Cone emission from laser-pumped two-level atoms. I. Quantum theory of resonant light propagation

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This is the first of a series of papers on the theory of cone emission from a laser-pumped two-level atomic medium. Starting from microscopic quantum theory for both the active medium and the field, equations for the slowly varying macroscopic quantities are derived. In the steady-state limit, which experimentally corresponds to cw excitation, we find that the sources for the paraxial fields of interest have a long coherence length despite the random nature of spontaneous emission. These radiation sources enter into the equations for the fields in the same form as the source generated by four-wave mixing and therefore may be considered as a "spontaneous four-wave mixing," since the photons emitted in the Rabi sidebands are correlated. This has far-reaching consequences on the physics of cone emission and also sheds some new light on the fluoresence from strongly driven atomic systems.

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I. INTRODUCTION

Cone emission has been the subject of numerous experiments and theoretical considerations in recent years [1–8]. In general, to observe the phenomenon from two-level atoms, the laser beam must be strong enough to form filaments due to self-focusing, and the frequency of the laser beam should be close to, but greater than, that of the atomic resonance transition. The propagating laser beam leads to emission of other beams separated in frequency and angular distribution from the original strong beam (see Fig. 1). Of particular interest is a beam of lower frequency (redshifted component) which forms a cone in the far-field intensity distribution. A blueshifted beam is also formed. The blueshifted component is emitted in the forward direction only, and not as a cone.

Several effects leading to cone emission have been discussed [2–8], see for example the recent paper by Valley *et al.* [2], which is probably the most complete to date. None of these papers provides a fully satisfactory description. It is believed that the process of formation and propagation of the three beams is a complicated one and involves several effects. Each process is simple in principle; however, in combination, they become quite complicated. It is questionable whether one simple effect can adequately describe the complete process. Careful theoretical analysis is therefore needed. The paper by Valley *et al.* [2] ascribes cone emission to an interplay between four-wave mixing (4WM) and the effects of diffractive spreading during propagation. However, other authors [3] have invoked Cherenkov emission to explain the phenomenon.

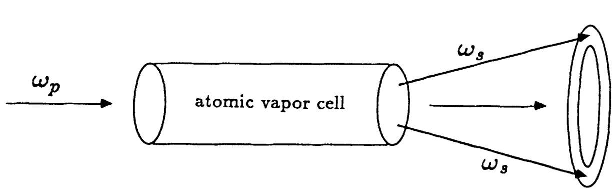

FIG. 1. Typical experimental geometry for observing cone emission in the far field.

In a recent paper [9], we pointed out that a Cherenkov-type emission, not considered by Valley *et al.* [2], should also be taken into account when discussing cone emission. This effect relates to the initiation and generation of the frequency-shifted fields and is intrinsically quantum in nature. This effect comes from the spatial correlation at different positions of the polarization of the medium generated by spontaneous emission at the relevant frequencies, and is in many ways analogous to a spontaneous four-wave mixing (at least in the absence of collisions) since the photons emitted in the Rabi sidebands are correlated. We show by explicit calculation that due to the interaction with the electromagnetic field, the polarization of the medium has a large correlation length, contrary to the δ-correlated assumption of Valley *et al.* [2]. The in-phase polarization of the medium acts as a Cherenkov-type source of the two beams (blueshifted and redshifted). Our results show that the source, although quantum in nature, is closer to a coherent source than to noise. This has far reaching consequences, especially for the angular distribution of the beams.

In this and the following paper, we provide more details on the theory we have developed [9]. In this first paper, the formulation of the problem is presented, and the propagation equation for the slowly varying field operators are derived. The important source terms coming from the incoherent scattering of the pump light are identified, which are shown to have a long coherence length under the paraxial approximation. In the second paper we analytically study various models to clarify the physics of cone emission, and we compare and unify our results with previous model studies. The 4WM and the Cherenkov-type radiation model controversy is resolved.

We plan to implement numerical simulation schemes based on our work in a future publication, and we will then try to make comparisons with available experimental work.

This paper is organized as follows. We describe the formulation of the problem in Sec. II, followed in Sec. III by the derivation of the equations for the collective atomic operators, which are the key to our formulation. In Sec. IV we solve the atomic operator equations perturbatively to first order in the generated weak fields $\hat{E}_s$ and $\hat{E}_4$. Section V is devoted to a detailed discussion of the long coherence length quantum noise. In Sec. VI the field operator equations are derived, which have the macroscopic polarizations as their source terms. We summarize in Sec. VII.

The system under consideration is modeled by the electromagnetic field and a medium formed by two-level atoms. The electromagnetic field will be decomposed into several components (see Fig. 2). One of the fields has large intensity. It will be called the pump, or laser beam, and will be treated as a classical quantity, and its interaction with the medium is described semiclassically with all the effects due to propagation included. Under the influence of the strong pump beam the medium responds by forming two weak beams of different frequencies at the Rabi sidebands. These two fields will be described by quantum-mechanical field operators, $\hat{E}_s$ and $\hat{E}_4$.

It is assumed that the medium has a pencil-like shape with the Fresnel number close to one or smaller. This assumption allows the description of the propagation of waves in the framework of the paraxial approximation. In order to treat correctly the spontaneous emission of photons by the atoms forming the medium we also have to include all the modes of the free electromagnetic field. Although important features of the spontaneous generation of the fields may be obtained from simple two-mode models [10], previous work has not considered cone formation. The fields $\hat{E}_s$ and $\hat{E}_4$ are smoothed macroscopic fields with the appropriate quantum noise due to spontaneous emission as their source. Most of the work regarding cone emission has been done under the assumption that the pump beam is self-focused and forms filaments. This assumption is an important one since it allows one to treat the pump field as truly strong, i.e., the resonant transition in the atoms is saturated. It has also been shown by Harter *et al.* [4] that in a self-focused filament the Rabi frequency is of the order of the detuning. In this paper we will not discuss the details of the self-focusing, even though it is also included in the formulation.

