Three-level atom in a broadband squeezed vacuum field. I. General theory

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A complete treatment of a three-level ladder system interacting with a broadband squeezed vacuum field is presented. It is assumed that the ground state and the second excited state are decoupled in the electric-dipole approximation, and are tuned close to the sum frequency of the incident squeezed vacuum field. Using Zwanzig's projection-operator techniques, we derive the master equation, assuming that the system interacts with a broadband squeezed vacuum field in one or more dimensions. It is shown that, in the first case, the squeezed vacuum introduces new decay constants and frequency-shift parameters. These have the same dependence on the atom-radiation coupling parameter as the ordinary vacuum decay rate and frequency shift, the only major difference being that they are multiplied by the squeezing parameters M and N. In more dimensions, the decay constants and frequency-shift parameters depend on the solid angle Ω over which the squeezing is propagated. For $\Omega=0$ these correspond to the usual Einstein A coefficients and Lamb shifts of the atomic levels, while for large Ω they are similar to those for the one-dimensional squeezed vacuum.

I. INTRODUCTION

With the recent successful observations of squeezed states of the electromagnetic field,[1,2] a great deal of attention has been given to possible applications to radiation-matter interactions. Squeezing is the noise reduction that can occur in a quantum field when the quantum fluctuations in one of the field quadrature phases are reduced below the usual vacuum level. This, of course, is at the expense of increased noise in the conjugate quadrature. New theories of the interaction of two-level atoms with the vacuum field have been recently developed[3,4] based on the assumption that the atoms interact with a multimode broadband squeezed-vacuum field. This is generated by squeezed light, which has zero average electric field, but reduced fluctuations in one quadrature, and a bandwidth much larger than the natural linewidth of the atoms. These theories have assumed an ideal coupling between the atoms and the squeezed vacuum field. The earliest work assumed that the atoms interact only with squeezed modes of the radiation field, with no interactions or spontaneous emission into ordinary (unsqueezed) vacuum modes. This is a significant practical problem, which Gardiner pointed out in his original paper,[4] stating the need for either an incoming squeezed electric-dipole wave, or an appropriate one-dimensional situation. Following this suggestion Parkins and Gardiner[5] extended the theory to three dimensions, assuming that the two-level atom was located in a microscopic plane-mirror Fabry-Pérot cavity, and interacted with a squeezed input field incident over some finite solid angle. All other modes were in an ordinary vacuum state. Such a finite and focused beam of squeezed light can be produced using squeezed light together with a system of lenses and phase plates. With this modification they have demonstrated that a significant reduction in fluctuations experienced by the atom can be achieved in one quadrature when the atom is located at the point in which the input squeezed field is focused. The reduction in fluctuations depends also on the solid angle Ω over which squeezing is propagated and increases with increasing Ω.

In this paper we shall consider a three-level atom in a cascade configuration (ladder system) interacting with a broadband squeezed vacuum field. In practice this model can be realized by the pumping of the atomic transitions by correlated light beams as, for example, can be obtained from the output of a parmetric amplifier.[6] In a parametric amplifier an intense laser beam at frequency 2ω—the pump beam—illuminates a suitable nonlinear medium. The nonlinearity couples the pump beam to other modes of the electromagnetic field in such a way that a pump photon at frequency 2ω can be annihilated to create strongly correlated pairs of photons at frequencies $\omega\pm\varepsilon$. These correlations lead to the unequal partition of the quantum noise between two quadrature components $E_1(t)$ and $E_2(t)$ of the electromagnetic field $E(t)$ emitted by the parametric amplifier. In our theoretical model an effective three-level atom interacts with squeezed light. The squeezed light is assumed to be broadband relative to the natural linewidth of the individual atomic transitions. In Sec. II we discuss the Hamiltonian in a one-dimensional squeezed vacuum. In Sec. III we derive the master equation for the reduced density operator ρ of the three-level atom interacting with the incident squeezed light. We assume that the squeezed light is a multimode one-dimensional field, and there is no spontaneous emission into unsqueezed modes. In Sec. IV we extend this model to an experimentally more realistic three-dimensional model in which the incident squeezed light is propagated over a solid angle Ω. We also allow spontaneous emission into the unsqueezed modes and discuss in detail the effect of such a "nonideal" coupling between the atom and the squeezed field on the coefficients for spontaneous emission and the frequency shifts. Finally, some concluding remarks are made in Sec. V.

The formalism developed in this paper will be applied in the following paper to investigate the spontaneous-emission properties of a three-level system. We will study in detail the transient and steady-state solutions for the populations of the atomic levels.

II. HAMILTONIAN IN A ONE-DIMENSIONAL SQUEEZED VACUUM

We consider a three-level atom in the cascade configuration with unequally spaced levels ($E_3 > E_2 > E_1$), coupled to a quantized multimode electromagnetic field (Fig. 1). The transition frequencies from the ground state $|1\rangle$ to the first excited state $|2\rangle$ and from the state $|2\rangle$ to the second excited state $|3\rangle$ are ω_{21} and ω_{32}, respectively. The transitions are connected by electric-dipole moments $\boldsymbol{\mu}_{12}$ and $\boldsymbol{\mu}_{23}$, respectively, whereas the transition $|1\rangle \to |3\rangle$ is forbidden in the electric-dipole approximation.

The Hamiltonian for the three-level atom interacting with the quantized one-dimensional multimode radiation field is given by[7]

$$H = H_A + H_F + H_{\text{in}} \ . \tag{1}$$

The atomic part of the Hamiltonian has the eigenvalue equation

$$H_A |m\rangle = E_m |m\rangle \ , \quad m = 1,2,3 \tag{2}$$

where

$$\hbar\omega_{ij} = E_i - E_j \ .$$

The field part of the Hamiltonian is the usual expression

$$H_F = \hbar \int d\omega_\lambda \omega_\lambda a^\dagger(\omega_\lambda) a(\omega_\lambda) \ , \tag{3}$$

where $a(\omega_\lambda)$ and $a^\dagger(\omega_\lambda)$ are bosonic operators for the electromagnetic field.

The interaction part of the Hamiltonian can be written in the electric-dipole approximation in the form

$$H_{\text{in}} = i\hbar \int d\omega_\lambda \left[\left[\sum_i \sum_{\substack{j \\ i\neq j}} g_{ij}(\omega_\lambda) S_{ij} \right] a(\omega_\lambda) - \text{H.c.} \right] \ , \tag{4}$$

where $S_{ij} = |i\rangle\langle j|$, $(i,j = 1,2,3)$ are the atomic operators satisfying the usual commutation relations

$$[S_{ij}, S_{pq}] = S_{iq}\delta_{jp} - S_{pj}\delta_{qi} \ , \tag{5a}$$

and the closure property

$$S_{11} + S_{22} + S_{33} = 1 \ . \tag{5b}$$

The coefficients $g_{21}(\omega_\lambda)$ and $g_{32}(\omega_\lambda)$ describe the coupling of the atomic transitions $|1\rangle \to |2\rangle$ and $|2\rangle \to |3\rangle$, respectively, with the electromagnetic field. For comparison with other notation, we sometimes use the compact notation of $\omega_i \equiv \omega_{i+1,i}$ and $g_i \equiv g_{i+1,i}$. In Eq. (4) we have explicitly taken $g_{31}(\omega_\lambda)$ to be zero, assuming that the transition $|1\rangle \to |3\rangle$ is forbidden in electric-dipole approximation. The integration in Eq. (4) extends over a relevant frequency interval around the optical frequencies ω_{21} and ω_{32}, and the coupling coefficients $g_{ij}(\omega_\lambda)$ are assumed to vary smoothly over this frequency band. In practical terms, some type of waveguide would be required to obtain this interaction Hamiltonian and one-dimensional mode structure.

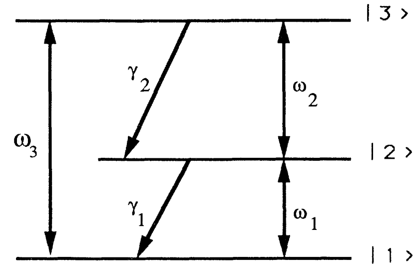

FIG. 1. Energy-level diagram of a three-level atom in a cascade configuration with possible transitions.

