

## Nuclear superfluorescence: A feasibility study based on the generalized Haake-Reibold theory

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A theory of nuclear superfluorescence (SF) is presented which includes electronic attenuation, competing transitions, homogeneous and inhomogeneous broadening, and finite pumping times. The effects of the nuclear and atomic parameters on the expected emitted SF pulse shape are analyzed in physically realizable regimes. A number of explicit calculations are made to illustrate the behavior of the pulse under various conditions. The feasibility of observing superfluorescence using the 58.59 keV transition in  $^{60}\text{Co}$ , is examined.

### I. INTRODUCTION

#### A. Background

Superfluorescence (SF) and superradiance<sup>1-4</sup> are both examples of the cooperative, spontaneous emission of coherent radiation by identical radiators such as atoms, molecules, or nuclei. Intense, directed pulses of duration much shorter than the spontaneous emission lifetime of an individual radiator characterize the radiation, which has been observed experimentally in molecular and atomic systems,<sup>2</sup> e.g.,  $\text{CH}_3\text{F}$ , HF, Na, Cs, Tl,  $\text{KClO}_2$ , Sr, and Li. Theoretical models that successfully explain the observations have been developed.<sup>5</sup>

The possibility of nuclear SF was suggested by Terhune and Baldwin<sup>6</sup> as early as 1965. However, nuclear SF has not been observed, and models used to study the nuclear phenomena are not as sophisticated as those developed for atomic SF nor do they treat all of the relevant phenomenology.

Trammell and Hannon<sup>7</sup> used a modification of the steady-state laser theory to treat amplified spontaneous emission (ASE) and early versions of the mean-field theory of SF to derive conditions for the existence of nuclear SF. They concluded that the conditions for nuclear SF are just as restrictive as those for ASE or pulsed nuclear lasers.

Recently Baldwin and Feld<sup>8</sup> used results obtained from a semiclassical theory of SF to derive conditions for nuclear superfluorescence. The dynamics of the phenomena are not fully developed in their treatment: they assume instantaneous inversion, and some of the more important experimentally determined parameters are treated in an *ad hoc* fashion or ignored.

We believe that a more sophisticated full treatment of nuclear SF that includes experimentally achievable parameters interacting in a consistent way is required to determine feasibility of nuclear SF. These parameters and their importance for nuclear as opposed to atomic SF are discussed in Sec. II. For our theoretical treatment we rely on the Maxwell-Bloch equations given by Haake and

Reibold<sup>9</sup> to treat SF phenomena for a multistate system of emitters. Our version of the model has modifications required for the nuclear case and is discussed here in Sec. III.

#### B. Theoretical models and requirements for nuclear superfluorescence

The version of the Haake-Reibold model used here treats one-dimensional propagation for the four-level system of emitters. Since quantum electrodynamics plays a major role in its derivation, it should also cover the initial stage in the generation of SF adequately. The model also takes into account certain effects, such as losses due to internal conversion, that are negligible in the atomic case but are important in the nuclear case.

The detailed characteristics of nuclear inversion pumping, radiation emission, and transport through media have an impact on the generation of nuclear SF pulses. In particular, the following special issues that differentiate nuclear SF from the well-studied atomic SF are important.

Atomic transition energies used in lasers are generally on the order of 1 to  $10^3$  eV (wavelengths on the order of  $10\text{--}10^4$  Å) but nuclear transitions of interest are on the order of  $10^3\text{--}10^5$  eV (wavelengths on the order of 0.1–10 Å). Trammel<sup>10</sup> has discussed the issue of short wavelengths of the emitted radiation as compared to the inter-nuclear spacing. It is generally ignored by other authors dealing with nuclear SF,<sup>7,8</sup> although it is certainly controversial, and often misunderstood, as pointed out in the series of letters to the editor following an article on the subject by Lipkin.<sup>11</sup>

A typical atom with resonance energy on the order of an eV will recoil with an energy which is much smaller than the typical atomic linewidths. On the other hand, the same atom in a typical nuclear transition will recoil with an energy which is orders of magnitude larger than the range of typical nuclear linewidths. The result is that while the emitting atom is in resonance with the other atoms in the active region, the emitting nucleus will be

out of resonance with the other nuclei. Emission with recoil will be treated in our formalism as a competing process which has the effect of reducing the excited-state population. Under certain conditions (Mössbauer effect)<sup>12</sup> a nucleus can emit without recoil, strongly resonate with other nuclei and participate in SF.

The transitions of interest in nuclear SF are generally of high multipolarity ( $M1, E2, M2$ , or higher) unlike atomic transitions of interest, for which the electric dipole is assumed to be responsible for the emission. All of the theories of SF known to the authors up to now have dealt with electric dipole transitions. Although complications with high multipoles could arise in cases where competing transitions occur, problems of high multipolarity SF transitions will not be considered in this paper. A recent work by Huang and Eberly<sup>13</sup> suggests some of the complications that may be involved.

Reference 9 considers the effect of several transitions competing for the depopulation of a level in theoretical SF calculations for atomic systems. In nuclear systems internal conversion is not only common but strongly competes with the radiation transition in the depopulation of low-energy nuclear states.<sup>14,15</sup> Internal conversion, analogous to the Auger effect in atoms, plays an especially important role in low-energy nuclear transitions. It must be considered in discussing nuclear SF. Other competing electromagnetic transitions are also common in nuclear systems and can be treated similarly.

For the cases of interest, the linear attenuation of the propagating electric field is much higher for the energy range involved in nuclear transitions ( $10^3$  to  $10^5$  eV) than in the energy range involved in atomic transitions (1 to  $10^3$  eV). Multiple passes in an optical cavity of a few centimeters can be achieved in the atomic case, while nuclear radiation is often limited to a path length of less than a millimeter.<sup>16</sup> Fortunately, even though the attenuation of short-wavelength radiation is high, commonly for typical materials on the order of  $10^2$ – $10^3$   $\text{cm}^{-1}$  for the wavelengths of interest, in crystals for special propagation directions the Borrmann effect<sup>16,17</sup> can reduce the attenuation by two or three orders of magnitude.

The pumping problem for SF is severe, and pumping times on the order of the lifetime of the lasing level are usually required. Instantaneous (shorter than the lifetime) pumping to inversion is not expected to be feasible in the nuclear case. Realistic pumping times are important and must be included in the analysis of nuclear SF.

Although for the convenience of analysis, an acicular shape (long thin cylinder) for the active region that radiates SF pulses is useful, it may not be realistic in terms of the attainable experimental conditions. The acicular geometry permits the restriction of analysis to a single mode emission and when the Fresnel number  $F=1$  guarantees the optimum diffraction limited condition.<sup>18</sup> To achieve a condition for the active region such that  $F=1$  may be difficult experimentally.

Collisions with electrons, finite pump bandwidth, and broadband crystal phonons contribute to homogeneous line broadening. In addition, inhomogeneous broadening due to several causes occurs more severely in the case of nuclear than in atomic transitions.

In this paper we will discuss the effect on nuclear SF of (1) competing transitions (e.g., internal conversion, emission with recoil, branching ratios),<sup>19</sup> (2) transport effects (electronic attenuation), (3) finite pumping times using incoherent sources, and (4) homogeneous and inhomogeneous line broadening. These issues are of secondary interest for atomic SF, but they present critical problems for the development of nuclear SF.

## II. HAAKE-REIBOLD STOCHASTIC SOURCE APPROACH TO NUCLEAR SF

The Haake-Reibold model of SF described in this section is a generalization of the one introduced by those authors in Ref. 9. Their paper, which treats multilevel systems, refers to earlier work concerned with two-level systems as the basis for their approach, particularly that reported in Ref. 20.