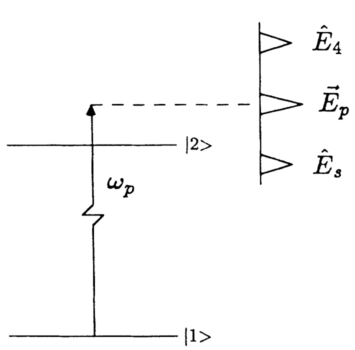

FIG. 2. The energy-level scheme of the two-level medium together with the sideband field distribution (i.e., the Mollow-triplet spectrum).

II. FORMULATION OF THE PROBLEM

The Hamiltonian of the system is given by

$$\hat{H}=\hat{H}_{\text{atom}}+\hat{H}_{\text{field}}+\hat{H}_{\text{int}} ,$$

with

$$\hat{H}_{\text{atom}}=\sum_{j=1}^{N_t}\sum_{\mu=1}^{2}\hbar\omega_\mu^{(j)}\hat{\sigma}_{\mu\mu}^{(j)} ,$$

$$\hat{H}_{\text{field}}=\sum_\lambda \hbar\omega_\lambda \hat{a}_\lambda^\dagger \hat{a}_\lambda+\frac{1}{8\pi}\int_V (E_pE_p^*+B_pB_p^*)d\mathbf{r} ,$$

$$\hat{H}_{\text{int}}=-\sum_{j=1}^{N_t}\frac{\hbar}{2}(\hat{\sigma}_{21}^{(j)}\Omega_p^* e^{-i(\omega_p t-k_p z_j)}+\Omega_p\hat{\sigma}_{12}^{(j)}e^{i(\omega_p t-k_p z_j)})$$
$$-\sum_{j=1}^{N_t}\sum_\lambda i\hbar(g_\lambda\hat{\sigma}_{21}^{(j)}\hat{a}_\lambda e^{i\mathbf{k}_\lambda\cdot\mathbf{r}_j}-\hat{a}_\lambda^\dagger g_\lambda^*\hat{\sigma}_{12}^{(j)}e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}_j}) .$$

$\hat{H}_{\text{atom}}$ is the Hamiltonian of the free two-level atoms forming the medium. $\hat{\sigma}_{\mu\nu}(t=0)=|\mu\rangle\langle\nu|$ are the atomic projection operators in the Heisenberg picture. They form a Lie algebra,

$$[\hat{\sigma}_{\mu\nu}^{(j)},\hat{\sigma}_{\mu'\nu'}^{(j')}]=\delta_{jj'}(\delta_{\nu\mu'}\hat{\sigma}_{\mu\nu'}^{(j)}-\delta_{\nu'\mu}\hat{\sigma}_{\mu'\nu}^{(j)}) . \tag{2.1}$$

In the above, greek letters are used to index the energy levels, while roman letters denote the atoms. Level 2 is the excited state, and level 1 is the ground state. N_t is the total number of atoms.

$\hat{H}_{\text{field}}$ is the Hamiltonian for the total field, the first term is that of the quantized fields, the operators $\hat{a}_\lambda^\dagger$ and $\hat{a}_\lambda$ are the creation and annihilation operators of photons in the mode λ. The second term is the energy of the strong pump semiclassical field, and can be neglected.

The total field is

$$\hat{\mathbf{E}}=\mathbf{E}_p+\sum_\lambda \hat{\mathbf{E}}_\lambda , \tag{2.2}$$

which consists of the classical pump field plus quantized field. The pump field can be written as

$$\mathbf{E}_p(\mathbf{r},t)=\mathbf{E}_p^{(+)}(\mathbf{r},t)e^{-i\omega_p t+ik_p z}+\mathbf{E}_p^{(-)}(\mathbf{r},t)e^{i\omega_p t-ik_p z} , \tag{2.3}$$

which can have any polarization. However, since we are using a two-level model system, we will take it to be orthogonal to the propagation direction, which we choose to be along the z axis. As previously mentioned we will treat the pump field classically. Thus $\mathbf{E}_p^{(+)}=(\mathbf{E}_p^{(-)})^*$ are not quantum operators. The quantum field part is

$$\sum_\lambda \hat{\mathbf{E}}_\lambda=\sum_\lambda \hat{\mathbf{E}}_\lambda^{(+)}+\sum_\lambda \hat{\mathbf{E}}_\lambda^{(-)} , \tag{2.4}$$

with

$$\sum_\lambda \hat{\mathbf{E}}_\lambda^{(+)}(\mathbf{r},t)=\sum_\lambda i(2\pi\hbar\omega_\lambda/V)^{1/2}\boldsymbol{\epsilon} e^{i\mathbf{k}_\lambda\cdot\mathbf{r}}\hat{a}_\lambda \ . \qquad (2.5)$$

Here V is the quantization volume and $\lambda=(\mathbf{k},\boldsymbol{\epsilon})$; $\mathbf{k}$ and $\boldsymbol{\epsilon}$ are wave and polarization vectors, respectively. At $t=0$, the quantum field reduces to the vacuum field, the evolution of which is described by the Heisenberg equation of motion, and any deviation from the vacuum field is due to the interaction with the medium. This quantized field is, in principle, in all directions as is evident from the above expansion. However, due to the particular geometry, the modes that correspond to propagation along the axis are more important. This will become clearer when we introduce the effective coupling fields.

