

## Quantum theory of nondegenerate multiwave mixing: General formulation

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In this first paper of a series on the quantum theory of nondegenerate multiwave mixing applicable to traveling-wave interaction geometries, we describe our problem formulation. We consider the explicit dynamics of a subset of the field quantization modes interacting with a system of stationary two-level atoms contained in a volume much smaller than the field quantization volume. Because we make the realistic assumption of leaving the remaining infinite set of field modes as a common thermal-field reservoir, the resulting Langevin equations contain extra decay terms due to collective spontaneous emission or super-radiance. We show that all but one of these super-radiance terms are negligible in the following two limits: (a) when the number of atoms in a diffraction volume is small; (b) when the atoms are pumped far off resonance. There is, however, an anomalous decay term which does not appear in a classical model of super-radiance based upon coherently phased atomic dipoles. The magnitude of this anomalous term is neither dependent upon the number of atoms nor on the pump-frequency detuning and may not be negligible at a low pump intensity. Neglecting the super-radiance terms, we then present a general Fourier-expansion solution technique for obtaining the atomic polarization in the presence of any number of field modes. The expansion is shown to be convergent in a commonly occurring situation in which all the strong pump modes are frequency degenerate and the remaining nondegenerate modes are all weak compared to the atomic saturation intensity. In subsequent papers of this series, we will present methods to treat, with some rigor, the spatial propagation of an interacting multimode quantum field and apply these methods to traveling-wave squeezed-state generation experiments.

### I. INTRODUCTION

Squeezed light is a state of the electromagnetic field at optical frequencies whose electric field measurement uncertainty is phase dependent, with a minimum falling below the level set by the coherent state of light.<sup>1</sup> Squeezed light is, in fact, a macroscopic quantum state which has no classical analog in the sense that it cannot be produced by random superposition of coherent states.

Recently, several groups have demonstrated the generation of squeezed light employing different systems, each obtaining a different degree of squeezing over a different bandwidth. Slusher *et al.*<sup>2</sup> were the first to generate squeezed light demonstrating 17% squeezing in light emitted by a cavity containing the probe and conjugate fields of a four-wave mixer undergoing nearly degenerate four-wave mixing in a sodium atomic beam. Thereafter, Shelby *et al.*<sup>3</sup> observed 12% squeezing via nearly degenerate forward four-wave mixing in a single-mode optical fiber. Kimble and collaborators<sup>4,5</sup> are, by far, the most successful ones, demonstrating squeezing in two different systems. With their first system,<sup>4</sup> they have set a

much higher benchmark for squeezing by observing over 50% squeezing in light emitted by a cavity containing the signal and idler fields of a parametric amplifier undergoing nearly degenerate optical parametric amplification in a MgO:LiNbO<sub>3</sub> crystal. Their other system exploits the large coupling strength of a small-volume high-finesse optical cavity to an atomic beam of two-level atoms.<sup>5</sup> Maeda *et al.*<sup>6</sup> have also observed squeezing via nearly degenerate forward four-wave mixing in a sodium vapor cell, demonstrating squeezing via interaction with a Doppler broadened medium for the first time. And most recently, Machida *et al.*<sup>7</sup> have generated amplitude squeezed light directly out of a constant-current driven semiconductor laser.

Out of the six experiments which have generated squeezed light, three have involved the basic resonant interaction of the electromagnetic field with a system of two-level atoms. The experiments of Slusher *et al.*<sup>2</sup> and Kimble and collaborators<sup>5</sup> differ in one important aspect from the Maeda *et al.*<sup>6</sup> experiment in that they employed optical cavities to enhance the atom-field interaction, thus making electromagnetic field propagation issues ir-

relevant. Such issues are, however, crucial to a proper theoretical description of the Maeda *et al.*<sup>6</sup> experiment, which employed a traveling-wave configuration. With these issues in mind, we have recently formulated a quantum theory for nondegenerate multiwave mixing in an atomic medium and applied it to traveling-wave squeezed-state generation experiments via four-photon mixing. A summary of this formulation and its application was recently reported.<sup>8</sup> In this and in a series of forthcoming publications, we present details of this theory and its application to squeezed-state generation experiments.

Our theory predicts squeezing in resonance fluorescence,<sup>9</sup> degenerate four-wave mixing,<sup>10–12</sup> and nondegenerate four-wave mixing<sup>13–15</sup> in a consistent manner. In the squeezing calculations the effects of spontaneous emission, propagation loss, atomic collisions, and phase mismatch due to propagation are properly taken into account for the first time. A preliminary analysis of the effect of super-radiance has also been carried out.

The experiments<sup>16</sup> following the first proposal of squeezed-state generation via backward degenerate four-wave mixing by Yuen and Shapiro<sup>10</sup> encountered difficulties partly due to loss and mainly due to atomic fluorescence. Because these experiments were performed in a near resonantly-pumped atomic medium, single-photon excitation resulted in population inversion, which led to spontaneous emission at the squeezed-mode frequency. A phenomenological analysis by Kumar and Shapiro<sup>11</sup> indicated that loss was less critical in a forward four-wave mixing geometry. However, a proper account of the spontaneous-emission noise, which required quantization of the atomic medium along with the electromagnetic field, was first given by Reid and Walls.<sup>12</sup> They showed that propagation loss and spontaneous emission severely limit the amount of squeezing obtainable via degenerate four-wave mixing in both forward and backward configurations. On the other hand, a study of the spectrum of squeezing<sup>17</sup> in resonance fluorescence from a single atom or a thin layer of atoms shows that most of the squeezing occurs at nondegenerate frequencies near the Rabi sidebands,<sup>9,18</sup> implying that a nondegenerate analysis of the four-wave mixing process is required. The total amount of squeezing obtainable from a thin layer of atoms is of course very small, therefore, we need to consider a thick medium in which case a proper treatment of multimode quantum field propagation and wave coupling becomes absolutely essential.

Recently, Reid and Walls,<sup>14</sup> Sargent and collaborators,<sup>15,19</sup> and Agarwal<sup>20</sup> have also developed quantum theories for nondegenerate four-photon mixing and applied them to the intracavity atomic-beam squeezed-state generation experiment of Slusher *et al.*<sup>2</sup> Because these theories consider electromagnetic fields inside optical cavities, they are not directly applicable to the traveling-wave experiment of Maeda *et al.*<sup>6</sup> The formulation of Reid and Walls<sup>14</sup> is based on the quantum statistical methods developed by Haken<sup>21</sup> and later adapted for optical bistability by Drummond and Walls.<sup>22</sup> Our formulation, although based on the same techniques, is much more general. Not only does it give a more general ex-

pression for the atomic polarizability, it also differs from theirs in an essential way in that we handle the slowly varying amplitude approximation in the frequency domain. This approach more rigorously justifies the adiabatic approximation for the elimination of atomic variables and leads to extra terms which are important for a consistent treatment of spatial propagation. Instead of adopting the heuristic  $t \rightarrow z/c$  transformation used in almost all the previous works, we have developed two equivalent methods to treat, with some rigor, the spatial propagation of the multimode traveling-wave quantum field. The slowly varying envelope method places a much better limit on the validity of the squeezed-noise calculations using the  $t \rightarrow z/c$  transformation,<sup>12,20</sup> whereas the quantum-mode method provides physical insight into how the time evolution of the usual annihilation operators leads to spatial propagation of the wave.

In the frequency-degenerate limit, without consideration of the pump-probe phase mismatch due to nonlinear dispersion, our theory agrees with the earlier four-wave mixing results of Reid and Walls,<sup>12</sup> and in the thin-medium approximation it correctly reproduces the results of Heidmann and Reynaud.<sup>9</sup> Our inclusion of the pump-probe phase mismatch, collisions such as those between the two-level atoms and those between the two-level atoms and the atoms of any externally added buffer gas, and super-radiance give rise to additional effects not considered elsewhere.

In this paper, we begin in Sec. II by setting up the atom-field Hamiltonian for a system of  $N$  two-level atoms interacting with the electromagnetic field quantized in a box whose volume is much larger than that occupied by the two-level atoms. We follow the explicit dynamics of a subset of the field quantization modes and leave the remaining infinite set of modes as a common thermal-field reservoir. In addition, to model the soft collisions between the atoms, each two-level atom is coupled to a separate phase-damping reservoir.

In Sec. III we follow the master-equation techniques described by Louisell<sup>23</sup> to derive a reduced density operator equation of motion for the above system via the Markov approximation. The coefficients of some of the terms in this equation of motion are dependent upon the phase coherence of the atomic dipoles. These super-radiance terms arise because of cooperative atomic behavior due to our usage of a common thermal-field reservoir for all atoms. In the case of a thick medium, the condition for the validity of the Markov approximation for the super-radiance terms is shown to be much more stringent than that for the usual spontaneous-decay terms.

In Sec. IV the reduced density operator equation of motion is transformed to a  $c$ -number equation of motion for the associated distribution function using the usual technique of choosing an operator ordering. For applications such as squeezed-state generation, where correlations of the field only up to the second order are needed, this  $c$ -number equation reduces to a Fokker-Planck equation, from which a corresponding set of Langevin equations are obtained. Besides the usual spontaneous decay and noise terms, the Langevin equations for the atomic variables are shown to contain extra decay and noise

terms due to super-radiance.

In Sec. V a preliminary analysis of the effect of super-radiance is carried out by assuming that the atomic dipoles are coherently excited at some initial time. Our analysis shows that in the zero mean-field limit, all but one of the additional decay and fluctuation terms due to super-radiance are negligible in the following two limits: (a) when the number of atoms in a diffraction volume is small and (b) when the atoms are pumped far from resonance. Both of these limits are dependent upon the length of the medium. There is, however, an anomalous decay term which is not accounted for by a classical coherently phased dipole model and whose magnitude is neither dependent upon the number of atoms nor the pump-frequency detuning. This anomalous decay term is shown to be negligible at high pump intensity when the total atomic state approaches an incoherent mixture of atomic-number states. However, at pump intensities below the saturation intensity for the two-level atoms, this term is *not negligible*. Thus, contrary to popular belief, our model shows that the super-radiance effects may not be negligible at low pump intensities even when the number of atoms in a diffraction volume is small. To the best of our knowledge this effect has not been studied before.

In Sec. VI after neglecting the super-radiance terms, we solve the set of Langevin equations by using a Fourier expansion technique. The resulting equations in the frequency domain, which are recursive in nature, are solved iteratively under the commonly occurring condition that the frequency-degenerate pump modes are all strong, whereas the remaining nondegenerate modes are all weak compared to the atomic saturation intensity. Using this method, a general expression is obtained for the  $c$ -number atomic polarization variable for any number of interacting field modes.

We will continue this development in the forthcoming papers of this series on the quantum theory of nondegenerate multiwave mixing. In the second paper, a frequency domain method will be formulated to treat the usual adiabatic approximation. A slowly varying envelope method will be developed to treat, with some rigor, the spatial propagation of an interacting multimode quantum field. This method will be applied in the third paper to obtain the spectrum of squeezing for a single beam propagating through the two-level medium. In the fourth paper, we will present another method to treat the spatial propagation, using a quantum-mode evolution technique.

## II. ATOM-FIELD MODEL

In this theory we consider a system of  $N$  stationary two-level atoms interacting with an infinite number of plane-wave electromagnetic field modes quantized with periodic boundary conditions in a box of volume  $V_Q$ . We consider the explicit dynamics of  $q$  of these modes with wave vectors  $\mathbf{k}_1, \dots, \mathbf{k}_q$  and leave the remaining infinite set of modes as a common thermal-field reservoir. The reservoir modes are assumed to be in thermal equilibrium with the environment at the initial time. To neglect Ray-

leigh scattering, the atoms are assumed to be uniformly distributed over a volume  $V_M$ , which in general is much smaller than  $V_Q$ . The soft collisions, such as those between the two-level atoms and the atoms of any additional buffer gas, are modeled by coupling the atoms to phase-damping reservoirs, thereby increasing their transverse relaxation rate.