The Maxwell-Bloch equations derived in Ref. 20 are similar to the semiclassical equations of Ref. 1, which included an *ad hoc* noise source associated with the atomic polarization. The source, which dies out before the nonlinear interaction between the electric field and the polarization becomes effective, is responsible for an initial dipole moment that triggers the SF pulse.

More rigorously, Ref. 20 proves that the quantum electrodynamic fluctuations of the initial electric-field vacuum state are, in fact, equivalent to a stochastic noise source that decays quickly, thus supporting the Ref. 1 concept in a general sense. However, the proof goes further: it demonstrates that the source represents zero mean, Gaussian distributed noise and derives the associated variance as a function of time. In the course of this analysis Ref. 20 also predicts the effect of homogeneous broadening and gives an explicit value for the natural lifetime of the system of emitters.

The Haake-Reibold model also introduces a stochastic, zero mean, Gaussian distributed noise source to replace the quantum vacuum state electric-field fluctuations as a means of starting the collective spontaneous emission that characterizes SF, using the result of Ref. 20 for this purpose. In addition, the introduction of other source terms of the same type provides the multilevel model with incoherent depletion effects at lower energy levels.

Reference 9 determines the second-order statistics of the quantum electric field and polarization operators at early times when the Maxwell-Bloch equations are effectively linear. Then, treating the Maxwell-Bloch equations classically, (i.e., the polarization and electric field are regarded as  $c$  numbers), it uses the results to generate a statistical distribution of initial values for calculating a corresponding distribution of solutions that are valid at later times when quantum effects are no longer important but the nonlinear effects are. Essentially, this is a Monte Carlo approach to estimating various emitted radiation parameters, such as the delay time of the radiated pulse.

Although restricted to one spatial dimension, the Haake-Reibold model accounts explicitly for propagation of the emitted electromagnetic field as long as the geometrical configuration of the emitters limits the radiation to

an endfire mode. For this purpose the assumed shape of the active region is acicular with an associated Fresnel number of the order of one. The effect is to restrict the field to plane waves propagating to the left or to the right in the longitudinal direction. The model assumes that these waves do not interact (i.e., that it is only necessary to consider one of them in calculating the radiation at a single end of the active region). Figure 1 shows the angular distribution of radiation expected from an acicular geometry for both SF emissions and emission due to ordinary spontaneous decay.

The most general multistate system of interest is one with four states for which the energy diagram is depicted in Fig. 2. In this scheme at the beginning state 4 is totally inverted relative to state 1. The decay of state 4 populates state 3, whose subsequent decay produces the radiations of interest.

The corresponding Haake-Reibold (Maxwell-Bloch) equations in dimensionless units are

$$\frac{\partial}{\partial t} N_4 = -\gamma N_4 + \xi_4, \quad (1a)$$

$$\frac{\partial}{\partial t} N_3 = -(E_{32}^+ R_{32}^+ + E_{32}^- R_{32}^-) - \Gamma_3 N_3 + \gamma N_4 - \xi_4, \quad (1b)$$

$$\frac{\partial}{\partial t} N_2 = +(E_{32}^+ R_{32}^+ + E_{32}^- R_{32}^-) - \Gamma_2 N_2 + \Gamma_3 N_3, \quad (1c)$$

$$\frac{\partial}{\partial t} N_1 = +\Gamma_2 N_2, \quad (1d)$$

$$\frac{\partial}{\partial t} R_{32}^\pm = (N_3 - N_2) E_{32}^\mp - \frac{1}{2} [\Gamma_3 + \Gamma_2 + \Gamma_\phi] R_{32}^\pm + \xi_{32}^\pm, \quad (1e)$$

$$\frac{\partial}{\partial x} E_{32}^\pm = g R_{32}^\mp - \frac{1}{2} \mu E_{32}^\pm, \quad (1f)$$

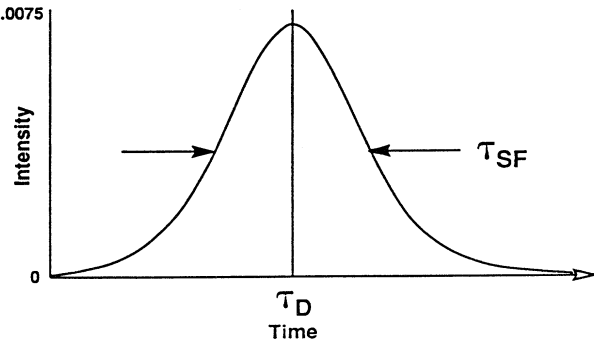


FIG. 1. Superfluorescent pulse shape and angular distribution of radiation. We show a single pulse here for simplicity although, under some conditions, multiple pulses can occur.

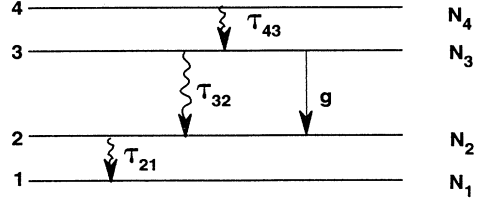


FIG. 2. Energy-level structure assumed in the calculations described in the paper. The SF transition is between upper state 3 and lower state 2 as indicated by  $g$ . Level 4 populates level 3, providing the inversion. The decay rates  $\gamma$ , and  $\Gamma_3$ ,  $\Gamma_2$ , are given by  $\tau_{43}^{-1}$ ,  $\tau_{32}^{-1}$ , and  $\tau_{21}^{-1}$ , respectively. Other depopulation transitions are generally allowed. We indicate only those of interest at present.

which are a generalized version of analogous equations introduced in Ref. 9. In (1) the  $N_i$  are population number density operators, the  $R_{32}^\pm$  are polarization operators, and the  $E_{32}^\pm$  are photon field operators. The time is retarded, so that

$$t = t' - \frac{x'}{c}, \quad x = x', \quad \frac{\partial}{\partial x} = \frac{\partial}{\partial x'} + \frac{1}{c} \frac{\partial}{\partial t'}, \quad \frac{\partial}{\partial t} = \frac{\partial}{\partial t'},$$

where  $x'$  and  $t'$  are the space and time coordinates, normalized to the superfluorescence time given by<sup>9</sup>

$$\tau_{\text{SF}} = \frac{8\pi\tau_r}{3\rho\lambda^2 l}. \quad (2)$$

In (2)  $\rho$  is the density of cooperating radiators,  $\lambda$  is the wavelength of the emitted photons,  $l$  is the length of the active region, and  $\tau_r$  is the radiative lifetime, which is given by

$$\tau_r = (1 + \alpha)\tau_0 \quad (3)$$

in terms of the natural lifetime  $\tau_0$  and the internal conversion parameter  $\alpha$ .

In Fig. 2,  $\tau_{ij}$  is the spontaneous emission lifetime of the transition from state  $i$  to state  $j$ . The corresponding emission rates in (1) are  $\gamma$ ,  $\Gamma_3$ , and  $\Gamma_2$  from states 4, 3, and 2 and  $g$  is the coupling constant associated with the transition from state 3 to state 2. The quantity  $\Gamma_i$  is a spontaneous decay rate that is not due to the collective emission process; but takes into account the effect of non-collective processes, such as internal conversion and fluorescence. The parameter  $\mu$ , is a spatial attenuation coefficient and  $\Gamma_\phi$  is a generalized collisional homogeneous line broadening decay constant, which acts as a dephasing parameter. In addition, the coupling parameter  $g$ , which in the Haake-Reibold theory is constant, in effect becomes a time-dependent quantity  $g'$  in the presence of inhomogeneous broadening.<sup>3,21,22</sup> When the line broadening statistical distribution is Lorentzian, the time-dependent parameter is given by  $g'(t) = g \exp[-1/2\Gamma_\theta t]$ , where  $\Gamma_\theta$  is the inhomogeneous broadening decay constant.<sup>23</sup>

The decay constants  $\Gamma_\phi$  and  $\Gamma_\theta$  can be written as proportional to the natural line width broadening parameters  $b$  and  $a$ , i.e.,  $\Gamma_\phi = b\Gamma$  and  $\Gamma_\theta = a\Gamma$  where  $\Gamma = \Gamma_3 + \Gamma_2$ .