$\hat{H}_{\rm int}$ is the interaction Hamiltonian between the fields and the atoms under the local field dipole approximation. For the pump field part of $\hat{H}_{\rm int}$ this amounts to using the pump field at a particular location, with effects due to propagation included. The rotating-wave approximation (RWA) has also been used. The dipole moments of the atoms are assumed to be the same irrespective of location, such that $\mathbf{d}_{11}=\mathbf{d}_{22}=\mathbf{0}$ (no permanent dipole), and $\mathbf{d}_{12}=\mathbf{d}_{21}^*$. The Rabi frequency for the pump field is defined as

$$\Omega_p=2\mathbf{d}_{12}\cdot\mathbf{E}_p^{(-)}/\hbar \ ,$$

and the coupling constant as

$$g_\lambda=(2\pi\hbar\omega_\lambda/V)^{1/2}\mathbf{d}_{21}\cdot\boldsymbol{\epsilon}/\hbar \ .$$

A zero-temperature ($T=0$) approximation has also been assumed for both the field and the medium.

In the above, we have separated the field into two parts, the classical pump field and the quantum field. The quantum field carries all the information about the statistics of the vacuum fluctuations. The propagation of the pump field is included because important features like self-focusing, filamentation, etc. have to be taken into account in order to describe correctly the spatial characteristics of the new frequency fields that are generated. The generated quantum fields are introduced under a general rotating-wave approximation (RWA) which gives rise to three slowing varying components centered at various frequencies (namely the pump frequency and the two Rabi sidebands). In this Heisenberg picture formulation, we will use the space-time-dependent operators to represent the corresponding classical quantities. This is in contrast to many other systems in quantum optics, since we do not have a good optical cavity to clearly define the modes for the field. Moreover, we are dealing with one-way propagation, like a swept gain amplifier [11], x-ray laser [12], superfluorescence [13,14], or Raman scattering [15]. The generated fields very often are not in resonance with the cavity, and they propagate out of the cell (no bouncing at the exit mirrors) after generation. These space-time-dependent operators usually contain many modes and therefore a single-mode theory will not be applicable. Despite the considerable amount of work on quantum optics in recent years, much work remains to be done on the theory of propagation. Graham and Haken [16] first considered the propagation problem in an interesting series of papers with emphasis on the laser theory. In the quantum theory of superfluorescence [13,14], propagation is included and is found to be essential. Later propagation aspects have also been included in a quantum theory that unifies Raman scattering [15]. More recently many new investigations on the quantum theory of light propagation based on various approaches have appeared [17–20]. Throughout this paper, we will present our formulation and important findings for near-resonant quantum light propagating in a two-level atomic medium. In the companion paper, we find that cone emission is a direct result of the quantum nature of the light propagation.

III. ATOMIC OPERATORS EQUATIONS

All the operators obey Heisenberg equations of motion

$$\frac{d\hat{O}}{dt}=\frac{i}{\hbar}[H,\hat{O}] \ , \qquad (3.1)$$

where $\hat{O}$ is a generic operator in the Heisenberg picture.

For the atomic operators, with normal ordering for the field operators, we have

$$\begin{aligned}\frac{d\hat{\sigma}_{21}^{(l)}}{dt}&=i\omega_{21}\sigma_{21}^{(l)}-i\frac{\Omega_p}{2}e^{i(\omega_p t-k_p z_l)}\hat{w}^{(l)}\\&\quad-\sum_\lambda g_\lambda^*\hat{a}_\lambda^\dagger e^{-\mathbf{k}_\lambda\cdot\mathbf{r}_l}\hat{w}^{(l)} \ ,\\ \frac{d\hat{\sigma}_{12}^{(l)}}{dt}&=-i\omega_{21}\hat{\sigma}_{12}^{(l)}+i\hat{w}^{(l)}\frac{\Omega_p^*}{2}e^{-i(\omega_p t-k_p z_l)}\\&\quad-\sum_\lambda g_\lambda\hat{w}^{(l)}\hat{a}_\lambda e^{i\mathbf{k}_\lambda\cdot\mathbf{r}_l} \ ,\\ \frac{d\hat{w}^{(l)}}{dt}&=\frac{d(\hat{\sigma}_{11}^{(l)}-\hat{\sigma}_{22}^{(l)})}{dt}\\&=i(\Omega_p\hat{\sigma}_{12}^{(l)}e^{i(\omega_p t-k_p z_l)}-\hat{\sigma}_{21}^{(l)}\Omega_p^*e^{-i(\omega_p t-k_p z_l)})\\&\quad+\sum_\lambda 2(g_\lambda\hat{\sigma}_{21}^{(l)}\hat{a}_\lambda e^{i\mathbf{k}_\lambda\cdot\mathbf{r}_l}+g_\lambda^*\hat{a}_\lambda^\dagger\hat{\sigma}_{12}^{(l)}e^{-\mathbf{k}_\lambda\cdot\mathbf{r}_l}) \ ,\end{aligned} \qquad (3.2)$$

where normal (antinormal) ordering refers to putting the $\hat{a}_\lambda$ ($\hat{a}_\lambda^\dagger$) to the rightmost, and $\hat{a}_\lambda^\dagger$ ($\hat{a}_\lambda$) to the leftmost position of an expression. This is important only when the formal solutions for $\hat{a}_\lambda$ and $\hat{a}_\lambda^\dagger$ are substituted into a given expression. In general, the field operator $\hat{a}_\lambda$ and the atomic operator $\hat{\sigma}_{\mu\nu}^{(j)}$ act on different subspaces, and therefore commute, i.e., $[\hat{a}_\lambda(t),\hat{\sigma}_{\mu\nu}^{(j)}(t)]=0$. However, the formal solution for $\hat{a}_\lambda$ [see Eq. (3.4)] contains many terms, and each of these individual terms does not commute with $\hat{\sigma}_{\mu\nu}^{(j)}$, thus we have to be consistent in using a particular ordering after the substitution.