In order to derive correct expressions for the spontaneous-decay and super-radiant decay rates, we follow the vector field treatment as given by Ho, Kumar, and Shapiro.<sup>24</sup> We assume that each atom is characterized by four states: a ground state  $|g\rangle$  and three degenerate excited states  $|x\rangle$ ,  $|y\rangle$ , and  $|z\rangle$  which couple to the  $x$ ,  $y$ , and  $z$  components, respectively, of the thermal-field modes. The  $q$  field modes, however, are all assumed  $x$  polarized so that they only couple to the state  $|x\rangle$ . Let us define the following atomic operators:

$$\hat{V}_{li} \equiv |g\rangle_i \langle l|, \quad (2.1)$$

$$\hat{V}_{li}^\dagger \equiv |l\rangle_i \langle g|, \quad (2.2)$$

$$\hat{n}_{li} \equiv |l\rangle_i \langle l| = \hat{V}_{li}^\dagger \hat{V}_{li}, \quad (2.3)$$

$$\hat{n}_{gi} \equiv |g\rangle_i \langle g| = \hat{V}_{li} \hat{V}_{li}^\dagger, \quad (2.4)$$

$$\hat{n}_{di} \equiv \sum_l \hat{n}_{li} - \hat{n}_{gi}, \quad (2.5)$$

where  $l \in \{x, y, z\}$ ,  $i$  labels the states and operators for the  $i$ th atom,  $\{\hat{V}_{li}\}$  are the atomic down-transition operators,  $\{\hat{V}_{li}^\dagger\}$  are the atomic up-transition operators,  $\{\hat{n}_{li}, \hat{n}_{gi}\}$  are the occupation operators for the respective atomic states, and  $\{\hat{n}_{di}\}$  are the population inversion operators.

We denote the annihilation operators of the electromagnetic field by  $\hat{a}_j$ , with  $j = 1, 2, \dots, q$  representing the  $q$  field modes of interest and  $j = q+1, q+2, \dots, \infty$  representing the thermal-field reservoir modes having wave vectors  $\mathbf{k}_j$ . We use  $\omega$  to denote exclusively the free-space angular frequencies  $\omega_j = |\mathbf{k}_j|c$ . The optical resonance frequency of the atoms is denoted by  $\omega_a$ .

Thus the Hamiltonian of this atom-field system is given by

$$\hat{H} = \hat{H}_0 + \hat{H}_I + \hat{H}_R + \hat{H}_C, \quad (2.6)$$

where the free part is

$$\hat{H}_0 = \sum_j \hbar\omega_j \hat{a}_j^\dagger \hat{a}_j + (\hbar\omega_a/2) \sum_i \hat{n}_{di}, \quad (2.7)$$

and the atom-field interaction part is

$$\hat{H}_I = \sum_{j,i} [i\hbar C_{xj}(\mathbf{r}_i) \hat{a}_j \hat{V}_{xi}^\dagger + \text{H.c.}], \quad (2.8)$$

with H.c. denoting the Hermitian conjugate. The thermal-field reservoir term consists of a free part and an interacting part

$$\hat{H}_R = \hat{R} + \hat{V}, \quad (2.9)$$

with the former being

$$\hat{R} = \sum_s \hbar\omega_s \hat{a}_s^\dagger \hat{a}_s, \quad (2.10)$$

and the latter being

$$\hat{V} = \sum_{l,s,i} [i\hbar C_{ls}(\mathbf{r}_i)\hat{a}_s\hat{V}_{li}^\dagger + \text{H.c.}] . \quad (2.11)$$

The phase-damping term due to collisions is given by

$$\hat{H}_C = \sum_i \hat{\Gamma}_{pi}\hat{n}_{di} , \quad (2.12)$$

where  $\{\hat{\Gamma}_{pi}\}$  are the operators of the phase-damping reservoirs. Under the summation signs of the above equations,  $j$  denotes the sum over the  $q$  field modes:  $j \in \{1, 2, \dots, q\}$ ;  $s$  denotes the sum over the thermal-field reservoir modes:  $s \in \{q+1, q+2, \dots, \infty\}$ ;  $l$  denotes the sum over the atomic excited states:  $l \in \{x, y, z\}$ ; and  $i$  denotes the sum over the number of atoms:  $i \in \{1, 2, \dots, N\}$ . The coefficients are given by

$$C_{lj}(\mathbf{r}_i) = g_j \mu_{lj} \exp(i\mathbf{k}_j \cdot \mathbf{r}_i) , \quad (2.13)$$

$$g_j = (\omega_a^2 / 2\hbar\epsilon_0\omega_j V_Q)^{1/2} , \quad (2.14)$$

where  $\mathbf{r}_i$  is the position of the  $i$ th atom and  $\mu_{lj}$  is a component of the atomic dipole  $\langle l | e\hat{\mathbf{r}}_i | g \rangle_i$  coupled to the  $j$ th field mode with  $e$  being the electron charge. We assume that  $\langle l | e\hat{\mathbf{r}}_i | g \rangle_i$  has the same value for each atom. If the polarization vector of the  $j$ th field mode is  $\mathbf{e}_j$ , then

$$\mu_{lj} \equiv_i \langle l | e\hat{\mathbf{r}}_i | g \rangle_i \cdot \mathbf{e}_j . \quad (2.15)$$

For later reference in treating propagation, we note that the interaction term of Eq. (2.8) has been obtained under dipole and rotating-wave approximations from the exact nonrelativistic interaction energy

$$H_I = \sum [e^2 \mathbf{A}_\perp(\mathbf{r}_{ei}) \cdot \mathbf{A}_\perp(\mathbf{r}_{ei}) - 2e \mathbf{P}_{ei} \cdot \mathbf{A}_\perp(\mathbf{r}_{ei})] / 2m_e , \quad (2.16)$$

where  $m_e$  and  $\mathbf{P}_{ei}$  are the mass and generalized momentum, respectively, of the interacting electron in the  $i$ th atom, and  $\mathbf{A}_\perp(\mathbf{r}_{ei})$  is the transverse vector potential at position  $\mathbf{r}_{ei}$  of the electron. The  $\mathbf{A} \cdot \mathbf{P}$  form of  $H_I$  is used instead of the  $\mathbf{E} \cdot \mathbf{r}$  form because in the Coulomb gauge it corresponds to the usual Hilbert space for quantization of the electromagnetic field.<sup>25</sup> This form of  $H_I$  also leads to the factor  $\omega_a^2/\omega_j$  instead of  $\omega_j$  in the coefficient  $g_j$  of Eq. (2.14). The  $\mathbf{A} \cdot \mathbf{A}$  term has been neglected while obtaining Eq. (2.8).

### III. REDUCED DENSITY OPERATOR EQUATION OF MOTION

Using the Markov approximation we employ the master-equation technique described by Louisell<sup>23</sup> to derive the equation of motion for the reduced density operator  $\hat{\rho}_T$  of the atom-field system comprised of  $N$  atoms and  $q$  field modes.  $\hat{\rho}_T$  is obtained from the density operator  $\hat{\rho}$  of the entire system by tracing over the thermal-field and collisional phase-damping reservoirs. The equation of motion in the Schrödinger picture is given by

$$\frac{\partial \hat{\rho}_T}{\partial t} = (i\hbar)^{-1} [\hat{H}_0 + \hat{H}_I, \hat{\rho}_T] + L_F(\hat{\rho}_T) + L_C(\hat{\rho}_T) , \quad (3.1)$$

where  $L_C(\hat{\rho})$  is the collisional damping term given by

$$L_C(\hat{\rho}) = - \sum_i \{ [\hat{n}_{di}, \hat{n}_{di} \hat{\rho}] + [\hat{\rho} \hat{n}_{di}, \hat{n}_{di}] \} \gamma_{ph} \quad (3.2)$$

and  $L_F(\hat{\rho})$  is the field damping term given by

$$L_F(\hat{\rho}) = \sum_{i,i'} L_{ii'}(\hat{\rho}) , \quad (3.3)$$

$$L_{ii'}(\hat{\rho}) = - \sum_{l,l'} \sum_{k,k'} \{ [\hat{V}_{li}^{(k)}, \hat{V}_{l'i'}^{(k')} \hat{\rho}] W_{ll'ii'}^{+kk'} + [\hat{\rho} \hat{V}_{l'i'}^{(k')}, \hat{V}_{li}^{(k)}] W_{l'lii'}^{-k'k} \} \times \delta(\omega^{(k)}, -\omega^{(k')}) . \quad (3.4)$$

In the above summations,  $(l, l') \in \{x, y, z\}$ ,  $(k, k') \in \{0, 1\}$ , and  $(i, i') \in \{1, 2, \dots, N\}$ . The collision-induced phase-damping rate is denoted by  $\gamma_{ph}$  and the  $\hat{V}$ 's,  $\omega$ 's, and  $W$ 's are defined by

$$\hat{V}_{li}^{(0)} \equiv \hat{V}_{li} , \quad (3.5)$$

$$\hat{V}_{li}^{(1)} \equiv \hat{V}_{li}^\dagger , \quad (3.6)$$

$$\omega^{(1)} \equiv \omega_a = -\omega^{(0)} , \quad (3.7)$$

$$W_{ll'ii'}^{+kk'} \equiv \int_0^{t_u} \exp(i\omega^{(k)}\tau) \langle \hat{F}_{li}^{(k)}(\tau) \hat{F}_{l'i'}^{(k')}(0) \rangle_R d\tau , \quad (3.8)$$

$$W_{l'lii'}^{-k'k} \equiv \int_0^{t_u} \exp(-i\omega^{(k')}\tau) \langle \hat{F}_{l'i'}^{(k')}(0) \hat{F}_{li}^{(k)}(\tau) \rangle_R d\tau , \quad (3.9)$$

$$\hat{F}_{li}^{(0)}(\tau) = \sum_s iC_{ls}^*(\mathbf{r}_i) \hat{a}_s^\dagger(\tau) , \quad (3.10)$$

$$\hat{F}_{li}^{(1)}(\tau) = - \sum_s iC_{ls}(\mathbf{r}_i) \hat{a}_s(\tau) , \quad (3.11)$$

where  $\hat{a}_s(\tau)$  is given by the free motion of the reservoir field mode

$$\hat{a}_s(\tau) = \hat{a}_s(0) \exp(-i\omega_s\tau) . \quad (3.12)$$

We see that the  $W$ 's are given by the two-time correlation functions of the thermal-field  $\langle \hat{F}_{li}^{(k)}(\tau) \hat{F}_{l'i'}^{(k')}(0) \rangle_R \equiv K_{ii'}$ , which depend upon phase factors of the form  $\exp[\pm i\mathbf{k}_s \cdot (\mathbf{r}_i - \mathbf{r}_{i'})]$ . As shown later, the spontaneous decay of the atoms is governed by the terms  $K_{ii}$  which are independent of  $\mathbf{r}_i$  and represent the temporal correlation of the thermal field at a fixed point in space. Let  $\tau_{th}$  be the characteristic time of this correlation. Then the Markov approximation<sup>23</sup> allows us to extend the upper limit of the integrals in Eqs. (3.8) and (3.9) to  $\infty$ , subject to the restriction that we look at a coarse-grained time  $\Delta t \gg \tau_{th}$ . This restriction is generally not too stringent because the relatively broad thermal spectrum usually makes  $\tau_{th}$  much smaller than the characteristic decay time of the atomic variables.

We shall see later that super-radiance occurs when the atoms are coherently pumped near resonance so that

$\hat{V}_i^\dagger \hat{V}_i$  has a spatial phase factor  $\exp[i\mathbf{k}_p \cdot (\mathbf{r}_{i'} - \mathbf{r}_i)]$ , where  $|\mathbf{k}_p|c \simeq \omega_a$ . As noted above,  $K_{ii'}$  also has a similar mode-dependent spatial phase factor. Therefore, when the sum over  $i$  and  $i'$  is carried out in Eq. (3.3), only a finite number of thermal-field modes in  $\hat{F}_{ii'}^{(k)}(\tau)$  contribute; those having wave vectors in the vicinity of  $\mathbf{k}_p$ , say  $\mathbf{k}_p \pm \Delta\mathbf{k}$ . This finite number of thermal-field modes have a spectral width  $\Delta\omega_{SR} \sim |\Delta\mathbf{k}|c$  or a characteristic correlation time  $\tau_{SR} \sim 1/\Delta\omega_{SR}$ . It will be shown that  $\tau_{SR}$  is determined by the length of the medium  $L_M$  to be  $\tau_{SR} = L_M/c$ , giving a much more stringent condition for the assumption of a coarse-grained time in the case of a thick medium.