We wish to derive from the Hamiltonian (1) the master equation for the reduced operator $\rho_s(t)$ of the three-level system. The normal treatment of the interaction between the atom and the quantized radiation field assumes that the field is in the vacuum state. Here we assume that the quantized radiation field is in a broadband *squeezed* vacuum state with a carrier frequency ω which is tuned closed to half the frequency of the atomic transition $|1\rangle \to |3\rangle$, i.e., $2\omega \simeq \omega_{21} + \omega_{32}$. The bandwidth of squeezing is assumed to be sufficiently broad that the squeezed vacuum appears as δ-correlated squeezed white noise to the atom. The correlation function for the field operators $a(\omega_\lambda)$ and $a^\dagger(\omega_\lambda)$ can then be written as[3,4]

$$\begin{aligned} \langle a(\omega_\lambda) a^\dagger(\omega_\lambda') \rangle &= [N(\omega_\lambda)+1]\delta(\omega_\lambda - \omega_\lambda') \ , \\ \langle a^\dagger(\omega_\lambda) a(\omega_\lambda') \rangle &= N(\omega_\lambda)\delta(\omega_\lambda - \omega_\lambda') \ , \\ \langle a(\omega_\lambda) a(\omega_\lambda') \rangle &= M(\omega_\lambda)\delta(2\omega - \omega_\lambda - \omega_\lambda') \\ &= M(2\omega - \omega_\lambda)\delta(2\omega - \omega_\lambda - \omega_\lambda') \ , \end{aligned} \tag{6}$$

where $N(\omega_\lambda)$ and $M(\omega_\lambda)$ are slowly varying functions of the frequency and characterize the squeezing such that (see Appendix A)

$$|M(\omega_\lambda)|^2 \le N(\omega_\lambda) N(2\omega - \omega_\lambda) + \min[N(\omega_\lambda), N(2\omega - \omega_\lambda)] \ . \tag{7}$$

In the above equation the equality holds for a minimum-uncertainty squeezed state, and $M(\omega_\lambda) = |M(\omega_\lambda)| \exp(i\phi_v)$, where ϕ_v is the phase of the squeezed vacuum. Note that the field mode at frequency ω_λ is correlated with the mode at frequency $(2\omega - \omega_\lambda)$ through the phase-sensitive term characterized by the factor $M(\omega_\lambda)$. For $|M(\omega_\lambda)| = 0$, Eq. (6) describes a thermal field (blackbody field) at a temperature T, with $N(\omega_\lambda)$ the mean occupation number of the mode λ with the frequency ω_λ.

The inequality (7) holds for the general case in which the intensities $N(\omega_\lambda)$ and $N(2\omega - \omega_\lambda)$ may be different as, for example, in the output from a parametric amplifier with frequency-dependent losses. In the case of equal intensities, the usual inequality is recovered, in the form[3,4]

$$|M(\omega_\lambda)|^2 \le N(\omega_\lambda)[1 + N(2\omega - \omega_\lambda)] \ . \tag{8}$$

This inequality is less stringent than Eq. (7), which is a more suitable form for describing correlations of fields that might have unequal intensities.

III. ONE-DIMENSIONAL MASTER EQUATION

Our aim is to derive from the Hamiltonian (1) the master equation for a reduced system density operator

$$\rho_s(t)=\mathrm{tr}_b W(t) , \tag{9}$$

where the trace is taken over a squeezed reservoir and $W(t)$ is the density operator of the total system. The density operator $W(t)$ obeys the equation

$$\dot{W}(t)=-(i/\hbar)[H,W(t)]=-iLW(t) \tag{10}$$

with the initial condition $W(t_0)=\rho_s(t_0)\rho_b(t_0)$, where $\rho_s(t_0)$ and $\rho_b(t_0)$ are density operators corresponding to the atomic system and squeezed reservoir, respectively. In Eq. (10), L is the Liouville operator.

The master equation for the reduced density operator $\rho_s(t)$ of the three-level atom interacting with the squeezed-vacuum field can be derived by using any of a number of traditional techniques as long as the relevant reservoir is assumed to be squeezed. Here, we will use Zwanzig's projection-operator techniques.[8,9] Let P be a projection operator defined by

$$P\cdots=G\,\mathrm{tr}_b\cdots , \tag{11}$$

where the operator G should be such that $P^2=P$.

It is clear from (9) and (11) that

$$PW(t)=G\rho_s(t) , \tag{12}$$

and if we choose $G=\rho_s(0)$, then

$$PW(t)=\rho_b(0)\rho_s(t) \tag{13}$$

and

$$\rho_s(t)=\mathrm{tr}_b PW(t) . \tag{14}$$

Employing Eq. (10) and the Hamiltonian (1), after some algebra we derive[10]

$$\frac{\partial}{\partial t}[PW(t)]+\int_0^t d\tau\, PL_{\mathrm{in}}^I(t)L_{\mathrm{in}}^I(t-\tau)PW^I(t-\tau)=0 , \tag{15}$$

where the superscript I stands for the operators in the interaction picture and

$$L_{\mathrm{in}}^I\cdots\cdot=[H_{\mathrm{in}},\cdots\cdot]/\hbar . \tag{16}$$

After a Laplace transform over time t, with (6), (14), and (16), Eq. (15) reduces to

$$\begin{aligned}\tilde{\rho}(0)-z\tilde{\rho}(z)=&\sum_{i,j=1}^{2}[\eta_{ij}^{(+)}(z)-i\Delta\omega_{ij}(z)]\{[S_j^+,\tilde{\rho}(z)S_i^+]+[S_i^+\tilde{\rho}(z),S_j^+]\}\\&+\sum_{i,j=1}^{2}[\eta_{ij}^{(+)*}(z)+i\Delta\omega_{ij}^*(z)]\{[S_j^-,\tilde{\rho}(z)S_i^-]+[S_i^-\tilde{\rho}(z),S_j^-]\}\\&+\sum_{i,j=1}^{2}\gamma_{ij}^{(+)}(z)[\tilde{\rho}(z)S_j^-S_i^++S_j^-S_i^+\tilde{\rho}(z)-2S_i^+\tilde{\rho}(z)S_j^-]\\&+\sum_{i,j=1}^{2}[\gamma_{ij}^{(+)}(z)+\gamma_{ij}^{(-)}(z)][\tilde{\rho}(z)S_i^+S_j^-+S_i^+S_j^-\tilde{\rho}(z)-2S_j^-\tilde{\rho}(z)S_i^+]\\&+i\sum_i[\Delta\omega_i^0(z)+\Delta\omega_i(z)][|i\rangle\langle i|,\tilde{\rho}(z)] .\end{aligned} \tag{17}$$

Here $\tilde{\rho}(z)$ is the Laplace transform of $\rho_s(t)$, and the parameters are given by

$$\begin{aligned}\eta_{ij}^{(+)}(z)&=\int d\omega_\lambda g_i(\omega_\lambda)g_j(2\omega-\omega_\lambda)\frac{M(\omega_\lambda)z}{[z^2+(\omega_i-\omega_\lambda)^2]} ,\\ \gamma_{ij}^{(+)}(z)&=\int d\omega_\lambda g_i(\omega_\lambda)g_j^*(\omega_\lambda)\frac{N(\omega_\lambda)z}{[z^2+(\omega_i-\omega_\lambda)^2]} ,\\ \gamma_{ij}^{(-)}(z)&=\int d\omega_\lambda g_i(\omega_\lambda)g_j^*(\omega_\lambda)\frac{z}{[z^2+(\omega_i-\omega_\lambda)^2]} ,\\ \Delta\omega_{ij}(z)&=\int d\omega_\lambda g_i(\omega_\lambda)g_j(2\omega-\omega_\lambda)\frac{M(\omega_\lambda)(\omega_i-\omega_\lambda)}{[z^2+(\omega_i-\omega_\lambda)^2]} ,\\ \Delta\omega_i^0(z)&=\sum_{j\neq i}\int d\omega_\lambda|g_{ij}(\omega_\lambda)|^2\frac{(\omega_{ij}-\omega_\lambda)}{[z^2+(\omega_{ij}-\omega_\lambda)^2]} ,\\ \Delta\omega_i(z)&=\sum_{j\neq i}\int d\omega_\lambda|g_{ij}(\omega_\lambda)|^2N(\omega_\lambda)\\&\quad\times\left[\frac{(\omega_{ij}-\omega_\lambda)}{[z^2+(\omega_{ij}-\omega_\lambda)^2]}+\frac{(\omega_{ij}+\omega_\lambda)}{[z^2+(\omega_{ij}+\omega_\lambda)^2]}\right] ,\end{aligned} \tag{18}$$

