Cooperative effects in a system of strongly driven three-level atoms

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Cooperative effects in a system of three-level atoms in cascade configuration interacting with intense resonant fields are investigated. Analytical expressions for atomic populations in the excited levels, fluorescence intensities, and normalized intensity correlations are presented. The intensity correlations are used to test the violation of the Cauchy-Schwarz inequalities. The analytical results predict in the cooperative limit $N \rightarrow \infty$, a discontinuous behavior of all atomic observables with respect to a parameter related to the field amplitudes and the atomic decay coefficients. This discontinuous behavior is analogous to a typical nonequilibrium first-order phase transition.

I. INTRODUCTION

Cooperative behavior of a system of atoms interacting with common coherent field and the vacuum of radiation is a subject of continuing interest in quantum optics. In particular, superradiance and superfluorescence have received a considerable theoretical and experimental attention[1–30] since Dicke[1] first introduced the concept of collective spontaneous emission. For a system of N excited atoms, the signature of superradiant emission is an intensity proportional to N^2; the release of energy takes place with a delay which is proportional to $1/N$ and within a short time τ/N, τ being the natural lifetime of the excited state. In contrast, independent emission of spontaneous photons would give a linear dependence on the number of excited atoms. Cooperative emission from a Dicke superradiant state is analogous to the radiation emitted from a classical dipole prepared by an initial phasing of the atomic state.

The first experimental demonstration of superradiance was made in optically pumped HF gas by Skribanowitz *et al.*[2] This and subsequent experiments[3–9] essentially involved a creation of a totally inverted state of two-level atoms by incoherent excitation from a three-level configuration. With complete initial inversion there is no initial macroscopic polarization, instead it must build up from the quantum noise of spontaneous emission. Dicke superradiance from such an inverted sample is known as superfluorescence[11] and all experimental observations are of this type. Also, all observations pertain to samples with linear dimensions that are large compared with the wavelength λ of the emitted radiation. Dependence of superradiance on the shape and size of the sample was pointed out by Dicke[1] and several others.[12–15] The relevance of optically thick samples in order that coherent decay prevails over the incoherent decay has been emphasized by Friedberg and Hartman.[16]

These experiments[2–9] established the general properties of superfluorescence, viz., that the pulse intensities proportional to N^2 and pulse delays proportional to $1/N$. However, the emitted radiation showed interesting new features like strong reshaping, ringing, and frequency chirping which could be attributed to "nonlinear propagation and diffraction" aspects[16–19,26,27] of the phenomenon not considered in the early theoretical papers.[10,11,20,21] More specifically, the experimental results could be interpreted by the semiclassical propagation model[2,19] based on the coupled Maxwell-Bloch equations. The semiclassical predictions were later confirmed by the quantized-field treatments[22–24] of superfluorescence which rigorously included propagation effects along with the quantum initiation process. On the other hand, mean-field theory[10,11] which excludes propagation effects failed to interpret the experiments[2–9] which showed strong ringing and could not explain quantitatively even the single pulse observations[25] subsequently made on cesium atomic vapor.[26] The suppression of ringing in the later experiments[25] was accounted for by further refinement of semiclassical propagation models which included transverse effects (non-plane-wave field variations).[27] Several review articles surveying the theoretical models[28] and comparing in detail the theoretical predictions and experimental observations[29–32] exist.

All reported experiments on superfluorescence utilize optical pumping on a manifold of a minimum of three atomic or molecular energy levels to create an inverted population of two-level atoms. Apart from the mean-field analysis of Bowden and Sung,[33] most of the theoretical papers deal with the relaxation process from a prepared state of complete inversion in a two-level manifold of atomic energy levels. Theoretical analysis including coherent pump dynamics initiation was first presented by Bowden and Sung[34] and Sobolewska.[35] This work has been the forerunner of subsequent more complete analysis of three-level superfluorescence including the dynamical and propagational aspects of the pump field discussed in a recent article by Matter *et al.*[36] A theory supporting the experimental observations of Florian *et al.*[37] on two-color superfluorescence has been discussed by Haake and Riebold.[38] Further, a stochastic theory of cooperative cascade emission interpreting the experimental observations on two-photon excited lithium vapor has been developed by Ikeda *et al.*[39] Also, cooperative effects in stimulated Raman scattering including the propagation effects and their consequences on the superfluorescence from the Stokes transition have been reported.[40]

An extension of the Dicke model for superradiance is widely used[41–51] to discuss cooperative resonance fluorescence. Here N two-level atoms are confined to a volume of dimension smaller than the radiation wavelength and coherently driven on resonance by an intense laser light. Incidentally, resonance fluorescence from a single coherently driven two-level atom shows some predominantly quantum features, viz., the dynamical Stark effect reflected in the triplet fluorescence spectrum,[52] antibunching and subPoissonian photon statistics,[53] and squeezing.[54] Cooperative resonance fluorescence combines both resonance fluorescence and collective decay. While superradiance is a transient effect, the long-time evolution drives the atomic system to a steady state in the present case. Further, the continuous presence of the driving field includes a phased dipole moment. At first sight, classical radiation from this collective dipole would imply fluorescent intensities proportional to N^2. This explanation, however, holds only for low fields. At sufficiently high laser intensities, the N-atom system is saturated and in this limit the mean dipole moment vanishes; the persistent N^2 dependence of the fluorescent intensity arises from the quantum fluctuations as in the case of superradiance. Indeed, the theoretically predicted[44,45,48] phase transition[55] at $\Theta=2\Omega/N\gamma=1$ (2Ω is the Rabi frequency; 2γ is the Einstein A coefficient) distinctly shows the onset of saturation in the collective dipole moment and the subsequent role played by quantum fluctuations into the underlying dynamics. However, just as in superfluorescence, modifications in this simple picture would be expected due to dipole-dipole interactions and geometric or propagational effects. The consequences of dipole-dipole interactions for a special symmetric arrangement of an arbitrary number N of atoms in a sample[28] are known.[56,57] It has been shown[56,57] that dipole-dipole interaction even in this restricted case may, under certain conditions, alter the nature of the phase transition from being of second order to first order.

More recently, collective effects in resonant Raman scattering of intense optical waves have been discussed by Bogolubov *et al.*[58] Their model essentially considers a point Dicke model of three-level atoms in Raman configuration with two coherent fields resonantly driving the Rayleigh and Stokes transitions. In this paper, we have formulated a theory of collective effects in a system of N three-level atoms in cascade configuration continuously interacting with two intense resonant fields. We describe the system by means of the quantum-mechanical master equation and employ the secular approximation valid for strong fields. The resulting approximate master equation admits a steady-state solution. This solution is used to derive analytical expressions for the steady-state fractional atomic populations in the excited levels, normalized intensities, and normalized-intensity correlations. In the cooperative limit $N\to\infty$, these atomic observables are found to show a discontinuous behavior with respect to a parameter X related to the amplitudes of the two driving fields and the coefficients for the spontaneous emission from the two excited levels. This discontinuous behavior is similar to a nonequilibrium first-order phase transition.