The detailed derivation of the  $W$ 's is given in Appendix A. It is shown that under Markov approximation, when small frequency shifts due to the imaginary parts of the  $W$ 's are neglected, we get

$$W_{ii'}^{+10} = W_{i'i}^{-10*} = \gamma_{ii'}(\mathbf{R}_{ii'}) (n_{\text{th}} + 1), \quad (3.13)$$

$$W_{ii'}^{+01} = W_{i'i}^{-01*} = \gamma_{ii'}(\mathbf{R}_{ii'}) n_{\text{th}}, \quad (3.14)$$

$$\gamma_{ii'}(\mathbf{R}_{ii'}) = \int_{-\infty}^{\infty} \exp(i\mathbf{k} \cdot \mathbf{R}_{ii'}) \bar{\gamma}_{ii'}(\mathbf{k}) d\mathbf{k} / (2\pi)^3, \quad (3.15)$$

$$\begin{aligned} \bar{\gamma}_{ii'}(\mathbf{k}) &= \frac{3}{4} \lambda_a^2 \gamma \delta(\omega_a/c - |\mathbf{k}|) \\ &\times \sum_{\sigma=1,2} [\mathbf{e}_i \cdot \mathbf{e}_\sigma(\mathbf{k})][\mathbf{e}_{i'} \cdot \mathbf{e}_\sigma(\mathbf{k})], \end{aligned} \quad (3.16)$$

where  $n_{\text{th}}$  is the average number of thermal photons at  $\omega_a$ ,  $\mathbf{R}_{ii'} = \mathbf{r}_i - \mathbf{r}_{i'}$ ,  $\lambda_a = 2\pi c/\omega_a$ ,  $\mathbf{e}_\sigma(\mathbf{k})$  for  $\sigma=1,2$  are the two orthogonal field-polarization vectors for the mode with wave vector  $\mathbf{k}$ ,  $\gamma = 2\omega_a^3 |\mu_d|^2 / 3(4\pi\epsilon_0)\hbar c^3$  is the usual spontaneous-decay rate with  $\mu_d = \langle x | e\hat{x} | g \rangle$  being the dipole matrix element, and  $\epsilon_0$  is the free-space permittivity.

After summing over  $k, k'$  in Eq. (3.4), we can write  $L_F(\hat{\rho})$  as

$$\begin{aligned} L_F(\hat{\rho}) &= - \sum_{l,l'} \sum_i \{ [\hat{V}_{li}^\dagger, \hat{V}_{l'i} \hat{\rho}] (n_{\text{th}} + 1) \gamma_{ll'}(\mathbf{R}_{ii'}) \\ &\quad + [\hat{V}_{li}, \hat{V}_{l'i}^\dagger \hat{\rho}] n_{\text{th}} \gamma_{ll'}(\mathbf{R}_{ii'}) \} + \text{H.c.} \end{aligned} \quad (3.17)$$

In Appendix B we show that  $\gamma_{ll'}(\mathbf{0}) = \gamma \delta_{ll'}$ , where  $\delta_{ll'}$  is the Kronecker  $\delta$  function, which allows us to write Eq. (3.17) as a sum of two terms: a spontaneous-decay term  $L_{FS}(\hat{\rho})$  and a super-radiant decay term  $L_{FSR}(\hat{\rho})$ ,

$$L_F(\hat{\rho}) = L_{FS}(\hat{\rho}) + L_{FSR}(\hat{\rho}), \quad (3.18)$$

$$\begin{aligned} L_{FS}(\hat{\rho}) &= - \sum_l \sum_i \{ [\hat{V}_{li}^\dagger, \hat{V}_{li} \hat{\rho}] \gamma (n_{\text{th}} + 1) \\ &\quad + [\hat{V}_{li}, \hat{V}_{li}^\dagger \hat{\rho}] \gamma n_{\text{th}} \} + \text{H.c.}, \end{aligned} \quad (3.19)$$

$$\begin{aligned} L_{FSR}(\hat{\rho}) &= - \sum_{l,l'} \sum_{i' \neq i} \{ [\hat{V}_{li}^\dagger, \hat{V}_{l'i} \hat{\rho}] (n_{\text{th}} + 1) \gamma_{ll'}(\mathbf{R}_{ii'}) \\ &\quad + [\hat{V}_{li}, \hat{V}_{l'i}^\dagger \hat{\rho}] n_{\text{th}} \gamma_{ll'}(\mathbf{R}_{ii'}) \} + \text{H.c.} \end{aligned} \quad (3.20)$$

As mentioned in Appendix B,  $\gamma_{ll'}(\mathbf{R}_{ii'})$  is in general not  $\delta$  correlated in  $l$  and  $l'$ .

#### IV. $c$ -NUMBER FOKKER-PLANCK AND LANGEVIN EQUATIONS

The equation of motion for the reduced density operator  $\hat{\rho}_T$ , Eq. (3.1), can be transformed into an equivalent  $c$ -number equation for the associated distribution function  $\rho_c$  by using the standard technique described by Louisell.<sup>23</sup> This  $c$ -number transformation is not unique in that it depends upon the choice of ordering of the atom and field operators. Moreover, in order to derive a set of Langevin equations linear in the  $c$ -number variables, we have to define a  $c$ -number variable for each of the following operators:

$$\hat{V}_{li}^\dagger, \hat{V}_{li}, \hat{n}_{li}, \hat{n}_{gi}, \hat{V}_{li}^\dagger \hat{V}_{l'i'} (i \neq i'), \hat{a}_j^\dagger, \hat{a}_j. \quad (4.1)$$

However, in our preliminary investigation of the effect of super-radiance, we will not define  $c$ -number variables for  $\hat{V}_{li}^\dagger \hat{V}_{l'i'} (i \neq i')$ . The  $c$ -number variables for the other operators will be denoted by the same symbols without the carets, and with pluses in lieu of the daggers. The operator ordering we use is

$$\{ \hat{V}_{li}^\dagger \} \{ \hat{n}_{li} \} \{ \hat{n}_{gi} \} \{ \hat{V}_{li} \} \{ \hat{a}_j^\dagger \} \{ \hat{a}_j \}.$$

We note that the operators for different  $i$  and different  $j$  commute with each other and the operators for the atoms commute with those for the field at all times.

The equation of motion for  $\rho_c$  contains derivatives of infinite order in the  $\{n_{li}\}$  and  $\{n_{gi}\}$  variables. We approximate it by keeping only the first- and second-order derivatives. This approximation is usually justified with the use of collective atomic variables summed over a microscopic volume element with linear dimensions smaller than a wavelength and by assuming Gaussian fluctuations for them in the limit when the number of atoms in the microscopic volume element is large.<sup>12,22</sup> The assumption of a large number of atoms in the microscopic volume element may be overly restrictive.<sup>26</sup> However, since we are ultimately concerned only with the two-time correlations of the atomic variables, the third- and higher-order derivatives in the generalized Fokker-Planck equation do not matter,<sup>27</sup> just as the second-order derivatives are irrelevant when only the mean values of single atomic variables are of interest.<sup>28</sup>

Further discarding the  $n_{\text{th}}$ -proportional terms in Eqs. (3.18)–(3.20), which amounts to neglecting the thermal-noise-induced atomic decay effects, we obtain the following equation of motion for  $\rho_c$ :

$$\begin{aligned}
\frac{\partial \rho_c}{\partial t} = & \left\{ \sum_j i\omega_j \frac{\partial}{\partial a_j} a_j + \sum_l \sum_i i\omega_a \frac{\partial}{\partial V_{li}} V_{li} + \sum_j \sum_i C_{xj}^*(\mathbf{r}_i) \left[ -\frac{\partial}{\partial a_j} V_{xi} + \frac{\partial}{\partial V_{xi}^+} (n_{gi} - n_{xi}) a_j^+ - \left[ \frac{\partial}{\partial n_{gi}} - \frac{\partial}{\partial n_{xi}} \right] a_j^+ V_{xi} \right. \right. \\
& \left. \left. - \frac{1}{2} \left[ \frac{\partial^2}{\partial n_{gi}^2} + \frac{\partial^2}{\partial n_{xi}^2} - 2 \frac{\partial^2}{\partial n_{gi} \partial n_{xi}} \right] a_j^+ V_{xi} + \frac{\partial^2}{\partial V_{xi}^{+2}} V_{xi}^+ a_j^+ \right] \right. \\
& + \sum_l \sum_i \gamma_{ph} \left[ \frac{\partial^2}{\partial V_{li}^+ \partial V_{li}} n_{li} + \frac{\partial}{\partial V_{li}} V_{li} \right] \\
& + \sum_l \sum_i \gamma \left[ -\left[ \frac{\partial}{\partial n_{gi}} - \frac{\partial}{\partial n_{li}} \right] n_{li} + \frac{1}{2} \left[ \frac{\partial^2}{\partial n_{gi}^2} + \frac{\partial^2}{\partial n_{li}^2} - 2 \frac{\partial^2}{\partial n_{gi} \partial n_{li}} \right] n_{li} + \frac{\partial}{\partial V_{li}} V_{li} \right] \\
& + \sum_{l,l'} \sum_{i' \neq i} \sum_i \gamma_{ll'}(\mathbf{R}_{ii'}) \left[ -\left[ \frac{\partial}{\partial n_{gi}} - \frac{\partial}{\partial n_{li}} \right] V_{li}^+ V_{l'i'} - \frac{1}{2} \left[ \frac{\partial^2}{\partial n_{gi}^2} + \frac{\partial^2}{\partial n_{li}^2} - 2 \frac{\partial^2}{\partial n_{gi} \partial n_{li}} \right] V_{li}^+ V_{l'i'} \right. \\
& \left. + \frac{\partial}{\partial V_{li}} (n_{gi} - n_{li}) V_{l'i'} - \frac{\partial^2}{\partial V_{li}^2} V_{li} V_{l'i'} \right] + \text{c.c.} \left. \right\} \rho_c, \tag{4.2}
\end{aligned}$$

where c.c. denotes complex conjugation of the numerical coefficients plus the mutual interchange of  $V_{li}^+$  with  $V_{li}$  and  $a_j^+$  with  $a_j$ , respectively.

Essentially by reading off the coefficients in the Fokker-Planck equation, as described by Louisell,<sup>23</sup> we get the equivalent following set of Langevin equations:

$$\frac{\partial a_j}{\partial t} = -i\omega_j a_j + \sum_i C_{xj}^*(\mathbf{r}_i) V_{xi}, \tag{4.3}$$

$$\frac{\partial V_{xi}}{\partial t} = -i\omega_a V_{xi} - \sum_j C_{xj}(\mathbf{r}_i) a_j (n_{gi} - n_{xi}) - (\gamma + \gamma_{ph}) V_{xi} - \sum_{l'} \sum_{i' (\neq i)} \gamma_{xl'}(\mathbf{R}_{ii'}) (n_{gi} - n_{xi}) V_{l'i'} + f_{V_{xi}}, \tag{4.4}$$

$$\frac{\partial V_{xi}^+}{\partial t} = i\omega_a V_{xi}^+ - \sum_j C_{xj}^*(\mathbf{r}_i) a_j^+ (n_{gi} - n_{xi}) - (\gamma + \gamma_{ph}) V_{xi}^+ - \sum_{l'} \sum_{i' (\neq i)} \gamma_{xl'}(\mathbf{R}_{ii'}) (n_{gi} - n_{xi}) V_{l'i'}^+ + f_{V_{xi}^+}, \tag{4.5}$$

$$\frac{\partial n_{xi}}{\partial t} = - \sum_j [C_{xj}^*(\mathbf{r}_i) a_j^+ V_{xi} + C_{xj}(\mathbf{r}_i) a_j V_{xi}^+] - 2\gamma n_{xi} - \sum_{l'} \sum_{i' (\neq i)} \gamma_{xl'}(\mathbf{R}_{ii'}) (V_{xi}^+ V_{l'i'} + V_{l'i'}^+ V_{xi}) + f_{n_{xi}}, \tag{4.6}$$

$$n_{gi} + n_{xi} = 1, \tag{4.7}$$

where  $f_{V_x}$ ,  $f_{V_x^+}$ , and  $f_{n_x}$  denote the various Langevin forces with the following correlations:

$$\langle f_{V_{xi}^+}(t) f_{V_{xi'}(t')} \rangle = 2\gamma_{ph} n_{xi} \delta_{ii'} \delta(t-t'), \tag{4.8}$$

$$\langle f_{V_{xi}}(t) f_{V_{xi'}(t')} \rangle = \left[ \sum_j C_{xj}(\mathbf{r}_i) a_j V_{xi} - \sum_{l'} \sum_{i' (\neq i)} \gamma_{ll'}(\mathbf{R}_{ii'}) V_{xi} V_{l'i'} \right] \delta_{ii'} \delta(t-t'), \tag{4.9}$$

$$\langle f_{V_{xi}^+}(t) f_{V_{xi'}^+}(t') \rangle = \left[ \sum_j C_{xj}^*(\mathbf{r}_i) a_j^+ V_{xi}^+ - \sum_{l'} \sum_{i' (\neq i)} \gamma_{ll'}(\mathbf{R}_{ii'}) V_{xi}^+ V_{l'i'}^+ \right] \delta_{ii'} \delta(t-t'), \tag{4.10}$$

$$\begin{aligned}
\langle f_{n_{xi}}(t) f_{n_{xi'}(t')} \rangle &= \left[ - \sum_j [C_{xj}^*(\mathbf{r}_i) a_j^+ V_{xi} + C_{xj}(\mathbf{r}_i) a_j V_{xi}^+] + 2\gamma n_{xi} - \sum_{l'} \sum_{i' (\neq i)} \gamma_{ll'}(\mathbf{R}_{ii'}) [V_{xi}^+ V_{l'i'} + V_{l'i'}^+ V_{xi}] \right] \delta_{ii'} \delta(t-t') \\
&= \langle f_{n_{gi}}(t) f_{n_{gi'}(t')} \rangle = \langle f_{n_{gi}}(t) f_{n_{xi'}(t')} \rangle. \tag{4.11}
\end{aligned}$$

