The role of quantum fluctuations in superradiant decay of extended systems

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The kinetics of superradiance (SR) by an extended system of two-level radiators is investigated. A closed set of integrodifferential equations is obtained for the space-time SR kinetics. It takes into account fluctuations in the population difference. It is shown that the theory provides a much better description of the observed SR pulse shape than the space-time SR theory that ignores fluctuations, or the mean field theory.

1. INTRODUCTION

The method of excluding the electromagnetic-field boson operators will be used below to obtain a closed chain of quantum-mechanical equations for atomic correlators. The method proposed below for decoupling the three-particle correlators was developed as a result of an analysis of the statistical properties of atomic operators. This new method takes into account quantum fluctuations in the population difference during superradiance (SR). This leads to a reduction in the rate of decay at the peak of the SR pulse and, as a consequence, to an increase in its length.

A new approach will also be given to the description of the spatial kinetics of superradiance, with account taken of the difference between the left and the right electromagnetic-field fluxes everywhere with the exception of the center of the specimen. This ensures that the rate of decay at any given point in the specimen at a selected direction of the cylinder axis depends not only on the density of the electromagnetic field, but also on the energy flux in the particular direction. The inclusion of longitudinal effects in the SR process leads to a still greater broadening of the SR pulse.

At present, SR research is being concentrated on the development of theories capable of providing a complete description of all the basic parameters of the SR pulse. Since existing theories provide only a qualitative interpretation of experimental data, it will be useful to examine the shortcomings of these approaches. Since SR arises as a result of the phasing of individual atoms, a rigorous analysis of the initial stage of superradiance must be based on a quantum-mechanical description of the vacuum electromagnetic field and the radiators. The next stage in the SR process is frequently analyzed semiclassically, i.e., the problem of interaction between a classical electromagnetic field and quantum oscillators is solved. This approach is a consequence of the approximate decoupling of the chain of quantum-mechanical equations describing the SR process.[1,2]

We note that the solutions obtained below for the quantum-mechanical equations improve the agreement between theory and experiment.[3,4]

2. BASIC EQUATIONS FOR THE SR PROCESS

The Hamiltonian for a two-level set of atoms interacting through electromagnetic radiation fields is

$$H=\hbar\omega_0\sum_{j=1}^{N}R_{3j}+\sum_{\mathbf{k}}\hbar\omega_{\mathbf{k}}a_{\mathbf{k}}^+a_{\mathbf{k}}+i\frac{\omega_0 d}{C}\sum_{j=1}^{N}\{A^+(\mathbf{r}_j)R_j^- - R_j^+A^-(\mathbf{r}_j)\},\qquad(1)$$

where d is the dipole moment of the transition, ω_0 is the transition frequency, R_j^+ (R_j^-) is the creation (annihilation) operator for the excited state of the jth atom, which satisfies the commutation relations

$$[R_j^+,R_i^-]=2R_{3j}\delta_{j,i},\qquad [R_j^+,R_{3i}]=-R_j^+\delta_{i,j},\qquad(2)$$

R_{3j} is the population difference operator for the jth atom, $a_{\mathbf{k}}^+$ ($a_{\mathbf{k}}$) is the creation (annihilation) operator for a photon of momentum $\hbar\mathbf{k}$ and energy $\hbar\omega_{\mathbf{k}}$, and $A^+(\mathbf{r}_j)$ ($A^-(\mathbf{r}_j)$) is the negative (positive)-frequency part of the vector potential.