The expressions for the intensity correlations are further used to study a nonclassical feature of cascade emission, viz., the possibility of violating the Cauchy-Schwarz (CS) inequality.[59] Incidently the violation of CS inequality has been observed by Clausser[60] in the two-photon cascade emission in an optical double-resonance experiment. This effect has also been predicted in the two-photon laser[61] and parametric amplifiers.[62,63] Our analysis predicts a strong violation of the CS inequality over a regime of the parameter X for all finite N as previously reported.[64] The inequality continues to be violated even in the cooperative limit $N\to\infty$. Thus, unlike antibunching, the violation of the CS inequality persists even for a large number of atoms and, in that sense, appears to be a macroscopic quantum effect.

In Sec. II, we present the basic formulation leading to the derivation of the master equation in the high-field limit and its steady-state solution. Sec. III is devoted to the derivation and discussion of the analytical expressions for the steady-state values of the atomic populations and their fluctuations, fluorescent intensities, and intensity-intensity correlations. Finally, some concluding remarks are added in Sec. IV.

II. BASIC MASTER EQUATION

We consider a system of N identical three-level atoms in cascade configuration shown in Fig. 1. The atomic transitions $|2\rangle$ to $|1\rangle$ and $|3\rangle$ to $|2\rangle$ are resonantly driven by two single-mode continuous-wave (cw) lasers of

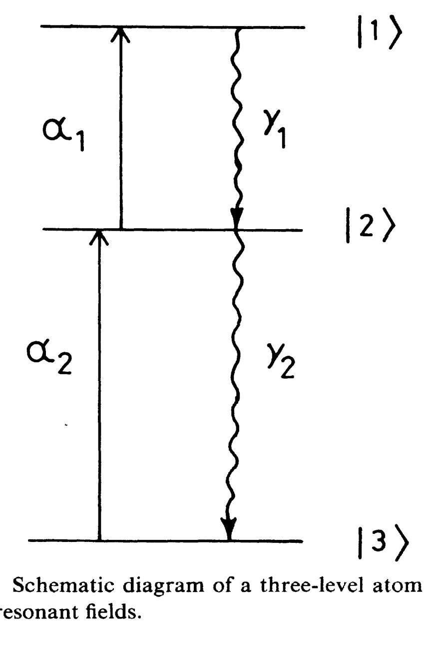

FIG. 1. Schematic diagram of a three-level atom interacting with two resonant fields.

respective Rabi frequencies $2\alpha_1$ and $2\alpha_2$. The Einstein coefficients A_1 and A_2 for spontaneous emission from the upper ($|1\rangle$) and lower ($|2\rangle$) excited levels are denoted by $2\gamma_1$ and $2\gamma_2$, respectively. We assume that the average distance between the atoms is such that the system of N atoms can be described by collective operators

$$A_{ij}=\sum_l A_{ij}^{(l)} , \tag{2.1}$$

where $A_{ij}^{(l)}$ is the operator $|i\rangle_{l\,l}\langle j|$ for the lth atom. The collective operators obey the commutation relations

$$[A_{ij}, A_{kl}]=A_{il}\delta_{jk}-\delta_{il}A_{kj} \tag{2.2}$$

and the conservation condition $\sum_i A_{ii}=N$. The master equation describing the stimulated and spontaneous transitions for the collective system can be obtained in the standard way[65] and reads as

$$\frac{d\rho}{dt}=-i[H_0,\rho]-\gamma_1(A_{12}A_{21}\rho+\rho A_{12}A_{21}-2A_{21}\rho A_{12}) -\gamma_2(A_{23}A_{32}\rho+\rho A_{23}A_{32}-2A_{32}\rho A_{23}) , \tag{2.3}$$

where ρ is the reduced atomic density operator and H_0 is the effective Hamiltonian given by

$$H_0=\alpha_1(A_{12}+A_{21})+\alpha_2(A_{23}+A_{32}) . \tag{2.4}$$

The master equation (2.3) involves the usual electric dipole and rotating-wave approximations. Further, the Born and Markov approximations with respect to the interaction with the continuum modes of the radiation field are inherent in the derivation. Lastly, the equation is written in a frame rotating with respect to the laser frequencies.

This equation cannot be solved analytically, in general. A considerable simplification arises if we assume the fields to be strong. In this case, it is convenient to go to a dressed-state picture and derive an approximate equation. For this purpose, we introduce the dressed states which are normalized eigenstates of the single-atom Hamiltonian $H_0^{(l)}$,

$$H_0^{(l)}|\psi_i^{(l)}\rangle=\lambda_i|\psi_i^{(l)}\rangle . \tag{2.5}$$

Diagonalizing the Hamiltonian $H_0^{(l)}$, we can obtain explicitly the dressed states,

$$\begin{aligned}|\psi_1^{(l)}\rangle&=\Gamma_2|1\rangle_l-\Gamma_1|3\rangle_l, \quad \lambda_1=0\\ |\psi_{2,3}^{(l)}\rangle&=\frac{1}{\sqrt{2}}(\Gamma_1|1\rangle_l\pm|2\rangle_l+\Gamma_2|3\rangle_l), \quad \lambda_{2,3}=\pm\Omega\end{aligned} \tag{2.6}$$

where $\Omega=(\alpha_1^2+\alpha_2^2)$, $\Gamma_{1,2}=\alpha_{1,2}/\Omega$. We may therefore write

$$\begin{aligned}|i\rangle_l&=\sum_{j-1}^{3}U_{ij}|\psi_j^{(l)}\rangle ,\\ |\psi_i^{(l)}\rangle&=\sum_{j=1}^{3}U_{ji}|j\rangle_l ,\end{aligned} \tag{2.7}$$

where the transformation matrix U is given by

$$U=\begin{bmatrix}\Gamma_2 & \Gamma_1/\sqrt{2} & \Gamma_1/\sqrt{2}\\ 0 & 1/\sqrt{2} & -1/\sqrt{2}\\ -\Gamma_1 & \Gamma_2/\sqrt{2} & \Gamma_2/\sqrt{2}\end{bmatrix} . \tag{2.8}$$

Note that the transformation matrix is real and orthogonal.