We note that the Fokker-Planck equation is obtained by commuting  $\hat{V}_{li}^\dagger$  and  $\hat{V}_{li}$  through the chosen ordering until they meet to become  $\hat{n}_{li}$  or  $\hat{n}_{gi}$  (see, e.g., Louisell,<sup>23</sup> p. 381). This is the origin of the  $2\gamma n_{xi}$  terms in Eqs. (4.6) and (4.11). Instead, if we commute  $\hat{V}_{li}^\dagger$  and  $\hat{V}_{li}$  to their own positions in the chosen ordering, we would obtain a term  $-2\gamma V_{xi}^+ V_{xi}$  in place of  $-2\gamma n_{xi}$  in Eq. (4.6) and a

term  $-2\gamma V_{xi}^+ V_{xi}$  in place of  $2\gamma n_{xi}$  in Eq. (4.11). This makes both Eqs. (4.6) and (4.11) look symmetric with respect to terms with  $i' \neq i$  and terms with  $i' = i$ . Also the sign of the  $2\gamma n_{xi}$  term in Eq. (4.11) is opposite to those of the  $i' \neq i$  terms due exactly to this reason. From Eqs. (4.8)–(4.11) we see that the correlations of the noise forces for different atoms, such as  $\langle f_{V_{xi}}(t) f_{V_{xi'}(t')} \rangle$  for  $i' \neq i$ , are

all zero. This is due to our assumption of a zero-temperature thermal-field reservoir and is not true if terms proportional to  $n_{th}$  are included in Eq. (4.2).

### V. EFFECT OF SUPER-RADIANCE

Our intent in this section is not to study super-radiance in detail but to obtain a qualitative understanding of the super-radiance terms in Eqs. (4.4)–(4.6), viz., those involving the sums  $\sum_{l'} \sum_{i' (\neq i)}$ . To study the free decay of the atomic variables in the absence of any mean field, let us consider the case in which all the  $q$  field modes are initially in the vacuum state, whereas the atomic polarization variables  $\{V_{xi}\}$  are uniformly excited in the form of a traveling wave propagating in the  $z$  direction.

Thus we assume that the expectation values of  $\hat{V}_{li}^\dagger(t)\hat{V}_{l'i'}(t)$  for  $i \neq i'$  are of the form

$$\langle \hat{V}_{li}^\dagger(t)\hat{V}_{l'i'}(t) \rangle = \mathcal{C}_{ll'}(t) \exp(-i\mathbf{k}_D \cdot \mathbf{R}_{ii'}), \quad (5.1)$$

where  $\mathbf{k}_D = k_D \mathbf{e}_z$ , and the amplitudes  $\{\mathcal{C}_{ll'}(t)\}$  are independent of  $i$  and  $i'$  but can, in general, be time dependent. This would be the case, for example, if the atoms

were pumped initially by a coherent wave with wave vector  $\mathbf{k}_D$  traveling in the  $z$  direction. Taking expectation values on both sides of Eq. (4.6), we get

$$\frac{\partial \langle n_{xi} \rangle}{\partial t} = -2\gamma \langle n_{xi} \rangle - T_i^{-1}, \quad (5.2)$$

where

$$T_i^{-1} = \sum_{l'} \sum_{i' (\neq i)} \gamma_{xl'}(\mathbf{R}_{ii'}) (\langle V_{xi}^+ V_{l'i'} \rangle + \langle V_{l'i'}^+ V_{xi} \rangle) \quad (5.3)$$

is a collective spontaneous-decay rate. Note that Eq. (5.2) describes free decay of the  $i$ th atom without the pump field. Therefore, the steady state is reached when  $\langle n_{xi} \rangle$  decays to zero, at which time the atomic polarizations  $\{\langle V_{xi} \rangle\}$  are also zero causing  $T_i^{-1}$  to vanish.

We assume that the atoms are uniformly distributed with  $N/V_M$  atoms per unit volume and that the medium is infinite in the  $x$  and  $y$  directions but finite in the  $z$  direction from  $z = -L_M/2$  to  $z = L_M/2$ . Then, as shown in Appendix C, under the approximation that  $|\omega_a/c - k_D| \ll \omega_a/c$ , which is also the condition for rotating-wave approximation, we obtain the following expression for  $T_i^{-1}$ :

$$T_i^{-1} \simeq (N/V_M) (\mathcal{C}_{xx}/2\pi) (3\lambda_a^2 \gamma L_M/2) \exp[i(\omega_a/c - k_D)z_i] \times \left[ \frac{\exp[i(\omega_a/c - k_D)L_M/2] - \exp[-i(\omega_a/c - k_D)L_M/2]}{i(\omega_a/c - k_D)L_M} \right] - 2\gamma \mathcal{C}_{xx}, \quad (5.4)$$

where  $z_i$  is the  $z$  coordinate of the  $i$ th atom. On resonance,  $k_D = \omega_a/c$  prevails, and the above equation reduces to

$$T_i^{-1} = [(N/V_M)(3/8\pi)\lambda_a^2 L_M - 1] 2\gamma \mathcal{C}_{xx}. \quad (5.5)$$

When the number of atoms  $N_D$  in a diffraction volume  $\lambda_a^2 L_M$  is large, i.e.,  $N_D = N\lambda_a^2 L_M/V_M \gg 1$ , the first term of Eq. (5.5) dominates the collective decay rate  $T_i^{-1}$ . A classical reason for the enhancement of the decay rate is reviewed in Appendix D. In fact, in the above limit,  $T_i^{-1}$  agrees with  $T_R^{-1}$  of Eq. (D5) if we identify  $\mathcal{C}_{xx} = |V_x^{\text{cl}}|^2$ . Therefore, we rewrite Eq. (5.5) as

$$T_i^{-1} = T_R^{-1} - 2\gamma \mathcal{C}_{xx}. \quad (5.6)$$

We note that the extra decay rate accompanies extra noise because  $T_i^{-1}$ -like terms [cf. Eq. (5.3)] also appear in the correlations  $\langle f_{n_{xi}}(t)f_{n_{xi'}}(t') \rangle$ ,  $\langle f_{n_{gi}}(t)f_{n_{gi'}}(t') \rangle$ , and  $\langle f_{n_{gi}}(t)f_{n_{xi'}}(t') \rangle$  as given by Eq. (4.11). The double sum  $\sum_{l'} \sum_{i' (\neq i)}$  terms in  $\langle f_{V_{xi}}(t)f_{V_{xi'}}(t') \rangle$  and  $\langle f_{V_{xi}^+}(t)f_{V_{xi'}^+}(t') \rangle$  of Eqs. (4.9) and (4.10), respectively, turn out to be negligible with our assumptions because they are proportional to  $V_{xi}V_{l'i'}$  and  $V_{xi}^+V_{l'i'}^+$ , respectively, instead of  $V_{xi}^+V_{l'i'}$ .

From Eqs. (5.4) and (5.6), we see that  $T_R^{-1}$  is negligible either when the atoms are pumped far from resonance so that  $|\omega_a/c - k_D| \gg N_D/L_M$ , or when  $N_D$  is small. In

the former case  $\mathcal{C}_{xx}$  is expected to be negligible, implying that  $T_i^{-1} \simeq 0$ , whereas in the latter case  $T_i^{-1} \simeq -2\gamma \mathcal{C}_{xx}$ , giving an anomalous gain correction to the decay rate of  $\langle n_{xi} \rangle$ . This anomalous gain term is negligible compared with the spontaneous-decay term if  $\mathcal{C}_{xx} \ll \langle n_{xi} \rangle$ , which is expected to be so only when the pump intensity is larger than the saturation intensity of the atoms.<sup>29</sup>

In order to obtain further insight into the validity of the above treatment and to compare with other treatments of super-radiance, let us examine from an operator-equation perspective how the Markov approximation leads to the various decay terms in the Langevin equations.

Using the Heisenberg equation of motion, we can derive the following set of operator equations for the atom-field system governed by the system Hamiltonian of Eqs. (2.6)–(2.12):

$$\frac{\partial \hat{a}_j}{\partial t} = -i\omega_j \hat{a}_j + \sum_i C_{xj}^*(\mathbf{r}_i) \hat{V}_{xi}, \quad (5.7)$$

$$\frac{\partial \hat{V}_{xi}}{\partial t} = -i\omega_a \hat{V}_{xi} - \sum_j C_{xj}(\mathbf{r}_i) \hat{a}_j (\hat{n}_{gi} - \hat{n}_{xi}), \quad (5.8)$$

$$\frac{\partial \hat{n}_{xi}}{\partial t} = - \sum_j [C_{xj}^*(\mathbf{r}_i) \hat{a}_j^\dagger \hat{V}_{xi} + C_{xj}(\mathbf{r}_i) \hat{a}_j \hat{V}_{xi}^\dagger], \quad (5.9)$$

where  $\sum_j$  denotes the sum over all the field modes, i.e.,

$j \in \{1, 2, \dots, \infty\}$ . In quantum statistical treatments of atom-field interactions, there are several equivalent approaches, e.g., the density matrix method and the Langevin noise operator method.<sup>30</sup> In all of these approaches, one starts by solving for the time evolution of the reservoir modes  $\{\hat{a}_s\}$ ,  $s \in \{q+1, q+2, \dots, \infty\}$ , in terms of the atomic operators and the initial conditions of the reservoir modes. Then by assuming that the reservoir modes are initially in thermal equilibrium with the environment, one traces away the reservoir modes. For example, in the Langevin noise operator method,  $\hat{a}_s$  is obtained first by formally integrating Eq. (5.7), then, after substituting the solution into Eqs. (5.8) and (5.9), it is subsequently traced away. In the end, with the Markov or the Wigner-Weisskopf approximations, this method then converts the  $j$  sum in Eqs. (5.8) and (5.9) into decay and noise terms. In principle, this conversion is correct only if all the electromagnetic field modes are taken to be reservoir modes, i.e.,  $s \in \{1, 2, \dots, \infty\}$  leaving no field terms in Eqs. (5.8) and (5.9). However, if we want to study the time evolution of some  $q$  field modes explicitly or if the  $q$  field modes are strongly excited initially instead of being in thermal equilibrium, then we must separate them out from the tracing procedure. For the ordinary spontaneous-decay and noise terms, the separation of a small subset of modes has little effect on the value of the decay constant  $\gamma$ . For super-radiant decay, however, the situation is completely different. From the above treatment and the justification of Markov approximation given in Sec. III we see that only a small finite subset of the field modes with  $\mathbf{k} \simeq \mathbf{k}_p \pm \Delta\mathbf{k}$ , where  $|\Delta\mathbf{k}| = 2\pi/L_M$ , is involved in determining the super-radiant decay and noise terms. In fact, for the case in which the quantization volume  $V_Q$  is equal to the medium volume  $V_M$ , there is only one mode contributing to these terms. Thus, we cannot blindly separate the  $q$  field modes without proper justification.

Our treatment here is justifiable only if  $V_M \ll V_Q$ , so that a large number of modes are involved and only an insignificant fraction is separated out. Even so, as discussed in Sec. III, the validity of the Markov approximation then requires that we only look at a coarse-grained time with  $\Delta t \gg L_M/c$ .