Expanding the electromagnetic field potential in terms of plane waves in the main part of the two-level system, and solving the Heisenberg equation for the electromagnetic field operators, we can readily express $A^+(\mathbf{r}_l,t)$ ($A^-(\mathbf{r}_l,t)$) in terms of atomic operators:

$$\check{A}^+(\mathbf{r}_l,t)=\check{A}_v^+(\mathbf{r}_l,t)+\frac{\omega_0\mathbf{d}}{c\hbar}\sum_{\mathbf{k}}\sum_{j=1}^{N}g_{\mathbf{k}}^2C_{lj}(\mathbf{k})\int_0^\infty G_k(t-t')\check{R}_j^+(t')\,dt',\qquad(3)$$

where

$$G_k(t-t')=\exp\{-i(\omega_{\mathbf{k}}-\omega_0-i\alpha(k))(t-t')\}\theta(t-t')$$

is the retarded Green function,

$$C_{lj}(\mathbf{k})=\exp\{i(\mathbf{k},\mathbf{r}_l-\mathbf{r}_j)\},\qquad \alpha(\mathbf{k})=\frac{c}{2}\left\{\frac{\alpha_x}{L_x}+\frac{\alpha_y}{L_y}+\frac{\alpha_z}{L_z}\right\}$$

is a coefficient describing the loss of photons from the active region of the two-level system, $\alpha_i=|k_i|/k$ ($i=x,y,z$),

$$g_k=\left(\frac{2\pi c^2\hbar}{\omega_k v}\right)^{1/2}\left(\mathbf{e}_\delta,\frac{\mathbf{d}}{d}\right),$$

$\mathbf{e}_\delta$ is the photon polarization vector, $\delta=1,2$, $v=L_xL_yL_z$ is the volume of the two-level system, and $A_v(\mathbf{r}_l,t)$ is the vacuum part of the electromagnetic field operator [$\langle v|A_v^+=0$, $A_v^-|v\rangle=0$ and $|v\rangle$ is the vacuum wave function]. The operators $\check{A}$, $\check{R}$ are written in the interaction representation and

$$\check{B}(t)=\exp\left(-\frac{i}{\hbar}H_0t\right)B(t)\exp\left(\frac{i}{\hbar}H_0t\right),$$

$$H_0=\hbar\omega_0\left[\sum_j R_{3j}+\sum_k a_{\mathbf{k}}^+a_{\mathbf{k}}\right].$$

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We shall suppose that the radiating atoms occupy a volume with Fresnel number $F \lesssim 1$. Consider the evolution of the system from the initially uncorrelated state of the two-level system and the vacuum state of the electromagnetic field. The average of $\langle R_{3l}(t)\rangle$ must then satisfy the following equation:

$$\frac{\partial}{\partial t}\langle R_{3l}(t)\rangle=-\frac{1}{\tau_0}\left(\langle R_{3l}(t)\rangle+\frac{1}{2}\right)-V^+(\mathbf{r}_l,t)-V^-(\mathbf{r}_l,t), \tag{4}$$

where

$$V^+(\mathbf{r}_l,t)=\sum_{\mathbf{k}(k_x>0)}V_\mathbf{k}(\mathbf{r}_l,t),\quad V^-(\mathbf{r}_l,t)=\sum_{\mathbf{k}(k_x<0)}V_\mathbf{k}(\mathbf{r}_l,t),$$

$$V_k(r_l,t)=\beta g_k^2\sum_{\substack{m=1\\ n\neq l}}^{N}C_{ml}(k)\left\{\int_0^\infty dt'G_k(t-t')\right.$$

$$\left.\times\langle R_l^+(t)R_m^-(t')\rangle+\text{H.c.}\right\},\quad \frac{1}{\tau_0}=\frac{4}{3}\frac{\omega_0^3d^2}{c^3\hbar},$$

$$\beta=(d\omega_0/c\hbar)^2,$$

and $V^+(r_l,t)$ $(V^-(r_l,t))$ is the rate of SR decay of the l the atom from left to right (right to left) along the cylinder axis $(k_x\|L_x)$.