We now define the dressed operators $B_{ij}^{(l)}$ for the lth atom and the collective operators B_{ij} as

$$B_{ij}^{(l)}=|\psi_i^{(l)}\rangle\langle\psi_j^{(l)}| , \tag{2.9}$$

$$B_{ij}=\sum_l B_{ij}^{(l)} . \tag{2.10}$$

The new collective operators B_{ij} satisfy the same commutation relations and the closure property as the old operators A_{ij}. The transformation between the A_{ij} and B_{ij} operators reads as

$$A_{ij}=\sum_{k,l}U_{ik}U_{jl}B_{kl} , \tag{2.11}$$

$$B_{ij}=\sum_{k,l}U_{ki}U_{lj}A_{kl} . \tag{2.12}$$

It is important to point out here that under the collective Hamiltonian H_0, the operators B_{ij} evolve in time as

$$B_{ij}(t)=B_{ij}(0)\exp[i(\lambda_i-\lambda_j)t]. \tag{2.13}$$

The diagonal operators B_{ii} are then slowly varying in time while the off-diagonal operators B_{ij} ($i\neq j$) are rapidly oscillating in time. Hence, under intense field condition, that is, when $\Omega\gg N\gamma_1,N\gamma_2$, we may insert the transformation (2.11) into (2.3) and neglect the rapidly oscillating terms (secular approximation) to arrive at the approximate master equation

$$\begin{aligned}\frac{d\rho}{dt}=&-i\Omega[Q,\rho]-e([Q,Q\rho]+[\rho Q,Q])-([B_{23},B_{32},\rho]+[\rho B_{23},B_{32}]+[B_{32},B_{23}\rho]+[\rho B_{32},B_{23}])\\ &-f_1([B_{21},B_{12}\rho]+[\rho B_{21},B_{12}]+[B_{31},B_{13}\rho]+[\rho B_{31},B_{13}])\\ &-f_2([B_{12},B_{21}\rho]+[\rho B_{12},B_{21}]+[B_{13},B_{31}\rho]+[\rho B_{13},B_{31}]) ,\end{aligned} \tag{2.14}$$

where

$$Q=B_{22}-B_{33}\ , \tag{2.15}$$

$$e=(\gamma_1\Gamma_1^2+\gamma_2\Gamma_2^2)/4\ , \tag{2.16}$$

$$f_1=\frac{\gamma_2\Gamma_1^2}{2},\quad f_2=\frac{\gamma_1\Gamma_2^2}{2}\ . \tag{2.17}$$

The steady-state solution of the master equation (2.14) is obtained by using detailed balance. It has the form

$$\rho^{\rm SS}=D^{-1}\exp(-\mu B_{11})\ , \tag{2.18}$$

which has also been obtained in Ref. 66. Inserting (2.18) in the steady-state form $(d\rho/dt=0)$ of Eq. (2.14) one determines μ, while D is obtained by the requirement $\mathrm{Tr}\rho^{\rm SS}=1$. Hence, μ and D are given by

$$\mu=\ln\left[\frac{f_2}{f_1}\right],\quad D=\mathrm{Tr}[\exp(-\mu B_{11})]\ . \tag{2.19}$$

The analytical expressions for the various physical quantities of interest presented in the subsequent section are based on the steady-state solution (2.18) of the approximate master equation (2.14). Note that the assumption of the intense field limit is inherent in our results. A correction to these results will be of the order of $(N\gamma/\Omega)^2$ where γ is the larger of γ_1,γ_2. It is clear from the nature of the steady-state solution that the expectation values of all off-diagonal products of B_{ij} operators will vanish. The expectation values are obtained by introducing the collective states[67] $|N,n_1,n_2\rangle$ corresponding to SU(3) algebra of the dressed operators. Here, in the state $|N,n_1,n_2\rangle$, n_1 atoms are in the dressed state $|\psi_1\rangle$, n_2-n_1 in the state $|\psi_2\rangle$, and $N-n_2$ in the state $|\psi_3\rangle$. The matrix elements of the collective operators can be obtained from the set of relations

$$\begin{aligned}
&B_{11}|N,n_1,n_2\rangle=n_1|N,n_1,n_2\rangle\ ,\\
&B_{22}|N,n_1,n_2\rangle=(n_2-n_1)|N,n_1,n_2\rangle\ ,\\
&B_{33}|N,n_1,n_2\rangle=(N-n_2)|N,n_1,n_2\rangle\ ,
\end{aligned} \tag{2.20}$$

$$\begin{aligned}
&B_{12}|N,n_1,n_2\rangle=[(n_2-n_1)(n_1+1)]^{1/2}|N,n_1+1,n_2\rangle\ ,\\
&B_{13}|N,n_1,n_2\rangle=[(N-n_2)(n_1+1)]^{1/2}|N,n_1+1,n_2+1\rangle\ ,\\
&B_{23}|N,n_1,n_2\rangle=[(N-n_2)(n_2-n_1+1)]^{1/2}|N,n_1,n_2+1\rangle .
\end{aligned}$$

Also, for a given N, the allowed values of n_1, n_2 are $n_1=0,1,2,\ldots,N$ while $n_2=n_1,\ n_1+1,\ldots,N$. In particular, it is easy to derive the following results:

$$D=\sum_{n_1=0}^{N}\sum_{n_2=n_1}^{N}X^{n_1}=\frac{(N+1)-(N+2)X+X^{N+2}}{(1-X)^2}\ , \tag{2.21}$$

$$\langle B_{11}^r\rangle=\frac{1}{D}\left[X\frac{d}{dX}\right]^r D\ , \tag{2.22}$$

where

$$X=e^{-\mu}=\frac{\gamma_2}{\gamma_1}\left[\frac{\alpha_1}{\alpha_2}\right]^2\ . \tag{2.23}$$

We may also note here some explicit results for $X=1$ which are used in the next section:

$$D=\tfrac{1}{2}(N+1)(N+2)\ , \tag{2.24}$$

$$\langle B_{11}\rangle=\frac{N}{3},\quad \langle B_{11}^2\rangle=\frac{N(N+1)}{6}\ , \tag{2.25}$$

$$\begin{aligned}
&\langle B_{11}^3\rangle=\frac{1}{30}N(3N^2+6N+1)\ ,\\
&\langle B_{11}^4\rangle=\frac{1}{30}N(N+1)(2N^2+4N-1)\ .
\end{aligned} \tag{2.26}$$

It is also straightforward to show that

$$\lim_{N\to\infty}\frac{\langle B_{11}^n\rangle}{N^n}=\begin{cases}0, & X<1\\ 2/[(n+1)(n+2)], & X=1\\ 1, & X>1\end{cases} \tag{2.27}$$

for all n. This discontinuous limiting behavior of the expectation values of the powers of the dressed operator (B_{11}/N) results in the corresponding discontinuous behavior of the atomic observables discussed in the subsequent section.

III. STEADY-STATE EXPECTATION VALUES AND COOPERATIVE LIMITS

In this section we discuss the steady-state behavior of the atomic populations in the upper and lower excited levels, the corresponding fluorescence intensities $G_1^{(1)},G_2^{(1)}$ and the intensity correlations $g_{ij}^{(2)}(0)$. All these quantities can be expressed in terms of the expectation values of various powers of B_{11}. Further, they are functions of X and also depend on the ratio $\beta=\gamma_1/\gamma_2$. We shall also examine and discuss the limiting behavior as $N\to\infty$.