Our treatment of super-radiance can be compared with that of a super-radiant laser given by Bonifacio *et al.*<sup>31</sup> using the laser master-equation approach. In their treatment,  $V_M = V_Q$  and the super-radiance term is, in fact, due only to one mode. Their equation can be obtained from our Eqs. (4.3)–(4.7) by setting  $V_M = V_Q$  and dropping the super-radiance decay terms, since the single mode which is responsible for them is explicitly solved for. However, the condition  $V_M = V_Q$  is equivalent to assuming an infinite medium because of the periodic boundary condition. Hence, in their analysis, they had to assume that this single mode decayed in time of order  $L_M/c$  in order to correctly model the radiation out from the medium of finite length. They did so by adding an extra decay term to the field equation, our Eq. (4.3). The inclusion of a rapid field-decay time then allowed them to assume that the field correlation had no memory over the coarse-grained time  $\Delta t \gg L_M/c$ , thus justifying the Mar-

kov approximation. In addition, it also allowed them to solve adiabatically for the field in terms of the atomic variables.

We are, however, more interested in the  $V_Q \gg V_M$  case where the radiation from the finite length of the medium is properly taken care of by spatial propagation through the medium using a multimode treatment. There is thus no need to assume rapidly decaying field modes. The coarse-grained time ( $\Delta t \gg L_M/c$ ) requirement now arises from the *finite number* of modes contributing to super-radiant decay.

## VI. SOLUTION FOR THE ATOMIC POLARIZATION

In this section we solve for the atomic polarization  $V_i(t)$  after neglecting the super-radiance terms in Eqs. (4.3)–(4.11). As pointed out in Sec. V, when the intensity of at least one of the field modes (usually the pump mode) with wave vector  $\mathbf{k}_p$  is larger than the saturation intensity of the atomic medium, the super-radiance terms can be shown to be negligible in either of the following two limits: (a) when the number of atoms in a diffraction volume  $N_D = \lambda_p^2 L_M N / V_M \ll 1$ , where  $\lambda_p \equiv 2\pi/|\mathbf{k}_p|$  and  $L_M$  is the length of the medium; and (b) when the atoms are pumped far from resonance, i.e.,  $|(\mathbf{k}_p - \omega_a/c)| L_M \gg N_D$ . With this approximation, the Langevin equations (4.3)–(4.6) reduce to

$$\frac{\partial a_j}{\partial t} = -i\omega_j a_j + \sum_i C_j^*(\mathbf{r}_i) V_i, \quad (6.1)$$

$$\frac{\partial V_i}{\partial t} = -i\omega_a V_i - a(\mathbf{r}_i, t)(n_{gi} - n_i) - \gamma_{\perp} V_i + f_{V_i}, \quad (6.2)$$

$$\frac{\partial V_i^+}{\partial t} = i\omega_a V_i^+ - a^+(\mathbf{r}_i, t)(n_{gi} - n_i) - \gamma_{\perp} V_i^+ + f_{V_i^+}, \quad (6.3)$$

$$\frac{\partial n_i}{\partial t} = -[a^+(\mathbf{r}_i, t)V_i + a(\mathbf{r}_i, t)V_i^+] - \gamma_{\parallel} n_i + f_{n_i}, \quad (6.4)$$

where  $n_{gi} + n_i = 1$ , we have omitted the subscript  $x$  from all the variables,  $\gamma_{\perp} \equiv \gamma + \gamma_{\text{ph}}$  is the transverse relaxation rate,  $\gamma_{\parallel} \equiv 2\gamma$  is the longitudinal relaxation rate, and the multimode field variables  $a(\mathbf{r}_i, t)$  and  $a^+(\mathbf{r}_i, t)$  are given by

$$a(\mathbf{r}_i, t) \equiv \sum_j C_j(\mathbf{r}_i) a_j(t), \quad (6.5)$$

$$a^+(\mathbf{r}_i, t) \equiv \sum_j C_j^*(\mathbf{r}_i) a_j^+(t). \quad (6.6)$$

The nonzero correlations of the Langevin forces become

$$\langle f_{V_i^+}(t) f_{V_i}(t') \rangle = 2\gamma_{\text{ph}} n_i \delta(t - t'), \quad (6.7)$$

$$\langle f_{V_i}(t) f_{V_i}(t') \rangle = a(\mathbf{r}_i, t) V_i \delta(t - t'), \quad (6.8)$$

$$\langle f_{V_i^+}(t) f_{V_i^+}(t') \rangle = a^+(\mathbf{r}_i, t) V_i^+ \delta(t - t'), \quad (6.9)$$

$$\langle f_{n_i}(t) f_{n_i}(t') \rangle = \{-[a^+(\mathbf{r}_i, t) V_i + a(\mathbf{r}_i, t) V_i^+] + \gamma_{\parallel} n_i\} \delta(t - t'), \quad (6.10)$$



$$\begin{aligned} \langle f_{n_{gi}}(t)f_{n_{gi}}(t') \rangle &= -\langle f_{n_{gi}}(t)f_{n_i}(t') \rangle \\ &= \langle f_{n_i}(t)f_{n_i}(t') \rangle . \end{aligned} \quad (6.11)$$

From here on in this section, we drop the subscript  $i$  from all the atomic variables to simplify the notation. To solve the Langevin equations (6.1)–(6.4) we express the temporal variations of  $a(\mathbf{r}, t)$  and  $a^+(\mathbf{r}, t)$  in terms of a pair of Fourier series:

$$a(\mathbf{r}, t) \equiv \sum_m A_m(\mathbf{r}) \exp(-i\nu_m t) , \quad (6.12)$$

$$a^+(\mathbf{r}, t) \equiv \sum_m A_m^+(\mathbf{r}) \exp(i\nu_m t) , \quad (6.13)$$

where  $m \in \{1, 2, \dots, \infty\}$  denotes the sum over an infinite number of Fourier coefficients  $A_m$  and  $A_m^+$ , respectively, and  $\nu_m \equiv 2\pi m/T$  with period  $T$  which can be chosen arbitrarily long so that it does not cause any undesirable periodicity within the observation time. These series expansions are made only for calculational convenience and should be distinguished from the expansion of  $a(\mathbf{r}, t)$  in terms of the  $q$  modes  $\{a_j(t)\}$  as in Eqs. (6.5) and (6.6). The relation between these two expansions will become clear in the following paper of this series.

The atomic polarization variable  $V_i(t)$  can be solved in terms of  $\{A_m\}$  using Eqs. (6.2)–(6.4) by the Fourier transform technique because we are only interested in the steady-state response. We define the Fourier transform of any variable  $X(t)$  by

$$X(\omega) \equiv \int_{-\infty}^{\infty} (dt/2\pi) X(t) \exp(i\omega t) , \quad (6.14)$$

where the transformed variable is denoted by the same symbol with  $\omega$  as the argument. Then we have, e.g.,

$$a(\omega) = \sum_m A_m \delta(\omega - \nu_m) , \quad (6.15)$$

$$a^+(\omega) = \sum_m A_m^+ \delta(\omega + \nu_m) . \quad (6.16)$$

The Langevin equations (6.2)–(6.4) are then transformed to

$$\begin{aligned} -i\omega V(\omega) &= -\sum_m A_m [\delta(\omega - \nu_m) - 2n(\omega - \nu_m)] \\ &\quad -(\gamma_{\perp} + i\omega_a)V(\omega) + f_V(\omega) , \end{aligned} \quad (6.17)$$

$$\begin{aligned} -i\omega V^+(\omega) &= -\sum_m A_m^+ [\delta(\omega + \nu_m) - 2n(\omega + \nu_m)] \\ &\quad -(\gamma_{\perp} - i\omega_a)V^+(\omega) + f_{V^+}(\omega) , \end{aligned} \quad (6.18)$$

$$\begin{aligned} -i\omega n(\omega) &= -\sum_m [A_m^+ V(\omega + \nu_m) + A_m V^+(\omega - \nu_m)] \\ &\quad -2F\gamma_{\perp} n(\omega) + f_n(\omega) , \end{aligned} \quad (6.19)$$

where we have used  $n_g = 1 - n$  to eliminate  $n_g(\omega)$ , and

defined a parameter  $F$  by

$$\gamma_{\parallel} = 2F\gamma_{\perp} , \quad (6.20)$$

so that  $\gamma_{\text{ph}} = \gamma_{\perp}(1 - F)$  for  $0 \leq F \leq 1$ . In the absence of collisions when  $\gamma_{\text{ph}} = 0$ ,  $F = 1$ . We introduce the following shorthand notation:

$$X_{\pm k \pm l \pm m \pm n} \equiv X(\omega \pm \nu_k \pm \nu_l \pm \nu_m \pm \nu_n) , \quad (6.21)$$

$$X_0 \equiv X(\omega) , \quad (6.22)$$

where  $X \in \{V, V^+, n, f_V, f_{V^+}, f_n, \delta\}$ , and  $k, l, n$  are dummy indices just like  $m$ . Using Einstein's summation convention, Eqs. (6.17)–(6.19) can be rewritten as

$$\begin{aligned} -i\omega V_0 &= -A_m(\delta_{-m} - 2n_{-m}) - (\gamma_{\perp} + i\omega_a)V_0 + f_{V_0} , \\ &\quad (6.23) \end{aligned}$$

$$\begin{aligned} -i\omega V_0^+ &= -A_m^+(\delta_m - 2n_m) - (\gamma_{\perp} - i\omega_a)V_0^+ + f_{V_0^+} , \\ &\quad (6.24) \end{aligned}$$

$$-i\omega n_0 = -A_m^+ V_m - A_m V_{-m}^+ - 2\gamma_{\parallel} n_0 + f_{n_0} . \quad (6.25)$$

A straightforward algebraic manipulation which eliminates the  $V$  and  $V^+$  variables from the above equations leads to the following recursion formula for  $n(\omega)$ :

$$\begin{aligned} -i\omega n_0 &= [A_l^+ A_m(\delta_{l-m} - 2n_{l-m})]/D_l^- \\ &\quad + [A_l A_m^+(\delta_{-l+m} - 2n_{-l+m})]/D_{-l}^+ - 2F\gamma_{\perp} n_0 \\ &\quad + f_{n_0} - A_l^+ f_{V_l}/D_l^- - A_l f_{V_{-l}^+}/D_{-l}^+ , \end{aligned} \quad (6.26)$$

where

$$D_{\pm l}^{\pm} \equiv D^{\pm}(\omega \pm \nu_l) , \quad (6.27)$$

$$D^+(\omega) \equiv -i(\omega + \omega_a) + \gamma_{\perp} , \quad (6.28)$$

$$D^-(\omega) \equiv -i(\omega - \omega_a) + \gamma_{\perp} , \quad (6.29)$$

and the repeated triple indices in Eq. (6.26) are to be summed over also.

