PHYSICAL REVIEW A VOLUME 53, NUMBER 5 MAY 1996

Dipole-dipole interaction in squeezed vacua

E. V. Goldstein and P. Meystre
Optical Sciences Center, University of Arizona, Tucson, Arizona 85721

(Received 24 October 1995)

We study the near-resonant dipole-dipole interaction between two atoms in a broadband squeezed vacuum. We show that this ''vacuum'' modifies the rate of spontaneous emission of the atoms so that it may depend not merely on their relative position, as is normally the case, but also on their center-of-mass position. The explicit form of this latter dependence is a function of the way squeezing is achieved. We also show that in contrast to the spontaneous decay rate, squeezed vacua do not modify the dipole-dipole potential between the atoms as compared to its usual form. Finally, we develop a Monte Carlo wave-function description of the interaction between atoms with an arbitrary internal structure and a squeezed vacuum and apply this formalism to the case of off-resonant excitation of the atoms. We find that in addition to the usual light shift, the squeezed vacuum results in an additional contribution to the effective potential governing the evolution of the atomic ground states.

PACS number(s): 42.50.Dv, 42.50.Fx, 34.20.−b

I. INTRODUCTION

It is now well established that a number of radiative properties of atoms can be modified by tailoring the electromagnetic environment with which the atoms interact. In particular the rate, or even the character, of spontaneous emission can be modified by changing the density of electromagnetic modes coupled to the atom. A related situation occurs if an atom interacts with a squeezed vacuum, in which case the usual single decay rate $1/T_2$ of the atomic polarization ceases to give a correct description of atomic relaxation, and must be replaced by two decay rates [1].

In this paper we extend these results to the study of two-body interactions between atoms. Since two-body interactions are mediated by the electromagnetic field, it should be expected that a tailored electromagnetic vacuum can have significant consequences there too. A trivial example of how this can occur is the near-resonant dipole-dipole interaction between excited and ground atoms in high-Q cavities. The physical origin of this interaction is the reabsorption by one of the atoms of a photon spontaneously emitted by the other one, so that if spontaneous emission is inhibited, respectively enhanced, the near-resonant dipole-dipole interaction will clearly also be suppressed, respectively enhanced.

More interesting perhaps is the situation of a ''squeezed vacuum.'' While the density of modes of the electromagnetic field is not modified here, correlations are introduced between various modes at the expense of having a nonvanishing mean photon number in the field. In this context we note that the dipole-dipole interaction in a thermal reservoir at nonzero temperature has been previously analyzed [2]. It was found that the dipole-dipole interaction is not changed from its zero-temperature expression, due to the exact balance between the effects of stimulated absorption and emission: the dipole-dipole interaction is truly an effect of the electromagnetic vacuum. We find that this result carries over to the case of a squeezed vacuum. More surprising is that the quantum correlations between field modes, while leading to significant qualitative and quantitative changes in the cooperative emission rate of the atoms, do not change the dipole-dipole potential.

There is a large amount of literature dealing with the interaction between single atoms and squeezed vacua, including the study of driven and nondriven two-level and multilevel atoms, see e.g., [1,3–7]. A few publications have also addressed issues related to two atoms [8–11]. In most cases, however, this work considers extensions of the Dicke model where the atoms are assumed to be separated by much less than an optical wavelength. A notable exception is the work of Ficek [10], who also considered the situation of large interatomic separations. However, his emphasis was not so much on the study of two-body collisions as on the creation of correlated atomic states. We try in this paper to make contact with Ficek's results whenever possible.

Section II defines our model of the interaction between two atoms, a classical light field and the free-space continuum of field modes of the quantized electromagnetic field. Section III is the central section of this paper. It outlines the derivation of the cooperative spontaneous emission rate of the atoms and of the near-resonant dipole-dipole interaction. They both result from the elimination of the continuum of field modes in the Born-Markov approximation. We show explicitly the dependence of the spontaneous emission rate on the relative as well as on the center-of-mass coordinates of the atoms. The physical origin of this double dependence is discussed. We also show that the coupling of the atoms to a squeezed vacuum does not change the form of the dipole-dipole potential as compared to the usual situation. Section IV then introduces a Monte Carlo wave-function formalism for atoms in a squeezed bath. Section V applies these results to the case of off-resonant excitation of the atoms, where the excited electronic states can be adiabatically eliminated. We show that in addition to the usual light shifts, the squeezed vacuum results in an additional contribution to the effective potential governing the evolution of the atomic ground states. Finally Sec. VI is a summary and conclusion. Calculational details are relegated to two appendixes.

1050-2947/96/53(5)/3573(9)/$10.00

II. PHYSICAL MODEL

Our model is an extension of the theory of the dipole-dipole interaction to account for a squeezed vacuum produced either by four-wave mixing [12,13] or by parametric amplification [14,15]. The atoms-field system is described by the Hamiltonian

$$\mathcal{H}=\mathcal{H}_S+\mathcal{H}_F+\mathcal{H}_{A-F}, \tag{1}$$

where $\mathcal{H}_S=\mathcal{H}_A+\mathcal{H}_{A-L}$ describes a system of two atoms and their electric dipole coupling to some laser fields of frequency ω_L which form, e.g., an optical lattice. (The presence of these classical fields is actually not necessary until Sec. V, where they will be used to justify the adiabatic elimination of the upper electronic levels of the atoms.) In a frame rotating at frequency ω_L, the Hamiltonian of the two-atom system takes the form

$$\mathcal{H}_A=\sum_{i=1}^{2}\mathcal{H}_{A,i}=\sum_{i=1}^{2}\left[\frac{\mathbf{p}_i^2}{2M}+\hbar(\omega_0-\omega_L)P_{e,i}\right], \tag{2}$$

where $\mathbf{p}_i$ is the center-of-mass momentum of the ith atom and ω_0 denotes the Bohr frequency between the degenerate ground and degenerate excited magnetic sublevels of the $J_g \rightarrow J_e$ atomic transition under consideration, the ground state energy being taken equal to 0. The operators $P_{e,i}$, and $P_{g,i}$ which will be introduced shortly, are projectors onto the subspaces of the excited, respectively ground electronic atomic states [16],

$$P_e=\sum_{m=-J_e}^{J_e}|e_m\rangle\langle e_m|, \tag{3}$$

and similarly for P_g. The electric dipole interaction between the atoms and the laser fields may be expressed compactly as

$$\mathcal{H}_{A-L}=\sum_{i=1}^{2}\mathcal{H}_{A-L,i}=-\sum_{i=1}^{2}[\mathbf{d}^+\cdot\mathbf{E}_L^+(\mathbf{r}_i)+\mathbf{d}^-\cdot\mathbf{E}_L^-(\mathbf{r}_i)], \tag{4}$$

where $\mathbf{r}_i$ is the center-of-mass position operator of the ith atom, $\mathbf{E}_L(\mathbf{r}_i)=\mathbf{E}_L^+(\mathbf{r}_i)e^{-i\omega_L t}+\text{c.c.}$ is the laser field at the position of that atom, and we have introduced the atomic raising operator $\mathbf{d}^+=P_e\mathbf{d}P_g$, $\mathbf{d}$ being the atomic dipole moment. The last two terms in Eq. (1) describe the dipole coupling of the atoms to the free-space continuum of modes of the quantized electromagnetic field, with

$$\mathcal{H}_F=\sum_s \hbar\omega_s a_s^\dagger a_s \tag{5}$$

and

$$\mathcal{H}_{A-F}=\sum_{i=1}^{2}\sum_s \mathcal{E}_0(k)[\mathbf{d}_i^+\cdot\hat{\epsilon}_\ell(\hat{\mathbf{k}})]a_s e^{i\mathbf{k}\cdot\mathbf{r}_i}+\text{H.c.} \tag{6}$$