Reference 22 shows how the inhomogeneous broadening effect can be reduced through relaxation or time-dependent hyperfine interactions.

The inhomogeneous term  $\xi_4$  in (1a) and (1b) is a noise source<sup>24</sup> associated with pumping that produces incoherent fluctuations in the populations of energy levels 3 and 4. The inhomogeneous terms of the form  $\xi_{32}^\pm$  represent the noise sources characterizing the initially important quantum field vacuum state fluctuations that trigger the SF process.<sup>25</sup>

Reference 9 assumes that the quantities  $\xi_4$  and  $\xi_{32}^\pm$ , although quantum-mechanical operators, are stochastic in the classical sense<sup>25</sup> with the following properties. They are statistically independent and Gaussian, with zero means and the second moments

$$\langle \xi_{32}^+ \xi_{32}^+ \rangle = \langle \xi_{32}^- \xi_{32}^- \rangle = \langle \xi_{32}^- \xi_{32}^+ \rangle = 0, \quad (4a)$$

$$\begin{aligned} \langle \xi_4(x, t) \xi_4(x', t') \rangle &= \langle \xi_{32}^+(x, t) \xi_{32}^-(x', t') \rangle \\ &= (1/N) \langle N_4(t) \rangle \gamma \delta(x - x') \delta(t - t') \\ &= (1/N) e^{-\gamma t} \gamma \delta(x - x') \delta(t - t'). \end{aligned} \quad (4b)$$

A comparison of (4a) with (4b) verifies that the quantities  $\xi_{32}^+$  and  $\xi_{32}^-$  are, in fact, operators rather than  $c$  numbers, since they do not commute. The term  $\gamma$  in (4b), (1a), and (1b) is dimensionless and is defined by the ratio of the superfluorescence time to the natural decay time from level 4 to level 3:

$$\gamma = \tau_{\text{SF}} / \tau_{43}. \quad (5)$$

The quantity  $N$  in (4b) is the number of radiators.

The initial state  $|0\rangle$  is the vacuum state for the electromagnetic field and an occupation number eigenstate of the number density operators  $N_2, N_3, N_4$ . Initial conditions for these operators are given by

$$\begin{aligned} E_{31}^\pm(x, 0)|0\rangle &= E_{32}(x, 0)|0\rangle = N_2(x, 0)|0\rangle \\ &= N_3(x, 0)|0\rangle = 0, \\ N_4(x, 0)|0\rangle &= |0\rangle. \end{aligned} \quad (6)$$

In addition, boundary conditions implying that no external signals impinge on the system are imposed:

$$E^\pm(0, t) = 0. \quad (7)$$

The objective is to calculate the mean radiation intensity  $I(t)$  at the right end of the active region where

$$I(t) = \langle E^-(l, t) E^+(l, t) \rangle.$$

Although the polarization sources  $\xi_{32}^\pm$  and the population source  $\xi_4$  are statistically independent, Ref. 8 assumes that the parameters defining their probability distributions (i.e., the first and second moments) are identical. This is presumably justified by the fact that the same radiation field reservoir, consisting initially of the vacuum state electric field, generates both.

### III. CALCULATED RESULTS

In this section we describe some results obtained by means of numerical (Monte Carlo) calculations applied to

the system of Eqs. (1). The calculations were designed to investigate and predict the SF pulse dependence on experimentally meaningful parameters. The ultimate purpose is to select isomer candidates for nuclear SF experiments.

The study concentrated on the effects of a few important parameters. One is the linear attenuation coefficient  $\mu$  ( $\text{cm}^{-1}$ ). Another is the lasing level spontaneous emission rate  $\Gamma_3$ , which depends on both radiative (photon) emission and internal conversion (atomic electron ejection). Others are the number of cooperating nuclei  $N$  and the inhomogeneous broadening coefficient  $a$  as well as the homogeneous broadening coefficient  $b$ , which determine the effective increase in linewidth of the total system in units of the natural linewidth. All of the calculated results are presented in normalized units consistent with the usage in (1). The unit of time is the superfluorescence time  $\tau_{\text{SF}}$  defined by (2).

Figure 1 shows a typical SF pulse and illustrates the definition of the delay time  $\tau_D$ . Comparing our results with the available literature we find that the delay time derived in different developments depends differently on the number  $N$  of cooperating nuclei. Bonifacio and Lugiato<sup>3</sup> and Gross and Haroche<sup>5</sup> use

$$\tau_D = \frac{1}{2} \tau_{\text{SF}} \ln(N); \quad (8)$$

on the other hand, Polder, Schuurmans, and Vreken<sup>20</sup> derived the expression

$$\tau_D = \frac{1}{4} \tau_{\text{SF}} [\ln \sqrt{2\pi N}]^2 \quad (9)$$

for the delay time. They state that the  $\ln^2(N)$  dependence is due to taking into account propagation effects that Bonifacio-Lugiato do not include in their model. From our calculations of the Maxwell-Bloch equations (1), with random sources and including propagation effects, we find that the time delay is given by

$$\bar{\tau}_D = c_1 [1 + c_2 \ln(N)] \ln(N) \quad (10)$$

with  $c_1 = 2.43$  and  $c_2 = 0.02$  providing the best fit to the data resulting from the calculations. The time delays obtained from the three models are compared in Fig. 3. For large  $N$  the effect of the  $\ln^2(N)$  term in (10) becomes important but for  $N = 10^5$  this term contributes only about 20% of the  $\ln(N)$  term's contribution.

We examined the variation in the individual pulses as the seed of the random number generation was varied for the Monte Carlo calculation. For our work, averaging 25–100 pulses appeared sufficient. The statistics of the variation in delay time  $\tau_D$ , and peak intensity  $I_0$ , are discussed in Ref. 9.

We now consider the pulse characteristics as a function of the pumping rate  $\gamma$ , the linear attenuation coefficient  $\mu$ , and the inhomogeneous broadening parameter  $a$  for different numbers of cooperating nuclei and then present some results that show both the spatial distribution of fields in the active region and the temporal pulse evolution under different conditions. In Fig. 4 we show the pulse emission delay time as a function of the pumping rate for three values of the cooperation number:

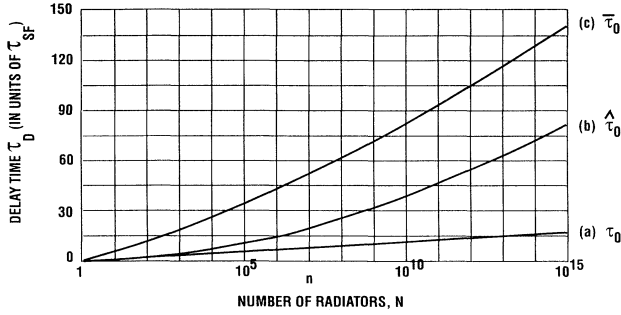


FIG. 3. Comparison of pulse delay times calculated from different theories as in (a) Bonifacio and Lugiato (Ref. 3) and Gross and Haroche (Ref. 5) result [ $\tau_D$  of Eq. (8)], (b) Polder, Schuurmans, and Vreken (Ref. 17) result [ $\tau_D$  of Eq. (9)], (c) Haake and Reibold (Ref. 15) result [ $\tau_D$  of Eq. (10)].

