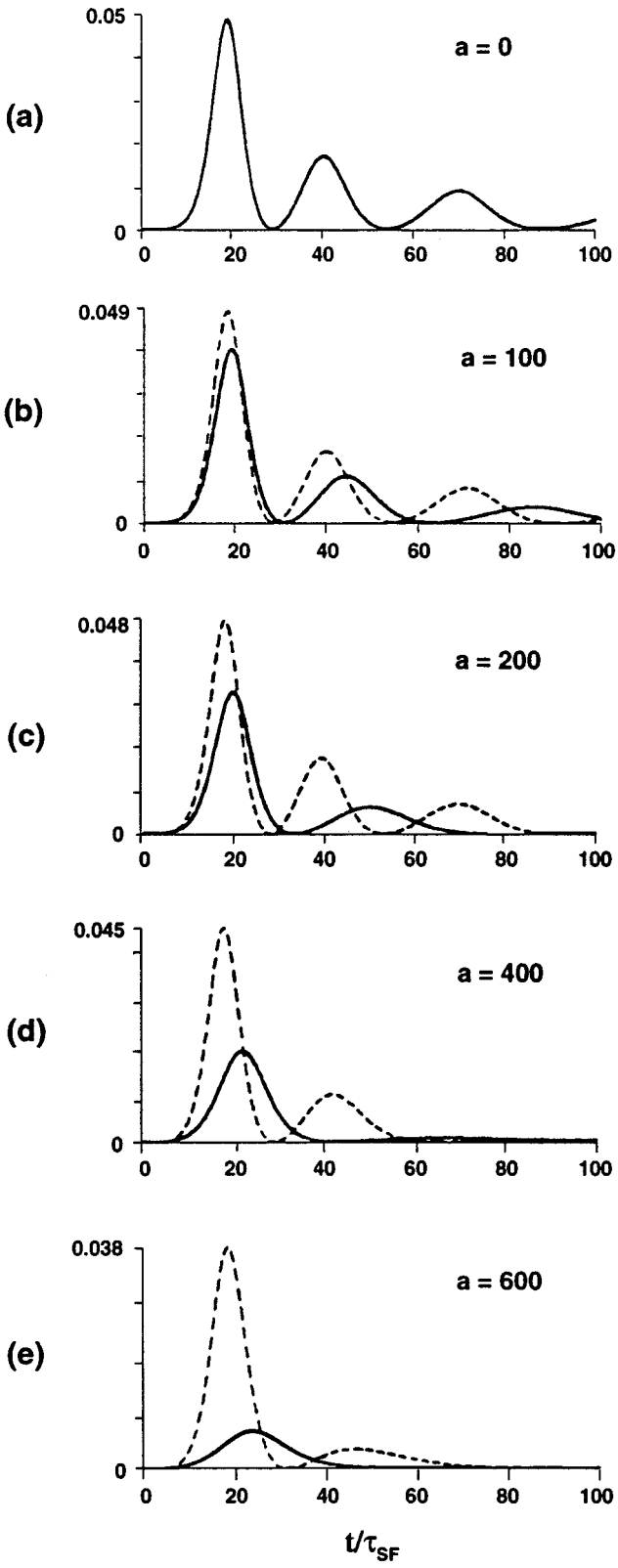


FIG. 4. SF pulse shape in the presence of inhomogeneous broadening. (a)–(e) show the results for increasing broadening characterized by the full width at half maximum in units of the natural unbroadened linewidth $\Gamma$ or the inhomogeneous broadening parameter $a$. The solid lines give the results for a Lorentzian distribution, and the dashed lines give the results for a Gaussian distribution.