As usual, the mode index $s=\{\mathbf{k},\ell\}$ runs over both a continuum of wave vectors and two orthogonal polarizations $\ell=\pm$ for each $\mathbf{k}$. We chose circular polarizations characterized by orthogonal vectors $\hat{\epsilon}_\ell(\hat{\mathbf{k}})$ in the plane perpendicular to the direction of propagation $\hat{\mathbf{k}}=\mathbf{k}/k$ of mode s. The annihilation and creation operators satisfy the boson commutation relation $[a_s,a_{s'}^\dagger]=\delta(\mathbf{k}-\mathbf{k}')\delta_{\ell,\ell'}$, $\mathcal{E}_0(k)=\sqrt{\hbar ck/2\epsilon_0 V}$ is the ''electric field per photon'' of mode s, and V is the quantization volume.

III. DIPOLE-DIPOLE EFFECTS

If the continuum of modes of the electromagnetic field is initially in the vacuum, then its elimination in the Born-Markov approximation leads to several familiar effects. The first, and best known of these, is single-atom spontaneous emission, and a radiative shift that will be ignored in the following. The presence of two atoms instead of one leads in addition to the appearance of a two-body dipole-dipole interaction between excited and ground atoms and to a modulation of the spontaneous emission rate [17–19]. Its physical origin is the reabsorption by one atom of a photon spontaneously emitted by the other one, while the modulation of the spontaneous emission rate is a quantum interference effect which reduces to ''Dicke superradiance'' if both atoms are within one optical wavelength.

The coupling of the atoms to a squeezed vacuum leads to more complex dynamics, even for a single atom. In particular, it is no longer possible to describe the decay of the atomic polarization by a single rate $1/T_2$ [1]. We consider explicitly two concrete ways to achieve a squeezed vacuum, near-degenerate four-wave mixing [12,13] and near-degenerate parametric amplification [14,15]. While both mechanisms lead to similar reservoir properties as far as frequencies are considered, and hence lead to similar single-atom dynamics, the pairs of correlated modes that are generated differ significantly with respect to their coordinate dependence, with important consequences for two-body interactions. Specifically, squeezing leads to the following nonvanishing field correlation functions between modes of the electromagnetic field [7]:

$$\langle a_s^\dagger a_{s'}\rangle=[N(\omega)/\omega\omega']\delta(\omega-\omega')\delta(\hat{\mathbf{k}}-\hat{\mathbf{k}}')\delta_{\ell,\ell'}, \tag{7}$$

$$\langle a_s a_{s'}\rangle=[M(\omega,\omega')/\omega(2\omega_0-\omega)]\delta(2\omega_0-\omega-\omega')\times\delta(\hat{\mathbf{k}}\pm\hat{\mathbf{k}}')\delta_{\ell,\ell'}, \tag{8}$$

with similar expressions for $\langle a_s a_{s'}^\dagger\rangle$ and $\langle a_s^\dagger a_{s'}^\dagger\rangle$. In these expressions, $N(\omega)$ are the mean photon numbers at frequency $\omega=ck$ in the squeezed ''vacuum,'' $M(\omega,\omega')$ is the so-called squeezing parameter, and the frequency of the pump beam(s) is taken to be equal to the atomic transition frequency ω_0. The $+$ sign in the argument of the δ function in Eq. (8) corresponds to four-wave mixing, and the $-$ sign to parametric amplification. The squeezing parameter $M(\omega,\omega')$ is related to the mean photon number per mode $N(\omega)$ by the inequality $|M(\omega,\omega')|^2\leq N(\omega)N(\omega')+\min(N(\omega),N(\omega'))$. In the case of ''perfect squeezing'' $N(\omega)=N(\omega')$ and $|M(\omega,\omega')|^2=N(\omega)[N(\omega)+1]$ [20].

The elimination of the electromagnetic field modes in the Born-Markov approximation leads to an atomic master equation of the usual form

$$\dot{\rho} = -\frac{i}{\hbar}[\mathcal{H}_s,\rho] + \mathcal{L}\rho, \tag{9}$$

where the Liouvillian

$$\begin{aligned}\mathcal{L}\rho = -\frac{1}{\hbar^2}\int_0^t d\tau \mathrm{Tr}_F\{ &\mathcal{H}_{A-F}(t)\mathcal{H}_{A-F}(\tau)\rho_{A-F}(\tau)\\ &-\mathcal{H}_{A-F}(t)\rho_{A-F}(\tau)\mathcal{H}_{A-F}(\tau)\\ &-\mathcal{H}_{A-F}(\tau)\rho_{A-F}(\tau)\mathcal{H}_{A-F}(t)\\ &+\rho_{A-F}(\tau)\mathcal{H}_{A-F}(\tau)\mathcal{H}_{A-F}(t)\}\end{aligned} \tag{10}$$

results from the elimination of the field degrees of freedom. As is now well established [17–19] in the case of two atoms the contribution $\mathcal{L}\rho$ to the master equation (9) cannot be interpreted as a simple damping term: it also includes a Hamiltonian contribution V_{dd}, which is the dipole-dipole potential resulting from the reabsorption by one atom of a photon spontaneously emitted by the other one.

In taking the partial trace Tr_F over the field variables, we assume that the reservoir is in a squeezed vacuum. This leads to the appearance of several new terms as compared to the thermal equilibrium situation, due to the nonvanishing second-order correlation functions involving pairs of annihilation or creation operators, see Eq. (7). Details of the derivation of the master equation are given in Appendix A. An important result is that the dipole-dipole potential V_{dd} is not modified as compared to its value for a usual vacuum, or, for that matter, of a thermal reservoir at nonzero temperature. Specifically, one finds [18,19]

$$V_{dd} = \frac{\hbar\gamma_f}{2}\sum_{q,q'=0,\pm}\alpha_{qq'}(\mathbf{D}_1^+\cdot\hat{\epsilon}_q)(\mathbf{D}_2^-\cdot\hat{\epsilon}_{q'}) + \text{H.c.}, \tag{11}$$

where the coefficient $\alpha_{qq'}$ is given explicitly by

$$\alpha_{qq'} = \frac{3}{2\pi\omega_0^3}\mathrm{P}\int d\omega\frac{\omega^3}{\omega_0-\omega}\int\frac{d\Omega(\hat{\mathbf{k}})}{4\pi}\mathcal{D}_{q,q'}(\hat{\mathbf{k}},\hat{\mathbf{k}})e^{i\omega\hat{\mathbf{k}}\cdot\mathbf{r}/c}, \tag{12}$$

and P stands for ‘‘principal part.’’ In this last expression,

$$\mathcal{D}_{q,q'}(\hat{\mathbf{k}},\hat{\mathbf{k}}') = \sum_{\ell=\pm}[\hat{\epsilon}_\ell(\hat{\mathbf{k}})\cdot\hat{\epsilon}_q]^*[\hat{\epsilon}_\ell(\hat{\mathbf{k}}')\cdot\hat{\epsilon}_{q'}] \tag{13}$$

gives the usual dipole-dipole radiation pattern, $\hat{\epsilon}_q, q=0,\pm$ are unit vectors with respect to the quantization axis $0z$, and $\mathbf{r}=\mathbf{r}_1-\mathbf{r}_2$ is the relative coordinate of the two atoms.