A closed set of equations can be obtained from the Heisenberg equation for $V^+(\mathbf{r}_l,t)$ $(V^-(\mathbf{r}_l,t))$:

$$\partial V^+(\mathbf{r}_l,t)/\partial t=Q_1^+(\mathbf{r}_l,t)+Q_2^+(\mathbf{r}_l,t), \tag{5}$$

where

$$Q_1^+(\mathbf{r}_l,t)=2\beta\sum_{\mathbf{k}(k_x>0)}\sum_{\mathbf{k}'}g_k^2g_{k'}^2$$

$$\times\sum_{n=1}^{N}\sum_{\substack{m=1\\ m\neq l}}^{N}\int_0^\infty dt'G_k(t-t')C_{lm}(\mathbf{k})C_{mn}(\mathbf{k}')$$

$$\times\int_0^\infty dt''G_{k'}(t'-t'')\langle\check{R}_l^+(t)\check{R}_{3m}(t')\check{R}_n^-(t'')\rangle+\text{H.c.}$$

$$Q_2^+(\mathbf{r}_l,t)=2\beta\sum_{\mathbf{k}(k_x>0)}\sum_{\mathbf{k}'}g_\mathbf{k}^2g_{\mathbf{k}'}^2\sum_{n=1}^{N}\sum_{\substack{m=1\\ m\neq l}}^{N}\int_0^\infty dt'G_k(t-t')$$

$$\times C_{lm}(\mathbf{k})C_{nl}(\mathbf{k}')\int_0^\infty dt''G_k(t-t'')\langle\check{R}_n^+(t'')\check{R}_{3l}(t)\check{R}_m^-(t')\rangle+\text{H.c.}$$

The method of decoupling the chain of equations given by (4) and (5) is sometimes indicated by the Hamiltonian of the interaction where it contains a small parameter. However, when the interpolation theory that is valid in the limit of the SR peak is developed, any discussion of accurate decoupling becomes meaningless because the problem does not then have a small parameter that can be used at any stage in the evolution of the system. There are at present no known internal criteria for the "quality" of decoupling, but it is clear that the symmetry properties of the Pauli operators must be preserved under any type of decoupling.

It is important to emphasize that the operators R_j^+ and R_l^- are Fermi operators when they correspond to a single atom (in this sense, they satisfy anticommutation relations):

$$R_j^+R_j^-+R_j^-R_j^+=1, \tag{6}$$

and Bose operators when they correspond to different atoms, since they satisfy the commutation relations

$$(R_j^+R_l^--R_l^-R_j^+)|_{j\neq l}=0. \tag{7}$$

Since the operators in the chain (5) appear in the normal ordering (this ordering was obtained by excluding the vacuum part of the operators), we shall try to decouple the system without affecting the disposition of the operators in the correlator. Bearing in mind the properties (6), (7), and (2), we decouple our three-particle correlators by analogy with Wick's theorem for boson and fermion operators:

$$\langle R_j^+(t)R_m^+(t')R_m^-(t')R_l^-(t'')\rangle$$

$$=\langle R_m^+(t')R_m'(t')\rangle\langle R_j^+(t)R_l^-(t'')\rangle$$

$$-\langle R_j^+(t)R_m^-(t')\rangle\langle R_m^+(t')R_l^-(t'')\rangle, \tag{8a}$$

$$\langle R_j^+(t)R_m^-(t')R_m^+(t')R_l^-(t'')\rangle$$

$$=\langle R_m^-(t')R_m^+(t')\rangle\langle R_j^+(t)R_l^-(t'')\rangle$$

$$+\langle R_j^+(t)R_m^-(t')\rangle\langle R_m^+(t')R_l^-(t'')\rangle. \tag{8b}$$

The decoupling (8) differs from the traditional decoupling of three-particle correlators[1]

$$\langle R_j^+(t)R_{3m}(t')R_l^-(t'')\rangle=\langle R_{3m}(t')\rangle\langle R_j^+(t)R_l^-(t'')\rangle \tag{9}$$

by the fact that it preserves the invariance of the integral of the motion

$$\langle R_{3l}^2\rangle=[\langle R_{3l}R_j^+R_m^-\rangle+{}^1/_2\langle R_{3l}\rangle]|_{l=j=m}={}^1/_4, \tag{10}$$

whereas (9) approximates (10) by the expression $\langle R_{3l}\rangle^2$. Hence it follows that the decoupling (9) is deficient in that it does not take into account fluctuations in the population difference operator R_{3j}, which is definitely unsatisfactory in the region of the SR intensity maximum when $\langle R_{3l}\rangle\to 0$, whereas (8) has a tendency to reduce these fluctuations in the course of the SR process.