For the field operator, we have

$$\frac{d\hat{a}_\lambda}{dt}=-i\omega_\lambda\hat{a}_\lambda+\sum_{j=1}^{N_t}g_\lambda^*\hat{\sigma}_{12}^{(j)}e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}_j} \ . \qquad (3.3)$$

We first write the above field equation in terms of the integral equations,

$$\hat{a}_\lambda(t)=\hat{a}_\lambda(0)e^{-i\omega_\lambda t}+g_\lambda^* e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}_l}\int_0^t \hat{\sigma}_{12}^{(l)}(t')e^{-i\omega_\lambda(t-t')}dt' + \sum_{j\,(\neq l)} g_\lambda^* e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}_j}\int_0^t \hat{\sigma}_{12}^{(j)}(t')e^{-i\omega_\lambda(t-t')}dt' . \quad (3.4)$$

In the above we have represented the fields by three separate terms. The first term is the free vacuum field, while the second term is the self-field (the radiated field of this particular dipole), which in the normal-ordered case, will be responsible for the radiative damping. The last term contains a summation over all the other atoms; it is the dipole field, which is the radiated field due to all the other dipoles. In the atomic-operator equations, the appropriate $\hat{E}$ field is that due to all the other atoms, whereas the field equation describes the total field. The difference gives the Lorenz-Lorentz corrections. When we put the above expressions [Eq. (3.4)] into the equation for the lth atom, we have

$$-\sum_\lambda g_\lambda^* e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}_l}\hat{a}_\lambda^\dagger \hat{w}^{(l)}=s_{21}^{(l)}+\hat{m}_{21}^{(l)}+\hat{f}_{21}^{(l)} , \quad (3.5)$$

where

$$\hat{s}_{21}^{(l)}=-\sum_\lambda |g_\lambda|^2\int_0^t \hat{\sigma}_{21}^{(l)}(t')e^{i\omega_\lambda(t-t')}dt'\hat{w}^{(l)}(t) ,$$

$$\hat{m}_{21}^{(l)}=-\sum_\lambda |g_\lambda|^2 e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}_l}\sum_{j\neq l} e^{i\mathbf{k}_\lambda\cdot\mathbf{r}_j}\int_0^t \hat{\sigma}_{21}^{(j)}(t')e^{i\omega_\lambda(t-t')}dt'\hat{w}^{(l)}(t) , \quad (3.6)$$

$$\hat{f}_{21}^{(l)}=-\sum_\lambda g_\lambda^* e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}_l}\hat{a}_\lambda^\dagger(0)e^{i\omega_\lambda t}\hat{w}^{(l)}(t) .$$

We first work out the $\hat{s}_{21}^{(l)}$ term, with the Markov approximation (MA) [21] introduced as (see Appendix A)

$$\hat{\sigma}_{21}^{(l)}(t')=\hat{\sigma}_{21}^{(l)}(t)e^{i\omega_{21}(t'-t)} . \quad (3.7)$$

We get

$$\hat{s}_{21}^{(l)}=-\tilde{\gamma}\hat{\sigma}_{21}^{(l)}(t)\hat{w}^{(l)}(t)=-\tilde{\gamma}\hat{\sigma}_{21}^{(l)}(t) , \quad (3.8)$$

where

$$\tilde{\gamma}=\frac{\gamma}{2}+i\tilde{\Delta}(\omega_{21}) . \quad (3.9)$$

The Lamb shift $\tilde{\Delta}(\omega_{21})$ is due to virtual processes, and a renormalization procedure has to be employed to obtain convergence of the integral. We will neglect this shift in the following.

Similarly we have

$$\sum_\lambda 2(g_\lambda\hat{\sigma}_{21}^{(l)}\hat{a}_\lambda e^{i\mathbf{k}_\lambda\cdot\mathbf{r}_l}+g_\lambda^*\hat{a}_\lambda^\dagger\hat{\sigma}_{12}^{(l)}e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}_l}) =\hat{s}^{(l)}+\hat{m}^{(l)}+\hat{f}^{(l)} , \quad (3.10)$$

where the details are given in Appendix B. Thus we can rewrite Eq. (3.2) as

$$\frac{d\hat{\sigma}_{21}^{(l)}}{dt}=i\omega_{21}\hat{\sigma}_{21}^{(l)}-\frac{\gamma}{2}\hat{\sigma}_{21}^{(l)}-i\frac{\Omega_p}{2}e^{i(\omega_p t-k_p z_l)}\hat{w}^{(l)}+\hat{m}_{21}^{(l)}+\hat{f}_{21}^{(l)} ,$$

$$\frac{d\hat{\sigma}_{12}^{(l)}}{dt}=-i\omega_{21}\hat{\sigma}_{12}^{(l)}-\frac{\gamma}{2}\hat{\sigma}_{12}^{(l)}+i\frac{\Omega_p^*}{2}e^{-i(\omega_p t-k_p z_l)}\hat{w}^{(l)}+\hat{m}_{12}^{(l)}+\hat{f}_{12}^{(l)} , \quad (3.11)$$

$$\frac{d\hat{w}^{(l)}}{dt}=-\gamma(\hat{w}^{(l)}-\hat{w}_0)+i(\Omega_p\hat{\sigma}_{21}^{(l)}e^{i(\omega_p t-k_p z_l)}-\hat{\sigma}_{21}^{(l)}\Omega_p^* e^{-i(\omega_p t-k_p z_l)})+\hat{m}^{(l)}+\hat{f}^{(l)} .$$