where z is the complex Laplace transform parameter. To obtain Eq. (17) we have used the commutation relations (5a) and made the rotating-wave approximation,[11] i.e., we neglected rapidly oscillating terms (the so-called counter-rotating terms). In Eq. (17) we have also introduced a shorter notation for the atomic operators, i.e.,

$$S_1^+=S_{21}=|2\rangle\langle 1|,\quad S_1^-=(S_1^+)^\dagger\ ,$$
$$S_2^+=S_{32}=|3\rangle\langle 2|,\quad S_2^-=(S_2^+)^\dagger\ . \tag{19}$$

Now we employ the Markov approximation. This neglects retardation effects and is valid in the long-time limit $t \gg \omega_\lambda^{-1}, \omega_i^{-1}$, providing this is short compared with the typical relaxation times of the system. With this approximation we can replace the $\eta^{(+)}(z)$, $\gamma^{(\pm)}(z)$, and $\Delta\omega(z)$ parameters by their limiting values as $z \to 0^+$. After this, the inverse Laplace transform of Eq. (17) leads to the master equation

$$\begin{aligned}\frac{\partial\rho}{\partial t} = &-\sum_{i,j}[M(\omega_i)\eta_{ij} - i\Delta\omega_{ij}]([S_i^+\rho, S_j^+] + [S_j^+, \rho S_i^+])e^{i(\omega_i+\omega_j-2\omega)t}\\ &-\sum_{i,j}[M^*(\omega_i)\eta_{ij}^* + i\Delta\omega_{ij}^*]([S_i^-\rho, S_j^-] + [S_j^-, \rho S_i^-])e^{-i(\omega_i+\omega_j-2\omega)t}\\ &-\sum_{i,j}N(\omega_i)\gamma_{ij}(\rho S_j^- S_i^+ + S_j^- S_i^+\rho - 2S_i^+\rho S_j)e^{i(\omega_i-\omega_j)t}\\ &-\sum_{i,j}[N(\omega_i)+1]\gamma_{ij}(\rho S_i^+ S_j^- + S_i^+ S_j^-\rho - 2S_j^-\rho S_i^+)e^{i(\omega_i-\omega_j)t} - i\sum_i(\Delta\omega_i^0 + \Delta\omega_i)[|i\rangle\langle i|, \rho]\ ,\end{aligned} \tag{20}$$

where the coefficients in the equation are

$$\begin{aligned}\eta_{ij} &= \pi g_i(\omega_i) g_j(2\omega - \omega_i)\ ,\\ \gamma_{ij} &= \pi g_i(\omega_i) g_j^*(\omega_i)\ ,\\ \Delta\omega_{ij} &= \int d\omega_\lambda \frac{g_i(\omega_\lambda) g_j(2\omega-\omega_\lambda)}{(\omega_\lambda - \omega_i)} M(\omega_\lambda)\ ,\\ \Delta\omega_i^0 &= \sum_{j\neq i}\int d\omega_\lambda \frac{|g_{ij}(\omega_\lambda)|^2}{(\omega_{ij}-\omega_\lambda)}\ ,\\ \Delta\omega_i &= \sum_{j\neq i}\int d\omega_\lambda |g_{ij}(\omega_\lambda)|^2 N(\omega_\lambda)\left[\frac{1}{(\omega_{ij}-\omega_\lambda)} + \frac{1}{(\omega_{ij}+\omega_\lambda)}\right]\ .\end{aligned} \tag{21}$$

The general master equation (20) includes both terms that are time independent and terms that have an explicit time dependence of the form $e^{i\tilde{\omega}t}$. Here $\tilde{\omega}$ is a linear combination of atomic frequencies and of the input reference frequency ω. Terms of this type are generalized level-shift and decay constants, and are known to occur in multilevel atomic master equations[7,9,12] in the ordinary vacuum case. When $\tilde{\omega}$ is small, as can occur with degenerate transition frequencies, such terms must be included in the equations of motion. In cases when $\tilde{\omega}$ is large, these terms are rapidly oscillating, and are usually neglected, in a consistent way with the rotating-wave approximation that is already made here. The new feature of the present equation is in the terms that oscillate as $2\omega - \omega_i - \omega_j$, which must clearly be included under conditions of two-photon resonance, whenever $2\omega \simeq \omega_i + \omega_j$.

The coefficients γ_{ii} ($i=1,2$), which appear in Eq. (20), are equal to half the radiative spontaneous-emission rates, respectively, for $|2\rangle \to |1\rangle$ and $|3\rangle \to |2\rangle$ transitions, whereas γ_{ij} for $i \neq j$ are generalized decay constants that arise from the coupling between these two transitions. In general, the γ_{12} and γ_{21} are nonzero, but the time dependence of the terms in which they appear includes an oscillation at the frequency $\omega_i - \omega_j$. For a large difference between the atomic transition frequencies such terms are rapidly oscillating and can be neglected.[7] This cannot be done for equally spaced atomic levels when $\omega_1 = \omega_2$.

The squeezed vacuum introduces new damping constants η_{ii} ($i=1,2$) that have the same dependence on the coupling coefficients $g_i(\omega_i)$ as the decay rates γ_{ii}, the only major difference being the presence of the squeezing parameter M. These include, as a special case, the modification to the polarization decay found by Gardiner[4] in the two-level atom. Moreover, Eq. (20) also contains generalized decay constants η_{12} and η_{21}, which arise from the coupling between these two transitions induced by the squeezed vacuum. In the following paper we shall show that these terms play an important role in the spontaneous emission from a three-level atom interacting with a squeezed-vacuum field.

The parameters $\Delta\omega_i$ and $\Delta\omega_i^0$ are generalized energy-level shift terms. The terms $\Delta\omega_i$ and $\Delta\omega_i^0$ are familiar in atomic spectroscopy, being the intensity-dependent Stark shift and Lamb shift, respectively. Here the Stark shift only depends on the field intensity $N(\omega)$, and is independent of the squeezing parameter M. This is in agreement with previous treatments of level shifts. The term $\Delta\omega_i^0$ represents the part of the Lamb shift induced by the first-order coupling in the Hamiltonian of Eq. (1). It is well known that to obtain a complete calculation of the Lamb shift, it is necessary to include a second-order, multilevel Hamiltonian including electron mass renormalization.[13] If these are included the standard nonrelativistic vacuum-Lamb-shift result is obtained, although still in one dimension.

The remaining term $\Delta\omega_{ij}$ is due to the interaction with a squeezed vacuum. This is proportional to the cross correlation M, and has the effect of coupling different levels together. This term was neglected in some earlier treatments of level shifts in a two-level atom interacting with a squeezed vacuum.[13] In Appendix B we show that the term $\Delta\omega_{ij}$ is different from zero only when the ith transition is outside of resonance with the squeezing carrier frequency ω. This is true independently of the exact spectral distribution of $M(\omega)$, and is due primarily to the symmetry properties of M. We note here that some results in Appendix B are obtained using a flat distribution for $M(\omega)$ for purposes of illustration. In practice it is more likely that $M(\omega)$ would have a Lorenzian spectrum at the resonant frequencies of the cavity producing the squeezed radiation. If propagative methods are used, the squeezing would be restricted in frequency width by the phase-matching and dispersion properties of the nonlinear medium used to generate the correlated photons. This can still be compatible with our broadband assumptions, but clearly modifies the shifts $\Delta\omega_i$ and $\Delta\omega_{ij}$, which are very sensitive to the broadband characteristics of the input field.