A. Atomic populations and fluctuations

The expectation value of the diagonal operator A_{ii} yields the population in the ith level. According to Eq. (2.11)

$$\langle A_{ii}\rangle=\sum_k U_{ik}U_{ik}\langle B_{kk}\rangle\ .$$

The fractional populations in the upper and lower excited levels are given by

$$\frac{\langle A_{11}\rangle}{N}=\frac{1}{2(1+\beta X)}\left[\left[1-\frac{\langle B_{11}\rangle}{N}\right]\beta X+\frac{2\langle B_{11}\rangle}{N}\right]\ , \tag{3.1}$$

$$\frac{\langle A_{22}\rangle}{N}=\frac{1}{2}\left[1-\frac{\langle B_{11}\rangle}{N}\right]\ . \tag{3.2}$$

The expectation value $\langle B_{11}\rangle$ may be readily evaluated using Eq. (2.22). It is of interest to derive the limiting behavior of $\langle A_{11}\rangle/N$ and $\langle A_{22}\rangle/N$ as $N\rightarrow\infty$. The resulting expressions read as

$$\lim_{N\rightarrow\infty}\frac{\langle A_{11}\rangle}{N}=\begin{cases}\dfrac{\beta X}{2(1+\beta X)}, & X<1\\ \frac{1}{3}, & X=1\\ \dfrac{1}{1+\beta X}, & X>1\end{cases} \tag{3.3}$$

and

$$\lim_{N\rightarrow\infty}\frac{\langle A_{22}\rangle}{N}=\begin{cases}\frac{1}{2}, & X<1\\ \frac{1}{3}, & X=1\\ 0, & X>1\ .\end{cases} \tag{3.4}$$

Note that in cooperative limit ($N\rightarrow\infty$) the atomic populations are equally divided among the three levels at the critical point $X=1$, while for $X>1$ the population in the intermediate level $|2\rangle$ tends to zero. It is clear that the fractional population show in the limit $N\rightarrow\infty$ a discontinuous behavior at $X=1$. This behavior is similar to that of a nonequilibrium first-order phase transition at the critical point $X=1$ where the ratio of the intensities of the driving fields is equal to β. The plots of $\langle A_{11}\rangle/N$ as functions of X for some infinite values of N and for $\beta=1$ are shown in Figs. 2(a) and 2(b), respectively. The dashed curves in Fig. 2 show the behavior as $N\rightarrow\infty$. Note that for finite values of N, the curves intersect at point $X=1$ where $\langle A_{11}\rangle/N$ and $\langle A_{22}\rangle/N=\frac{1}{3}$. Also, the discontinuous behavior of $\langle A_{22}\rangle/N$ is independent of β while that of $\langle A_{11}\rangle/N$ is dependent on β. Curiously enough, the discontinuity in $\langle A_{11}\rangle/N$ disappears for $\beta=2$ as seen from Eq. (3.3).

It may be of some interest to examine the behavior of the fluctuations in the atomic populations. We have

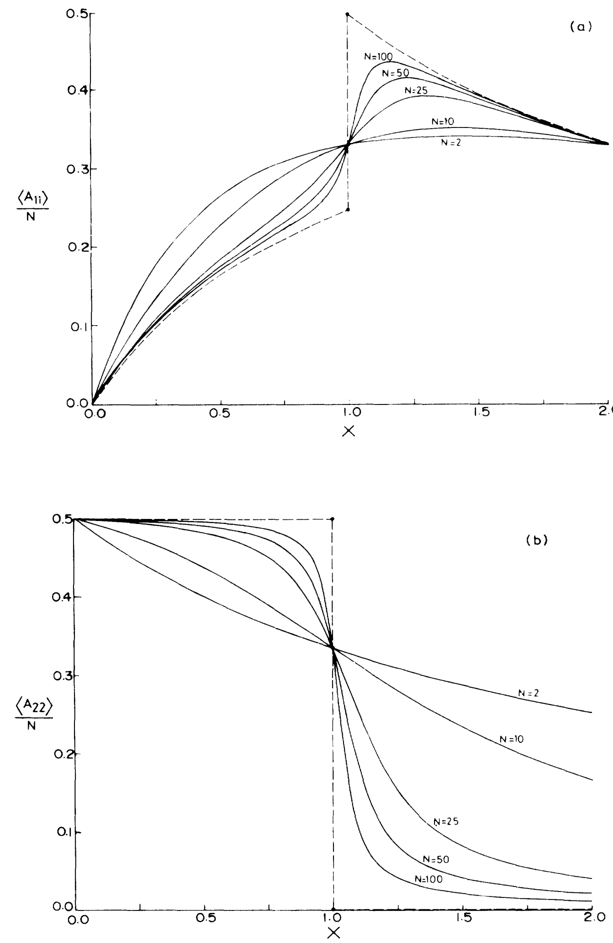

FIG. 2. Steady-state values of the fractional atomic populations in the (a) upper and (b) lower excited levels as a function of the parameter X for $\beta=1$. The solid (dashed) curves indicate the behavior for finite N ($N\rightarrow\infty$).

$$\begin{aligned}\sigma_1^2&=\langle A_{11}^2\rangle-\langle A_{11}\rangle^2\\&=\frac{1}{(1+\beta X)^2}\left[\frac{N}{2}\left\{\frac{N+2}{6}(\beta X)^2+\beta X\right\}\right.\\&\qquad\left.+[\tfrac{1}{2}(2N+1)\beta X-\tfrac{1}{6}(N+1)(\beta X)^2]\langle B_{11}\rangle+[\tfrac{1}{3}(\beta X)^2-2(\beta X)+1]\langle B_{11}^2\rangle-[\tfrac{1}{4}(\beta X)^2-\beta X+1]\langle B_{11}\rangle^2\right],\end{aligned} \tag{3.5}$$

$$\sigma_2^2=\langle A_{22}^2\rangle-\langle A_{22}\rangle^2=\tfrac{1}{2}[N(N+2)-2(N+1)\langle B_{11}\rangle+4\langle B_{11}^2\rangle-3\langle B_{11}\rangle^2]\ . \tag{3.6}$$

We may now take the cooperative limit $N\rightarrow\infty$ and show that

$$\lim_{N\rightarrow\infty}\frac{\sigma_1^2}{N^2}=\begin{cases}\dfrac{1}{2}\left[\dfrac{\beta X}{1+\beta X}\right]^2, & X<1\\ \frac{1}{18}, & X=1\\ 0, & X>1\end{cases} \tag{3.7}$$

$$\lim_{N\rightarrow\infty}\frac{\sigma_2^2}{N^2}=\begin{cases}\frac{1}{12}, & X<1\\ \frac{1}{18}, & X=1\\ 0, & X>1\ .\end{cases} \tag{3.8}$$

It is interesting to note here that the quantum fluctuations σ_1^2/N^2 and σ_2^2/N^2 are finite and nonzero in the regime $0<X<1$, which is characteristic of the cooperative behavior of the system in this regime. On the other hand, the vanishing of the fluctuations above the threshold $(X>1)$ indicates typically a single-atom behavior. Curves in Figs. 3(a) and 3(b) show the behavior of the fluctuations σ_i^2/N^2 $(i=1,2)$ with respect to the parameter X for finite N as well as for $N\rightarrow\infty$.