The result that neither a thermal bath [2] nor a squeezed vacuum modifies the dipole-dipole potential V_{dd} may appear surprising at first, as its form is, after all, very ‘‘classical looking.’’ One might therefore have expected that the stimulated effects associated with a nonzero mean photon number in a reservoir at finite temperature, or with nonvanishing correlation functions between pairs of annihilation operators in a squeezed vacuum, would lead to a modification of V_{dd}. That the *stimulated* absorption and emission effects associated with these nonvanishing correlation functions compensate each other exactly clearly demonstrates that despite its classical-looking form, the dipole-dipole interaction is indeed truly a ‘‘vacuum effect,’’ intimately connected to spontaneous emission.

Turning now to the dissipative part of the master equation (9), we note that it includes several types of contributions. The first ones are ‘‘single-atom’’ effects, and account both for usual spontaneous emission as well as for its modifications due to the coupling of the atoms to a squeezed vacuum [1]. In addition, dissipation includes two-atom effects whose origin can be traced back both to ‘‘thermal’’ and to ‘‘squeezed’’ contributions. The ‘‘thermal’’ contributions account for the spatially modulated rate of spontaneous emission resulting from the interference between the two decay channels of the atomic system, including the stimulated processes associated with the nonvanishing mean photon number in a squeezed vacuum. Finally, the two-atom Liouvillian includes a contribution proportional to the squeezing parameter $M(\omega_0,\omega_0)$. Its form depends on the way the squeezed vacuum has been produced. In particular, it exhibits an explicit dependence on the center-of-mass coordinate $\mathbf{R}=(\mathbf{r}_1+\mathbf{r}_2)/2$ of the atomic system in the case of squeezing via parametric amplification.

A lengthy calculation outlined in Appendix A yields for the dissipative part $\mathcal{L}_d\rho$ of the two-atom master equation

$$\begin{aligned}\mathcal{L}_d\rho = &-\frac{\gamma_f}{2}\sum_{j=1,2}\sum_{q=0,\pm}\{[N(\omega_0)+1](\mathbf{D}_j^+\cdot\hat{\epsilon}_q)(\mathbf{D}_j^-\cdot\hat{\epsilon}_{-q})\rho + N(\omega_0)(\mathbf{D}_j^-\cdot\hat{\epsilon}_{-q})(\mathbf{D}_j^+\cdot\hat{\epsilon}_q)\rho\}\\ &-\frac{\gamma_f}{2}M(\omega_0,\omega_0)\sum_{j=1,2}\sum_{q,q'=0,\pm}\xi_{qq'}[(\mathbf{D}_j^+\cdot\hat{\epsilon}_q)(\mathbf{D}_j^+\cdot\hat{\epsilon}_{q'})\rho + \rho(\mathbf{D}_j^+\cdot\hat{\epsilon}_q)(\mathbf{D}_j^+\cdot\hat{\epsilon}_{q'})]\\ &-\frac{\gamma_f}{2}[N(\omega_0)+1]\sum_{q,q'=0,\pm}\gamma_{qq'}[(\mathbf{D}_1^+\cdot\hat{\epsilon}_q)(\mathbf{D}_2^-\cdot\hat{\epsilon}_{q'})\rho + \rho(\mathbf{D}_1^+\cdot\hat{\epsilon}_q)(\mathbf{D}_2^-\cdot\hat{\epsilon}_{q'})]\\ &-\frac{\gamma_f}{2}N(\omega_0)\sum_{q,q'=0,\pm}\gamma_{qq'}[(\mathbf{D}_1^-\cdot\hat{\epsilon}_q)(\mathbf{D}_2^+\cdot\hat{\epsilon}_{q'})\rho + \rho(\mathbf{D}_1^-\cdot\hat{\epsilon}_q)(\mathbf{D}_2^+\cdot\hat{\epsilon}_{q'})]\\ &-\gamma_f M(\omega_0,\omega_0)\sum_{q,q'=0,\pm}\kappa_{qq'}[(\mathbf{D}_1^+\cdot\hat{\epsilon}_q)(\mathbf{D}_2^+\cdot\hat{\epsilon}_{q'})\rho + \rho(\mathbf{D}_1^+\cdot\hat{\epsilon}_q)(\mathbf{D}_2^+\cdot\hat{\epsilon}_{q'})]\end{aligned} \tag{14}$$

$$+\frac{3\gamma_f}{2}\sum_{j,\ell=1,2}\sum_{q,q'=\pm}\int\frac{d\Omega(\hat{\mathbf{k}})}{4\pi}\{[N(\omega_0)+1]\mathscr{D}_{q,q'}(\hat{\mathbf{k}},\hat{\mathbf{k}})(\mathbf{D}_j^-\cdot\hat{\epsilon}_{-q})e^{-i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{r}_j}\rho e^{i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{r}_\ell}(\mathbf{D}_\ell^+\cdot\hat{\epsilon}_{q'})$$

$$+N(\omega_0)\mathscr{D}_{q,-q'}(\hat{\mathbf{k}},\hat{\mathbf{k}})(\mathbf{D}_j^+\cdot\hat{\epsilon}_{-q})e^{i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{r}_j}\rho e^{-i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{r}_\ell}(\mathbf{D}_\ell^-\cdot\hat{\epsilon}_{q'})$$

$$+M(\omega_0,\omega_0)\mathscr{D}_{q,-q'}(\hat{\mathbf{k}},\pm\hat{\mathbf{k}})(\mathbf{D}_j^+\cdot\hat{\epsilon}_{q})e^{i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{r}_j}\rho e^{i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{r}_\ell}(\mathbf{D}_\ell^+\cdot\hat{\epsilon}_{q'})\}+\text{H.c.}$$

The last contribution to this Liouvillian, proportional to a solid angle integral, would usually be interpreted as a ''fill-up'' term which describes how the atomic ground states are populated via spontaneous emission. The terms including products of raising and lowering operators can indeed be interpreted in this way, the exponentials such as, e.g., $\exp(-i\omega_0\hat{\mathbf{k}}\cdot\mathbf{r}_j/c)$ describing the atomic recoil associated with atomic decay. In addition to these familiar fill-up terms, this integrand also includes additional terms containing products of two atomic raising or lowering operators. Their physical origin is the absorption of two photons from, or emission of two photons into, the squeezed vacuum, with the associated atomic recoil. Such terms disappear for an electromagnetic reservoir in thermal equilibrium $[M(\omega_0,\omega_0)=0]$. While these two-atom and two-photon emission or absorption terms can obviously not be interpreted as ''fill-up'' terms, we shall still loosely call the integral in the master equation (14) the fill-up part.