Despite this, it is always true that $\Delta\omega_{ij}$ vanishes when the ith transition is resonant with the squeezing carrier frequency ω. This provides a justification for the neglect of these terms in the case of a two-level atom excited at resonance, since our model can certainly be used for two-level transitions. Under conditions of two-photon resonance, $\omega_i+\omega_j=2\omega$, we also show in Appendix B that $\Delta\omega_{ij}=-\Delta\omega_{ji}$. This is consistent with $\Delta\omega_{ij}$ vanishing when $\omega=\omega_i$, since two-photon resonance then requires that $\omega=\omega_j$. It should be clear from this that $\Delta\omega_{ij}$ has symmetry properties that are different to those of η_{ij}, but it has a similar effect as the η_{ij} coupling term. In general, this term does not vanish or rapidly oscillate in the interaction picture and needs to be included in the equation of motion not merely as a phase but as a dynamical coupling.

In summary, master equation (20) will be the fundamental equation of motion in the theory of interaction between a three-level atom and a one-dimensional broadband squeezed vacuum field. It can, for example, be used to discuss how the atom decays, or to solve problems concerning population fluctuations, development of atomic correlations, etc. In the following paper this equation will be used to study the transient and steady-state populations of the atomic levels. An essential feature of this master equation is the assumption that M,N are slowly varying over the two relevant frequency bands near the transition frequencies ω_1 and ω_2. In experimental practice, this might imply that the squeezing is produced in cavity modes that are broadband relative to the atomic linewidth. Other methods of producing squeezing include pulse propagation in nonlinear media. In either case, it is possible for the radiation at frequencies between ω_1,ω_2 to be in the ordinary vacuum state, as long as the squeezing bandwidth near the two resonant frequencies is large compared to the decay rates.

IV. MASTER EQUATION IN A THREE-DIMENSIONAL SQUEEZED VACUUM

All of the above analyses in Sec. III have assumed an ideal coupling between the three-level atom and a one-dimensional squeezed vacuum field. Thus the atom interacts only with squeezed modes of the radiation field. From the experimental point of view this one-dimensional model could be realized in an appropriate waveguide, at whose termination one would locate the three-level atom. However, this would be difficult at visible wavelengths, though it might be possible at the wavelengths corresponding to transitions in Rydberg atoms. The situation looks more complicated in free space, where the atom interacts with a three-dimensional electromagnetic field. In this case an ideal coupling between the atom and the squeezed-vacuum field is difficult to achieve in experimental generation of a three-dimensional squeezed-vacuum field.

To avoid these difficulties Parkins and Gardiner[5] have proposed a "nonideal" coupling between the two-level atom and the squeezed vacuum field. The nonideal coupling means that the atom is coupled to the squeezed as well as to the unsqueezed modes of the electromagnetic field. In the Parkins and Gardiner model only those modes whose propagation vectors lie within a solid angle over which the input field is propagated are assumed to be squeezed. All other modes are unsqueezed. This model seems to be more acceptable for experimental realization since, for example, the output of a nondegenerate parametric amplifier could be passed through a system of lenses and phase plates in order to produce a focused beam of squeezed light at the point where the atom is located.

Following Parkins and Gardiners suggestion we shall examine how the interaction between a three-level atom and a three-dimensional squeezed vacuum field can affect the master equation for the reduced atomic operator ρ. First of all, we derive the general master equation for the reduced density operator ρ of the three-level atom interacting with a three-dimensional electromagnetic field. We assume that the three-dimensional field is squeezed and allow spontaneous emission into the unsqueezed modes. We start with the assumption that the radiation field is a generalized Gaussian state. This includes all possible types of squeezed radiation field. We then specialize to a model like that of Parkins and Gardiner, in which only a part of the incoming radiation field is squeezed.

The Hamiltonian (1) for a three-level atom interacting with an electromagnetic field in three dimensions has the following form:[7]

$$H=H_A+\hbar c\sum_s\int|\mathbf{k}|a^\dagger(\mathbf{k},s)a(\mathbf{k},s)d^3\mathbf{k} +i\hbar\sum_s\sum_{\substack{i,j=1\\ i\neq j}}^{3}\int[\boldsymbol{\mu}_{ij}\cdot\mathbf{g}_{\mathbf{k}s}(\mathbf{r})a(\mathbf{k},s)S_{ij}+\text{H.c.}]d^3\mathbf{k}\ , \tag{22}$$

where H_A is the Hamiltonian for the atom given by Eq. (2), and $S_{ij}=|i\rangle\langle j|$ are the atomic operators satisfying relations (5). In Eq. (22), $\boldsymbol{\mu}_{ij}=\langle i|\boldsymbol{\mu}|j\rangle$ is the electric-dipole moment associated with the transition between the states $|i\rangle$ and $|j\rangle$, and $\mathbf{g}_{\mathbf{k}s}(\mathbf{r})$ is the appropriate mode function, evaluated at the position $\mathbf{r}$ of the atom. For a three-dimensional multimode field in free space, $\mathbf{g}_{\mathbf{k}s}(\mathbf{r})$ is defined as [in *Système International* (SI) units]

$$\mathbf{g}_{\mathbf{k}s}(\mathbf{r})=\left[\frac{c|\mathbf{k}|}{2\epsilon_0\hbar(2\pi)^3}\right]^{1/2}\hat{\mathbf{e}}_{\mathbf{k}s}e^{i\mathbf{k}\cdot\mathbf{r}}\ , \tag{23}$$

where $\hat{\mathbf{e}}_{\mathbf{k}s}$ is the unit polarization vector. We assume, as in Secs. II and III, that we have a three-level atom in ladder configuration ($E_3>E_2>E_1$) located at the point $\mathbf{r}=0$ with a dipole-forbidden transition from level $|1\rangle$ to level $|3\rangle$, i.e., $|\boldsymbol{\mu}_{13}|=0$.

The operators $a(\mathbf{k},s)$ and $a^\dagger(\mathbf{k},s)$, which appear in Eq. (22), describe a three-dimensional electromagnetic field, which can be in a squeezed vacuum state. In the general case such a three-dimensional field can be defined as having commutation relations and correlations given by

$$\begin{aligned}&[a(\mathbf{k},s),a^\dagger(\mathbf{k}',s')]=\delta^3(\mathbf{k}-\mathbf{k}')\delta_{ss'}\ ,\\&\langle a^\dagger(\mathbf{k},s)a(\mathbf{k}',s')\rangle=n(\mathbf{k}s,\mathbf{k}'s')\ ,\\&\langle a(\mathbf{k},s)a(\mathbf{k}',s')\rangle=m(\mathbf{k}s,\mathbf{k}'s')\ .\end{aligned}\tag{24}$$

Here $n(\mathbf{k}s,\mathbf{k}'s')$ and $m(\mathbf{k}s,\mathbf{k}'s')$ are photon number and squeezing densities, respectively, in momentum space, whose explicit form depends on the preparation of the squeezed vacuum. These correlations completely define a generalized Gaussian state, which corresponds to a wide choice of possible inputs.[14,15] In particular, it includes as inputs all the possible parametric amplifier configurations below threshold. It also can be used to describe a pulsed squeezing experiment.