We remind ourselves that up to this point we have been dealing with one particular atom located at $\mathbf{r}_l$, and the $\hat{m}$ and $\hat{f}$ terms have not yet been treated. Now we introduce the collective operators as [11,14]

$$\hat{Q}_{\mu\nu}(\mathbf{r})=\frac{1}{N_{[l]}}\sum_{l\in[l]}\hat{\sigma}_{\mu\nu}^{(l)}\delta(\mathbf{r}-\mathbf{r}_l) , \quad (3.12)$$

where $[l]$ refers to the lth coarse-graining volume element centered around $\mathbf{r}_l$, which has a linear dimension that is less than a wavelength λ_p of the pump field, but large enough to contain at least one atom. This is the so-called physically small, microscopically large coarse graining. $N_{[l]}$ is the effective number density of atoms in this volume element. With this definition, the collective atomic operators are going to be dimensionless. Under this collective variable description approximation, we will limit the discussion to the density regime $n\lambda_p^3 \ll 1$, such that we can neglect the near-field dipole-dipole interaction and frequencies shifts, i.e., we do not distinguish between the field at the atom and the total field. This is consistent with neglecting the Lorenz-Lorentz correction. We also assume that the field and atomic operators decorrelate when the field is determined by the far fields of the other atoms. Then

$$\frac{1}{N_{[l]}}\sum_{l\in[l]}\hat{E}(\mathbf{r}_l)\hat{\sigma}_{\mu\nu}^{(l)}\delta(\mathbf{r}-\mathbf{r}_l)=\hat{E}(\mathbf{r})\frac{1}{N_{[l]}}\sum_{l\in[l]}\hat{\sigma}_{\mu\nu}^{(l)}\delta(\mathbf{r}-\mathbf{r}_l) =\hat{E}(\mathbf{r})\hat{Q}_{\mu\nu}(\mathbf{r}) . \quad (3.13)$$

According to Eq. (3.12), we have

$$\hat{Q}_{21}(\mathbf{r},t)=\frac{1}{N_{[l]}}\sum_{l\in[l]}\hat{\sigma}_{21}^{(l)}(t)\delta(\mathbf{r}-\mathbf{r}_l) ,$$

$$\hat{Q}_{12}(\mathbf{r},t)=\frac{1}{N_{[l]}}\sum_{l\in[l]}\hat{\sigma}_{12}^{(l)}(t)\delta(\mathbf{r}-\mathbf{r}_l) ,$$

$$\hat{Q}(\mathbf{r},t)=\frac{1}{N_{[l]}}\sum_{l\in[l]}\hat{w}^{(l)}(t)\delta(\mathbf{r}-\mathbf{r}_l) , \quad (3.14)$$

$$\frac{d\hat{Q}_{\mu\nu}(\mathbf{r},t)}{dt}=\frac{1}{N_{[l]}}\sum_{l\in[l]}\frac{d\hat{\sigma}_{\mu\nu}^{(l)}}{dt}\delta(\mathbf{r}-\mathbf{r}_l) .$$

The commutation relations for the new collective variable description approximation atom variables take on the following form [11]:

$$[\hat{Q}_{21}^{\dagger}(\mathbf{r},t),\hat{Q}_{21}(\mathbf{r}',t)]=\frac{1}{N(\mathbf{r})}\hat{Q}(\mathbf{r},t)\delta(\mathbf{r}-\mathbf{r}')\ ,$$
$$[\hat{Q}_{21}(\mathbf{r},t),\hat{Q}(\mathbf{r}',t)]=\frac{2}{N(\mathbf{r})}\hat{Q}_{21}(\mathbf{r},t)\delta(\mathbf{r}-\mathbf{r}')\ , \tag{3.15}$$

with the initial conditions at $t=0$ exactly the same as in superfluorescence (SF) [12,13], since with $|\Psi\rangle$ as the state vector for the system, we have

$$\langle\Psi|\hat{Q}(\mathbf{r},0)\delta(\mathbf{r}-\mathbf{r}')|\Psi\rangle=-\langle\Psi|\hat{N}(\mathbf{r})\delta(\mathbf{r}-\mathbf{r}')|\Psi\rangle\ , \tag{3.16}$$

where $\hat{N}(\mathbf{r})$ is the total number density operator of the atom, which is a conserved constant.

As indicated above, we evolved our system from a Hamiltonian, thus the above commutation relations should be conserved at all later times. Our collective variable description approximation is consistent with this so far. In the following, we will attempt to solve the system under various other approximations, some of which might violate these commutation relations. However, as long as the errors are higher order than the terms we keep, the physics involved should not be affected. In particular, when we adiabatically eliminate the atomic variables, we will be left with a Hilbert space that is spanned by the field alone. Nevertheless, our important results do not significantly violate these relations.

Depending on the preparation of the medium and also the geometry, the time variable t is not necessarily the most convenient natural choice of an independent variable, as is the case here. Our medium has the shape of a pencil with the pump propagating along the axis. Under the approximation that the dispersion of the pump can be neglected, we can easily see that depending on the location of the atom along the axis, the natural choice of time is the retarded local time $\tau=t-z/c$. It is with respect to this local time that the dynamics of the atoms located at different positions along the axis will be the same. This is a crucial point, for each individual atom the above choice is as good as any other choice. However, after we introduce the collective variable description approximation, we have to deal with the atomic operator field variables (we have let the coarse-grained spatial coordinates become continuous). In order to specify these field-variable envelopes, we have to use a time that is the same for all the atoms. We will also neglect the linear dispersion of the nonresonant atoms (background gas), which in practice, might also contribute. By choosing $\tau=t-z/c$ [15], rather than $\tau=t-z/v(\omega)$, we are able to handle more than one spectral component, and for a dilute medium the nonlinear dispersion is accounted for via the polarization of the medium, so that the errors will be negligible.

As we mentioned earlier, the $\hat{m}$ terms (material field) are proportional to the product of the dipole operators of all the other atoms with the atomic operators of the one of interest. It describes the interaction of the fields emitted by all the other atoms with the particular atomic dipole. When we go to a collective variable description approximation, under the decorrelation approximation, the volume average of the dipole operators of all the other atoms outside of this particular element will be effectively described by the electromagnetic field operators, the spectral components of which, of course, will be determined by the dynamics of the medium (since this is the radiation of the medium). The atomic dipole operators due to atoms inside the same volume element will be neglected as discussed earlier, thus m terms will give the generated fields interacting with collective atomic operators. The exact form of the generated fields depends on the detailed dynamics of the atomic system, and also on the geometry when the slowly varying macroscopic quantities are introduced.