Substituting (8) in (5) and assuming that, firstly, $R_{3m}(t')$ is a smooth function of spatial coordinates and, secondly, most of the radiation is emitted into the solid angle $\delta\Omega\sim\lambda^2/s$ (s is the cross section of the specimen) since $F\lesssim 1$, we can readily obtain the following expression for the first term in (5):

$$Q_1^+(x_l,t)=\frac{1}{\tau_1\tau_0 L}\int_0^{\theta_F}\sin\theta\,d\theta\sum_{n=1}^{N_l}\left\{\exp\left[i\frac{\omega_0}{c}\cos\theta r_{ln}\right]\right.$$

$$\times\left\langle R_n^+\left(t-\frac{x_l-x_n}{c}\right)R_l^-(t)\right\rangle+\text{H.c}\Big\}$$

$$\times\int_{t-(x_l-x_n)/c}^{t}R_3(t_1)\,dt_1+\frac{1}{\tau_0}\int_0^{\theta_F}\sin\theta\,d\theta\cdot$$

$$\times\sum_{\substack{m=1\\ m\neq l}}^{N_l}\left\{\exp\left[i\frac{\omega_0}{c}\cos\theta r_{ln}\right]\right.$$

$$\times\left\langle R_m^+\left(t-\frac{x_l-x_m}{c}\right)R_l^-(t)\right\rangle+\text{H.c}\Big\}$$

$$\times\frac{\partial\langle R_{3m}(t-(x_l-x_m)/c)\rangle}{\partial t};$$

$$R_3(t)=\sum_{m=1}^{N}R_{3m}(t),\quad \frac{1}{\tau_1}=\frac{\pi c^2}{\omega_0^2\tau_0 s},\quad s=L_yL_z,\quad \theta_F\sim\frac{\lambda}{s^{1/2}},\tag{11}$$

$r_{ln}\cos\theta\approx x_l-x_n$. The sum over n and m includes all atoms lying left of point x_l.

Differentiating the first term in (11) with respect to x, we can readily show that, when $L/c\tau_R<1$ ($\tau_R=\tau_1/N$), Eq. (11) assumes the form

$$Q_1^+(x,t)=\frac{2}{\tau_1 L}\int_0^x R_3\left(t-\frac{x-x'}{c}\right)\left\{V^+\left(x',t-\frac{x-x'}{c}\right)\right.$$

$$\left.+\frac{1}{2\tau_0}\left[R_3\left(x',t-\frac{x-x'}{c}\right)+\frac{1}{2}\right]\right\}dx'+2V^+(x,t)\frac{\partial R_3(x,t)}{\partial t}.\tag{12}$$

Substituting (8) into the expression of Q_2^+, we obtain the following expression

$$Q_2^+(x_l,t)=V^+(x_l,t)\frac{\partial R_{3l}}{\partial t}-\frac{1}{2\tau_0^2}\int_0^{\theta_F}\sin\theta\,d\theta$$

$$\times\int_0^{\theta_F}\sin\theta'\,d\theta'\left\{\sum_{n=1}^{N_l}\sum_{m=1}^{N_l}\exp\left[\frac{\omega_0}{c}r_{ln}\cos\theta\right]\right.$$

$$\times\exp\left[-i\frac{\omega_0}{c}r_{lm}\cos\theta'\right]$$

$$\times\langle R_{3l}(t)\rangle\left\langle R_m^+\left(t-\frac{x_l-x_m}{c}\right)R_n^-\left(t-\frac{x_l-x_n}{c}\right)\right\rangle+\text{H.c}\Big\}.\tag{13}$$