$N = 10^3, 10^8, 10^{12}$ . Even if the pumping is essentially over before the SF pulse peak occurs, i.e., before  $\tau_D$ , the pumping rate has an effect on the pulse shape. In units of  $\tau_{SF}$  for larger  $N$  the delay time increases, which seems counter intuitive; however, in constant physical units (e.g., seconds) the delay time actually decreases because  $\tau_{SF}$  varies as  $1/N$ .

There are two first-order effects expected from a finite linear attenuation. One effect arises because of losses in the electromagnetic field as it propagates through the medium. This would be exhibited in the experimental results as a decrease in the intensity of the emitted SF pulse. The other effect is a result of the suppressed development of the electric field. For increasing values of  $\mu$ , one expects that a longer time would be required for the generation of a large enough  $E$  to initiate the SF pulse. This would be observed experimentally as an increase in the time delay  $\tau_D$ .

Figures 5(a) and 5(b) exhibit those trends, showing the decrease in the peak intensity, which eventually reaches

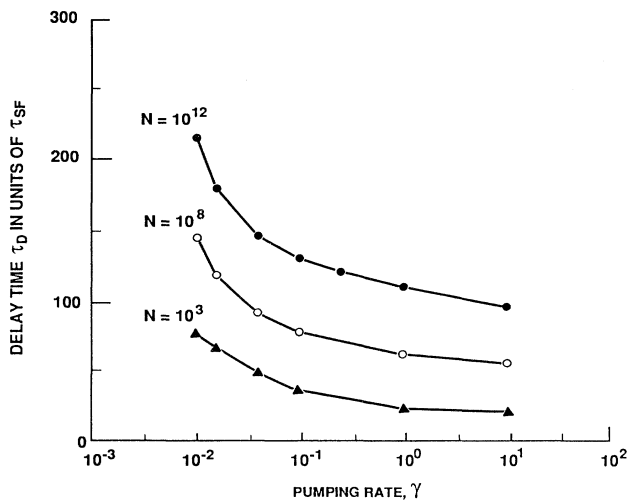


FIG. 4. SF pulse delay time  $\tau_D$  as a function of the pumping rate  $\gamma$  for different values of the cooperation number  $N$ .

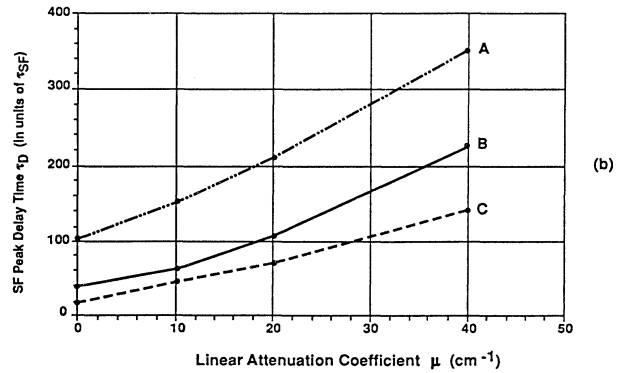
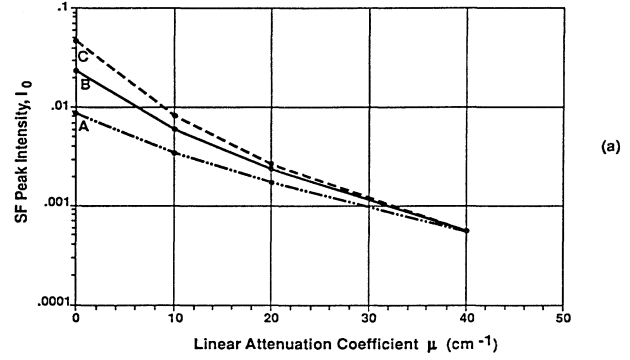


FIG. 5. SF pulse emission delay time  $\tau_D$  (a) and intensity  $I_0$  (b) as a function of the linear attenuation coefficient  $\mu$  for different values of the cooperation number  $N$ . For curves labeled (A)  $N = 10^{12}$ , (B)  $N = 10^6$ , and (C)  $N = 10^3$ .

the value of noncooperative spontaneous emission, and the increase of the delay. It should be kept in mind that the pulse shape broadens as  $N$  increases, which complicates the interpretation of the intensity curves shown.

There are no thresholdlike effects of the attenuation coefficient  $\mu$  on the SF phenomena.<sup>3</sup> If there are no dephasing mechanisms reducing the dipole moment triggered by the quantum fluctuations (i.e., the noise term  $\xi_{32}^{\pm}$ ), the SF pulse peak will decrease because of  $\mu$  but will not disappear completely. This will happen unless the population inversion is destroyed by some depopulation process, like internal conversion or some other competing transition. The gradual decrease in the peak due to  $\mu$  is characteristic of SF and other nonlinear radiation phenomena.<sup>26</sup>

It is instructive to further compare the effect of  $\mu$  on the Trammell-Hannon model<sup>7</sup> of ASE with the effect on SF. Our study shows that linear attenuation affects the development of lasing quite differently than it does the development of a SP pulse. The stimulation cross section  $\sigma_s$ , inversion  $\Delta n^*$ , and  $\mu$  are coupled in a linear relation forming the gain coefficient  $k$ , which characterizes the lasing phenomenon. The Schawlow-Townes condition demands that the gain coefficient  $k = \sigma_s \Delta n^* - \mu$  be positive for lasing to take place. The gain in a system exhibiting ASE is given by<sup>27</sup>

$$G = \frac{e^{kl} - 1}{kl} . \quad (11)$$

For  $k > 0$  gain is observed, but for  $k < 0$  only attenuation of the beam traversing the medium is possible. In SF, the inversion  $\Delta n^*$  and attenuation  $\mu$  interact through the nonlinear relationship in Eqs. (1), and thus their combined effect on SF is more complicated. For SF, positive inversion (i.e.,  $\Delta n^* > 0$ ) is required. However, the effect of  $\mu$  on the development of  $E$  is only to delay the SF pulse and modify its shape, not to prevent it. SF can occur as long as other effects, such as competing processes or dephasing, do not destroy the inversion. This can be seen from (4c) which is equivalent to

$$E_{32}^{\pm}(x) = E_{32}^{\pm}(0)e^{(-\mu/2)x} + \int_0^x gR_{32}(y)e^{-\mu/2(y-x)} dy . \quad (12)$$

For large  $\mu$  the first term is unimportant and only that part of the integration region where  $y \approx x$  contributes. However, there is a gradual decrease of the electric field and not a sharp cutoff or threshold effect. A quantitative investigation of this phenomenon using theory such as that presented here is necessary to obtain the optimum conditions for observing SF in specific nuclei under specific conditions.