With the Hamiltonian (22) and on using the correlations (24), the Laplace transform over time t of the master equation (15) takes a form identical to the general one-dimensional case, Eq. (17). The parameters $\eta^{(+)}$, $\gamma^{(\pm)}$, and $\Delta\omega$, are now given by

$$\begin{aligned}
\eta_{ij}^{(+)}(z)&=\frac{1}{c}\sum_{s,s'}\int\int[\boldsymbol{\mu}_i\cdot\mathbf{g}_{\mathbf{k}s}(0)][\boldsymbol{\mu}_j\cdot\mathbf{g}_{\mathbf{k}'s'}(0)]\frac{m(\mathbf{k}s,\mathbf{k}'s')(z/c)}{[(z/c)^2+(k_i-k)^2]}d^3\mathbf{k}\,d^3\mathbf{k}'\ ,\\
\gamma_{ij}^{(+)}(z)&=\frac{1}{c}\sum_{s,s'}\int\int[\boldsymbol{\mu}_i^*\cdot\mathbf{g}_{\mathbf{k}s}^*(0)][\boldsymbol{\mu}_j\cdot\mathbf{g}_{\mathbf{k}'s'}(0)]\frac{n(\mathbf{k}s,\mathbf{k}'s')(z/c)}{[(z/c)^2+(k_i-k)^2]}d^3\mathbf{k}d^3\mathbf{k}'\ ,\\
\gamma_{ij}^{(-)}(z)&=\frac{1}{c}\sum_{s}\int[\boldsymbol{\mu}_i^*\mathbf{g}_{\mathbf{k}s}^*(0)][\boldsymbol{\mu}_j\cdot\mathbf{g}_{\mathbf{k}s}(0)]\frac{(z/c)}{[(z/c)^2+(k_i-k)^2]}d^3\mathbf{k}\ ,\\
\Delta\omega_{ij}(z)&=\frac{1}{c}\sum_{s,s'}\int\int[\boldsymbol{\mu}_i\cdot\mathbf{g}_{\mathbf{k}s}(0)][\boldsymbol{\mu}_j\cdot\mathbf{g}_{\mathbf{k}'s'}(0)]\frac{m(\mathbf{k}s,\mathbf{k}'s')(k-k_i)}{[(z/c)^2+(k-k_i)^2]}d^3\mathbf{k}d^3\mathbf{k}'\ .\\
\Delta\omega_i^0(z)&=\frac{1}{c}\sum_{j\neq i}\sum_{s}\int|\boldsymbol{\mu}_{ij}\cdot\mathbf{g}_{\mathbf{k}s}(0)|^2\frac{(k_{ij}-k)}{[(z/c)^2+(k_{ij}-k)^2]}d^3\mathbf{k}\ ,\\
\Delta\omega_i(z)&=\frac{1}{c}\sum_{j\neq i}\sum_{s,s'}\int\int n(\mathbf{k}s,\mathbf{k}'s')[\boldsymbol{\mu}_{ij}^*\cdot\mathbf{g}_{\mathbf{k}s}^*(0)][\boldsymbol{\mu}_{ij}\cdot\mathbf{g}_{\mathbf{k}'s'}(0)]\\
&\quad\times\left[\frac{(k_{ij}-k)}{[(z/c)^2+(k_{ij}-k)^2]}+\frac{(k_{ij}+k)}{[(z/c)^2+(k_{ij}+k)^2]}\right]d^3\mathbf{k}d^3\mathbf{k}'\ ,
\end{aligned}\tag{25}$$

where $k_i=\omega_i/c$ and $k_{ij}=\omega_{ij}/c$. To simplify the notation, in Eq. (25) we have defined $\boldsymbol{\mu}_i\equiv\boldsymbol{\mu}_{i+1,i}$. In the above form, we have an exact description of the interaction of the atom with an arbitrary squeezed field, apart from using the rotating-wave approximation.

As an example of the interaction between a three-level atom and the three-dimensional squeezed-vacuum field we now consider the Parkins and Gardiner model.[5] In this model only those modes are squeezed whose propagation vectors $\mathbf{k}$ have an angle θ_k with the z axis less than a maximum value θ over which squeezing is propagated. All other modes are not squeezed. The squeezing is defined to correspond to the focused output field of a nondegenerate cavity,[16] resonant near ω_1 and ω_2, but with each resonance relatively broadband compared to the atomic relaxation rates. This can be achieved by defining the squeezing parameters $n(\mathbf{k}s,\mathbf{k}'s')$ and $m(\mathbf{k}s,\mathbf{k}'s')$ as[5]

$$\begin{aligned}n(\mathbf{k}s,\mathbf{k}'s')&=N'(k)U_s^*(\mathbf{k})U_{s'}(\mathbf{k}')\delta(k-k')/k^2\ ,\\m(\mathbf{k}s,\mathbf{k}'s')&=M'(k)U_s(\mathbf{k})U_{s'}(\mathbf{k}')\delta(2k_0-k-k')/kk'\ ,\end{aligned}\tag{26}$$

where $k_0=\omega/c$ and $M'(k)=M'(2k_0-k)$.

Here ω is the carrier frequency of the squeezing and $U_s(\mathbf{k})$ is defined as a square normalized mode function that includes only directions $\mathbf{k}$ confined to a solid angle $\Omega_k=(\theta_k,\phi_k)$ with equal amplitude. This mode function depends on the angle of propagation, and can have a different structure depending on the method of propagation and focusing of the squeezed light. In particular, it may be chosen to optimize the coupling. We now suppose that the two transition dipole moments are parallel. This is not an essential feature if the transitions are nondegenerate, since the two frequencies can be independently matched to two differently polarized transitions. However, in practice it is more likely that both transitions would have identical Δm values to obtain a true three-level submanifold. In this case, $\boldsymbol{\mu}_1$ and $\boldsymbol{\mu}_2$ are parallel. The procedure of mode matching to the atomic transition is then simplified, because $U_s(\mathbf{k})$ can be chosen to match both transitions, with the choice

$$U_s(\mathbf{k})=\begin{cases}\mathcal{N}_i(k)^{-1/2}\boldsymbol{\mu}_i^*\cdot\mathbf{g}_{\mathbf{k}s}^*(\mathbf{r})&\text{for } \theta_k\le\theta\\0\ \text{ for } \theta_k>\theta\ .\end{cases}\tag{27}$$

This corresponds to the coupling constant (23) coupling the electromagnetic field with the atomic transition dipole moment $\boldsymbol{\mu}_i$. In Eq. (27) the normalization constant

$\mathcal{N}_i$ is defined so that U is dimensionless, with

$$\mathcal{N}_i(k)=\int_\Omega d\Omega_k \sum_s |\boldsymbol{\mu}_i\cdot\mathbf{g}_{\mathbf{k}s}(\mathbf{r})|^2 , \tag{28}$$

where Ω is the total solid angle over which the squeezing is incident. This model describes possibly the simplest experiment, in which the input radiation is directly incident on the atom. More complex arrangements would modify Eq. (26). For simplicity, we have not included possible modifications due to a resonator structure. This can be readily treated within the general formalism of Eq. (25), as it simply changes the incident correlation functions.

With the squeezing parameters in Eq. (26), the master-equation coefficients of Eq. (25) take the form