Taking the average of (13) over the cross section of the cylinder containing the two-level system, and assuming that retardation is important only along the axis of the cylinder, we obtain

$$Q_2^+(x,t)=V^+(x,t)\frac{\partial R_3(x,t)}{\partial t}+\frac{2N}{\tau_1 L}R_3(x,t)$$

$$\int_0^x\left\{V^+\left(x',t-\frac{x-x'}{c}\right)+\frac{1}{2\tau_0}\left(R_3\left(x,t-\frac{x-x'}{c}\right)+0{,}5\right)\right\}dx'.\tag{14}$$

From (4) and (5) with allowance for (12) and (14), we readily obtain the following set of integro-differential equations:

$$\frac{\partial R_3(x,t)}{\partial t}=-\frac{1}{\tau_0}\left(R_3(x,t)+\frac{1}{2}\right)-V^+(x,t)-V^-(x,t),\tag{15}$$

$$\frac{\partial V^+(x,t)}{\partial t}=-\frac{V^+(x,t)}{T_2}+2V^+(x,t)\frac{\partial R_3(x,t)}{\partial t}$$

$$+\frac{2}{\tau_1 L}\int_0 dx'\left[NR_3(x,t)+R_3\left(t-\frac{x-x'}{c}\right)\right]$$

$$\times\left[V^+\left(x',t-\frac{x-x'}{c}\right)+\frac{1}{2\tau_0}\left(R_3\left(x',t-\frac{x-x'}{c}\right)+\frac{1}{2}\right)\right]$$

$$\times\theta\left(t-\frac{x-x'}{c}\right),\tag{16}$$

$$V^+(x,t)=V^-(L-x,t),\tag{17}$$

where T_2 is the time of transverse relaxation of the dipole moment. Equations (15)–(17) must be augmented by the following initial conditions:

3. SOLUTION OF THE SET OF EQUATIONS

When we ignore quantum fluctuations in the population inversion [i.e., when we replace the decoupling (8) and (9)], the set of equations describing the SR process has the following form:

$$\frac{\partial R_3(x,t)}{\partial t}=-\frac{1}{2\tau_0}\left(R_3(x,t)+\frac{1}{2}\right)-V^+(x,t)-V^-(x,t),\tag{18}$$

$$\frac{\partial V^+(x,t)}{\partial t}=-\frac{V^+(x,t)}{T_2}$$

$$+\frac{2}{\tau_1 L}\left\{\int_0^x dx'\left[NR_3(x,t)+R_3\left(t-\frac{x-x'}{c}\right)\right]\right.$$

$$\times\left[V^+\left(x',t-\frac{x-x'}{c}\right)+\frac{1}{2\tau_0}\left(R_3\left(x',t-\frac{x-x'}{c}\right)+\frac{1}{2}\right)\right]$$

$$\times\ \theta\left(t-\frac{x-x'}{c}\right),\tag{19}$$

$$V^+(x,t)=V^-(L-x,t).\tag{20}$$

The basic difference between (15)–(17) and (18)–(20) is that the former system acquired terms of the form