If we look at this from a different perspective, by studying the dynamics of a single atom, we can easily find the appropriate field operators to introduce. For a single atom located at $\mathbf{r}_l$, we have

$$\frac{d\hat{\sigma}_{21}^{(l)}}{dt}=i\omega_{21}\hat{\sigma}_{21}^{(l)}-\frac{\gamma}{2}\hat{\sigma}_{21}^{(l)}-i\frac{\Omega_p}{2}e^{i(\omega_p t-k_p z_l)}\hat{w}^{(l)}+\hat{f}_{21}^{(l)}\ ,$$
$$\frac{d\hat{\sigma}_{12}^{(l)}}{dt}=-i\omega_{21}\hat{\sigma}_{12}^{(l)}-\frac{\gamma}{2}\hat{\sigma}_{12}^{(l)}+i\frac{\Omega_p^*}{2}e^{-i(\omega_p t-k_p z_l)}\hat{w}^{(l)}+\hat{f}_{12}^{(l)}\ ,$$
$$\frac{d\hat{w}^{(l)}}{dt}=-\gamma(\hat{w}^{(l)}-\hat{w}_0)+i(\Omega_p\hat{\sigma}_{12}^{(l)}e^{i(\omega_p t-k_p z_l)}-\hat{\sigma}_{21}^{(l)}\Omega_p^*e^{-i(\omega_p t-k_p z_l)})+\hat{f}^{(l)}\ . \tag{3.17}$$

The expectation values of these equations describe a two-level atom driven by a strong near-resonant field (the resonance fluorescence Mollow spectrum problem) [22–24], for which the solutions are well known. After some time (the characteristic damping time of the atom) the system will settle down to a steady state, independent of the initial conditions. This particular atom will end up spontaneously emitting the Mollow-triplet fields in addition to the usual coherent Rayleigh scattering. Since we are dealing with an extended medium rather than a single atom, these fields will propagate and interact with the atoms downstream. We can simulate these as three additional macroscopic fields each centered at one of the Mollow-triplet positions. We have set up the equations with the pump field propagating in one direction, and, consistent with the one-way approximation, we will simulate the coherent component of the dipole fields as travel-

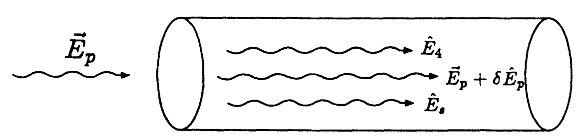

FIG. 3. The description under the paraxial approximation of the fields emitted by other atoms acting on the atom of interest are described by the fields indicated, which are classified according to their frequency and the direction of propagation.

ing in the same direction. Of course, the dipole fields are traveling in all directions according to the angular distribution pattern of the spontaneous radiation, but for our geometry coherent-phase-matched components are possible only in the forward direction. (See discussion in Sec. V, where such source terms are derived from first principles.) We will not treat the incoherent background in this paper although this could also play a role in a realistic comparison with experiments [25]. Under the slowly varying envelope approximation (SVEA) we can in principle approximate the coherent generated fields by (see Fig. 3) the following:

$$\left[\left[\sum_\lambda |g_\lambda|^2 e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}_l}\sum_{j\,(\neq 1)} e^{i\mathbf{k}_\lambda\cdot\mathbf{r}_j}\int_0^t \hat\sigma_{21}^{(j)}(t')e^{i\omega_\lambda(t-t')}dt'\right]\right] \approx i[\hat{\mathbf{E}}_s^{(-)}(\mathbf{r}_l,t)e^{i(\omega_s t-k_s z_l)}+\hat{\mathbf{E}}_4^{(-)}(\mathbf{r}_l,t)e^{i(\omega_4 t-k_4 z_l)}+\delta\hat{\mathbf{E}}_p^{(-)}(\mathbf{r}_l,t)e^{i(\omega_p t-k_p z_l)}]\ , \quad (3.18)$$

where [[ ]] denotes both a collective variable description approximation for atomic operators and a SVEA for the fields. The collective variable description approximation involves a volume average over a volume element of the order of interatomic dimensions about $\mathbf{r}_l$, which we introduced explicitly in Eq. (3.11). However, the SVEA for the field is not all that clear here, as it is the polarization of the medium (via the atomic dipole operators) that will radiate the field, so that the field will be determined by the distribution of the polarization, i.e., by the geometry of medium. The SVEA presupposes a given geometry for propagation close to the z axis, whose justification can actually only be made after the problem is solved. If valid, the SVEA is equivalent to averaging over a volume a^3 larger than or of the order λ_p^3, with the corresponding diffraction angle $\theta_d \sim 1/(k_j a)$. To describe the structure of cone angle θ_c correctly we also have to satisfy $a \ll 1/(k_j\theta_c)$.

In Eq. (3.18) $\hat{\mathbf{E}}_s(\mathbf{r},t)$ denotes the SVEA macroscopic field with frequency centered around the Mollow-triplet component to the red side of the pump, and similarly $\hat{\mathbf{E}}_4(\mathbf{r},t)$ denotes the field with frequency centered around the Mollow-triplet component to the blue side of the pump, and finally the change associated with the component resonant with the pump field is denoted by $\delta\hat{\mathbf{E}}_p(\mathbf{r},t)$. These fields are to be slowly varying over a volume of order λ_p^3 (however, we note that the collective operators are usually defined over a volume less than λ_p^3). Then we obtain