$$\begin{aligned}
\eta_{ij}^{(+)}(z)&=\frac{1}{c}\int dk\, k(2k_0-k)M'(k)\frac{(z/c)}{[(z/c)^2+(k_i-k)^2]}\\
&\quad\times\left[\int_\Omega d\Omega_k\sum_s U_s(\mathbf{k})[\boldsymbol{\mu}_i\cdot\mathbf{g}_{\mathbf{k}s}(0)]\right]\left[\int_\Omega d\Omega_{k'}\sum_{s'} U_{s'}(\mathbf{k}')[\boldsymbol{\mu}_j\cdot\mathbf{g}_{\mathbf{k}'s'}(0)]\right],\\
\gamma_{ij}^{(+)}(z)&=\frac{1}{c}\int dk\, k^2N'(k)\frac{(z/c)}{[(z/c)^2+(k_i-k)^2]}\left[\int_\Omega d\Omega_k\sum_s U_s^*(\mathbf{k})[\boldsymbol{\mu}_i^*\cdot\mathbf{g}_{\mathbf{k}s}^*(0)]\right]\left[\int_\Omega d\Omega_{k'}\sum_{s'} U_{s'}(\mathbf{k}')[\boldsymbol{\mu}_j\cdot\mathbf{g}_{\mathbf{k}'s'}(0)]\right],\\
\gamma_{ij}^{(-)}(z)&=\frac{1}{c}\int dk\, k^2\frac{(z/c)}{[(z/c)^2+(k_i-k)^2]}\int_{\text{sphere}} d\Omega_k\sum_s[\boldsymbol{\mu}_i^*\cdot\mathbf{g}_{\mathbf{k}s}^*(0)][\boldsymbol{\mu}_j\cdot\mathbf{g}_{\mathbf{k}s}(0)],\\
\Delta\omega_{ij}(z)&=\frac{1}{c}\int dk[k(2k_0-k)]M'(k)\frac{(k-k_i)}{[(z/c)^2+(k-k_i)^2]}\\
&\quad\times\left[\int_\Omega d\Omega_k\sum_s U_s(\mathbf{k})[\boldsymbol{\mu}_i\cdot\mathbf{g}_{\mathbf{k}s}(0)]\right]\left[\int_\Omega d\Omega_{k'}\sum_{s'} U_{s'}(\mathbf{k}')[\boldsymbol{\mu}_j\cdot\mathbf{g}_{\mathbf{k}'s'}(0)]\right],\\
\Delta\omega_i^0(z)&=\frac{1}{c}\sum_{j\neq i}\int dk\, k^2\frac{(k_{ij}-k)}{[(z/c)^2+(k_{ij}-k)^2]}\int_{\text{sphere}} d\Omega_k\sum_s|\boldsymbol{\mu}_{ij}\cdot\mathbf{g}_{\mathbf{k}s}(0)|^2,\\
\Delta\omega_i(z)&=\frac{1}{c}\sum_{j\neq i}\int dk\, k^2N'(k)\left[\frac{(k_{ij}-k)}{[(z/c)^2+(k_{ij}-k)^2]}+\frac{(k_{ij}+k)}{[(z/c)^2+(k_{ij}+k)^2]}\right]\\
&\quad\times\left[\int_\Omega d\Omega_k\sum_s U_s^*(\mathbf{k})[\boldsymbol{\mu}_{ij}^*\cdot\mathbf{g}_{\mathbf{k}s}^*(0)]\right]\left[\int_\Omega d\Omega_{k'}\sum_{s'} U_{s'}(\mathbf{k}')[\boldsymbol{\mu}_{ij}\cdot\mathbf{g}_{k's'}(0)]\right].
\end{aligned}\tag{29}$$

In deriving Eq. (29), we have assumed that the atom is located at the point $\mathbf{r}=0$ in which the incident squeezed light is focused.

Employing the Markov approximation in which we ignore retardation effects, and taking the long-time limit, i.e., $t\gg 1/\omega_1, 1/\omega_2$, we can replace the coefficients (29) by their limiting values as $z\to 0+$. This is still quite general, in terms of the squeezed mode function $U_s(\mathbf{k})$, although it requires that the correlation functions be now relatively broadband. Next, substituting Eq. (27) for $U_s(\mathbf{k})$ and Eq. (23) for $\mathbf{g}_{\mathbf{k}s}$, we derive

$$\begin{aligned}
\eta_{ij}^{(+)}&=\frac{[k_i(2k_0-k_i)]^{3/2}}{16\pi^2\epsilon_0\hbar}\mu_i\mu_jM'(k_i)\left[\int_\Omega d\Omega_k\sum_s|\hat{\boldsymbol{\mu}}_i\cdot\hat{\mathbf{e}}_{\mathbf{k}s}|^2\right]^{1/2}\left[\int_\Omega d\Omega_{k'}\sum_{s'}|\hat{\boldsymbol{\mu}}_j\cdot\hat{\mathbf{e}}_{\mathbf{k}'s'}|^2\right]^{1/2},\\
\gamma_{ij}^{(+)}&=\frac{k_i^3}{16\pi^2\epsilon_0\hbar}\mu_i\mu_jN'(k_i)\left[\int_\Omega d\Omega_k\sum_s|\hat{\boldsymbol{\mu}}_i\cdot\hat{\mathbf{e}}_{\mathbf{k}s}|^2\right]^{1/2}\left[\int_\Omega d\Omega_{k'}\sum_{s'}|\hat{\boldsymbol{\mu}}_j\cdot\hat{\mathbf{e}}_{\mathbf{k}'s'}|^2\right]^{1/2},\\
\gamma_{ij}^{(-)}&=\frac{k_i^3}{16\pi^2\epsilon_0\hbar}\mu_i\mu_j\int_{\text{sphere}} d\Omega_k\sum_s(\hat{\boldsymbol{\mu}}_i^*\cdot\hat{\mathbf{e}}_{\mathbf{k}s}^*)(\hat{\boldsymbol{\mu}}_j\cdot\hat{\mathbf{e}}_{\mathbf{k}s}),\\
\Delta\omega_{ij}&=\frac{\mu_i\mu_j}{16\pi^3\epsilon_0\hbar}\int\frac{dk\,[k(2k_0-k)]^{3/2}}{(k-k_i)}M'(k)\left[\int_\Omega d\Omega_k\sum_s|\hat{\boldsymbol{\mu}}_i\cdot\hat{\mathbf{e}}_{\mathbf{k}s}|^2\right]^{1/2}\left[\int_\Omega d\Omega_{k'}\sum_{s'}|\hat{\boldsymbol{\mu}}_j\cdot\hat{\mathbf{e}}_{\mathbf{k}'s'}|^2\right]^{1/2},\\
\Delta\omega_i^0&=\sum_{j\neq i}\frac{\mu_{ij}^2}{16\pi^3\epsilon_0\hbar}\int dk\frac{k^3}{(k_{ij}-k)}\left[\int_{\text{sphere}} d\Omega_k\sum_s|\hat{\boldsymbol{\mu}}_{ij}\cdot\hat{\mathbf{e}}_{\mathbf{k}s}|^2\right],\\
\Delta\omega_i&=\sum_{j\neq i}\frac{\mu_{ij}^2}{16\pi^3\epsilon_0\hbar}\int dk\, k^3N'(k)\left[\frac{1}{(k_{ij}-k)}+\frac{1}{(k_{ij}+k)}\right]\left[\int_\Omega d\Omega_k\sum_s|\hat{\boldsymbol{\mu}}_{ij}\cdot\hat{\mathbf{e}}_{\mathbf{k}s}|^2\right],
\end{aligned}\tag{30}$$

where $\hat{\mu}_i$ is the unit vector along the transition electric-dipole moment $\boldsymbol{\mu}_i$, and $\mu_i = |\boldsymbol{\mu}_i|$.

To carry out the polarization sums in Eq. (30) we can go over to a spherical representation in which the unit orthogonal polarization vectors $\hat{e}_{k1}$ and $\hat{e}_{k2}$ may be taken as[12]

$$\hat{e}_{k1} = (-\cos\theta_k \cos\varphi_k, -\cos\theta_k \sin\varphi_k, \sin\theta_k) , \quad \hat{e}_{k2} = (\sin\varphi_k, -\cos\varphi_k, 0) . \tag{31}$$

First we consider $\Delta m = 0$ transitions, in which the atomic dipole moments can be written

$$\boldsymbol{\mu}_i = \mu_i(0,0,1) . \tag{32}$$

With this choice of polarization vectors, the sums over s and integrals over $d\Omega_k$ in (30) lead to

$$\begin{aligned} \eta_{ij}^{(+)} &= \left[\frac{2k_0 - k_i}{k_j}\right]^{3/2} (\gamma_i\gamma_j)^{1/2} M'(k_i) v(\theta) , \\ \gamma_{ij}^{(+)} &= \left[\frac{k_i}{k_j}\right]^{3/2} (\gamma_i\gamma_j)^{1/2} N'(k_i) v(\theta) , \\ \gamma_{ij}^{(-)} &= \left[\frac{k_i}{k_j}\right]^{3/2} (\gamma_i\gamma_j)^{1/2} , \\ \Delta\omega_{ij} &= \frac{\mu_i\mu_j}{6\pi^2\epsilon_0\hbar} v(\theta) \int \frac{dk\,[k(2k_0-k)]^{3/2}}{(k-k_i)} M'(k) , \\ \Delta\omega_i^0 &= \sum_{j\neq i} \frac{\mu_{ji}^2}{6\pi^2\epsilon_0\hbar} \int \frac{dk\,k^3}{(k_{ij}-k)} , \\ \Delta\omega_i &= \sum_{j\neq i} \frac{\mu_{ji}^2}{6\pi^2\epsilon_0\hbar} v(\theta) \\ &\quad \times \int dk\,k^3 N'(k) \left[\frac{1}{(k_{ij}-k)} + \frac{1}{(k_{ij}+k)}\right] , \end{aligned} \tag{33}$$

where $\gamma_i = k_i^3\mu_i^2/6\pi\epsilon_0\hbar$. The function $v(\theta)$ depends on the angle θ over which squeezing is propagated, and is given by

$$v(\theta) = \tfrac{1}{2}[1 - \tfrac{1}{4}(3+\cos^2\theta)\cos\theta], \quad \theta \in (0,\pi) . \tag{34}$$