Turning now to the other contributions to Eq. (14), we observe that the first term, proportional to the spontaneous emission rate $\gamma_f=(1/3\pi\epsilon_0)(d^2\omega_0^3/\hbar c^3)$, describes single-atom spontaneous emission, and in addition the stimulated emission and absorption processes associated with the presence of a mean photon number $\langle N(\omega_0)\rangle$ in the squeezed vacuum. The second term, proportional to

$$\xi_{qq'}=\frac{3}{2}\int\frac{d\Omega(\hat{\mathbf{k}})}{4\pi}\mathscr{D}_{q,-q'}(\hat{\mathbf{k}},\hat{\mathbf{k}})e^{2i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{R}},\tag{15}$$

for the case where the squeezed vacuum results from parametric amplification, and to

$$\xi_{qq'}=\delta_{q,-q'}\tag{16}$$

in the case of near-degenerate four-wave mixing, describes the modification of the single-atom spontaneous decay rate due to the presence of a squeezed vacuum. The dependence of $\xi_{qq'}$ on the center-of-mass position of the atoms, in the case of parametric amplification, has previously been noted by Ficek [10]. It results from the fact that squeezing destroys the isotropy of space, as can be seen by expressing the squeezed vacuum correlation functions in coordinate space: Due to the resonant nature of the atom-photon exchange, the only pairs of modes which contribute to the atomic decay are

$$\langle a_s a_{s'}\rangle=[M(\omega_0,\omega_0)/\omega\omega_0]\,\delta(\omega-\omega_0)\,\delta(\hat{\mathbf{k}}\pm\hat{\mathbf{k}}')\,\delta_{\ell,\ell'},\tag{17}$$

where ± corresponds to parametric amplification and four-wave mixing, respectively. In coordinate space this correlation function becomes

$$\langle a_{\ell,\mathbf{r}_1}a_{\ell',\mathbf{r}_2}\rangle\equiv\int d\mathbf{k}d\mathbf{k}'\langle a_s a_{s'}\rangle e^{-i\mathbf{k}\cdot\mathbf{r}_1}e^{-i\mathbf{k}'\cdot\mathbf{r}_2}$$
$$=M(\omega_0,\omega_0)\int d\Omega(\hat{\mathbf{k}})e^{-i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot(\mathbf{r}_1\pm\mathbf{r}_2)}\delta_{\ell,\ell'}.\tag{18}$$

In the case of counterpropagating modes (four-wave mixing), it exhibits a dependence on the relative coordinate **r**,

$$\langle a_{\ell,\mathbf{r}_1}a_{\ell',\mathbf{r}_2}\rangle=M(\omega_0,\omega_0)\int d\Omega(\hat{\mathbf{k}})e^{-i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{r}}\delta_{\ell,\ell'},\tag{19}$$

while in the case of copropagating modes (parametric amplification) it depends on the center-of-mass coordinate **R**,

$$\langle a_{\ell,\mathbf{r}_1}a_{\ell',\mathbf{r}_2}\rangle=M(\omega_0,\omega_0)\int d\Omega(\hat{\mathbf{k}})e^{-2i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{R}}\delta_{\ell,\ell'},\tag{20}$$

which demonstrates how squeezing destroys the isotropy of space.

The next two terms in Eq. (14), proportional to $\gamma_{qq'}$, describe the spatial modulation of the atomic decay rates due to the presence of a second atom, and depend on their relative position **r**, as expected. Here

$$\gamma_{qq'}=\frac{3}{2}\int\frac{d\Omega(\hat{\mathbf{k}})}{4\pi}\mathscr{D}_{q,q'}(\hat{\mathbf{k}},\hat{\mathbf{k}})e^{i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{r}}.\tag{21}$$

The explicit forms of these coefficients are given in Appendix B. In the absence of squeezing or for a reservoir at zero temperature, we have $N(\omega_0)=0$ and we recover the modulated spontaneous decay discussed, e.g., in Refs. [18,19,21].

Finally, the ''squeezed'' contribution to the collective decay of the two-atom system is proportional to $\kappa_{qq'}$, whose form depends on the way squeezing has been achieved. For parametric amplification we have $\kappa_{qq'}=\xi_{qq'}$, and hence a dependence of the decay on the center-of-mass position of the atomic system, while for near-degenerate four-wave mixing we have

$$\kappa_{qq'}=\frac{3}{2}\int\frac{d\Omega(\hat{\mathbf{k}})}{4\pi}\mathscr{D}_{q,-q'}(\hat{\mathbf{k}},-\hat{\mathbf{k}})e^{i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{r}},\tag{22}$$

and hence a dependence on the relative position of the atoms. The explicit forms of these coefficients are also given in Appendix B.

We find, then, that in contrast to the dipole-dipole potential, the dissipative part of the two-atom Liouvillian depends sensitively on the spatial characteristics of the reservoir correlations. For a squeezed vacuum achieved via parametric amplification, the atomic decay depends both on the relative and center-of-mass positions of atoms, while in the case of four-wave mixing it depends only on the relative position of atoms, as in the case of a thermal bath.

IV. MONTE CARLO WAVE-FUNCTION APPROACH

The two-atom master equation derived in the preceding section is rather combersome to handle, in particular numerically, since a two-atom description leads to substantial computer memory requirements. It has recently been demonstrated that in case the master equation of a given problem is of the Lindblad form, it is possible to replace it by an equivalent formalism relying on an effective non-Hermitian Hamiltonian and Monte Carlo wave-function (MCWF) simulations [22–24]. For example, Ref. [22] explicitly developed such a formalism for the situation of a two-level atom in a squeezed reservoir. The goal of this section is to extend these ideas to the problem at hand, that is, to a system of two multilevel atoms with arbitrary internal structure and coupled to a squeezed vacuum, by introducing operators allowing expression of the master equation (14) in the Lindblad form

$$\mathscr{L}_d\rho=-\frac{1}{2}\sum_m (C_m^\dagger C_m\rho+\rho C_m^\dagger C_m)+\sum_m C_m\rho C_m^\dagger . \tag{23}$$

We proceed by first introducing the operators

$$C_{\ell}^{(1)}(\hat{\mathbf{k}})=\sum_{q=\pm}[\hat{\epsilon}_{\ell}(\hat{\mathbf{k}})\cdot\hat{\epsilon}_q][e^{-i\hat{\mathbf{k}}\cdot\mathbf{r}_1}(\mathbf{D}_1^-\cdot\hat{\epsilon}_q) \\ +e^{-i\hat{\mathbf{k}}\cdot\mathbf{r}_2}(\mathbf{D}_2^-\cdot\hat{\epsilon}_q)] \tag{24}$$

and

$$C_{\ell}^{(2)}(\hat{\mathbf{k}})=[C_{\ell}^{(1)}(\hat{\mathbf{k}})]^\dagger, \tag{25}$$

which permit reexpression of the master equation (14) as

$$\mathscr{L}_d\rho=\frac{1}{2}\sum_{i,j=1}^{2}\beta_{ij}\sum_{\ell=\pm}\int\frac{d\Omega(\hat{\mathbf{k}})}{4\pi}[2C_{\ell}^{(i)}(\hat{\mathbf{k}}_1)\rho C_{\ell}^{(j)}(\hat{\mathbf{k}}_2)^\dagger \\ -C_{\ell}^{(j)}(\hat{\mathbf{k}}_2)^\dagger C_{\ell}^{(i)}(\hat{\mathbf{k}}_1)\rho-\rho C_{\ell}^{(j)}(\hat{\mathbf{k}}_2)^\dagger C_{\ell}^{(i)}(\hat{\mathbf{k}}_1)] \\ \times\delta(\hat{\mathbf{k}}_1\pm\hat{\mathbf{k}}_2), \tag{26}$$

where the ± in the argument of the δ function corresponds to a preparation of the squeezed vacuum via parametric amplification and four-wave mixing, respectively. The coefficients β_{ij} can be thought of as elements of the (2×2) matrix

$$\beta=\frac{3\gamma_f}{2}\begin{pmatrix} N(\omega_0)+1 & -M^*(\omega_0,\omega_0)\\ -M(\omega_0,\omega_0) & N(\omega_0)\end{pmatrix}. \tag{27}$$