$$[[\hat m_{21}^{(l)}]]=\left[\left[\sum_\lambda |g_\lambda|^2 e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}_l}\sum_{j\,(\neq l)} e^{i\mathbf{k}_\lambda\cdot\mathbf{r}_j}\int_0^t \hat\sigma_{21}^{(j)}(t')e^{i\omega_\lambda(t-t')}dt'\hat w^{(l)}\right]\right]$$
$$\approx -\frac{i}{2}[\hat\Omega_s^{(-)}e^{i(\omega_s t-k_s k z_l)}+\hat\Omega_4^{(-)}e^{i(\omega_4 t-k_4 z_l)}+\delta\hat\Omega_p^{(-)}e^{i(\omega_p t-k_p z_l)}]\hat Q(\mathbf{r},\tau)\ ,$$
$$[[\hat m^{(l)}]]=\sum_\lambda\sum_{\substack{j,l\\ j(\neq l)}} 2|g_\lambda|^2\left[\left[\left[\hat\sigma_{21}^{(l)}e^{i\mathbf{k}_\lambda\cdot\mathbf{r}_l}e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}_j}\int_0^t\hat\sigma_{12}^{(j)}(t')e^{-i\omega_\lambda(t-t')}dt'+\text{H.c.}\right]\right]\right]$$
$$\approx i[\hat\Omega_s^{(-)}\hat Q_{12}e^{i(\omega_s t-k_s z_l)}-\hat Q_{21}\Omega_s^{(+)}e^{-i(\omega_s t-k_s z_l)}]+i[\hat\Omega_4^{(-)}\hat Q_{12}e^{i(\omega_4 t-k_4 z_l)}-\hat Q_{21}\Omega_4^{(+)}e^{-i(\omega_4 t-k_4 z_l)}]$$
$$+i[\delta\hat\Omega_p^{(-)}\hat Q_{12}e^{i(\omega_p t-k_p z_l)}-\hat Q_{21}\delta\Omega_p^{(+)}e^{-i(\omega_p t-k_p z_l)}]\ , \quad (3.19)$$

where

$$\delta\hat\Omega_p^{(-)}=2\delta\hat{\mathbf{E}}_p^{(-)}\cdot\mathbf{d}_{12}/\hbar\ ,$$
$$\hat\Omega_j^{(-)}=2\hat{\mathbf{E}}_j^{(-)}\cdot\mathbf{d}_{12}/\hbar\ \text{ for } j=s,4\ . \quad (3.20)$$

The $\hat f$ terms describe the vacuum fields interacting with the atomic operators. Under the collective variable description approximation for atomic operators and the SVEA for the field operators, we require this source term to be consistent with other terms in the equations for the slowly varying quantities. This implies that the source can be averaged over a finite volume element ($\gtrsim \lambda_p^3$) but the final result should not depend on the size of the volume chosen. At present we will not explicitly evaluate the vacuum field operators, and only replace the atomic operators by the corresponding collective ones in the above expression. The SVEA for the f terms will be carried out later. Thus we have

$$\hat{f}_{21}=-\sum_{\lambda} g_{\lambda}^{*} e^{-i\mathbf{k}_{\lambda}\cdot\mathbf{r}} \hat{a}_{\lambda}^{\dagger}(0) e^{i\omega_{\lambda}(\tau+z/c)} \hat{Q}(\mathbf{r},\tau) ,$$
$$\hat{f}=\sum_{\lambda} 2[g_{\lambda}\hat{Q}_{21}\hat{a}_{\lambda}(0)e^{-i\omega_{\lambda}(\tau+z/c)}e^{i\mathbf{k}_{\lambda}\cdot\mathbf{r}} + g_{\lambda}^{*}\hat{a}_{\lambda}^{\dagger}(0)e^{i\omega_{\lambda}(\tau+z/c)}\hat{Q}_{12}e^{-i\mathbf{k}_{\lambda}\cdot\mathbf{r}}] . \tag{3.21}$$

We then obtain the following set of equations which describe quantum mechanically the dynamics of the collective atomic operators:

$$\frac{d\hat{Q}_{21}}{d\tau}=i\omega_{21}\hat{Q}_{21}-\frac{\gamma}{2}\hat{Q}_{21}-i\frac{\hat{\Omega}^{(-)}}{2}\hat{Q}^{i\omega_p\tau}+\hat{f}_{21} ,$$
$$\frac{d\hat{Q}_{12}}{d\tau}=-i\omega_{21}\hat{Q}_{12}-\frac{\gamma}{2}\hat{Q}_{12}+i\hat{Q}\frac{\hat{\Omega}^{(+)}}{2}e^{-i\omega_p\tau}+\hat{f}_{12} ,$$
$$\frac{d\hat{Q}}{d\tau}=-\gamma(\hat{Q}-w_0)+i(\hat{\Omega}^{(-)}\hat{Q}_{12}e^{i\omega_p\tau}-\hat{Q}_{21}\hat{\Omega}^{(+)}e^{-i\omega_p\tau})+\hat{f} . \tag{3.22}$$

The general Rabi frequency operator is

$$\hat{\Omega}^{(-)}=\Omega_p+\delta\hat{\Omega}_p^{(-)}+\hat{\Omega}_s^{(-)}e^{-i\delta(t-z/c)}+\hat{\Omega}_4^{(-)}e^{i\delta(t-z/c)} , \tag{3.23}$$

where $\delta=\omega_p-\omega_s$, ω_p and ω_s are, respectively, the center frequencies of the pump and s fields.