In order to bring all the  $n_0$  terms in Eq. (6.26) to the left-hand side, we rewrite the double sum over  $l$  and  $m$  in the first two terms, for example, as

$$\begin{aligned} \sum_{l,m} A_l^+ A_m n_{l-m}/D_l^- &= \sum_k A_k^+ A_k n_0/D_k^- \\ &\quad + \sum_l \sum_{m (\neq l)} A_l^+ A_m n_{l-m}/D_l^- , \end{aligned} \quad (6.30)$$

where we have separated the  $l=m$  terms. Using the short form

$$\sum_l \sum_{m (\neq l)} A_l^+ A_m n_{l-m}/D_l^- \equiv A_l^+ A_{l''} n_{l-l''}/D_l^- \quad (6.31)$$

for the double sum, where the double prime denotes sum over  $l''$  with  $l'' \neq l$ , Eq. (6.26) can be written as

$$\begin{aligned}
 n_0 = & \frac{A_k^+ A_k \delta_0}{\pi(\omega)} \left[ \frac{1}{D_k^-} + \frac{1}{D_{-k}^+} \right] \\
 & + \frac{A_l^+ A_{l'} \delta_{l-l'}}{\pi(\omega)} \left[ \frac{1}{D_l^-} + \frac{1}{D_{-l'}^+} \right] \\
 & - \frac{2A_l^+ A_{l'} n_{l-l'}}{\pi(\omega)} \left[ \frac{1}{D_l^-} + \frac{1}{D_{-l'}^+} \right] \\
 & - \frac{A_l^+ f_{V_l}}{\pi(\omega) D_l^-} - \frac{A_l f_{V_{-l}^+}}{\pi(\omega) D_{-l}^+} + \frac{f_{n_0}}{\pi(\omega)}, \tag{6.32}
 \end{aligned}$$

where

$$\pi(\omega) = \left[ -i\omega + 2F\gamma_1 + 2A_k^+ A_k \left[ \frac{1}{D_k^-} + \frac{1}{D_{-k}^+} \right] \right]. \tag{6.33}$$

As in Eqs. (6.21) and (6.22), in the following we abbreviate:

$$\pi_0 \equiv \pi(\omega), \tag{6.34}$$

$$\pi_{\pm k} \equiv \pi(\omega \pm \nu_k). \tag{6.35}$$

Equation (6.32) can then be iterated to yield a power-series solution for  $n_0$  provided its convergence can be established. For example, the first iteration which neglects the third term in Eq. (6.32) gives the following solution for  $n_0$  up to second order in the Fourier amplitudes  $\{A_k\}$ :

$$\begin{aligned}
 n_0 = & \frac{A_k^+ A_k \delta_0}{\pi_0} \left[ \frac{1}{D_k^-} + \frac{1}{D_{-k}^+} \right] \\
 & + \frac{A_m^+ A_{m'} \delta_{m-m'}}{\pi_0} \left[ \frac{1}{D_m^-} + \frac{1}{D_{-m'}^+} \right] \\
 & - \frac{A_k^+ f_{V_k}}{\pi_0 D_k^-} - \frac{A_k f_{V_{-k}^+}}{\pi_0 D_{-k}^+} + \frac{f_{n_0}}{\pi_0}. \tag{6.36}
 \end{aligned}$$

From the above equation we obtain  $n_{l-l'}$ , which can be inserted back into Eq. (6.32) to give the fourth-order solution. Similarly, after one more iteration and some algebraic simplification we obtain the following solution for  $n_0$  up to sixth order in the Fourier amplitudes  $\{A_k\}$ :

$$\begin{aligned}
 n_0 = & \frac{A_k^+ A_k}{\pi_0} \left[ \frac{1}{D_k^-} + \frac{1}{D_{-k}^+} \right] \delta_0 + \frac{A_l^+ A_{l'}}{\pi_0} \left[ \frac{1}{D_l^-} + \frac{1}{D_{-l'}^+} \right] \frac{N_{l-l'}}{\pi_{l-l'}} \delta_{l-l'} \\
 & - \frac{2A_l^+ A_{l'}}{\pi_0} \left[ \frac{1}{D_l^-} + \frac{1}{D_{-l'}^+} \right] \frac{A_m^+ A_{m'}}{\pi_{l-l'}} \left[ \frac{1}{D_{l-l'+m}^-} + \frac{1}{D_{l'-l-m}^+} \right] \frac{N_{l-l'+m-m'}}{\pi_{l-l'+m-m'}} \delta_{l-l'+m-m'} \\
 & + \frac{2A_l^+ A_{l'}}{\pi_0} \left[ \frac{1}{D_l^-} + \frac{1}{D_{-l'}^+} \right] \frac{2A_m^+ A_{m'}}{\pi_{l-l'}} \left[ \frac{1}{D_{l-l'+m}^-} + \frac{1}{D_{l'-l-m}^+} \right] \frac{A_n^+ A_{n'}}{\pi_{l-l'+m-m'}} \\
 & \times \left[ \frac{1}{D_{l-l'+m-m'+n}^-} + \frac{1}{D_{l'-l-m-n}^+} \right] \frac{N_{l-l'+m-m'+n-n'}}{\pi_{l-l'+m-m'+n-n'}} \delta_{l-l'+m-m'+n-n'} + \Gamma_{n_0}, \tag{6.37}
 \end{aligned}$$

where

$$N_{\pm l \pm l' \pm m \pm m' \pm n \pm n'} \equiv N(\omega \pm \nu_l \pm \nu_{l'} \pm \nu_m \pm \nu_{m'} \pm \nu_n \pm \nu_{n'}), \tag{6.38}$$

$$N(\omega) \equiv 2F\gamma_1 - i\omega, \tag{6.39}$$

and the noise term is given by

$$\begin{aligned}
 \Gamma_{n_0} = & -\frac{A_k^+ f_{V_k}}{\pi_0 D_k^-} - \frac{A_k f_{V_{-k}^+}}{\pi_0 D_{-k}^+} + \frac{f_{n_0}}{\pi_0} - \frac{2A_l^+ A_{l'}}{\pi_0} \left[ \frac{1}{D_l^-} + \frac{1}{D_{-l'}^+} \right] \left[ -\frac{A_k^+ f_{V_{l-l'+k}}}{\pi_{l-l'} D_{l-l'+k}^-} - \frac{A_k f_{V_{l-l'-k}^+}}{\pi_{l-l'} D_{l-l'-k}^+} + \frac{f_{n_{l-l'}}}{\pi_{l-l'}} \right] \\
 & + \frac{2A_l^+ A_{l'}}{\pi_0} \left[ \frac{1}{D_l^-} + \frac{1}{D_{-l'}^+} \right] \frac{2A_m^+ A_{m'}}{\pi_{l-l'}} \left[ \frac{1}{D_{l-l'+m}^-} + \frac{1}{D_{l'-l-m}^+} \right] \\
 & \times \left[ -\frac{A_k^+ f_{V_{l-l'+m-m'+k}}}{\pi_{l-l'+m-m'} D_{l-l'+m-m'+k}^-} - \frac{A_k f_{V_{l-l'+m-m'-k}^+}}{\pi_{l-l'+m-m'} D_{l-l'+m-m'-k}^+} + \frac{f_{n_{l-l'+m-m'}}}{\pi_{l-l'+m-m'}} \right]. \tag{6.40}
 \end{aligned}$$

From  $n_0$  we can obtain  $n_{-m}$ , which when substituted into Eq. (6.23) yields the following solution for  $V_0$ :

$$V_0 = -\frac{A_m \delta_{-m}}{D_0^-} + \frac{2A_m}{D_0^-} \frac{A_k^+ A_k \delta_{-m}}{\pi_{-m}} \left[ \frac{1}{D_{k-m}^-} + \frac{1}{D_{-k-m}^+} \right] \delta_{-m}$$

$$\begin{aligned}
& + \frac{2A_m}{D_0^-} \frac{A_l^+ A_{l''}}{\pi_{-m}} \left[ \frac{1}{D_{l-m}^-} + \frac{1}{D_{-l''-m}^+} \right] \frac{N_{l-l''-m}}{\pi_{l-l''-m}} \delta_{l-l''-m} \\
& - \frac{2A_m}{D_0^-} \frac{A_l^+ A_{l''}}{\pi_{-m}} \left[ \frac{1}{D_{l-m}^-} + \frac{1}{D_{-l''-m}^+} \right] \frac{A_n^+ A_{n''}}{\pi_{l-l''-m}} \left[ \frac{1}{D_{l-l''+n-m}^-} + \frac{1}{D_{l-l''-n''-m}^+} \right] \\
& \times \frac{N_{l-l''+n-n''-m}}{\pi_{l-l''+n-n''-m}} \delta_{l-l''+n-n''-m} + \Gamma_V(\omega), \tag{6.41}
\end{aligned}$$

where

$$\Gamma_V(\omega) = -\frac{2A_m}{D_0^-} \frac{A_l^+ f_{V_{l-m}}}{\pi_{-m} D_{l-m}^-} - \frac{2A_m}{D_0^-} \frac{A_l f_{V_{-l-m}^+}}{\pi_{-m} D_{-l-m}^+} + \frac{2A_m}{D_0^-} \frac{f_{n-m}}{\pi_{-m}} + \frac{f_{V_0}}{D_0^-}, \tag{6.42}$$

and we have truncated the deterministic part after the fifth-order terms and the noise part after the second-order terms in the Fourier amplitudes  $\{A_k\}$ .

Finally, the following solution for  $V(t)$  is obtained by inverse Fourier transforming  $V_0 = V(\omega)$ :

$$\begin{aligned}
V(t) = & -\frac{(2F\gamma_1)A_m \exp(-i\nu_m t)}{D^-(\nu_m)\pi(0)} + \frac{(2F\gamma_1)2A_m \exp[i(\nu_l - \nu_{l''} - \nu_m)t] A_l^+ A_{l''}}{D^-(-\nu_l + \nu_{l''} + \nu_m)\pi(0)\pi(-\nu_l + \nu_{l''})} \left[ \frac{1}{D^-(\nu_{l''})} + \frac{1}{D^+(-\nu_l)} \right] \\
& - \frac{(2F\gamma_1)2A_m \exp[i(\nu_l - \nu_{l''} + \nu_n - \nu_{n''} - \nu_m)t] A_l^+ A_{l''}}{D^-(-\nu_l + \nu_{l''} - \nu_n + \nu_{n''} + \nu_m)\pi(0)\pi(-\nu_l + \nu_{l''})} \left[ \frac{1}{D^-(\nu_{l''})} + \frac{1}{D^+(-\nu_l)} \right] \frac{2A_n^+ A_{n''}}{\pi(-\nu_n + \nu_{n''} - \nu_l + \nu_{l''})} \\
& \times \left[ \frac{1}{D^-(\nu_{n''} - \nu_l + \nu_{l''})} + \frac{1}{D^+(-\nu_n - \nu_l + \nu_{l''})} \right] + \Gamma_V(t), \tag{6.43}
\end{aligned}$$

$$V(t) \equiv V_{\text{Det}}(t) + \Gamma_V(t), \tag{6.44}$$

where

$$\Gamma_V(t) = \int_{-\infty}^{\infty} d\omega \Gamma_V(\omega) \exp(-i\omega t). \tag{6.45}$$

A complete series solution for  $V(t)$ , expanded to all orders, can easily be derived using the above procedure and is given in Appendix E.

Now a few words about the convergence of the above series solutions are in order. The expansion for  $n_0$  in Eq. (6.37) does not appear to converge when the magnitudes of all  $\{A_k\}$  are large. For example, the numerator of the third term in Eq. (6.37) has a double sum

$$\sum_{m'' (\neq m)} \sum_m A_m^+ A_{m''} \left[ \frac{1}{D_{l-l''+m}^-} + \frac{1}{D_{l-l''-m''}^+} \right], \tag{6.46}$$

while the denominator has a single sum

$$\pi_{l-l''} \sim \sum_k 2A_k^+ A_k \left[ \frac{1}{D_{l-l''+k}^-} + \frac{1}{D_{l-l''-k}^+} \right], \tag{6.47}$$

making the former greater. Therefore, the higher-order terms could be progressively larger. If the magnitudes of  $m$  Fourier amplitudes are large, then the  $n$ th-order term in the series is  $(m-1)$  times greater than the  $(n-1)$ th-order term for  $n > 2$ . The divergence in the higher-order terms seems to remain even after mutual cancellation of some of the terms in Eq. (6.37). However, it is clear that the series converges for the case when  $m=1$  or when only one of the Fourier amplitudes is arbitrarily large.

We note that it may be possible to put  $V_{\text{Det}}(t)$ , the

deterministic part of  $V(t)$ , in a closed form. For example, the following expression for  $V_{\text{Det}}(t)$  gives a correct expansion up to fifth order [Eq. (6.43)] in the Fourier amplitudes  $\{A_k\}$ :

$$V_{\text{Det}}(t) = -\frac{(2\gamma_1 F)A_m \exp(-i\nu_m t)}{D^-(\nu_m)\Pi_m(\nu_m, t)}, \tag{6.48}$$

where  $\Pi_m(\omega, t)$  is defined recursively as

$$\begin{aligned}
\Pi_m(\omega, t) \equiv & 2\gamma_1 F - i(\omega - \nu_m) \\
& + \frac{2A_l^+ A_n \exp[i(\nu_l - \nu_n)t] \Pi_m(\omega, t) D^-(\omega)}{D^-(-\nu_l + \nu_m + \omega)\Pi_m(-\nu_l + \nu_n + \omega, t)} \\
& \times \left[ \frac{1}{D^-(\nu_n + \omega - \nu_m)} \right. \\
& \left. + \frac{1}{D^+(-\nu_l + \omega - \nu_m)} \right]. \tag{6.49}
\end{aligned}$$

This closed-form solution involving unrestricted sums over  $\{A_k\}$  will be useful for justifying the slowly varying amplitude approximation in the following paper of this series. It will be interesting if a closed-form solution can be obtained which gives a correct expansion for  $V_{\text{Det}}(t)$  up to all orders. Then analytic continuation implies that this solution will be valid for arbitrarily large values of the Fourier amplitudes  $\{A_k\}$  and the convergence of the

solutions will always be guaranteed.