To carry out these polarization sums we have used a linear representation for the polarization vectors $\hat{e}_{ks}$. It is not difficult to show that with a circular representation for the polarization vectors, i.e.,

$$\hat{e}_{k+} = \frac{1}{\sqrt{2}}(\hat{e}_{k1} + i\hat{e}_{k2}) , \quad \hat{e}_{k-} = \frac{1}{\sqrt{2}}(\hat{e}_{k1} - i\hat{e}_{k2}) , \tag{35}$$

and with parallel dipole moments $\hat{\mu}_i = \mp 2^{-1/2}(1, \pm i, 0)$ for $\Delta m = \pm 1$ transitions,[11] Eq. (30) leads to the same decay constants and frequency shifts as given in Eq. (33). In this situation, a true three-level submanifold can exist, as shown in Fig. 2.

With the parameters (33) the inverse Laplace transform of Eq. (17) leads to an equation of exactly the form found in Eq. (20), the one-dimensional case. However, the squeezing parameters, decay rates, and frequency shifts must now be replaced by their effective values in the three-dimensional squeezed vacuum. In the present case, the effective squeezing parameters M,N are now modified to new values that depend on the angle over which the squeezed field is propagated:

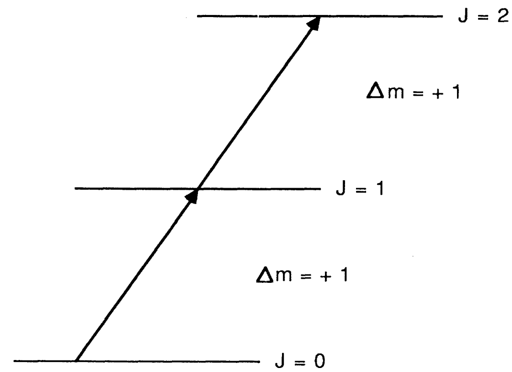

FIG. 2. A three-level atomic system with $\Delta m = 1$ transitions, in which only the indicated levels can become excited.

$$M(\omega_i) = M'(k_i)v(\theta), \quad N(\omega_i) = N'(k_i)v(\theta) , \tag{36}$$

while the coefficients in the equation are

$$\begin{aligned} \eta_{ij} &= \left[\frac{2\omega - \omega_i}{\omega_j}\right]^{3/2} (\gamma_i\gamma_j)^{1/2} , \\ \gamma_{ij} &= \left[\frac{\omega_i}{\omega_j}\right]^{3/2} (\gamma_i\gamma_j)^{1/2} , \\ \Delta\omega_{ij} &= \left[\frac{\gamma_i\gamma_j}{(\omega_i\omega_j)^3}\right]^{1/2} \int \frac{d\omega_k[\omega_k(2\omega-\omega_k)]^{3/2}}{(\omega_k-\omega_i)} M(\omega_k) , \\ \Delta\omega_i^0 &= \frac{1}{6\pi^2c^3\epsilon_0\hbar} \sum_{j\neq i} \mu_{ji}^2 \int \frac{d\omega_k\omega_k^3}{(\omega_{ij}-\omega_k)} , \\ \Delta\omega_i &= \frac{1}{6\pi^2c^3\epsilon_0\hbar} \sum_{j\neq i} \mu_{ji}^2 \int d\omega_k\omega_k^3 N(\omega_k) \left[\frac{1}{(\omega_{ij}-\omega_k)} + \frac{1}{(\omega_{ij}+\omega_k)}\right] , \end{aligned} \tag{37}$$

where $\omega_k = ck$.

The three-dimensional master equation has form identical to the master equation (20) derived for the one-dimensional field. However, the parameters M,N now depend on the angle θ over which the squeezing is propagated. For small θ ($\theta \ll 1$), $v(\theta) \approx 0$, and the master equation (36) reduces to the well-known equation[7,17] describing the interaction of the three-level atom with the ordinary (unsqueezed) vacuum field. We should point out that what is denoted as $2\gamma_i$ in Eq. (33) is the usual Einstein A coefficient for spontaneous emission from level $i+1$ to level i. On the other hand, for large θ, $v(\theta) \approx 1$, and then Eq. (36) reproduces the master equation (20) derived for the case when the atom interacts only with an idealized one-dimensional multimode squeezed vacuum field.

In order to show that more precisely, we note that for all angles θ, $v(\theta)\le 1$. Using the results from Appendix A, and integrating over all mode directions, it is possible to show that

$$|M'(k)|^2 \le N'(k)N'(2k_0-k)+\min[N'(k),N'(2k_0-k)] \ . \tag{38}$$

In terms of the effective one-dimensional squeezing parameters M,N, this leads to the result

$$|M(\omega_k)|^2 \le N(\omega_k)N(2\omega-\omega_k) + v(\theta)\min[N(\omega_k),N(2\omega-\omega_k)] \ . \tag{39}$$

This is a stronger inequality than the one-dimensional equation (7), and simply means that it is necessary to have $v(\theta)=1$ (i.e., $\theta=\pi$) in order for the effective three-dimensional squeezing correlation function $M(\omega)$ to have as large a value as in the idealized one-dimensional case.

Our results demonstrate that there is no essential distinction between the cases of a one-dimensional and higher-dimensional squeezed vacuum. A higher-dimensional squeezed vacuum, in which only some modes are correlated, behaves exactly as a one-dimensional squeezed vacuum with imperfect squeezing. In either case, the terms proportional to M, the effective cross-correlation coefficient, have a reduced size in the final master equation. In order to increase the effectiveness of the squeezed radiation, it may be beneficial to also include a cavity or arbitrary optical devices into the configuration. Generally speaking, this can increase the effective squeezing parameters (M), relative to the case treated here. However, these only change the vacuum correlation function, and hence can be treated using the general theory of Eq. (25).

We note that, as previously, the vacuum-Lamb-shift results require the addition of a mass-renormalization counterterm, and higher-order terms in the Hamiltonian, in order to obtain the correct results. Even then, an appropriate cutoff at $\omega_{\max}\sim c/r_0$ needs to be added at the atomic dimension of r_0, in order for the present approximation to be valid. In this respect, the present results do not differ from standard treatments of multilevel atoms without squeezing. However, the terms proportional to the squeezing parameter M are a unique feature of the new master equation.

V. DISCUSSION

We have considered here the problem of the interaction between a three-level atom in cascade configuration and a squeezed vacuum field. Starting from the Hamiltonian for the atom coupled to the continuum of quantized electromagnetic modes we have derived, within the Born and Markov approximations, the master equations for the reduced density operator of the atom interacting with a multimode broadband one-dimensional squeezed-vacuum field and a three-dimensional vacuum field. In the three-dimensional case we have treated both the case of an arbitrary squeezed field and one where only those modes are squeezed whose propagation vectors lie inside the solid angle Ω over which squeezing is propagated. Spontaneous emission into ordinary (unsqueezed) vacuum modes is allowed. Our master equations [(20) and (25)] can be applied to many experimental situations. Thus Eq. (20) holds when the atom interacts with a multimode one-dimensional squeezed-vacuum field, while Eq. (25) is generally applicable. Equation (20) with the parameters (36) and (37) holds when the multimode three-dimensional squeezed vacuum field is mode matched, and propagates over some solid angle Ω, as well as when the atom is embedded in a three-dimensional squeezed vacuum. The last situation can be realized when the mode-matched squeezed vacuum field propagates over the angle $\theta=\pi$, which corresponds to a perfect electric dipole wave.[4] Our equations also include the situation in which two incident correlated modes can have different intensities as, for example, in output from the degenerate parametric amplifier. There is no restriction to degenerate frequencies in the atomic transitions.