We now turn to the line broadening mechanisms. In addition to the natural decay rates  $\Gamma_2$  and  $\Gamma_3$ , homogeneous broadening appears in the Maxwell-Bloch equations (1) through the dephasing rate  $\Gamma_{\phi} = b\Gamma$ . As noted earlier, inhomogeneous broadening is taken into account in (1) through the time dependence of the coupling parameter  $g'(t)$ , that replaces  $g$  and decays with rate  $\Gamma_{\theta} = a\Gamma$ . Using (1) we study the production of the SF pulse as a function of  $b$  or  $a$ . The results are shown in Figs. 6(a) and 6(b) where the natural decay is assumed to be  $\Gamma = 10^{-4}$ . At low values of  $b$  or  $a$  (when  $\Gamma_{\phi}, \Gamma_{\theta} \ll \tau_D^{-1}$ ) there is no noticeable effect on either the delay time or the intensity. At around  $b, a = 10^2$  ( $\Gamma_{\phi}, \Gamma_{\theta} = 10^{-2}$ ) the pulse delay increases and reaches a maximum, while the intensity starts to decrease. For  $b, a > 10^3$  the delay time decreases but the intensity drops to the noncooperative spontaneous value. The increase in  $\tau_D$  occurs during the time when the dephasing effect of  $\Gamma_{\phi}$  or  $\Gamma_{\theta}$  partially neutralizes the correlations induced by the electric-field vacuum state fluctuations, and consequently more time is required to build up the SF pulse. The decrease of both  $\tau_D$  and  $I_0$  indicates the complete breakup of SF past a certain time. Thus only the initial part of the SF peak appears, but becomes smaller as  $a$  or  $b$  increases. The only differences between the effects of  $a$  and  $b$  on the SF pulse is the stronger reduction in the peak intensity  $I_0$  and the more pronounced increase in the delay time  $\tau_D$  due to the inhomogeneous broadening as compared with that due to the homogeneous broadening for the same amount of broadening, i.e., for  $a = b$ .

Figure 8 exhibits the effect of the two broadening mechanisms on the full SF pulse shape when  $a$  and  $b$  take on the values 200, 500, and 1000. In Figs. 7(a), (b), and (c)  $a = 0$  and in Figs. 7(d), (e), and (f)  $b = 0$ . In Fig. 7(a) the dashed line represents, for comparison, the pulse with

no broadening  $a = b = 0$ , and the solid line, the pulse when  $a = 0$  and  $b = 200$ . As expected, from Figs. 6(a) and (b) with inhomogeneous broadening the pulse intensity decreases more and the pulse spreads out more than with homogeneous broadening. The delay time is also affected differently at these values of the parameters.

We have so far discussed the emission of the SF pulse from the right face of the active medium as a function of the characteristics of the medium. This is what would be measured in an experiment. It is interesting and instructive to follow the activity in the medium as the pulse develops and propagates through it. The field inside the medium ( $0 < x < 1$ ) is a function of  $x$  and  $t$ . The field at  $x = 1$  determines the emitted pulse propagating in the positive  $x$  direction. The results of the study are summarized by the curves shown for different values of the critical parameters in Figs. 8, 9, and 10, which indicate the variety of results that can be obtained by varying the in-

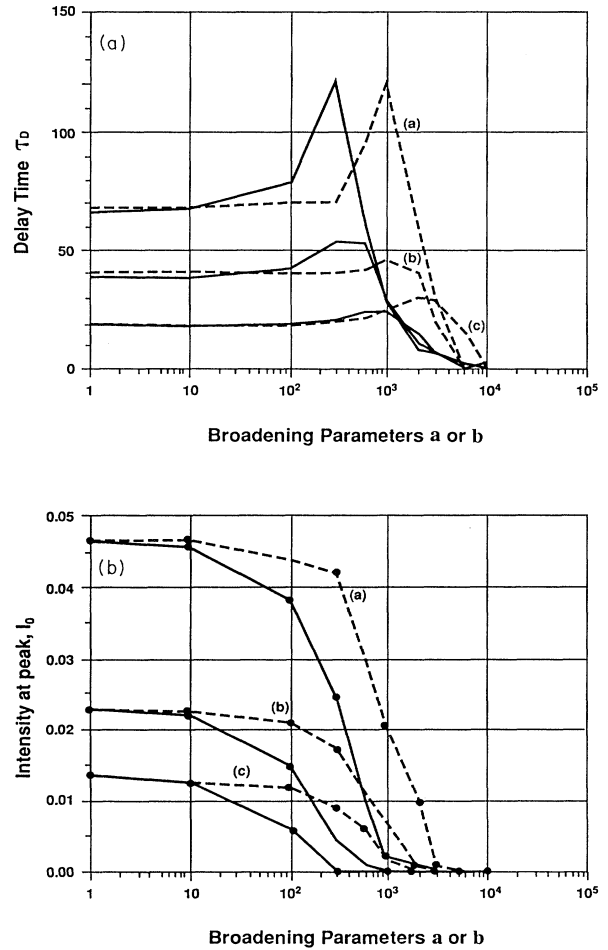


FIG. 6. (a) SF pulse emission delay time  $\tau_D$  as a function of the inhomogeneous broadening parameter  $a$  (solid line) and the homogeneous broadening parameter  $b$  (dashed line). Curves (a), (b), and (c) were obtained with  $N = 10^3, 10^6, 10^9$ , respectively. (b) SF pulse emission intensity  $I_0$  as a function of the broadening parameters  $a$  and  $b$ .

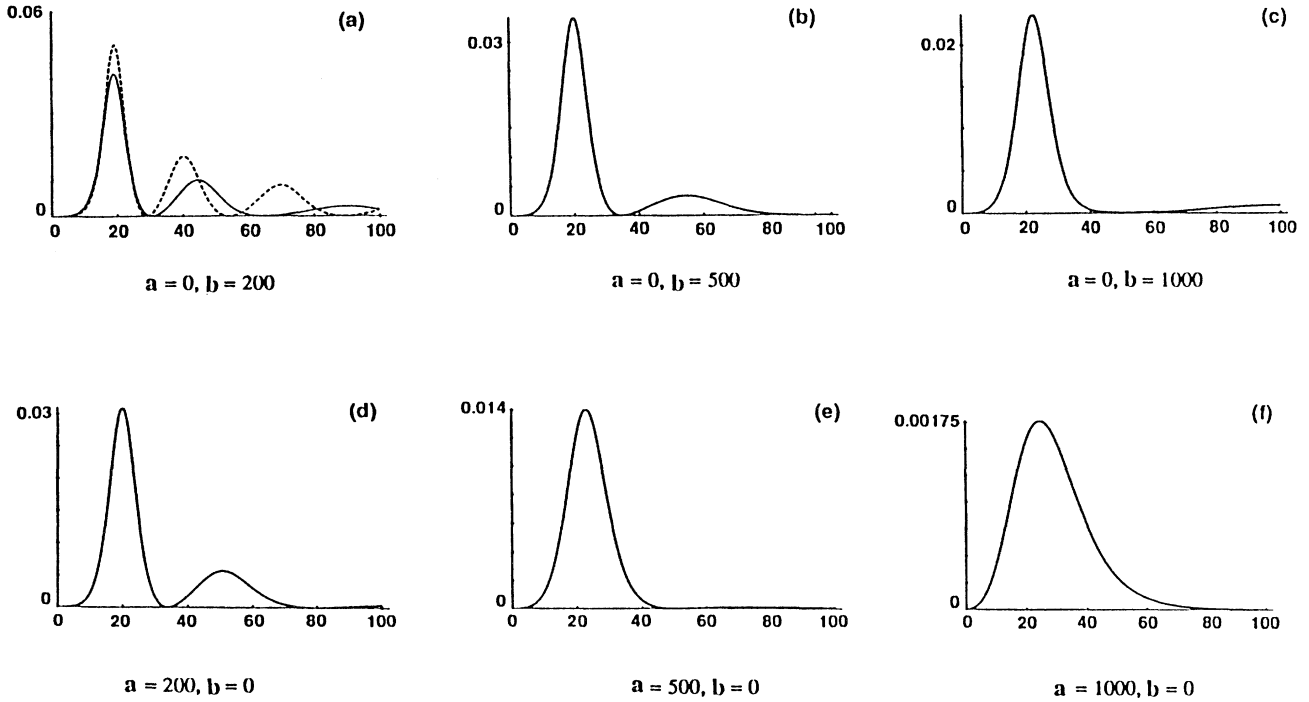


FIG. 7. SF Pulse assuming  $\gamma=10$ ,  $\Gamma=10^{-4}$ ,  $\mu=0$ ,  $N=10^3$ , and different values of the broadening parameters  $a$  and  $b$ . The dashed curve shows a pulse for the case when  $a=0$ ,  $b=0$ .