Equation (26) has the same form as Eq. (22) of Ref. [22], except that the physical interpretation of the operators involved is of course different. Nonetheless, it can be diagonalized in the same way, leading to

$$\mathscr{L}\rho=\frac{1}{2}\sum_{i=1}^{2}\sum_{\ell=\pm}\int\frac{d\Omega(\hat{\mathbf{k}})}{4\pi}[2A_{\ell}^{(i)}(\hat{\mathbf{k}}_1)\rho A_{\ell}^{(i)}(\hat{\mathbf{k}}_2)^\dagger-A_{\ell}^{(i)}(\hat{\mathbf{k}}_2)^\dagger \\ \times A_{\ell}^{(i)}(\hat{\mathbf{k}}_1)\rho-\rho A_{\ell}^{(i)}(\hat{\mathbf{k}}_2)^\dagger A_{\ell}^{(i)}(\hat{\mathbf{k}}_1)]\,\delta(\hat{\mathbf{k}}_1\pm\hat{\mathbf{k}}_2), \tag{28}$$

where the operators $A_{\ell}^{(i)}(\hat{\mathbf{k}})$ are given by

$$A_{\ell}^{(i)}(\hat{\mathbf{k}})=\sqrt{\lambda_i}\sum_{j=1}^{2}C_{\ell}^{(j)}(\hat{\mathbf{k}})U_{ji}\,. \tag{29}$$

Here $\lambda_{1,2}=3\gamma_f[2N(\omega_0)+1\pm\sqrt{1+4|M(\omega_0,\omega_0)|^2}]/4$, and the coefficients U_{ji} are the elements of the unitary matrix

$$\mathbf{U}=\begin{pmatrix}\cos(\theta/2)e^{-i\phi/2} & -\sin(\theta/2)e^{-i\phi/2}\\ \sin(\theta/2)e^{i\phi/2} & \cos(\theta/2)e^{i\phi/2}\end{pmatrix}, \tag{30}$$

with $\tan\theta=2|M(\omega_0,\omega_0)|$ and $M(\omega_0,\omega_0)=|M(\omega_0,\omega_0)|e^{i(\phi+\pi)}$. For two-level atoms coupled to a perfectly squeezed vacuum and $\mathbf{R}=\mathbf{r}=\mathbf{0}$ (Dicke model), the master equation (28) reduces to the results of Agarwal and Puri [8]. [Note that in that case, $\lambda_2=0$ and hence $A_{\ell}^{(2)}(\hat{\mathbf{k}})=0$.]

The effective non-Hermitian Hamiltonian describing two atoms coupled to a squeezed vacuum is therefore

$$\mathscr{H}_{\text{eff}}=\mathscr{H}_s+V_{dd}+\mathscr{H}_{\text{damp}}, \tag{31}$$

where the non-Hermitian Hamiltonian describing the atomic damping

$$\mathscr{H}_{\text{damp}}=\frac{i\hbar}{2}\sum_{i=1}^{2}\sum_{\ell=\pm}\int\frac{d\Omega(\hat{\mathbf{k}})}{4\pi}A_{\ell}^{(i)}(\hat{\mathbf{k}}_2)^\dagger A_{\ell}^{(i)}(\hat{\mathbf{k}}_1)\,\delta(\hat{\mathbf{k}}_1\pm\hat{\mathbf{k}}_2) \tag{32}$$

can be expressed as the sum of a single-atom contribution

$$\mathscr{H}_{1,\text{damp}}=\frac{i\hbar\gamma_f}{2}\sum_{i=1,2}\sum_{q=0,\pm}(\hat{\mathbf{D}}_i^+\cdot\hat{\epsilon}_q)(\hat{\mathbf{D}}_i^-\cdot\hat{\epsilon}_q^*) \tag{33}$$

and a two-body component $\mathscr{H}_{2,\text{damp}}=\mathscr{H}_{2,\text{damp}}^{(t)}+\mathscr{H}_{2,\text{damp}}^{(s)}$ with

$$\mathscr{H}_{2,\text{damp}}^{(t)}=\frac{i\hbar\gamma_f}{2}[2N(\omega_0)+1]\sum_{q,q'=0,\pm}\gamma_{qq'}[(\mathbf{D}_1^+\cdot\hat{\epsilon}_q) \\ \times(\mathbf{D}_2^-\cdot\hat{\epsilon}_{q'})+(\mathbf{D}_1^-\cdot\hat{\epsilon}_q)(\mathbf{D}_2^+\cdot\hat{\epsilon}_{q'})] \tag{34}$$

giving the ‘‘thermal’’ contributions and

$$\mathscr{H}_{2,\text{damp}}^{(s)}=i\hbar\gamma_f M(\omega_0,\omega_0) \\ \times\left(\sum_{q,q'=0,\pm}\kappa_{qq'}(\mathbf{D}_1^+\cdot\hat{\epsilon}_q)(\mathbf{D}_2^+\cdot\hat{\epsilon}_{q'})+\text{H.c.}\right) \tag{35}$$

giving the ‘‘squeezing’’ contributions. We observe that in contrast to the two-atom part, $\mathscr{H}_{1,\text{damp}}$ does not contain a ‘‘squeezing’’ contribution, due to the fact that

$\mathbf{D}_i^+ \cdot \mathbf{D}_i^+ = \mathbf{D}_i^- \cdot \mathbf{D}_i^- = 0$. Single-atom ''squeezing'' contributions appear only in the fill-up term of the master equation (14), as previously discussed.

V. LOW INTENSITY REGIME

The impact of a squeezed vacuum on the modulation of the decay rate is particularly transparent for atoms interacting with a low intensity near-resonant external laser field such that the subspace of excited states remains almost unpopulated. In this case, it is possible to adiabatically eliminate the excited states manifold. Similarly to the approach of Ref. [25], we decompose the atomic state vector $|\psi(t)\rangle$ as

$$
\begin{aligned}
|\psi(t)\rangle &= P_{e,1}P_{e,2}|\psi(t)\rangle + P_{e,1}P_{g,2}|\psi(t)\rangle + P_{g,1}P_{e,2}|\psi(t)\rangle \\
&\quad + P_{g,1}P_{g,2}|\psi(t)\rangle \\
&\equiv \psi_{ee} + \psi_{eg} + \psi_{ge} + \psi_{gg}\,, \qquad (36)
\end{aligned}
$$

where the projectors $P_{e,i}$ and $P_{g,i}$, $i=1,2$ are given by Eq. (3). Substituting this expression into the Schrödinger equation

$$
i\hbar \frac{d|\psi\rangle}{dt} = \mathscr{H}_{\rm eff}|\psi\rangle \qquad (37)
$$