B. Intensities of fluorescence

The expressions for the intensities of fluorescence from the upper $|1\rangle$ and lower $|2\rangle$ excited levels are defined by the usual expressions[65]

$$G_i^{(1)}=\langle E_i^{(-)}E_i^{(+)}\rangle,\quad i=1,2 \tag{3.9}$$

where $E_i^{(\pm)}$ are the positive and negative frequency parts of the fluorescent electric field operator E_i at the detector and are, in general functions of space and time. Now, in the far-field limit, the fields $E_{1,2}^{(\pm)}$ can be expressed in terms of the atomic operators A_{21} and A_{32} as[65]

$$E_1^{(+)}(r,t)=E_{01}^{(+)}(r,t)+\tfrac{1}{2}\psi_1(r)A_{21}(t-r/c)\ ,$$

$$E_2^{(+)}(r,t)=E_{02}^{(+)}(r,t)+\tfrac{1}{2}\psi_2(r)A_{32}(t-r/c)\ . \tag{3.10}$$

Here, $E_{01}^{(+)}$ and $E_{02}^{(+)}$ are the incident laser fields and

$$\psi_{1,2}^2(r)=\left[\frac{3\hbar\omega\gamma_{1,2}}{2cr^2}\right]\sin^2\Theta_{1,2}\ , \tag{3.11}$$

$\Theta_{1,2}$ being the angle between the observation direction $\mathbf{r}$ and the atomic transition dipole moment $\mathbf{d}_{1,2}$. In particular, if the measurements are carried out in such a way that the incident fields do not contribute, the intensities in the steady state $(t\rightarrow\infty)$ are given by $G_1^{(1)}=\langle A_{12}A_{21}\rangle$ and $G_2^{(1)}=\langle A_{23}A_{32}\rangle$ apart from an unimportant spatial factor. The evaluation of $G_1^{(1)}$ and $G_2^{(1)}$ yields the analytical expressions

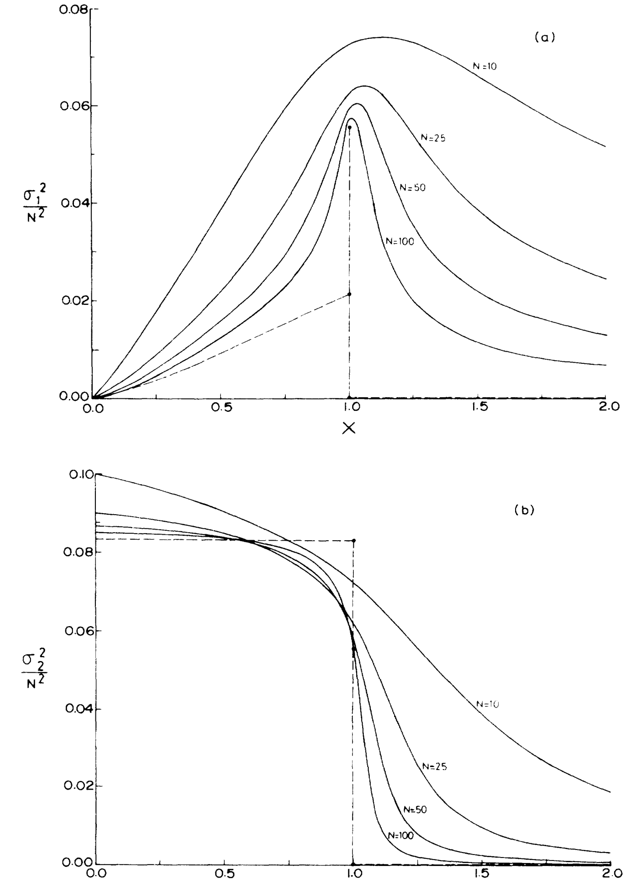

FIG. 3. Fluctuations σ_i^2/N^2 in the steady-state fractional population in the (a) upper and (b) lower excited levels as a function of X. Data same as in Fig. 2.

$$G_1^{(1)}=\langle A_{12}A_{21}\rangle$$

$$=\frac{1}{6(1+\beta X)}\{N(N+2)\beta X+[3(N+2)-2(N+1)\beta X]\langle B_{11}\rangle+(\beta X-3)\langle B_{11}^2\rangle\}\ , \tag{3.12}$$

$$G_2^{(1)}=\langle A_{23}A_{32}\rangle=\frac{1}{6(1+\beta X)}\{3N(\beta X)+N(N+2)+[3(N-1)\beta X-2(N+1)]\langle B_{11}\rangle+(1-3\beta X)\langle B_{11}^2\rangle\}\ . \tag{3.13}$$

It is clear from these results that

$$\lim_{N\rightarrow\infty}\frac{G_1^{(1)}}{N^2}=\begin{cases}\dfrac{\beta X}{6(1+\beta X)}, & X<1\\ \frac{1}{12}, & X=1\\ 0, & X>1\end{cases} \tag{3.14}$$

$$\lim_{N\rightarrow\infty}\frac{G_2^{(1)}}{N^2}=\begin{cases}\dfrac{1}{6(1+\beta X)}, & X<1\\ \frac{1}{12} & X=1\\ 0, & X>1\ .\end{cases} \tag{3.15}$$

Thus, below the bifurcation threshold $X=1$ the intensities $G_1^{(1)}$ and $G_2^{(1)}$ are proportional to N^2, indicating that the system behaves cooperatively. Above the threshold, the intensities vary as N and the system behaves as a collection of atoms radiating independently of each other.