In the following paper we shall apply the master equation (20) to study the transient and steady-state populations of the atomic levels. We shall compare the atomic population for the atom interacting with the thermal (blackbody) field with the population when the thermal field is replaced by the multimode broadband squeezed vacuum field.

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APPENDIX A

We wish to derive the inequality of Eq. (7). Accordingly, assume that we have two modes a_i and a_j, for which the correlation functions are

$$\langle a_i^\dagger a_j\rangle = N_i\delta_{ij} \ ,$$

and

$$\langle a_i a_j\rangle = M,\quad \langle a_i^\dagger a_j^\dagger\rangle = M^* \quad \text{for } i\neq j \ , \tag{A1}$$

where $N_i=N(\omega_i)$ is the intensity of the mode a_i with the frequency ω_i. This corresponds to a discrete case, where the quantization volume is finite. The relations between N_i and M are related to the squeezing and Cauchy-Schwarz inequalities, which are known from parametric amplifier theory.[16] For two operators X and $X^\dagger$ these limits are

$$\langle X^\dagger X\rangle \ge 0 \ , \tag{A2}$$

$$\langle X_1^\dagger X_1\rangle\langle X_2^\dagger X_2\rangle \ge |\langle X_1^\dagger X_2\rangle|^2 \ . \tag{A3}$$

Let us assume that

$$X=a_i+za_j^\dagger \ ,$$
$$X_1=a_i+za_i^\dagger \ , \tag{A4}$$
$$X_2=a_j+za_j^\dagger \ .$$

From (A2) and (A4) we have that

$$N_i+|z|^2(1+N_j)\geq 2|M|\,|z|\ . \quad (A5)$$

Letting $|z|=N_i$, then

$$2|M|\leq 1+N_i(1+N_j)\ , \quad (A6)$$

or

$$2|M|+N_j\leq (1+N_i)(1+N_j)\ . \quad (A7)$$

Inequality (A6) or (A7) must be true for all j, i.e.,

$$2|M|\leq 1+N_iN_j+\min(N_i,N_j)\ . \quad (A8)$$

On the other hand, from inequalities (A3) and (A4) we derive

$$[|z|^2+N_i(1+|z|^2)][1+N_j(1+|z|^2)]\geq |M(1+|z|^2)|^2\ . \quad (A9)$$

In particular, for $|z|=0$

$$|M|^2\leq N_i(1+N_j)\ , \quad (A10)$$

and for $|z|=1$

$$(1+2N_i)(1+2N_j)\geq 4|M|^2\ . \quad (A11)$$

Since, $\frac{1}{4}(1+2N_i)^2\geq N_i(1+N_i)$ the inequality (A11) is weaker than (A10). The inequality (A10), similar to (A8), must be true for all j, i.e.,

$$1+|M|^2\leq 1+N_iN_j+\min(N_i,N_j)\ . \quad (A12)$$

The squeezing limit (A2) leads to the inequality (A8) between the parameters N_i, N_j, and M, whereas the Cauchy-Schwarz inequality (A3) leads to (A12). But $(1+|M|^2)$ is always greater than $2|M|$. Thus (A12) is a stronger inequality than (A8). Further implications of this relationship are discussed elsewhere.[18] In this present paper, the infinite volume limit is taken to obtain the continuum mode relationship corresponding to (A12), given in Eq. (7).

APPENDIX B

We wish to estimate the greatest order of magnitude of the $\Delta\omega_{ij}$ parameter, which is due to the cross correlations between two modes ω_λ and $2\omega-\omega_\lambda$.

For a one-dimensional squeezed-vacuum field this parameter is defined in Eq. (21) and has the form

$$\Delta\omega_{ij}=\int d\omega_\lambda \frac{g_i(\omega_\lambda)g_j(2\omega-\omega_\lambda)}{(\omega_\lambda-\omega_i)}M(\omega_\lambda)\ . \quad (B1)$$

The exact value of this parameter depends on the explicit spectral form of $M(\omega_\lambda)$. Consistently with the white-noise approximation we assume a flat distribution

$$M(\omega_\lambda)=\begin{cases} M & \text{for } 0<\omega_\lambda\leq 2\omega \\ 0 & \text{for } \omega_\lambda>2\omega\ . \end{cases} \quad (B2)$$

With this approximation and using the relation (valid for electric-dipole coupling) that

$$g_i(\omega_\lambda)=\beta\mu_i(\omega_\lambda)^{1/2} \quad (B3)$$

where β is a constant, we obtain

$$\Delta\omega_{ij}=\beta^2\mu_i\mu_j M\int_0^{2\omega} d\omega_\lambda \frac{[\omega_\lambda(2\omega-\omega_\lambda)]^{1/2}}{(\omega_\lambda-\omega_i)}\ . \quad (B4)$$

A change of integration variable ($u=\omega_\lambda-\omega$) leads to

$$\Delta\omega_{ij}=\beta^2\mu_i\mu_j M\int_{-\omega}^{\omega} du\frac{(\omega^2-u^2)^{1/2}}{(u-\delta_i)}\ , \quad (B5)$$

where

$$\delta_i=(\omega_i-\omega)\ . \quad (B6)$$

Evaluating the integral in (B5) we obtain

$$\Delta\omega_{ij}=\pi\beta^2\mu_i\mu_j M\delta_i\ , \quad (B7)$$

or

$$\Delta\omega_{ij}=\eta_{ij}\frac{(\omega_i-\omega)M}{[\omega_i(2\omega-\omega_i)]^{1/2}}\ , \quad (B8)$$

where η_{ij} is defined in Eq. (21). It is evident from the above equation that the generalized level shift $\Delta\omega_{ij}$ induced by the cross correlation is exactly zero for a transition with $\omega=\omega_i$, i.e., when the transition is on resonance with the carrier frequency ω of the squeezed field.

For a three-dimensional squeezed-vacuum field the term $\Delta\omega_{ij}$ is defined in Eq. (37) and has the form

$$\Delta\omega_{ij}=\left[\frac{\gamma_i\gamma_j}{(\omega_i\omega_j)^3}\right]^{1/2}\int\frac{d\omega_k[\omega_k(2\omega-\omega_k)]^{3/2}}{(\omega_k-\omega_i)}M(\omega_k)\ . \quad (B9)$$

The integral appearing in Eq. (B9) can be evaluated similarly to the one-dimensional case and leads to

$$\Delta\omega_{ij}=\eta_{ij}\left[\frac{\omega_j}{2\omega-\omega_i}\right]^{3/2}\left[\frac{\omega_i-\omega}{(\omega_i\omega_j)^{1/2}}\right]\times\left[1+\frac{\omega^2}{2\omega_i\omega_j}\right]M\ . \quad (B10)$$

In all the above calculations we have assumed a flat distribution for $M(\omega_\lambda)$. Since $M(\omega_\lambda)$ is symmetric, i.e, $M(\omega_\lambda)=M(2\omega-\omega_\lambda)$ the integrals in (B1) and (B9) are equal to zero for the degenerate case independent of the spectral form of $M(\omega_\lambda)$. It is also noteworthy that the results (B7) and (B10) have the property that, on two-photon resonance where $\omega_i+\omega_j=2\omega$,

$$\Delta\omega_{ij}=-\Delta\omega_{ij}\ . \quad (B11)$$

More generally, for an arbitrary correlation function $M(\omega_\lambda)$, the symmetry properties of $M(\omega_\lambda)[=M(2\omega-\omega_\lambda)]$ imply that $\Delta\omega_{ij}$ can always be written in the form

$$\Delta\omega_{ij} = \int_{-\omega}^{\omega} \frac{\alpha(u)du}{u-\delta_i} , \quad \text{(B12)}$$

where $\alpha(u)$ is an even function for either dimensionality of the squeezed vacuum. This means that Eq. (B11) must be true whenever $\omega_i+\omega_j=2\omega$, as $\delta_i=-\delta_j$ on two-photon resonance. In this more general case, where $\alpha(u)$ has a frequency cutoff, $\Delta\omega_{ij}$ can be reduced substantially from the "worst-case" estimates of (B7) and (B10).

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