$$V^+(x,t)\frac{\partial R_3(x,t)}{\partial t}$$

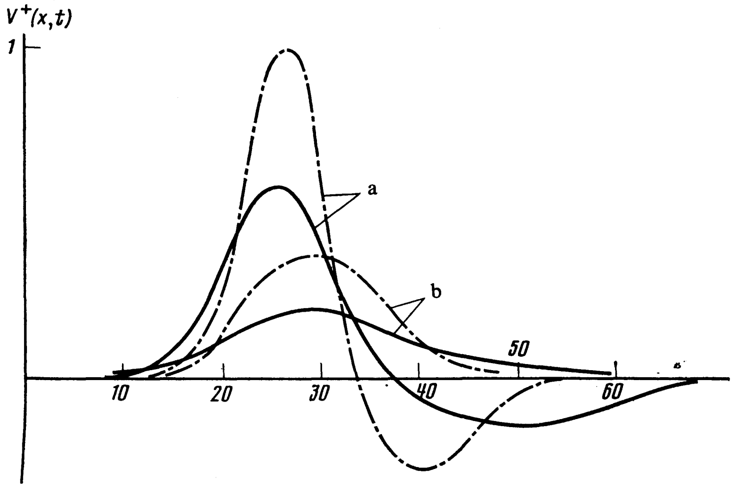

FIG. 1. Rate of SR decay $V^+(x, t)$ as a function of time: a—near the right edge of the specimen, b—near the center of the specimen $\tau_R/\tau_0 = 10^{-3}$, $c\tau_R = 3L$; dot-dash curves—solution obtained for (18)–(20); solid curves—solution obtained for (15)–(17).

which are important in the region of the SR intensity maximum. These terms produce a considerable broadening of the radiation pulse during the SR process. Figure 1 shows plots of the SR decay rate $V^+(x, t)$ near the center and near the right edge of a thin cylinder, calculated from (15)–(17) and (18)–(20). The abscissas are $2t/\tau_R$.

The following equation for SR has been obtained[1] for extended systems in the mean-field approximation (i.e., when the cooperative decay time is $\tau_R \gg L/c$):

$$\frac{d^2R_3}{dt^2}+\frac{1}{\tau_0}\frac{dR_3}{dt}-\frac{2}{\tau_1}R_3\frac{dR_3}{dt}=-\frac{2}{\tilde{\tau}_1{}^*\tau_0}R_3\left(R_3+\frac{N}{2}\right), \qquad (21)$$

where

$$\frac{1}{\tilde{\tau}_1{}^*}=\frac{1}{\tau_1}(1-\varkappa), \qquad \varkappa=\left(\frac{\tau_1}{\tau_0}\right)^{-1}, \qquad T_2\to\infty.$$

To compare the solution of (18)–(20) with the solution of (21), it is convenient to integrate (18)–(20) with respect to x. In the Markov approximation ($\tau_R \gg L/c$) the expressions for the collective terms in (18)–(20) then assume the form

$$\frac{2}{L^2\tau_1}R_3(t)\int_0^L dx\left[\int_0^x dx'\,V^+(x',t)+\int_x^L dx'\,V^-(x',t)\right]$$
$$=\frac{2}{\tau_1}R_3(t)\int_0^L dx'\left\{\frac{L-x'}{L^2}V^+(x',t)+\frac{x'}{L^2}V^-(x',t)\right\}. \qquad (22)$$

Replacement of the functions $L-x'$ and x' with their mean values within the interval $(0, L)$ (i.e., with $L/2$) reduces (22) to an expression that has the same form as the third term in (21).

Thus, (18)–(20) take into account longitudinal effects in the course of the SR process that are due to the variation in the electromagnetic field radiation density. In final analysis, this ensures that (18)–(20) describe a broader SR pulse than (21).

An approximation such as (22) can also be obtained for (16)–(17). In the mean-field approximation [i.e., when x' and $L-x'$ in the integrand in (22) are replaced with $L/2$], we then obtain the following equation for the population inversion (see Ref. 5 for further details):