put parameters. In each figure the two curves at the top give the temporal activity at the right end ( $x=1$ ) of the active region. The figure labeled  $a$  shows the time variation in the populations  $N_4$  and  $N_3$ , and the one labeled  $b$  the pulse intensity. In the bottom figures  $c$  gives the spatial variation of the polarization  $R^+(x)$ ,  $d$  the population density  $N_3(x)$ , and  $e$  the intensity  $I(x)$ . The curves are snapshots at different times in units of  $\tau_{SF}$  as indicated in the figures. Note the effect of slower pumping in Fig. 9, and the introduction of attenuation in Fig. 10.

With  $\mu=0$ , the spatial variations of the field, in particular the intensity, increase monotonically with an increasing slope, whereas for  $\mu \neq 0$  the electric field changes sign; consequently, at some point in the region the intensity goes to zero. According to (7) and (12) this happens when the polarization and the electric field are such that

$$E(x) = g \int_0^x R(x') dx' - \frac{\mu}{2} \int_0^x E(x') dx' = 0. \quad (13)$$

This indicates that for maximum radiation intensity out of the active region, the attenuation  $\mu$  in the medium and the coupling constant  $g$  between the nuclei and the electromagnetic field must be considered.

#### IV. SAMPLE CALCULATION: SF IN $^{60}\text{Co}$

Earlier researchers have considered lasing in nuclear systems such as  $^{180}\text{Ta}$  prepared by thermal neutron cap-

ture reactions<sup>28,29</sup> and nuclear SF in  $^{119}\text{Sn}$  and  $^{133}\text{Ba}$ .<sup>8</sup> It was found that lasing in  $^{180}\text{Ta}$  required an inaccessibly high thermal neutron flux ( $10^{28} \text{ cm}^{-2} \text{ s}^{-1}$ ) for inversion.<sup>30</sup> In another study SF in good Mössbauer isotopes  $^{119}\text{Sn}$  and  $^{133}\text{Ba}$  was discussed without consideration of a pumping mechanism, i.e., instantaneous inversion was assumed. On the other hand, other researchers<sup>31</sup> concentrated on the pumping or inversion mechanisms and did not investigate the details of the emission dynamics.

We feel that consideration of the pumping requirements and the emission dynamics are equally important for the selection of candidate nuclei. The general theory presented in Sec. II was developed to provide this capability for dealing with the various parameters of the problem consistently in the same calculation.

As an example of the application of our theory to a real nuclear system, we have selected the isomeric transition in  $^{60}\text{Co}$  as a sample system because, unlike  $^{119}\text{Sn}$  and  $^{133}\text{Ba}$ ,  $^{60}\text{Co}$  has a potential mechanism for inversion: thermal neutron pumping of  $^{59}\text{Co}$ . The characteristic parameters of  $^{60}\text{Co}$  are given in Table I and the energy-level diagram for the region of interest is given in Fig. 11. The long lifetime (906 sec) could permit longer pumping times and therefore lower thermal neutron fluxes to obtain inversion than those required for  $^{180}\text{Ta}$ . The required fluxes could possibly be obtained in reactors rather than from disruptive nuclear explosions. The difficulty with  $^{60}\text{Co}$ , as with all long-lived nuclei, is the inhomogeneous

broadening which acts to reduce the resonance effect and destroy SF unless it can be somehow mitigated.

We use our theory [Eqs. (1c)–(1f)] to determine how much inhomogeneous broadening can be tolerated at a particular pumping rate and electronic attenuation of the beam. We assume that  $^{60}\text{Co}$  can be prepared in the excited isomeric state by thermal neutron pumping of  $^{59}\text{Co}$ . Using parameters given in Table I we calculated the intensity of the SF pulse as a function of the inhomogeneous broadening parameter  $a$ , and attenuation coefficient  $\mu$  and various pumping rates. For these calculations we assumed  $l = 1$  cm, giving  $N = \rho\lambda^2 l = 1.1 \times 10^5$  and, from (2),  $\tau_{\text{SF}} = 2.4$  sec.

The possibility of observing nuclear SF is considered

by comparing the SF emission with the spontaneous emission from the natural decay of  $^{60}\text{Co}$  as discussed in Ref. 8. The ratio of the integrated photon count from SF pulses divided by the integrated spontaneously emitted photon count in the natural decay process is defined as the  $S/N$  ratio.

We assume the sample is prepared in a cylindrical shape, irradiated, and placed in front of a collimator so that the directional beam characteristics (see Fig. 1) can be used to discriminate against the natural decay. The small divergence of the SF beam<sup>32</sup> compared to the isotropic distribution of the natural decay enhances the signal to noise by  $4\pi/\theta_D^2 = 5 \times 10^9$ .