The normalized intensities $G_1^{(1)}/N^2$ and $G_2^{(1)}/N^2$ as functions of the parameter X for some finite values of N and for $\beta=1$ are shown in Figs. 4(a) and 4(b), respectively. The dotted curve in each of these figures corresponds to the limiting behavior as $N\rightarrow\infty$. Note that the mean dipole moment corresponding to the transition $|1\rangle\rightarrow|2\rangle$ $(|2\rangle\rightarrow|3\rangle)$ is proportional to $\langle A_{12}\rangle$ $(\langle A_{23}\rangle)$. Since the steady-state expectation values $\langle B_{ij}\rangle=0$ $(i\neq j)$ and $\langle B_{22}\rangle=\langle B_{33}\rangle$, it follows from the transformation equation (2.11) that the mean dipole moment corresponding to $|1\rangle\rightarrow|2\rangle$ $(|2\rangle\rightarrow|3\rangle)$ vanishes identically over the range

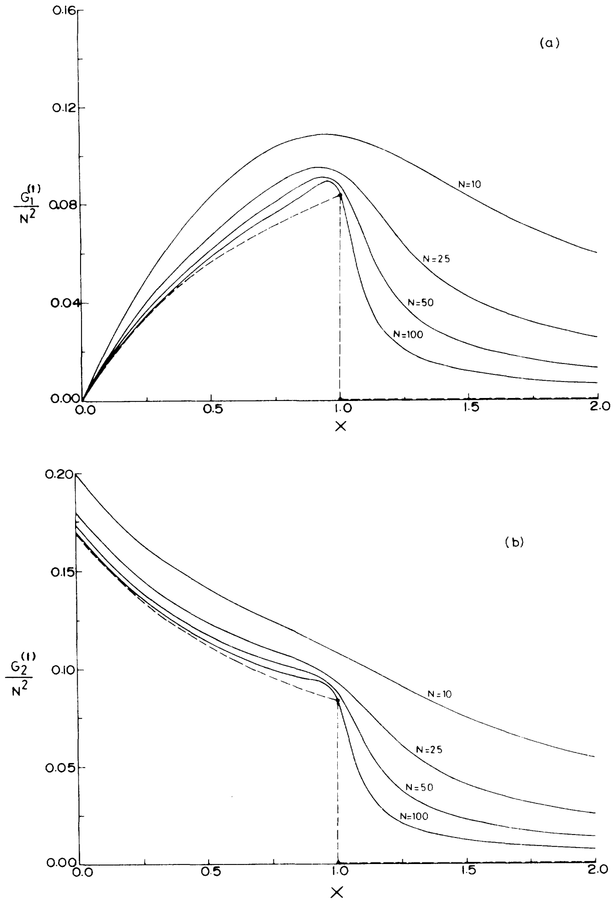

FIG. 4. Steady-state fluorescent intensities $G_i^{(1)}/N^2$ plotted against X for finite N and $N\rightarrow\infty$. (a) $G_1^{(1)}/N^2$ and (b) $G_2^{(1)}/N^2$ ($\beta=1$).

($0<X<\infty$). The superradiant behavior of G_1 and G_2 for $X<1$ thus arises solely from the quantum fluctuations. This is analogous to the intense field limit of cooperative resonance fluorescence from a system of two-level atoms.

C. Intensity-intensity correlation and violation of CS inequality

The nature of the fluorescent light emitted from the excited levels $|1\rangle$ and $|2\rangle$ may be understood from the normalized second-order intensity correlation functions which are defined by

$$g_{ij}^{(2)}(\tau)=\frac{\langle T_N[G_i^{(1)}(\tau)G_j^{(1)}(t+\tau)]\rangle}{\langle G_i^{(1)}(t)\rangle\langle G_j^{(1)}(t+\tau)\rangle}\ , \tag{3.16}$$

where T_N is the time-ordering normal ordering operator. In the far-field limit, we may use the relations (3.9) and (3.10) to express $g_{ij}^{(2)}(\tau)$ in terms of the atomic operators A_{ij}. In particular, we are interested in the steady-state correlations $g_{ij}^{(2)}(0)$ which are given by

$$g_{ij}^{(2)}(0)=\langle S_i^\dagger S_j^\dagger S_j S_i\rangle/(\langle S_i^\dagger S_i\rangle\langle S_j^\dagger S_j\rangle)\ , \tag{3.17}$$

where, for brevity, we have set $S_1=A_{21}$ and $S_2=A_{32}$. The quantity $g_{ij}^{(2)}(0)$ $(i,j=1,2)$ is a measure of the probability of detecting simultaneously a photon from the ith excited level and another photon emitted from the jth excited level. For a single atom $g_{11}^{(2)}(0)=g_{21}^{(2)}(0)=g_{22}^{(2)}(0)=0$, while $g_{12}^{(2)}=1/\langle A_{22}\rangle\neq 0$. It turns out that the intensity correlations $g_{ij}^{(2)}(0)$ can be expressed in terms of the expectation values $\langle B_{11}^n\rangle$ $(n=1,\ldots,4)$. The analytical expressions for $g_{ij}^{(2)}(0)$ are as follows:

$$g_{11}^{(2)}(0)=\frac{1}{(\beta X+1)^2}\frac{1}{(G_1^{(1)})^2}\times[(\beta X)^2\langle R_1\rangle+\beta X\langle R_2\rangle+\langle R_3\rangle]\ , \tag{3.18}$$

$$g_{22}^{(2)}(0)=\frac{1}{(\beta X+1)^2}\frac{1}{(G_2^{(1)})^2}\times[(\beta X)^2\langle R_4\rangle+\beta X\langle R_5\rangle+\langle R_1\rangle]\ , \tag{3.19}$$

$$g_{12}^{(2)}(0)=\frac{1}{(\beta X+1)^2}\frac{1}{G_1^{(1)}G_2^{(1)}}\times[(\beta X)^2\langle R_6\rangle+\beta X\langle R_7\rangle+\langle R_8\rangle]\ , \tag{3.20}$$

$$g_{21}^{(2)}(0)=\frac{1}{(\beta X+1)^2}\frac{1}{G_1^{(1)}G_2^{(1)}}\times[(\beta X)^2\langle R_9\rangle+\beta X\langle R_{10}\rangle+\langle R_{11}\rangle]\ . \tag{3.21}$$

The various expectation values $\langle R_i\rangle$ $(i=1,2,\ldots,11)$ have the following expressions:

$$\langle R_1\rangle=\tfrac{1}{30}[N(N-1)(N+2)(N+3)-2(N+1)(2N^2+4N-3)\langle B_{11}\rangle+(6N^2+12N+1)\langle B_{11}^2\rangle-4(N+1)\langle B_{11}^3\rangle+\langle B_{11}^4\rangle]\ , \tag{3.22}$$

$$\langle R_2\rangle=\tfrac{1}{3}[N(N+2)(N+3)\langle B_{11}\rangle-(3N^2+10N+6)\langle B_{11}^2\rangle+(3N+5)\langle B_{11}^3\rangle-\langle B_{11}^4\rangle]\ , \tag{3.23}$$

$$\langle R_3\rangle=\tfrac{1}{3}[-(N+2)(N+3)\langle B_{11}\rangle+(N^2+7N+11)\langle B_{11}^2\rangle-2(N+3)\langle B_{11}^3\rangle+\langle B_{11}^4\rangle]\ , \tag{3.24}$$

$$\langle R_4\rangle=\tfrac{1}{3}[2N(N-1)+(3N-1)(N-2)\langle B_{11}\rangle+(N^2-7N+5)\langle B_{11}^2\rangle-(N-2)\langle B_{11}^3\rangle+\langle B_{11}^4\rangle]\ , \tag{3.25}$$