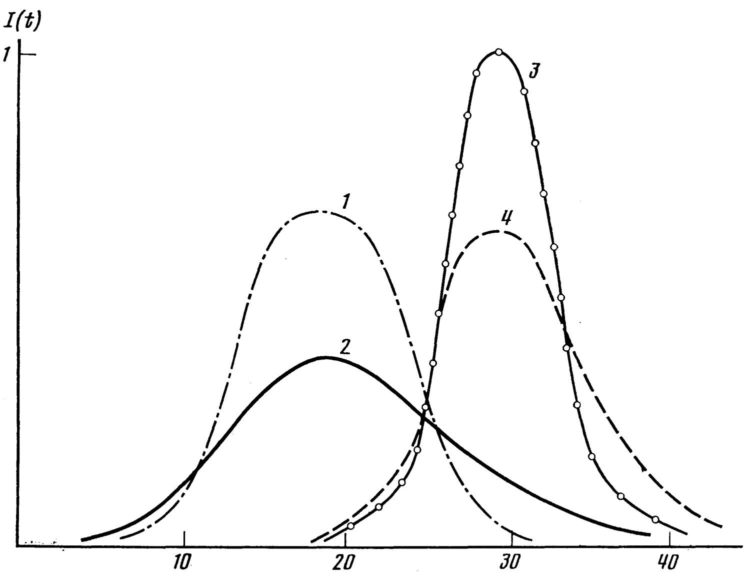

FIG. 2 SR intensity as a function of time deduced from (18)–(20)—curve 1, (15)–(17)—curve 2; Eq. (21)—curve 3, Eq. (23)—curve 4.

$$\frac{\partial^2 R_3}{\partial t^2} - \frac{2}{N}\frac{\partial R_3}{\partial t}\frac{\partial R_3}{\partial t} - \frac{2}{\tau_1'} R_3 \frac{dR_3}{dt} = \frac{2}{\tau_1}\frac{R_3}{\tau_0}\left(R_3 + \frac{N}{2}\right),$$

$$1/\tau_1' = 1/\tau_1 + 1/N\tau_0, \quad T_2 \to \infty. \tag{23}$$

Like (16)–(17), this equation takes into account fluctuations in the population difference in the two-level system, but ignores the variation in the field amplitude over the specimen. The effect of the second term in (23) on the shape and half-width of the SR pulse can be estimated analytically. Near the SR intensity maximum, the right-hand side of (23) can be neglected, so that, since the boundary conditions are

$$\left.\frac{dR_3}{dt}\right|_{t=0} = -\frac{N}{\tau_0}, \quad R_3(t)\,|_{t=0} = \frac{N}{2}$$

Eq. (23) becomes

$$\frac{dR_3}{dt} = \left(\frac{N^2}{\tau_1} - \frac{N}{\tau_0}\right)\exp\left[\frac{2}{N}\left(R_3 - \frac{N}{2}\right)\right] - \frac{N}{\tau_1}\left(R_3 + \frac{N}{2}\right). \tag{24}$$

Near the SR peak, when $R_3/N < 1$, the exponential can be replaced by the series:

$$\exp(2R_3/N) \approx 1 + 2R_3/N + 2(R_3/N)^2. \tag{25}$$

Substituting (25) in (24), we readily obtain the following solution:

$$R_3(t) \approx \frac{0{,}93}{2}\,\mathrm{th}\left(\frac{t-t_0}{2\tau_{Rf}^*}\right) + \frac{0{,}72N}{4}, \tag{26}$$

where $\tau_{Rf}^{*-1} = 0.68\tau_R^{-1}$ and t_0 is the delay time. Thus, τ_R^{*-1} turns out to be less than the value obtained under the generally accepted decoupling (9). The same conclusion is obtained for the point system by introducing the replacement $\tau_1 \to \tau_0$ into (24).

Figure 2 shows the results of a numerical integration for the SR intensity

$$I(t) = -\frac{\tau_0}{L} I_0 \int_0^L \frac{\partial R_3(x, t-x/c)}{\partial t}\, dx, \quad I_0 = \frac{N\hbar\omega_0}{\tau_1},$$

obtained by using (15)–(17), (18)–(20), and (21), (23). Thus, (15)–(17) leads to a broader and asymmetric curve for the SR intensity as compared with (21), (23) and (18)–(20).

4. CONCLUSIONS

Our theoretical investigation shows that the simultaneous inclusion of quantum fluctuations and longitudinal structure of the interaction field between the radiators results in a substantial improvement in the agreement between theory and experiment. Our new method of decoupling enables us to preserve some of the symmetry properties of kinetic equations for the two-level systems that influence the shape of the SR pulse. We have also shown that the separate allowance for the size of the system will also broaden the SR pulse.

It is clear from the foregoing discussion that fluctuations have an important effect on the shape and half-width of the SR pulse and that the decoupling (8) can be effectively used in equations for SR kinetics.

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