Our calculations show that for an ideal sample with no

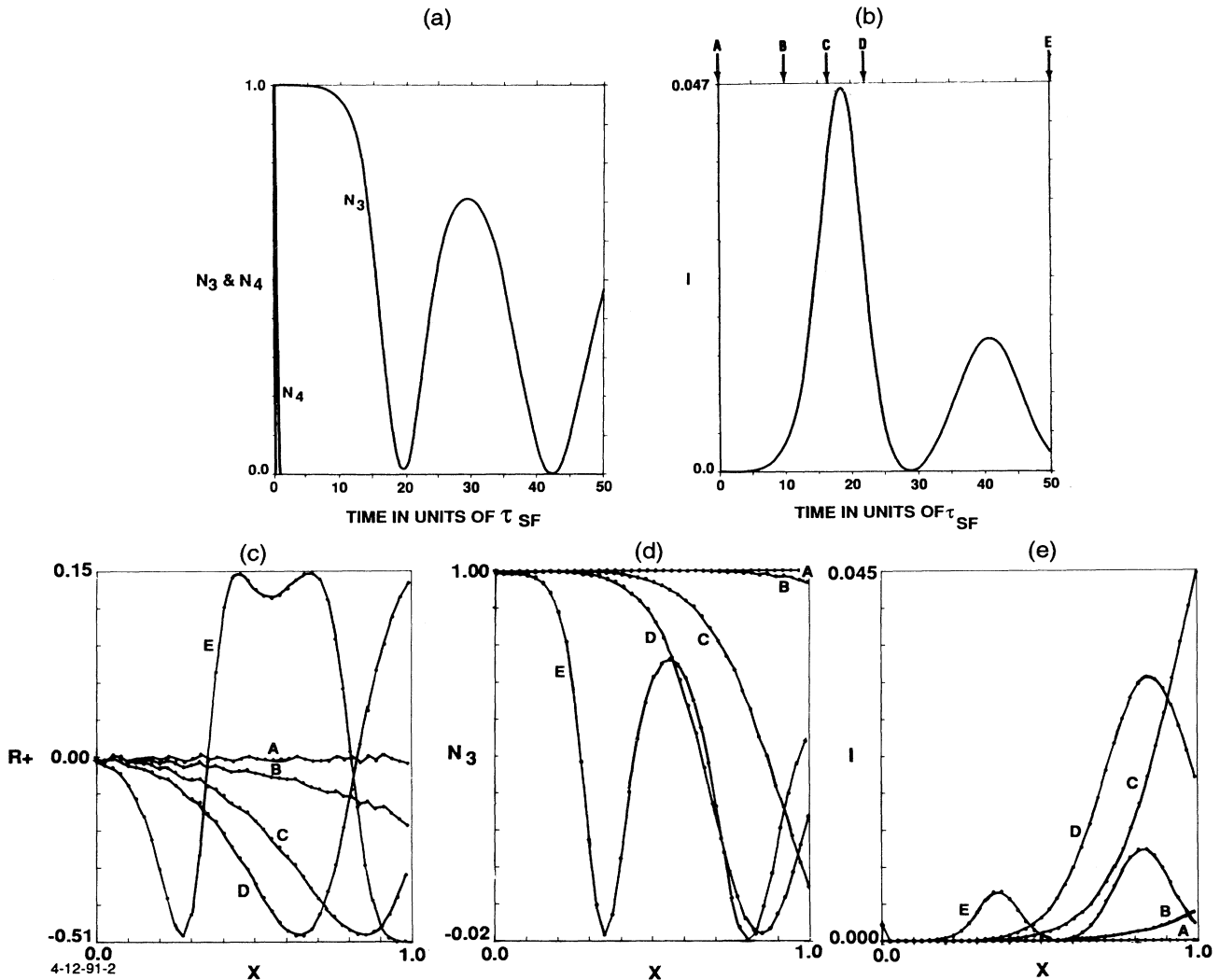


FIG. 8. SF pulse characteristics for input parameters  $\gamma = 10$ ,  $\Gamma_3 = 10^{-4}$ ,  $\mu = 0$ , and  $N = 10^3$ . (a) Shows the variation of level densities  $N_4$  and  $N_3$ , calculated at  $x = 1$  and (b) gives the pulse intensity  $i(t)$ . The bottom portions, (c), (d), and (e) give plots of the spatial variation of the polarization  $R^+(x)$ , number density  $N_3(x)$ , and intensity  $I(x)$  at different times in units of  $\tau_{\text{SF}}$  as follows: (A) = 0, (B) = 10, (C) = 17.5, (D) = 22.5, (E) = 50.



inhomogeneous broadening,  $a=0$ , and no attenuation of the beam,  $\mu=0$ , a thermal neutron flux of  $J=2\times 10^{17}$   $\text{cm}^{-2}\text{s}^{-1}$  will generate a SF pulse with a  $S/N=80$ . Deviations from ideal conditions erode the SF pulse emission quickly. For example, with  $a=20$  and  $\mu=12\text{ cm}^{-1}$  one requires a flux of  $J=4\times 10^{19}$   $\text{cm}^{-2}\text{s}^{-1}$  to produce a SF pulse with  $S/N=7$ .

Whether an experiment described here is feasible depends to a large extent on the ability to reduce inhomogeneous broadening and on the ability to generate the required thermal neutron fluxes. Inhomogeneous broadening in isomeric nuclei has been discussed in the literature.<sup>16</sup> Theoretical predictions using a static nuclear dipole-dipole interaction model give a value of  $a=10^5$  for  $^{109}\text{Ag}$ .<sup>33</sup> Subsequent measurements of self-attenuation in a  $^{109}\text{Ag}$  source by three different groups<sup>34-36</sup> have given much lower values of  $a$ . In addition to this, relaxation was proposed as a possible mechanism to explain the discrepancy between these measurements and the value

$10^5$  calculated from the static dipole-dipole interaction.<sup>37</sup> We discuss the effect of relaxation on SF in another publication.<sup>38</sup> We now turn to the availability of thermal neutron fluxes. The presently available thermal neutron fluxes are of the order of  $10^{16}$  neutrons/s (Ref. 39) and therefore not sufficient for generating SF from  $^{60}\text{Co}$  although concepts for providing  $10^{17}$  and  $10^{18}$  n/s were proposed as early as 1975.<sup>40</sup> The kind of fluxes required for the experiments discussed here do not seem to be out of reach of technology.

The experiment with  $^{60}\text{Co}$  is a difficult one and is presented here as an example of the application of the theory. It should be pointed out that our theory is generally applicable to other pumping schemes such as inter-level transfer or upconversion discussed by Collins *et al.*<sup>31</sup> As other concepts are introduced and other nuclei are presented as candidates for  $\gamma$ -ray lasing or nuclear SF, the theory developed here could be used to predict the outcome of such experiments.

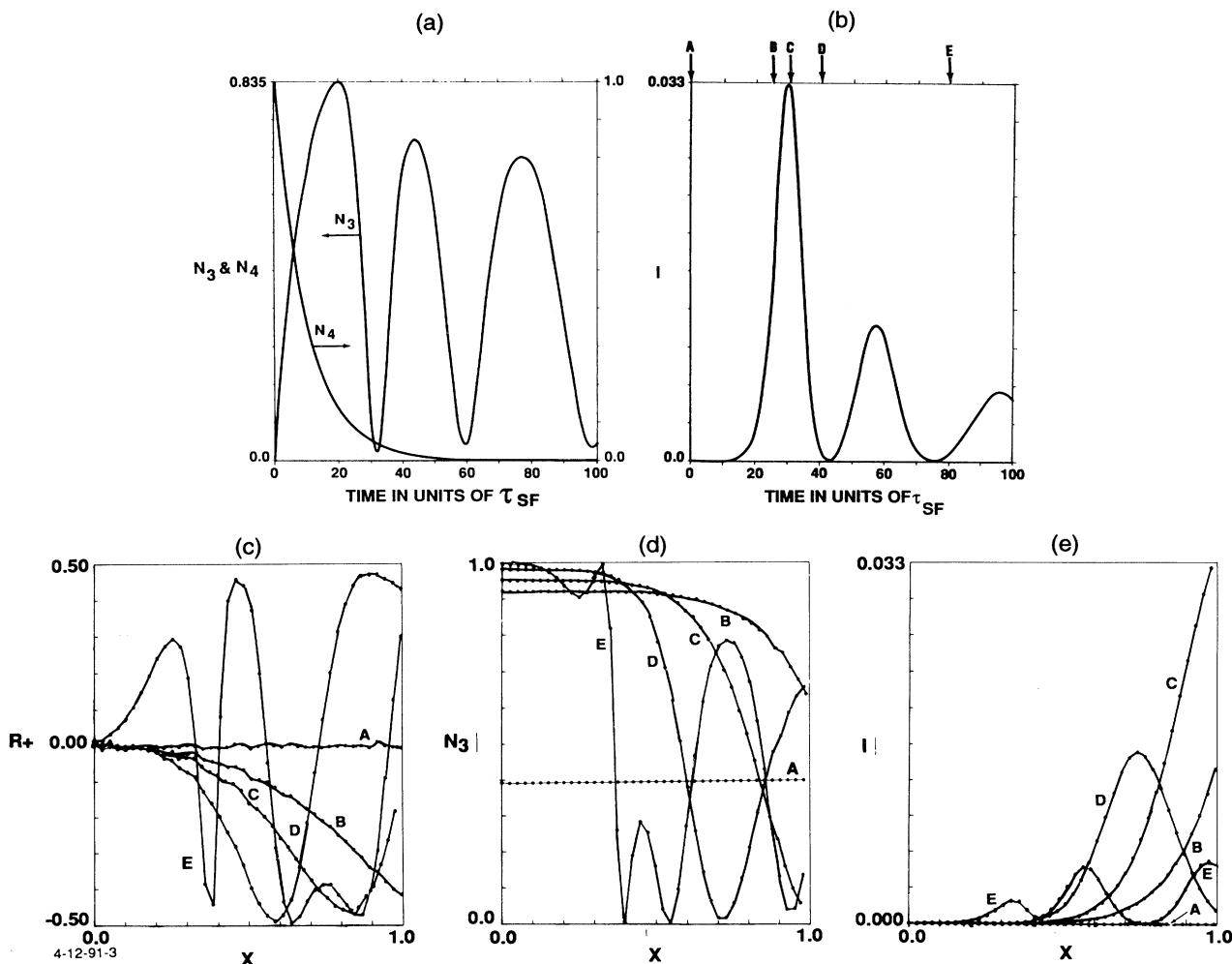


FIG. 9. SF pulse characteristics for the same parameters as Fig. 8 except with  $\gamma=0.1$  and the times are in units of  $\tau_{SF}$  as follows: (A) = 0, (B) = 25, (C) = 30, (D) = 40, and (E) = 80.