$$\langle R_5\rangle=\tfrac{1}{3}[N(N-1)(N+2)+(N^3-2N^2-4N+2)\langle B_{11}\rangle-(3N^2-N-3)\langle B_{11}^2\rangle+3N\langle B_{11}^3\rangle-\langle B_{11}^4\rangle]\ , \tag{3.26}$$

$$\langle R_6\rangle=\tfrac{1}{12}[N(N+1)(N+2)+(N^3-4N-2)\langle B_{11}\rangle-(3N^2+3N-1)\langle B_{11}^2\rangle+(3N+2)\langle B_{11}^3\rangle-\langle B_{11}^4\rangle]\ , \tag{3.27}$$

$$\langle R_7\rangle=\tfrac{1}{30}[N(N+2)(N^2+2N+2)-(N+1)(9N^2+18N+14)\langle B_{11}\rangle+(N+1)(31N+46)\langle B_{11}^2\rangle$$
$$-3(13N+18)\langle B_{11}^3\rangle+16\langle B_{11}^4\rangle]\ , \tag{3.28}$$

$$\langle R_8\rangle=\tfrac{1}{12}[(N+2)^2(N+3)\langle B_{11}\rangle-(N+2)(3N+8)\langle B_{11}^2\rangle+(3N+7)\langle B_{11}^3\rangle-\langle B_{11}^4\rangle]\ , \tag{3.29}$$

$$\langle R_9\rangle=\tfrac{1}{12}[N^2(N-1)+N(N^2-5N+3)\langle B_{11}\rangle-(3N-1)(N-2)\langle B_{11}^2\rangle+3(N-1)\langle B_{11}^3\rangle-\langle B_{11}^4\rangle]\ , \tag{3.30}$$

$$\langle R_{10}\rangle=\tfrac{1}{30}[(N-1)N(N^2+1)-(9N^3-8N^2+2N-1)\langle B_{11}\rangle+(31N^2-8N+1)\langle B_{11}^2\rangle$$
$$-(39N-1)\langle B_{11}^3\rangle+16\langle B_{11}^4\rangle]\ , \tag{3.31}$$

$$\langle R_{11}\rangle=\tfrac{1}{12}[N(N+1)(N+2)\langle B_{11}\rangle-(3N^2+6N+2)\langle B_{11}^2\rangle+3(N+1)\langle B_{11}^3\rangle-\langle B_{11}^4\rangle]\ . \tag{3.32}$$

The limiting behavior of $g_{ij}^{(2)}(0)$ as $N\to\infty$ resulting from Eqs. (3.22)–(3.32) can be shown to be given by

$$g_{ii}^{(2)}(0)=\begin{cases}1.2, & X<1\\ 1.6, & X=1 \quad (i=1,2),\\ 2.0, & X>1\end{cases} \tag{3.33}$$

$$g_{12}^{(2)}(0)=\begin{cases}1.2, & X<1\\ 0.8, & X=1\\ 1+X, & X>1\end{cases} \tag{3.34}$$

$$g_{21}^{(2)}(0)=\begin{cases}1.2, & X<1\\ 0.8, & X=1\\ 1+1/X, & X>1\ .\end{cases} \tag{3.35}$$

Thus, in the cooperative limit $N\to\infty$, the normalized intensity-intensity correlation functions show a discontinuous transition similar to a typical nonequilibrium first-order phase transition at the critical point $X=1$. The behavior of these correlation functions in the regime below and above the threshold is also interesting. For $X<1$, the intensity correlations show that the scattered radiation is partially coherent $[g_{ii}^{(2)}(0)<2]$, which is consistent with cooperative behavior in this regime. On the other hand, the limiting values of $g_{11}^{(2)}(0)=g_{22}^{(2)}(0)=2$ above the threshold imply that the radiation emitted from the excited levels $|1\rangle$ and $|2\rangle$ is totally incoherent. This is a manifestation of the fact that in the parameter regime $X>1$ the atoms tend to radiate independently, as is also evident from the limiting behavior of the intensities $G_1^{(1)}/N^2$ and $G_2^{(1)}/N^2$ discussed in Sec. II B. The curves in Figs. 5(a) and 5(b) show the behavior of $g_{ii}^{(2)}(0)$ $(i=1,2)$ for some finite values of N and in the limit $N\to\infty$. The cross correlations $g_{12}^{(2)}(0)$ and $g_{21}^{(2)}(0)$ are shown in Figs. 6(a) and 6(b) for the same data as in Fig. 5. Note that in Fig. 6 the antibunching behavior is shown by these correlations in the region around $X=1$ for some finite N. As $N\to\infty$, the region over which the antibunching is exhibited collapses to a single critical point $X=1$ (dashed curves in Fig. 6).

Incidentally, the analytical expressions for $g_{ij}^{(2)}(0)$ may be used to test the violation of Cauchy-Schwarz inequalities. For this purpose we introduce two quantities,

$$\chi_1=[g_{11}^{(2)}(0)g_{22}^{(2)}(0)]/[g_{12}^{(2)}(0)]^2\ , \tag{3.36}$$

$$\chi_2=[g_{11}^{(2)}(0)g_{22}^{(2)}(0)]/[g_{21}^{(2)}(0)]^2\ . \tag{3.37}$$

The CS inequality is violated if $\chi_{1,2}<1$. This would be the case if the cross correlations between the photons emitted from the two different excited levels are larger than the correlation between the photons emitted from the same level. Clearly for $N=1$ $\chi_1=0$ for all values of X and CS inequality is violated. We may also verify that for $N\le 3$, $\chi_1<1$ for all values of X. The behavior of $\chi_{1,2}$ in

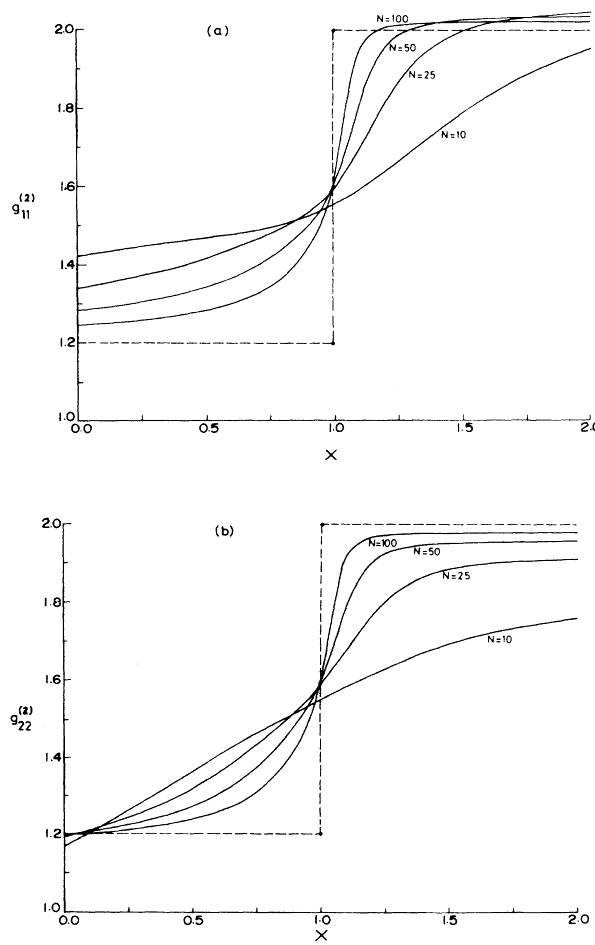

FIG. 5. Steady-state normalized intensity-intensity correlations (a) $g_{11}^{(2)}$ and (b) $g_{22}^{(2)}$ plotted against the paramter X $(\beta=1)$. The limiting behavior as $N\to\infty$ is shown by the dashed curve.