then yields the coupled differential equations

$$
\begin{aligned}
\dot{\psi}_{ee} &= \left[-\frac{iK}{\hbar} + 2\left(i\delta - \frac{\gamma_f}{2}[N(\omega_0)+1] \right) \right] \psi_{ee} + \frac{i\Omega}{2}[G^+(\mathbf{r}_1)\psi_{ge} + G^+(\mathbf{r}_2)\psi_{eg}] + \frac{i}{\hbar}\mathscr{H}^{(s)}_{2,\rm damp}\psi_{gg}\,, \\
\dot{\psi}_{eg} &= \left[-\frac{iK}{\hbar} + \left(i\delta - \frac{\gamma_f}{2}[N(\omega_0)+1] \right) \right] \psi_{eg} + \frac{i\Omega}{2}[G^+(\mathbf{r}_1)\psi_{gg} + G^-(\mathbf{r}_2)\psi_{ee}] + \frac{i}{\hbar}(V_{dd} + \mathscr{H}^{(t)}_{2,\rm damp})\psi_{ge}\,, \\
\dot{\psi}_{ge} &= \left[-\frac{iK}{\hbar} + \left(i\delta - \frac{\gamma_f}{2}[N(\omega_0)+1] \right) \right] \psi_{ge} + \frac{i\Omega}{2}[G^-(\mathbf{r}_1)\psi_{ee} + G^+(\mathbf{r}_2)\psi_{gg}] + \frac{i}{\hbar}(V_{dd} + \mathscr{H}^{(t)}_{2,\rm damp})\psi_{eg}\,, \\
\dot{\psi}_{gg} &= -\left[\frac{iK}{\hbar} + \frac{\gamma_f}{2}N(\omega_0) \right] \psi_{gg} + \frac{i\Omega}{2}[G^-(\mathbf{r}_1)\psi_{eg} + G^-(\mathbf{r}_2)\psi_{ge}] + \frac{i}{\hbar}\mathscr{H}^{(s)}_{2,\rm damp}\psi_{ee}\,, \qquad (38)
\end{aligned}
$$

where $K=(\mathbf{p}_1^2+\mathbf{p}_2^2)/2M$ is the kinetic energy of the atoms, and the local optical potentials, or light shifts, are given by $G^{\pm}(\mathbf{r}_i) = \mathbf{d}^{\pm}\cdot\mathbf{E}_L^{\pm}(\mathbf{r}_i)/\hbar\Omega$, Ω being the Rabi frequency [16]. Particularly noteworthy in these equations is the coupling between the states ψ_{ee} and ψ_{gg} via $\mathscr{H}^{(s)}_{2,\rm damp}$. This coupling is possible because in contrast to the usual vacuum, it is possible to simultaneously absorb two photons from, or emit two photons into, a squeezed vacuum.

In the case of large detunings $\delta = \omega_L - \omega_0$ between the laser frequency and the Bohr frequency of the atomic transition, the excited states remain weakly populated and it is possible to adiabatically eliminate them. To lowest order in the dipole-dipole interaction, the effective evolution of the ground state wave function ψ_{gg} is then given by

$$
\begin{aligned}
\dot{\psi}_{gg} &= -\frac{i}{\hbar}\left(\frac{\mathbf{p}_1^2}{2M} + \frac{\mathbf{p}_2^2}{2M} \right)\psi_{gg} - \gamma_f N(\omega_0)\psi_{gg} - i\delta s_0 \Bigg[(G_1^- G_1^+ \\
&\quad + G_2^- G_2^+) + \frac{2}{\hbar^2\Omega^2}(\mathscr{H}^{s}_{2,\rm damp})^2 \Bigg] \psi_{gg} + O(\delta^{-2}), \qquad (39)
\end{aligned}
$$

where $s_0 = (\Omega/2)^2/(\delta^2 + \{\gamma_f[N(\omega_0)+1]/2\}^2)$ is the saturation parameter. However, in the regime of large detunings considered here, $\delta \gg \Omega, \gamma_f N(\omega_0)$ it is adequate to approximate this parameter by $s_0 \sim (\Omega/2\delta)^2$. The other components of the wave function of the two-atom system adiabatically follow ψ_{gg} according to the algebraic relations $\psi_{eg} = -(1/\delta)G_1^+\psi_{gg}$, $\psi_{ge} = -(1/\delta)G_2^+\psi_{gg}$, and $\psi_{ee} = -(\mathscr{H}^{(s)}_{2,\rm damp}/2\hbar\delta)\psi_{gg}$. In addition to the usual optical potential, the potential energy appearing in Eq. (39) also includes a correction due to interaction of the atoms with the squeezed vacuum. It might at first appear unexpected that $\mathscr{H}^{(s)}_{2,\rm damp}$ should give a contribution to the potential. But as follows from Eqs. (38), the coupling to the squeezed vacuum results in the exchange of populations between excited and ground state manifolds of the atoms, which means that $\mathscr{H}^{(s)}_{2,\rm damp}$ plays the role of a parametric force. It was already shown in Ref. [25] that the dipole-dipole interaction contributes to the effective potential

$$
V_{dd-\rm eff} = -\frac{\Omega^2}{\delta^2}(G_1^- V_{dd} G_2^+ + G_2^- V_{dd} G_1^+), \qquad (40)
$$

only to second order in perturbation theory. In contrast, the presence of a squeezed vacuum contributes to the effective potential

$$
V^{q}_{dd-\rm eff} = \frac{1}{2\hbar\delta}(\mathscr{H}^{(s)}_{2,\rm damp})^2 \qquad (41)
$$

at the same order as the optical fields.

VI. SUMMARY AND CONCLUSION

In this paper we have analyzed the impact of a squeezed vacuum on the dynamics of a two-atom system. Surprisingly perhaps, we found that it does not modify the Hamiltonian part of the dipole-dipole interaction, due to a cancellation of the associated stimulated absorption and emission effects in a way analogous to the case of a thermal bath at nonzero temperature. It does, however, modify the decay rate of the atoms. In particular, for a squeezed vacuum obtained via parametric amplification, this rate depends not just on the relative position of the atoms, as is normally the case, but also on their center of mass. This results from the fact that squeezing destroys the isotropy of space. Finally, we found that in case the excited electronic states of the atoms can be adiabatically eliminated, the effects of a squeezed vacuum appear already in lowest order in perturbation theory, in the form of an effective potential resulting from the squeezed-vacuum-induced population exchange between the excited and ground atomic states. This exchange is possible since, in contrast to the situation of a thermal reservoir, it is now possible to simultaneously exchange two excitations between the atoms and the reservoir. It is interesting to note that this effect already appears to first order in perturbation theory, while the dipole-dipole interaction contributes to the effective potential only to second order in perturbation theory.

An experimental verification of the effects predicted in this paper requires one to achieve conditions similar to those needed to demonstrate the modification of single-atom radiative processes in squeezed light, with the additional difficulty that ideally, one would like to be able to keep the separation between the atoms under control. The major difficulty, both for single- and two-atom experiments, seems to be associated with the need to achieve a situation where the coupling of the atoms to the squeezed bath dominates over the coupling to ordinary vacuum. This requires that ‘‘mode matching’’ be achieved between the squeezed vacuum and the radiation pattern of the atoms. In free space, this implies that a squeezed vacuum must be achieved over a large fraction of 4 π steradians. Kimble and co-workers [26] have proposed several approaches toward this goal, involving, for instance, the use of atoms trapped in the Lamb-Dicke regime and irradiated by squeezed light from a beam of large numerical aperture, or an alternative approach using ‘‘one-dimensional’’ atoms in a high finesse cavity.