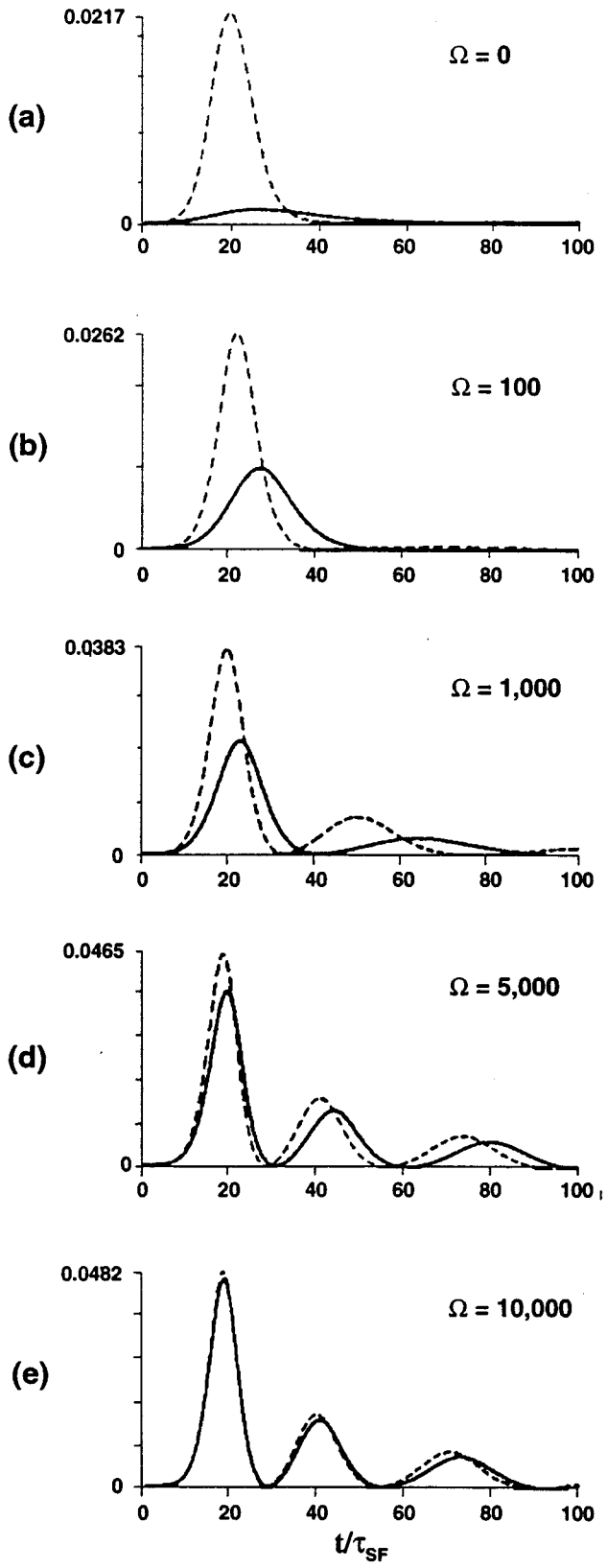


FIG. 5. SF line shape in the presence of inhomogeneous broadening and relaxation. (a) shows the SF pulses when inhomogeneous broadening with $a=1000$ is present. (b)–(e) show the pulses for increasing values of the relaxation rate $\Omega$ given in units of $\Gamma$. The solid lines give the results for a Lorentzian distribution, and the dashed lines give the results for a Gaussian distribution.

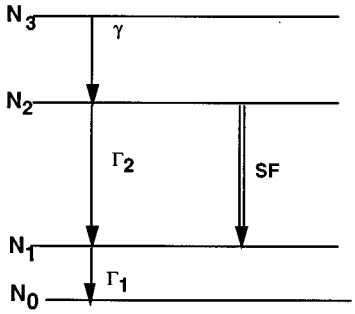


FIG. 6. Energy-level structure assumed in the calculation discussed in this paper.

over a large time range. These electronic relaxation effects are well known for their line-shape modification as observed in Mössbauer and nuclear magnetic resonance experiments.[9,14–18] For SF, relaxation effects provide a means of reducing the dephasing caused by inhomogeneous broadening and, thus, may be useful in overcoming one of the more difficult obstacles to nuclear SF.