In conclusion, we have presented a quantum theory of nondegenerate multiwave mixing applicable to traveling-wave interaction geometries. We have also carried out a preliminary analysis of the effect of super-radiance and identified regions where such effects are negligible. Neglecting the super-radiance terms, we have presented a general Fourier expansion method to solve for the atomic polarization and fluctuation in the presence of any number of field modes. In subsequent papers of this series, we will develop a frequency domain technique for adiabatic elimination of the atomic variables and then present methods to treat, with some rigor, the spatial propagation of an interacting multimode field for application to traveling-wave squeezed-state generation experiments.

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#### APPENDIX A

In this appendix we find an expression for  $W_{ll'}^{\pm kk'}$  in terms of a Fourier coefficient. Let the density operator of the thermal-field reservoir be  $\hat{\rho}_R$ , which at temperature  $T$  is given by

$$\hat{\rho}_R = \exp(-\hat{R}/k_B T) / \text{Tr}_R [\exp(-\hat{R}/k_B T)], \quad (\text{A1})$$

where  $\text{Tr}_R$  denotes trace over the reservoir operators and  $k_B$  is the Boltzmann constant. The reservoir averages in Eqs. (3.8) and (3.9) involve terms like

$$\langle \hat{a}_s^\dagger(\tau) \hat{a}_s(0) \rangle_R = \exp(i\omega_s \tau) n_s^{\text{th}} \delta_{ss'}, \quad (\text{A2})$$

$$\langle \hat{a}_s(\tau) \hat{a}_s^\dagger(0) \rangle_R = [\exp(-i\omega_s \tau)] (n_s^{\text{th}} + 1) \delta_{ss'}, \quad (\text{A3})$$

where

$$\begin{aligned} n_s^{\text{th}} &= \text{Tr}_R \{ \hat{\rho}_R [\hat{a}_s^\dagger(0) \hat{a}_s(0)] \} \\ &= [\exp(\hbar\omega_s/k_B T) - 1]^{-1}. \end{aligned} \quad (\text{A4})$$

Thus, for example,

$$\begin{aligned} W_{ll'}^{+10} &= \sum_s (n_s^{\text{th}} + 1) C_{l's}^* (\mathbf{r}_{l'}) C_{ls} (\mathbf{r}_l) \\ &\quad \times \int_0^\infty \exp[i(\omega_a - \omega_s)\tau] d\tau, \end{aligned} \quad (\text{A5})$$

where the Markov approximation has been made by letting  $t_u = \infty$  in Eq. (3.8). Using the identity

$$\int_0^\infty \exp(\pm i\Omega\tau) d\tau = \pi\delta(\Omega) \pm i\text{P} \frac{1}{\Omega}, \quad (\text{A6})$$

and discarding the imaginary part, which amounts to neglecting a small Lamb shift in frequency, we get after integration over  $\tau$ ,

$$W_{ll'}^{+10} = (n_{\text{th}} + 1) \gamma_{ll'}(\mathbf{R}_{ll'}), \quad (\text{A7})$$

where

$$n_{\text{th}} = [\exp(\hbar\omega_a/k_B T) - 1]^{-1}, \quad (\text{A8})$$

$$\mathbf{R}_{ll'} = \mathbf{r}_l - \mathbf{r}_{l'}, \quad (\text{A9})$$

and

$$\gamma_{ll'}(\mathbf{R}_{ll'}) = \pi \sum_s C_{ls}(\mathbf{r}_l) C_{l's}^*(\mathbf{r}_{l'}) \delta(\omega_a - \omega_s). \quad (\text{A10})$$

Let us assume that the sum  $\sum_{s=q+1}^\infty$  can be approximated by  $\sum_{s=1}^\infty$ , whose validity depends upon the case of interest, and replace  $\sum_{s=1}^\infty$  by the integral  $\int_{-\infty}^\infty [d\mathbf{k}/(2\pi)^3] V_Q \sum_\sigma$ , where  $\sum_\sigma$  denotes sum over the two orthogonal states of polarization for each  $\mathbf{k}$ . Then using Eq. (2.13), we get

$$\begin{aligned} \gamma_{ll'}(\mathbf{R}_{ll'}) &= \pi V_Q \int_{-\infty}^\infty [d\mathbf{k}/(2\pi)^3] g \delta(\omega_a - |\mathbf{k}|c) \\ &\quad \times \sum_\sigma \mu_{l'\sigma}^*(\mathbf{e}_\mathbf{k}) \mu_{l\sigma}(\mathbf{e}_\mathbf{k}) \exp(i\mathbf{k} \cdot \mathbf{R}_{ll'}), \end{aligned} \quad (\text{A11})$$

where

$$\mu_{l\sigma}(\mathbf{e}_\mathbf{k}) = {}_i \langle l | e^{\hat{\mathbf{r}}_l} | g \rangle_i \cdot \mathbf{e}_\sigma(\mathbf{k}) \quad (\text{A12})$$

and

$$g = \omega_a / 2\hbar\epsilon_0 V_Q. \quad (\text{A13})$$

By symmetry of the  $x$ ,  $y$ , and  $z$  directions, we have

$${}_i \langle l | e^{\hat{\mathbf{r}}_l} | g \rangle_i = {}_i \langle x | e^{\hat{x}} | g \rangle_i \mathbf{e}_l, \quad (\text{A14})$$

where  $\mathbf{e}_l, l \in \{x, y, z\}$  are the coordinate unit vectors. This allows us to write  $\gamma_{ll'}(\mathbf{R}_{ll'})$  in terms of an inverse Fourier transform

$$\begin{aligned} \tilde{\gamma}_{ll'}(\mathbf{k}) &\equiv \int_{-\infty}^\infty d\mathbf{R} \exp(-i\mathbf{k} \cdot \mathbf{R}) \gamma_{ll'}(\mathbf{R}) \\ &= \pi V_Q g |\mu_d|^2 \sum_\sigma \delta(\omega_a - |\mathbf{k}|c) [\mathbf{e}_l \cdot \mathbf{e}_\sigma(\mathbf{k})] \\ &\quad \times [\mathbf{e}_{l'} \cdot \mathbf{e}_\sigma(\mathbf{k})] \\ &= \frac{3}{4} \lambda_a^2 \gamma \delta(\omega_a/c - |\mathbf{k}|) \sum_\sigma [\mathbf{e}_l \cdot \mathbf{e}_\sigma(\mathbf{k})][\mathbf{e}_{l'} \cdot \mathbf{e}_\sigma(\mathbf{k})], \end{aligned} \quad (\text{A15})$$

where we have assumed all atoms to be identical so that  $\mu_d = {}_i \langle x | e^{\hat{x}} | g \rangle_i$ ,  $\lambda_a = 2\pi c/\omega_a$ , and  $\gamma = \omega_a^3 |\mu_d|^2 / 6\pi\epsilon_0 \hbar c^3$ . Therefore

$$\gamma_{ll'}(\mathbf{R}_{ll'}) = \int_{-\infty}^\infty \exp(i\mathbf{k} \cdot \mathbf{R}_{ll'}) \tilde{\gamma}_{ll'}(\mathbf{k}) d\mathbf{k} / (2\pi)^3. \quad (\text{A16})$$

Similarly,  $W_{ll'}^{+01} = n_{\text{th}} \gamma_{ll'}(\mathbf{R}_{ll'})$ ,  $W_{ll'}^{-10} = W_{ll'}^{+10*}$ , and  $W_{ll'}^{-01} = W_{ll'}^{+01*}$ .

#### APPENDIX B

In this appendix we calculate special cases of  $\gamma_{ll'}(\mathbf{R})$ . When  $l \neq l'$ ,  $\gamma_{ll'}(\mathbf{R})$  is in general not zero. However, we show that for  $l \neq l'$ ,  $\gamma_{ll'}(\mathbf{R}) = 0$  if  $\mathbf{R} \parallel \mathbf{e}_x$ ,  $\mathbf{e}_y$ , or  $\mathbf{e}_z$ . Consider, for example,  $\mathbf{R} = |\mathbf{R}| \mathbf{e}_z$  so that  $\exp(i\mathbf{k} \cdot \mathbf{R}) = \exp(ik_z |\mathbf{R}|)$ . If  $\mathbf{e}_l = \mathbf{e}_z$  and  $\mathbf{e}_{l'} = \mathbf{e}_x$ , then in Eq. (A11),

$$\mu_{l'\sigma}^*(\mathbf{e}_k)\mu_{l\sigma}(\mathbf{e}_k) \propto (\mathbf{e}_z \cdot \mathbf{e}_\sigma)(\mathbf{e}_x \cdot \mathbf{e}_\sigma).$$

Converting  $\int_{-\infty}^{\infty} d\mathbf{k}$  in Eq. (A11) to polar coordinates

$$\int_{-\infty}^{\infty} |\mathbf{k}|^2 d|\mathbf{k}| \int_0^\pi \sin\theta d\theta \int_0^{2\pi} d\phi,$$

we see that a change of variable  $\phi \rightarrow \phi + \pi$  leaves  $\exp(ik_z |\mathbf{R}|)$  unchanged while  $(\mathbf{e}_z \cdot \mathbf{e}_\sigma)(\mathbf{e}_x \cdot \mathbf{e}_\sigma)$  changes sign, implying that  $\gamma_{zx}(\mathbf{R})=0$ . Similarly, for  $\mathbf{e}_l = \mathbf{e}_x$  and  $\mathbf{e}_{l'} = \mathbf{e}_y$ , a change of variable  $\phi \rightarrow \phi + \pi/2$  implies  $\gamma_{xy}(\mathbf{R})=0$ . Using the same argument we can also show that  $\gamma_{ll'}(\mathbf{0})=0$  for  $l \neq l'$ .

To calculate  $\gamma_{ll}(\mathbf{0})$ , we note that  $\sum_{\sigma} (\mathbf{e}_l \cdot \mathbf{e}_\sigma)^2 = (\mathbf{e}_l \cdot \mathbf{e}_1)^2 + (\mathbf{e}_l \cdot \mathbf{e}_2)^2$  is just the square of the projection of  $\mathbf{e}_l$  onto the plane defined by  $\mathbf{e}_1$  and  $\mathbf{e}_2$ , so that

$$\sum_{\sigma} (\mathbf{e}_l \cdot \mathbf{e}_\sigma)^2 = \sin^2[\theta_l(\mathbf{e}_k)], \quad (\text{B1})$$

where  $\theta_l(\mathbf{e}_k)$  is the angle between the unit vector along  $\mathbf{k}$  denoted by  $\mathbf{e}_k$  and  $\mathbf{e}_l$ . Using Eq. (A16), we then get

$$\gamma_{ll}(\mathbf{0}) = \int_{-\infty}^{\infty} \frac{3}{4} \lambda_a^2 \gamma \sin^2[\theta_l(\mathbf{e}_k)] \delta(\omega_a/c - |\mathbf{k}|) d\mathbf{k} / (2\pi)^3 = \gamma, \quad (\text{B2})$$

where the integration can be carried out trivially in polar coordinates with  $\theta_l$  as the polar angle.

### APPENDIX C

In this appendix we evaluate  $T_i^{-1}$  as defined in Eq. (5.3). Adding and subtracting the  $i'=i$  term in the right

side of Eq. (5.3), we get

$$T_i^{-1} = \sum_{l'} \sum_{l''} \gamma_{xl'}(\mathbf{R}_{il'}) [\mathcal{E}_{xl'} \exp(-i\mathbf{k}_D \cdot \mathbf{R}_{il'}) + \mathcal{E}_{l'x} \exp(i\mathbf{k}_D \cdot \mathbf{R}_{il'})] - \gamma \mathcal{E}_{xx}, \quad (\text{C1})$$

where the last term is obtained by using  $\gamma_{xl'}(\mathbf{0}) = \gamma \delta_{xl'}$ , as derived in Appendix B. The sum over  $i'$  can now be converted into an integral over the volume  $V_M$  to give

$$T_i^{-1} = \sum_{l'} \int_{V_M} d\mathbf{R} \frac{N}{V_M} \gamma_{xl'}(\mathbf{r}_i - \mathbf{R}) \times \{ \mathcal{E}_{xl'} \exp[-i\mathbf{k}_D \cdot (\mathbf{r}_i - \mathbf{R})] + \mathcal{E}_{l'x} \exp[i\mathbf{k}_D \cdot (\mathbf{r}_i - \mathbf{R})] \} - \gamma \mathcal{E}_{xx}. \quad (\text{C2})$$

The validity of this conversion requires that the number of atoms in volume  $\lambda_a^3$  be large. Otherwise, a discrete Fourier transform can be used with the assumption that the atoms are randomly located in space, which is usually the case in a gaseous medium, to neglect the aliasing effect.