ACKNOWLEDGMENTS

This work is funded by the U.S. Office of Naval Research Contract No. N00014-91-J1205, by the National Science Foundation Grants No. PHY92-13762 and No. PHY95-07639, and by the Joint Services Optics Program.

APPENDIX A: DERIVATION OF THE MASTER EQUATION FOR ATOMS IRRADIATED BY SQUEEZED VACUUM

This appendix gives details of the derivation of the master equation (14), mostly emphasizing terms due to atom interaction with squeezed vacuum. Introducing the field operators

$$\Gamma_{jq}(t) \equiv \sum_{s} \Gamma_{jqs} = \sum_{s} \mathscr{E}_0(k)[\hat{\boldsymbol{\epsilon}}_{\ell}(\hat{\mathbf{k}}) \cdot \hat{\boldsymbol{\epsilon}}_q] a_s e^{i\hat{\mathbf{k}}\cdot\mathbf{r}_j} e^{i(\omega_0-\omega)t} \tag{A1}$$

permits one to express the interaction between the atoms and the field modes as

$$\mathscr{H}_{A-F} = \sum_{jq} (\mathbf{D}_j^{+} \cdot \hat{\boldsymbol{\epsilon}}_q)\Gamma_{jq} + \text{H.c.} \tag{A2}$$

In taking the trace Tr_F over the bath, we have to take into account that it is in a squeezed state, so that in addition to correlations of the type $\langle \Gamma_{1q}^{\dagger}\Gamma_{2q'}\rangle$, which are already nonzero for a thermal bath, the following nonzero correlation functions must also be considered:

$$\begin{aligned}\langle \Gamma_{1q}(t)\Gamma_{2q'}(\tau)\rangle &= \sum_{s_1,s_2} \langle \Gamma_{1qs_1}\Gamma_{2q's_2}\rangle \\ &= -\frac{3\gamma_f}{16\pi}\frac{c^3}{\omega_0^3}\frac{\epsilon_0 V}{2\pi\hbar}\sum_{\ell,\ell'=\pm} \int\int d\mathbf{k}_1 d\mathbf{k}_2 \mathscr{E}_0(k_1)\mathscr{E}_0(k_2) \\ &\quad \times \langle a_{s_1}a_{s_2}\rangle_r \mathscr{D}_{q,q'}(\hat{\mathbf{k}}_1,\hat{\mathbf{k}}_2) e^{\frac{i}{2}(\mathbf{k}_1-\mathbf{k}_2)\cdot\mathbf{r}} e^{i(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{R}} e^{i(ck_1-\omega_0)t} e^{i(ck_2-\omega_0)\tau}.\end{aligned}$$

Taking into account Eqs. (7) and (8) yields then, in the case of parametric amplification,

$$\begin{aligned}\langle \Gamma_{1q}(t)\Gamma_{2q'}(\tau)\rangle &= \sum_{s} [\langle \Gamma_{1qs_1}\Gamma_{2q's_2}\rangle e^{i(\omega_0-\omega)(t-\tau)} + \langle \Gamma_{1qs_2}\Gamma_{2q's_1}\rangle e^{-i(\omega_0-\omega)(t-\tau)}]\delta_{\ell,\ell'}\delta(2\omega_0-ck_1-ck_2)\delta(\hat{\mathbf{k}}_1-\hat{\mathbf{k}}_2) \\ &= -\frac{3\gamma_f}{4}\frac{1}{\omega_0^3}\int d\omega\, \omega^2\sqrt{\omega(2\omega_0-\omega)}[M(\omega,2\omega_0-\omega)e^{i(\omega_0-\omega)(t-\tau)} \\ &\quad + M(2\omega_0-\omega,\omega)e^{-i(\omega_0-\omega)(t-\tau)}]\int \frac{d\Omega(\mathbf{k})}{4\pi}\mathscr{D}_{q,-q'}(\mathbf{z}\hat{\mathbf{k}},\hat{\mathbf{k}}) e^{i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{r}} e^{2i\frac{\omega_0-\omega}{c}\hat{\mathbf{k}}\cdot\mathbf{R}}.\end{aligned}$$

while for four-wave mixing we find

$$\langle\Gamma_{1q}\Gamma_{2q'}\rangle=\sum_{s}\,[\langle\Gamma_{1qs_1}\Gamma_{2q's_2}\rangle e^{i(\omega_0-\omega)(t-\tau)}+\langle\Gamma_{1qs_2}\Gamma_{2q's_1}\rangle e^{-i(\omega_0-\omega)(t-\tau)}]\delta_{\ell,\ell'}\delta(2\omega_0-ck_1-ck_2)\,\delta(\hat{\mathbf{k}}_1+\hat{\mathbf{k}}_2)$$

$$=-\frac{3\gamma_f}{4}\frac{1}{\omega_0^3}\int d\omega\omega^2\sqrt{\omega(2\omega_0-\omega)}[M(\omega,2\omega_0-\omega)e^{i(\omega_0-\omega)(t-\tau)}$$

$$+M(2\omega_0-\omega,\omega)e^{-i(\omega_0-\omega)(t-\tau)}]\int\frac{d\Omega(\hat{\mathbf{k}})}{4\pi}\mathscr{D}_{q,-q'}(\hat{\mathbf{k}},-\hat{\mathbf{k}})e^{i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{r}}e^{2i\frac{\omega_0-\omega}{c}\hat{\mathbf{k}}\cdot\mathbf{R}}.$$

Performing the integral over the time difference τ in Eq. (10) then gives in the case of parametric amplification

$$\int_0^t\langle\Gamma_{1q}\Gamma_{2q'}\rangle d\tau=-\frac{3\gamma_f}{4}\frac{1}{\omega_0^3}\int d\omega\omega^2\sqrt{\omega(2\omega_0-\omega)}\left\{M(\omega,2\omega_0-\omega)\left[\delta(\omega_0-\omega)+\frac{i}{\pi}\mathrm{P}\left(\frac{1}{\omega_0-\omega}\right)\right]\right.$$

$$\left.+M(2\omega_0-\omega,\omega)\left[\delta(\omega_0-\omega)-\frac{i}{\pi}\mathrm{P}\left(\frac{1}{\omega_0-\omega}\right)\right]\right\}\int\frac{d\Omega(\hat{\mathbf{k}})}{4\pi}\mathscr{D}_{q,-q'}(\mathbf{k},-\mathbf{k})e^{2i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{R}}e^{i\frac{\omega_0-\omega}{c}\hat{\mathbf{k}}\cdot\mathbf{r}}$$

$$=-\frac{3\gamma_f}{2}M(\omega_0,\omega_0)\int\frac{d\Omega(\hat{\mathbf{k}})}{4\pi}\mathscr{D}_{q,-q'}(\hat{\mathbf{k}},\hat{\mathbf{k}})e^{2i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{R}},$$

while for four-wave mixing we get

$$\int_0^t\langle\Gamma_{1q}\Gamma_{2q'}\rangle d\tau=-\frac{3\gamma_f}{4}\frac{1}{\omega_0^3}\int d\omega\omega^2\sqrt{\omega(2\omega_0-\omega)}\left\{M(\omega,2\omega_0-\omega)\left[\delta(\omega_0-\omega)+i\frac{\mathrm{P}}{\pi}\left(\frac{1}{\omega_0-\omega}\right)\right]\right.$$

$$\left.+M(2\omega_0-\omega,\omega)\left[\delta(\omega_0-\omega)-i\frac{\mathrm{P}}{\pi}\left(\frac{1}{\omega_0-\omega}\right)\right]\right\}\int\frac{d\Omega(\hat{\mathbf{k}})}{4\pi}\mathscr{D}_{q,q'}(\hat{\mathbf{k}},-\hat{\mathbf{k}})e^{i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{r}}e^{2i\frac{\omega_0-\omega}{c}\hat{\mathbf{k}}\cdot\mathbf{R}}$$

$$=-\frac{3\gamma_f}{2}M(\omega_0,\omega_0)\int\frac{d\Omega(\hat{\mathbf{k}})}{4\pi}\mathscr{D}_{q,q'}(\hat{\mathbf{k}},-\hat{\mathbf{k}})e^{i\frac{\omega_0}{c}\hat{\mathbf{k}}\cdot\mathbf{r}}.$$