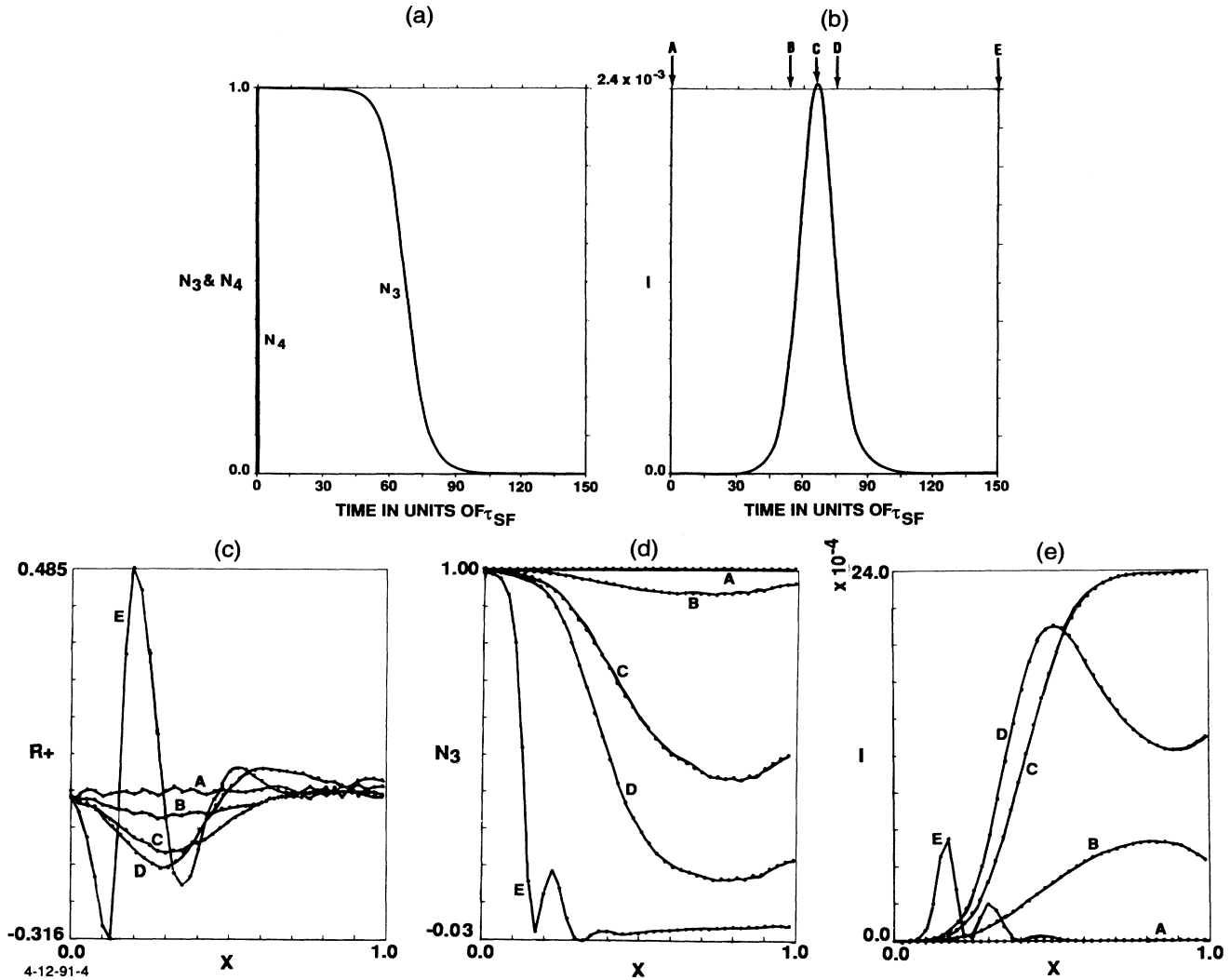


FIG. 10. SF pulse characteristics for the same parameters as Fig. 8 except the times are in units of  $\tau_{SF}$  as follows: (A) = 0, (B) = 52.5, (C) = 67.5, (D) = 75, and (E) = 150.

V. CONCLUSIONS

The spatial attenuation parameter  $\mu$  reduces the SF pulse peak predicted by the generalized version of the Haake-Reibold model of SF introduced in this paper and shifts it to longer times. The peak will decrease gradually but will remain positive if no dephasing mechanisms that

reduce the initial macroscopic dipole responsible for launching the SF pulse exist and population depletion mechanisms do not destroy the inversion.

Small values of the dephasing factor have no effect on the emitted pulse delay time or intensity, but when the dephasing rate due to either homogeneous or inhomogeneous broadening is comparable to the superfluorescence decay rate,  $1/\tau_{SF}$ , the delay time starts to increase with increasing values of the dephasing factor

TABLE I. Characteristic parameters for  $^{60}\text{Co}$ .

Parameter	Value	Symbol
transition energy	58.6 keV	$E_0$
Wave length on resonance	$0.2 \text{ \AA} (2 \times 10^{-9} \text{ cm})$	$\lambda$
Natural lifetime (half time)	906 sec (628 sec)	$\tau_0 (T_{1/2})$
Recoilless fraction	0.304	$f$
Linear attenuation coefficient	$12 \text{ cm}^{-1}$	$\mu$
Internal conversion coefficient	48.3	$\alpha'$
Partial density (in solid form)	$8.97 \times 10^{22} \text{ cm}^{-3}$	$\rho$

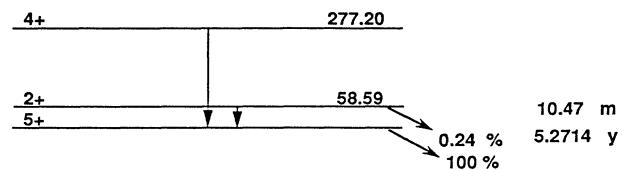


FIG. 11. Level structure of  $^{60}\text{Co}_{27}$ .

until it reaches a maximum, after which it decreases rapidly. The inhomogeneous effect is consistently stronger than the homogeneous effect for the same amount of broadening.

The active region length for which the SF pulse is a maximum depends on the values of the attenuation coefficient  $\mu$  and the coupling constant  $g$  associated with coupling between the collective polarization of the emitters in the region and the electric field.

The theory developed in this paper is applied to the 58 keV transition in  $^{60}\text{Co}$ . It is found that conditions for obtaining SF in  $^{60}\text{Co}$  taking into account pumping and including inhomogeneous broadening reduction, are not

quite within the capability of presently available technology.

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