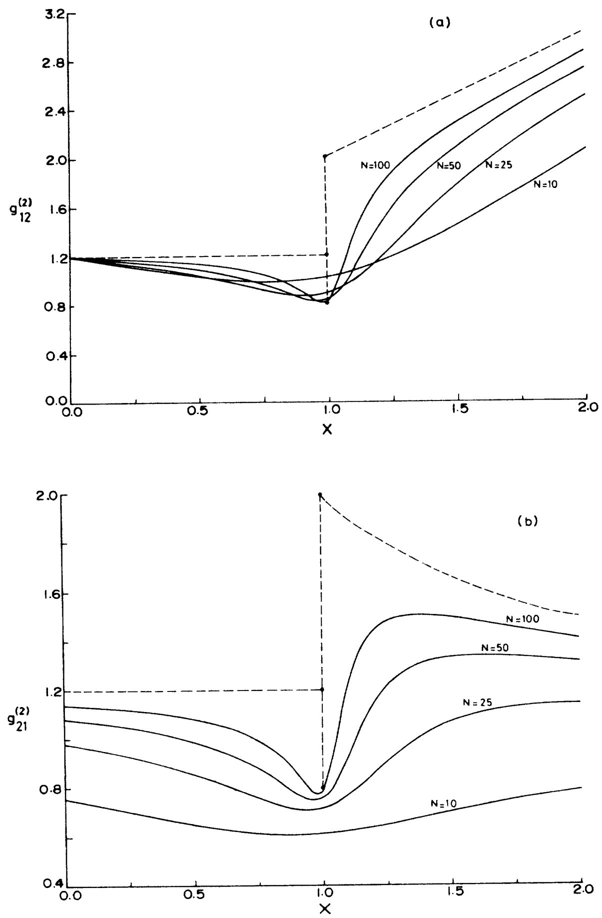

FIG. 6. Steady-state normalized cross-correlations (a) $g_{12}^{(2)}$ and (b) $g_{21}^{(2)}$ shown as functions of X ($\beta=1$). The behavior as $N\rightarrow\infty$ is shown by the dashed curve.

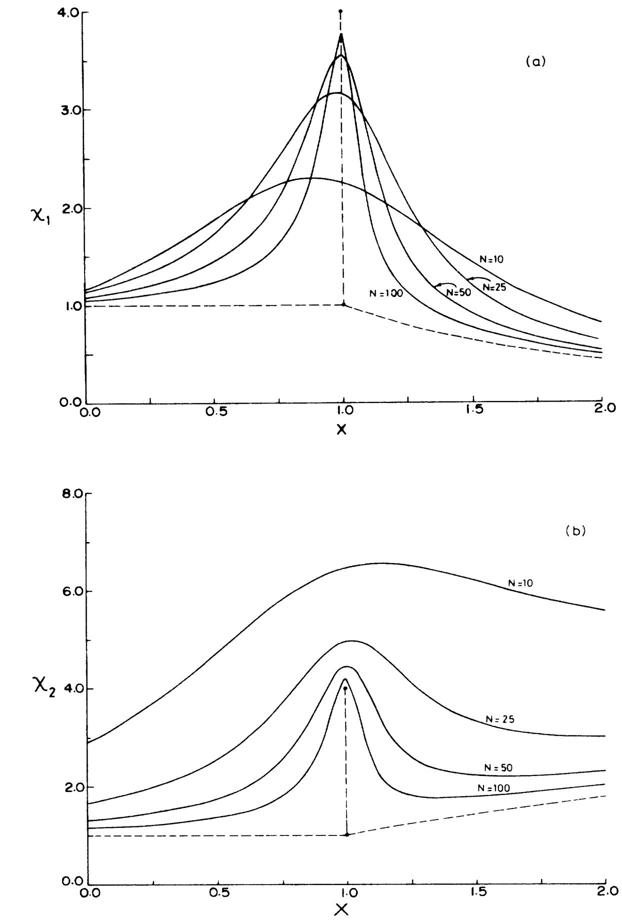

FIG. 7. Plots of CS inequality functions χ_i against X. (a) χ_1 and (b) χ_2 ($\beta=1$). The dashed curve indicates the limit $N\rightarrow\infty$.

the limit $N\rightarrow\infty$ follows from Eqs. (3.33)–(3.35). Figures 7(a) and 7(b) show the plots of χ_1 and χ_2 as functions of X for some finite values of N and for $\beta=1$. It is seen from the curves in Fig. 7(a) that $\chi_1<1$ for $X>1$, indicating that violation of CS inequality persists for increasing values of N. We may also mention here that the violation is more for lower values of β. In the cooperative limit $N\rightarrow\infty$, we have $\chi_1<1$ in the entire region $1<X<\infty$ manifesting, thereby, a strong macroscopic violation of the CS inequality [dashed curve in Fig. 7(a)]. On the other hand, χ_2 shows no violation of CS inequality for all finite values of N as well as for $N\rightarrow\infty$ [Fig. 7(b)].

IV. CONCLUSIONS

In this paper, we have examined analytically the steady-state behavior of a collection of strongly driven N identical three-level atoms in cascade configuration. Our analysis predicts that in the cooperative limit $N\rightarrow\infty$, the atomic observables have a discontinuous behavior with respect to a parameter X. The parameter X is characterized by the relative intensities of the two driving fields and the ratio of the spontaneous decay coefficients of the two excited levels. The critical behavior predicted by the present model at $X=1$ is similar to a nonequilibrium phase transition of first order. In particular, the threshold $X=1$ marks a transition from superradiant emission to incoherent emission. The superradiant character of the emitted radiation below the threshold arises essentially due to quantum fluctuations. This is also evident from the behavior of normalized intensity-intensity correlations $g_{11}^{(2)}(0)$ and $g_{22}^{(2)}(0)$ in the limit $N\rightarrow\infty$.

The analytical expressions for intensity-intensity correlations also predict a strong violation of the Cauchy-Schwarz inequality in the region above the threshold ($X>1$) for finite values of N and also in the limit $N\rightarrow\infty$. This interesting result emphasizes a macroscopic manifestation of a nonclassical feature of the cascade emission.

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