## ACKNOWLEDGMENT

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## APPENDIX: THE NUCLEAR SUPERFLUORESCENCE MODEL

In this appendix, we present a summary of the modified Haake-Reibold Model (Maxwell-Bloch equations) of nuclear SF for reference. The model is discussed in detail in Ref. 5. The nuclear transitions are shown in the energy-level diagram given in Fig. 6, where $N_0$, $N_1$, $N_2$, and $N_3$ represent the populations of the levels and $\gamma$, $\Gamma_1$, $\Gamma_2$, and the transition rates between levels as shown. The pumping mechanism is modeled by the transition from level 3 to level 2. The SF transition occurs between levels 2 and 1. The modified Haake-Reibold equations in dimensionless units are

$$\frac{\partial N_3}{\partial t}=\gamma N_3, \tag{A1}$$

$$\frac{\partial N_2}{\partial t}=-(E^+R^++E^-R^-)-\Gamma_2N_2+\gamma N_3, \tag{A2}$$

$$\frac{\partial N_1}{\partial t}=+(E^+R^++E^-R^-)+\Gamma_2N_2-\Gamma_1N_1, \tag{A3}$$

$$\frac{\partial N_0}{\partial t}=\Gamma_1N_1, \tag{A4}$$

$$\frac{\partial R^\pm}{\partial t}=(N_2-N_1)E^\mp-\frac{1}{2}(\Gamma_2+\Gamma_\phi)R^\pm+\xi^\pm, \tag{A5}$$

$$\frac{\partial E^\pm}{\partial x}=g'(t)R^\mp-\frac{1}{2}\mu E^\pm. \tag{A6}$$

The first set of equations, (A1)–(A4), govern the time rate of change of the level populations. Equation (A5) determines the rate of buildup of the system polarizations $R^{\pm}$, resulting from the electric fields $E^{\mp}$ and a noise source $\xi^{\pm}$. A representation of the noise source used in this model has been derived from quantum electrodynamic considerations by Polder, Schuurmanns, and Vrehen.[3] The last equation, Eq. (A6), governs the spatial transport of the fields and connects the system polarization with the electric-field gradient through a coupling parameter $g'(t)$. This coupling parameter is time dependent because we assume inhomogeneous broadening; otherwise, it would be a constant as in the original Haake-Reibold theory. When the line broadening statistical distribution is Lorentzian, the time-dependent parameter is given by an exponential function as discussed in Ref. 5. In the present paper we also deal with Gaussian distributions and relaxation effects which also modify $g'(t)$. The second term in Eq. (A6) models the attenuation of the beam as it propagates through the system. In the equation, $\mu$ is the linear attenuation coefficient, and $\Gamma_{\phi}$ is the homogeneous broadening dephasing rate. Inhomogeneous and homogeneous broadening play quite different and distinct roles in this model, unlike some of the earlier models where they were assumed to have essentially indistinguishable effects on the emission of SF.

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[11] The assumption of a Lorentzian line shape is a mathematical convenience and provides us with an exponential time dependence. It was also used in Ref. 3 to obtain the exponentially varying coupling factor $g(t)$. Other line shapes may be more appropriate in specific cases. In this paper, we compare the effects of Lorentzian and Gaussian distributions.

[12] The inhomogeneous broadening parameter characterizes the broadening in units of the natural linewidth so that the full width at half maximum of the broadened line (Lorentzian distribution) is $(1+a)\Gamma$. For a Gaussian distribution, it is related to the standard deviation, $\sigma$, by $a=\sqrt{2\ln 2}\,\sigma/\Gamma$.

[13] For a Lorentzian distribution, $\gamma(\alpha, a)=a\Gamma/2\pi/[\alpha^2+(a\Gamma/2)^2]$.

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