Using Eq. (A16) for  $\gamma_{xl'}(\mathbf{r}_i - \mathbf{R})$  and assuming that the medium is infinite in the  $x$  and  $y$  directions, we can carry out the integrals over  $X$ ,  $Y$ ,  $k_x$ , and  $k_y$  in the above equation to obtain

$$T_i^{-1} = \frac{N}{V_M} \sum_{l'} \int_{-L_M/2}^{L_M/2} dZ \int_{-\infty}^{\infty} \frac{dk_z}{2\pi} \tilde{\gamma}_{xl'}(k_z \mathbf{e}_z) \{ \mathcal{E}_{xl'} \exp[i(k_z - k_D)(z_i - Z)] + \mathcal{E}_{l'x} \exp[i(k_z + k_D)(z_i - Z)] \} - 2\gamma \mathcal{E}_{xx}. \quad (\text{C3})$$

Using arguments similar to those used in Appendix B we can show that the  $l' \neq x$  terms in Eq. (C3) vanish when the integral over  $k_z$  is performed. The  $l'=x$  term can be evaluated using a method similar to that used in deriving Eq. (B2) to give

$$T_i^{-1} = (N/V_M) (\mathcal{E}_{xx}/2\pi) (3\lambda_a^2 \gamma L_M/2) \times \left[ \frac{\exp[i(\omega_a/c - k_D)L_M/2] - \exp[-i(\omega_a/c - k_D)L_M/2]}{i(\omega_a/c - k_D)L_M} \exp[i(\omega_a/c - k_D)z_i] + \frac{\exp[i(\omega_a/c + k_D)L_M/2] - \exp[-i(\omega_a/c + k_D)L_M/2]}{i(\omega_a/c + k_D)L_M} \exp[i(\omega_a/c + k_D)z_i] \right] - 2\gamma \mathcal{E}_{xx}. \quad (\text{C4})$$

The second term within the large square brackets in the above equation can, in general, be neglected compared with the first if  $|\omega_a/c - k_D| \ll \omega_a/c$ , which is usually the case.

### APPENDIX D

In this appendix we review the collective decay of coherently phased classical dipoles. Consider an  $x$ -polarized array of  $N$  dipoles enclosed in a volume

$V_M = AL_M$  of length  $L_M$ , whose macroscopic polarization density  $P_x^{\text{cl}}(z, t)$  is in the form of a plane wave traveling in the  $z$  direction. Such a traveling wave, which can be created with approximate initial conditions on  $P_x^{\text{cl}}$  and  $\partial/\partial t P_x^{\text{cl}}$ , can be written as

$$P_x^{\text{cl}}(z, t) = \text{Re} \{ \mathcal{P}_x^{\text{cl}}(z, t) \exp[-i(\omega_a t - k_z z)] \}, \quad (\text{D1})$$

where  $\mathcal{P}_x^{\text{cl}} = (N/V_M) 2\mu_d^* |V_x^{\text{cl}}|$  and  $\omega_a = ck_z$  is the resonance frequency of the dipoles. If  $V_M$  is infinite in the  $x$

and  $y$  directions, then the total radiated electric field will also be a plane wave propagating in the  $z$  direction.

In terms of the slowly varying amplitudes  $\mathcal{E}_x^{\text{cl}}(z, t)$  and  $\mathcal{P}_x^{\text{cl}}(z, t)$  for the electric field and polarization, respectively, the wave equation in steady state reduces to

$$\frac{\partial \mathcal{E}_x^{\text{cl}}(z)}{\partial z} = \frac{ik_z}{2\epsilon_0} \mathcal{P}_x^{\text{cl}}(z). \quad (\text{D2})$$

This equation can be integrated trivially from  $z=0$  to  $L_M$  to give

$$\mathcal{E}_x^{\text{cl}}(L_M) = \left[ \frac{ik_z}{2\epsilon_0} \right] \left[ \frac{N}{V_M} \right] \mu_d^* 2 |V_x^{\text{cl}}| L_M. \quad (\text{D3})$$

The power radiated in an area  $A$  at  $z=L_M$  is  $P_R = (cA\epsilon_0/2) |\mathcal{E}_x^{\text{cl}}(L_M)|^2$ , which can be written as

$$P_R = \hbar\omega_a N / T_R, \quad (\text{D4})$$

where

$$T_R^{-1} = \left[ \frac{N}{V_M} \right] \left[ \frac{3}{4\pi} \lambda_a^2 L_M \gamma \right] |V_x^{\text{cl}}|^2. \quad (\text{D5})$$

Then by energy conservation, we expect  $\langle n_x \rangle$  to decay like

$$\frac{\partial \langle n_x \rangle}{\partial t} = -1/T_R. \quad (\text{D6})$$

Similar to the comment made after Eq. (5.3), the above equation describes free decay of the coherently phased classical dipoles in the absence of the pump field which created the traveling-wave polarization density of Eq. (D1). Therefore, the steady state is reached when  $V_x^{\text{cl}}=0$ , causing  $T_R^{-1}$  and  $\langle n_x \rangle$  to be zero as well.

## APPENDIX E

The expression for  $V(t)$ , expanded to all orders, is given as follows:

$$V(t) = \frac{2F\gamma_\perp A_m \exp(-iv_m t)}{D^-(v_m)\pi(0)} \left[ -1 + \sum_{l=0}^{\infty} W_l^{-m} \prod_{n=0}^l P_n^{-m} \right] + \Gamma_V(t), \quad (\text{E1})$$

where

$$W_l^{-m} = \frac{D^-(v_m) \exp \left[ i \sum_{p=0}^l (v_{m_p} - v_{m_p''}) t \right]}{D^- \left[ \sum_{p=0}^l (-v_{m_p} + v_{m_p''}) + v_m \right]}, \quad (\text{E2})$$

$$P_l^{-m} = \frac{2A_{m_l}^+ A_{m_l}''}{\pi(-v_{m_l} + v_{m_l}'')} \left[ \frac{1}{D^-(v_{m_l}'')} + \frac{1}{D^+(-v_{m_l})} \right], \quad (\text{E3})$$

and  $P_n^{-m}$  for  $0 \leq n < l$  is given by

$$P_n^{-m} = - \frac{2A_{m_n}^+ A_{m_n}''}{\pi \left[ \sum_{p=n}^l (-v_{m_p} + v_{m_p}'') \right]} \times \left[ \frac{1}{D^- \left[ \sum_{p=n}^l (-v_{m_p} + v_{m_p}'') + v_{m_n} \right]} + \frac{1}{D^+ \left[ \sum_{p=n}^l (-v_{m_p} + v_{m_p}'') - v_{m_n}'' \right]} \right]. \quad (\text{E4})$$

In the above equations  $\{m_p, m_p'' : p=0, 1, \dots, l\}$  are dummy indices like  $k, l, m$ , and  $n$  in Sec. IV. Repeated indices are to be summed over from 1 to  $\infty$  and a double prime implies that the term  $m_p'' = m_p$  should be ignored.

The noise part  $\Gamma_V(t)$  is obtained from its Fourier transform  $\Gamma_V(\omega)$  using Eq. (6.45), which when expanded to all orders can be written as follows:

$$\Gamma_V(\omega) = \frac{2A_m}{D_0^-} \Gamma_{n-m} + \frac{f_{V_0}}{D_0^-}, \quad (\text{E5})$$

$$\Gamma_{n-m} = - \frac{A_k^+ f_{V_{k-m}}}{\pi_{-m} D_{k-m}^-} - \frac{A_k f_{V_{k-m}^+}}{\pi_{-m} D_{-k-m}^+} + \frac{f_{n-m}}{\pi_{-m}} + \sum_{l=0}^{\infty} Z_l^{-m} \prod_{n=0}^l Y_n^{-m}. \quad (\text{E6})$$

Here

$$Z_l^{-m} = - \frac{A_k^+ f_V \sum_{p=0}^l (m_p - m_p'') + k - m}{\pi \sum_{p=0}^l (m_p - m_p'') - m D \sum_{p=0}^l (m_p - m_p'') + k - m} - \frac{A_k f_V^+ \sum_{p=0}^l (m_p - m_p'') - k - m}{\pi \sum_{p=0}^l (m_p - m_p'') - m D \sum_{p=0}^l (m_p - m_p'') - k - m} + \frac{f_n \sum_{p=0}^l (m_p - m_p'') - m}{\pi \sum_{p=0}^l (m_p - m_p'') - m}, \quad (\text{E7})$$

$$Y_0^{-m} = \frac{A_{m_0}^+ A_{m_0}''}{\pi_{-m}} \left[ \frac{1}{D_{m_0 - m}^-} + \frac{1}{D_{-m_0'' - m}^+} \right], \quad (\text{E8})$$

and  $Y_n^{-m}$  for  $0 < n \leq l$  is given by

$$Y_n^{-m} = - \frac{2 A_{m_n}^+ A_{m_n}''}{\pi \sum_{p=0}^{n-1} (m_p - m_p'') - m} \times \left[ \frac{1}{D \sum_{p=0}^{n-1} (m_p - m_p'') + m_n - m} + \frac{1}{D \sum_{p=0}^{n-1} (m_p - m_p'') - m_n'' - m} \right]. \quad (\text{E9})$$

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<sup>26</sup>The assumption may still hold for collective atomic variables defined over the entire medium with dimensions much larger than a wavelength, as long as the number of atoms being summed over is large. It is not straightforward to prove this for our case of a multimode field, although, for a single-mode field with wave vector  $\mathbf{k}$ , it is easily shown by absorbing the phase-factor  $\exp(i\mathbf{k} \cdot \mathbf{r}_i)$  in the definition of the state vectors  $|x\rangle_i$ ,  $|y\rangle_i$ , and  $|z\rangle_i$ .

<sup>27</sup>More precisely, the equations of motion for the second moments of the atomic variables are given in terms of the coefficients of the second-order derivatives in the Fokker-Planck equation together with the first- and/or second-order moments of the atomic variables, only when the Langevin equations are linear in these variables. This is true for Eqs. (4.3)–(4.7) if both the super-radiance and the feedback of the field fluctuation on the atomic variables are neglected.

<sup>28</sup>The coefficients of the third- or higher-order derivatives can be shown to be related to the correlations of three or more atomic variables. For the relation of these coefficients to the moments of the variables, see, e.g., M. Sargent III, M. O. Scully, and W. E. Lamb, Jr., *Laser Physics* (Addison-Wesley, Reading, MA, 1974), p. 312.

<sup>29</sup>This is because when the anomalous term in  $T_i^{-1}$  is neglected,  $V_{xi}$  and  $n_{xi}$ , in general, have solutions of the following form:

$$V_{xi} \sim \frac{2g_p a_p}{\gamma_{\parallel}(1+i\delta_p)(1+I/I_s)} + \Gamma_{V_{xi}},$$

$$n_{xi} \sim \frac{2g_p^2 a_p^+ a_p}{\gamma_{\parallel}^2(1+\delta_p^2)(1+I/I_s)} + \Gamma_{n_{xi}},$$

where

$$I/I_s \sim \frac{g_p^2 a_p^+ a_p}{\gamma_{\parallel} \gamma_{\perp} (1+\delta_p^2)},$$

$\delta_p$  is the pump-frequency detuning from the atomic resonance frequency normalized to  $\gamma_1$ ,  $a_p$  is the  $c$ -number variable for the pump mode,  $g_p$  is the atom-pump-mode coupling constant, and the correlation  $\langle \Gamma_{V_{xi}^+} \Gamma_{V_{xi'}} \rangle$  is zero for  $i' \neq i$ . It is clear from these solutions that  $\langle V_{xi}^+ V_{xi'} \rangle \ll n_{xi}$ ,  $i' \neq i$ , at high pump intensity where  $I \gg I_s$ , which in turn is consistent with the assumption that the anomalous decay term is negligible [cf. Eq. (5.1)]. However, when  $I \ll I_s$ , i.e., at low pump inten-

sity,  $\langle V_{xi}^+ V_{xi'} \rangle = 2n_{xi}$ , which is inconsistent with the assumption that the anomalous decay term and thus  $T_i^{-1}$  are negligible.

- <sup>30</sup>M. Sargent III, M. O. Scully, and W. E. Lamb, Jr., *Laser Physics* (Addison-Wesley, Reading, MA, 1974).  
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