For both parametric amplification and four-wave mixing, the principal parts cancel each other, which means that the coupling of the atoms to a broadband squeezed vacuum does not lead to ‘‘frequency shifts,’’ i.e., there are no new contributions to the Hamiltonian evolution of the atoms. The only effect of squeezing is to modify the various rates of spontaneous emission.

APPENDIX B: EXPLICIT EXPRESSIONS FOR THE COEFFICIENTS OF THE MASTER EQUATION

This appendix gives expressions for the coefficients (15)–(22) occurring in the two-atom master equation. The rates $\gamma_{qq'}$ are given by

$$\gamma_{qq'}=\gamma^*_{q'q},$$

$$\gamma_{++}=\gamma_{--}=j_0(kr)-\tfrac{1}{4}(3\cos^2\theta_r-1)j_2(kr),$$

$$\gamma_{00}=j_0(kr)+\tfrac{1}{2}(3\cos^2\theta_r-1)j_2(kr),$$

$$\gamma_{-0}=\gamma_{0+}=\frac{3}{2\sqrt{2}}j_2(kr)\sin\theta_r\,\cos\theta_re^{-i\phi_r},$$

$$\gamma_{-+}=\tfrac{3}{4}j_2(kr)(1-\cos^2\theta_r)e^{-2i\phi_r}.$$

In the case of parametric amplification, the coefficients $\kappa_{qq'}$ are

$$\kappa_{qq'}=\kappa^*_{-q'-q},$$

$$\kappa_{++}=\tfrac{3}{4}j_2(2kR)(1-\cos^2\theta_R)e^{-2i\phi_R},$$

$$\kappa_{00}=j_0(2kR)+\tfrac{1}{2}(3\cos^2\theta_R-1)j_2(2kR),$$

$$\kappa_{-0}=\kappa_{0-}=\frac{3}{2\sqrt{2}}j_2(2kR)\sin\theta_R\,\cos\theta_Re^{-i\phi_R},$$

$$\kappa_{+-}=\kappa_{-+}=j_0(2kR)-\tfrac{1}{4}(3\cos^2\theta_R-1)j_2(2kR),$$

and in the case of four-wave mixing they are

$$\kappa_{qq'}=\kappa^*_{q'q},$$

$$\kappa_{++}=\kappa_{--}=j_0(kr)-\tfrac{1}{4}(3\cos^2\theta_r-1)j_2(kr),$$

$$\kappa_{00}=j_0(kr)+\tfrac{1}{2}(3\cos^2\theta_r-1)j_2(kr),$$

$$\kappa_{-0}=\kappa_{0+}=\frac{3}{2\sqrt{2}}j_2(kr)\sin\theta_r\,\cos\theta_r e^{-i\phi_r},$$

$$\kappa_{+-}=\tfrac{3}{4}\,j_2(kr)(1-\cos^2\theta_r)e^{-2i\phi_r}.$$

In these expressions (r,θ_r,ϕ_r) and (R,θ_R,ϕ_R) are the spherical coordinates of the relative and center-of-mass position, respectively, expressed with respect to the quantization coordinate system, and the functions $j_i(kr)$ are spherical Bessel functions of the first kind.

---

[1] C. W. Gardiner, Phys. Rev. Lett. **56**, 1917 (1986).

[2] G. Lenz and P. Meystre, in *Quantum Optics V*, edited by D. F. Walls and J. D. Harvey (Springer-Verlag, Heidelberg, 1994).

[3] G. J. Milburn, Phys. Rev. **34**, 4882 (1986).

[4] C. W. Gardiner, A. S. Parkins, and M. J. Collet, J. Opt. Soc. Am. B **4**, 1683 (1987).

[5] G. M. Palma and P. L. Knight, Opt. Commun. **73**, 131 (1989).

[6] G. M. Palma and P. L. Knight, Phys. Rev. A **39**, 1962 (1989).

[7] V. Buzek, P. L. Knight, and I. K. Kudryavtsev, Phys. Rev. A **44**, 1931 (1991).

[8] G. S. Agarwal and R. R. Puri, Phys. Rev. A **41**, 3782 (1990).

[9] Z. Ficek, Opt. Commun. **82**, 130 (1991).

[10] Z. Ficek, Phys. Rev. A **44**, 7759 (1991).

[11] Z. Ficek, J. Mod. Opt. **40**, 2339 (1993).

[12] M. D. Reid and D. F. Walls, Phys. Rev. A **31**, 1622 (1985).

[13] R. E. Slusher, L. W. Hollberg, B. Yurke, J. C. Mertz, and J. F. Valley, Phys. Rev. Lett. **55**, 2409 (1985).

[14] M. J. Collet and C. W. Gardiner, Phys. Rev. A **30**, 1386 (1984).

[15] L. A. Wu, H. J. Kimble, J. L. Hall, and H. Wu, Phys. Rev. Lett. **57**, 2520 (1986).

[16] C. Cohen-Tannoudji, in *Fundamental Systems in Quantum Optics*, edited by J. Dalibard, J. M. Raimond, and J. Zinn-Justin (North-Holland, Amsterdam, 1992), p. 153.

[17] G. S. Agarwal, in *Quantum Optics*, edited by G. Höhler, Springer Tracts in Modern Physics Vol. 70 (Springer, New York, 1974).

[18] A. M. Smith and K. Burnett, J. Opt. Soc. Am. B **8**, 1592 (1991).

[19] G. Lenz and P. Meystre, Phys. Rev. A **48**, 3395 (1993).

[20] P. D. Drummond, Quantum Opt. **2**, 205 (1990).

[21] J. Guo and J. Cooper, Phys. Rev. A **51**, 3128 (1995).

[22] R. Dum, A. S. Parkins, P. Zoller, and C. W. Gardiner, Phys. Rev. A **46**, 4382 (1992).

[23] L. Tian and H. J. Carmichael, Phys. Rev. A **46**, R6801 (1992).

[24] K. Mølmer, Y. Castin, and J. Dalibard, J. Opt. Soc. Am. A **10**, 524 (1993).

[25] E. V. Goldstein, P. Pax, and P. Meystre, Phys. Rev. A **53**, 2604 (1996).

[26] H. J. Kimble, E. S. Polzik, N. Georgiades, and H. Mabuchi, in *Laser Spectroscopy*, edited by Louis Bloomfield, Thomas Gallagher, and Daniel Larson, AIP Conf. Proc. No. 290 (AIP, New York, 1993), p. 340.