Now we will introduce the RWA for the atomic collective operators in a manner similar to the field operators. Thus the RWA is here equivalent to the SVEA for the electromagnetic field operators. This again involves a volume average over a size $\gtrsim\lambda_p^3$, with

$$\hat{\sigma}_{12}(\mathbf{r},t)=\hat{Q}_{12}(\mathbf{r},t)e^{-ik_pz+i\omega_pt}\rightarrow\hat{Q}_{12}(\mathbf{r},\tau)e^{i\omega_p\tau} ,$$
$$\hat{\sigma}_{21}(\mathbf{r},t)=\hat{Q}_{21}(\mathbf{r},t)e^{ik_pz-i\omega_pt}\rightarrow\hat{Q}_{21}(\mathbf{r},\tau)e^{-i\omega_p\tau} , \tag{3.24}$$
$$\hat{w}(\mathbf{r},t)\rightarrow\hat{Q}(\mathbf{r},\tau) .$$

We finally get

$$\frac{d\hat{\sigma}_{21}}{d\tau}=-i\Delta\hat{\sigma}_{21}-\frac{\gamma}{2}\hat{\sigma}_{21}-i\frac{\hat{\Omega}^{(-)}}{2}\hat{w}+\hat{F}_2^{+} ,$$
$$\frac{d\hat{\sigma}_{12}}{d\tau}=i\Delta\hat{\sigma}_{12}-\frac{\gamma}{2}\hat{\sigma}_{12}+i\hat{w}\frac{\hat{\Omega}^{(+)}}{2}+\hat{F}_2 , \tag{3.25}$$
$$\frac{d\hat{w}}{d\tau}=-\gamma(\hat{w}-w_0)+i(\hat{\Omega}^{(-)}\hat{\sigma}_{12}-\hat{\sigma}_{21}\hat{\Omega}^{(+)})+\hat{F}_1 ,$$

with $\Delta=\omega_p-\omega_{21}$, the pump detuning, and

$$\hat{F}_2^{\dagger}(\mathbf{r},\tau)=\hat{f}_{21}(\mathbf{r},\tau)e^{-i\omega_p\tau} =\hat{\Omega}_{\rm vac}^{(-)}(\mathbf{r},\tau)\hat{w}(\mathbf{r},\tau)e^{-i\omega_p\tau} ,$$
$$\hat{F}_1(\mathbf{r},\tau)=\hat{f}=-2(\hat{\sigma}_{21}e^{i\omega_p\tau}\hat{\Omega}_{\rm vac}^{(+)}+\hat{\Omega}_{\rm vac}^{(-)}\hat{\sigma}_{12}e^{-i\omega_p\tau}) , \tag{3.26}$$
$$\hat{\Omega}_{\rm vac}^{(-)}(\mathbf{r},\tau)=-\sum_{\lambda}g_{\lambda}^{*}e^{-i\mathbf{k}_{\lambda}\cdot\mathbf{r}}\hat{a}_{\lambda}^{\dagger}(0)e^{i\omega_{\lambda}(\tau+z/c)} .$$

These equations are operator Bloch equations; however, they are different from the Bloch equations for the density-matrix elements. Equations (3.25) are for operators rather than for the c-number elements of the density matrix. As a result they should preserve the commutation relations between the atomic operators [Eq. (3.15)]. This is indeed the case in spite of the damping terms in the equations. This damping is compensated by the Langevin forces, denoted by $\hat{F}$'s. Thus the Langevin forces are crucial in sustaining the operator character of the variables.

A similar situation is encountered in the study of electromagnetic waves in cavities [26]. A mode of electromagnetic radiation, described by creation and annihilation operators, is damped due to leaking of the radiation from the cavity. The creation and annihilation operators, however, cannot be damped to zero without violating the commutation relations. Careful analysis of the damped mode shows that in addition to damping, a fluctuating Langevin force exists which drives the annihilation and creation operators in such a way that the commutation relations are preserved. In the atomic case we are discussing here, the situation is somewhat more complicated. Namely the Langevin forces are not simply additive, as in the case of electromagnetic modes in a cavity, but rather have the form of a multiplicative noise. This is because, as is clearly seen from Eq. (3.26), the Langevin forces are proportional to the collective atomic operators $\hat{w}(\mathbf{r},t)$ and $\hat{\sigma}(\mathbf{r},t)$ which themselves depend on Langevin forces. Because the Langevin forces are not additive the complete solution to these equations may be difficult to find. We will, therefore, in Sec. IV, use a simplified treatment in which we linearize the Langevin forces around the steady state (nonfluctuating) solution and effectively replace the multiplicative noise by an additive noise. This will be a good approximation in our case since to zeroth order the steady-state elements are determined by the strong (nonfluctuating) pump field.

IV. PERTURBATIVE SOLUTIONS FOR THE ATOMIC DYNAMICS

For clarity and convenience we rewrite Eq. (3.25) in matrix form,

$$\frac{d\hat{\boldsymbol{\sigma}}}{d\tau}=\mathcal{A}\hat{\boldsymbol{\sigma}}+\hat{\boldsymbol{\sigma}}_0+\mathcal{G}\hat{\boldsymbol{\sigma}}+\hat{\mathbf{F}} , \tag{4.1}$$

with the last two terms $\mathcal{G}\hat{\boldsymbol{\sigma}}$ and $\hat{\mathcal{F}}$ expressed in normal ordering, where

$$\hat{\boldsymbol{\sigma}}=\begin{bmatrix}\hat{\sigma}_{21}\\ \hat{\sigma}_{12}\\ \hat{w}\end{bmatrix} , \tag{4.2}$$

$$\hat{\boldsymbol{\sigma}}_0=\begin{bmatrix}0\\ 0\\ \gamma\hat{w}_0\end{bmatrix} , \tag{4.3}$$

$$\mathcal{A}=\begin{bmatrix} -i\Delta-\frac{\gamma}{2} & 0 & -i\frac{\Omega_p}{2} \\ 0 & i\Delta-\frac{\gamma}{2} & i\frac{\Omega_p^*}{2} \\ -i\Omega_p^* & i\Omega_p & -\gamma \end{bmatrix}, \tag{4.4}$$

$$\mathcal{G}=\begin{bmatrix} 0 & 0 & \mathcal{G}_{13} \\ 0 & 0 & \mathcal{G}_{23} \\ \mathcal{G}_{31} & \mathcal{G}_{32} & 0 \end{bmatrix}, \tag{4.5}$$

with

$$\begin{aligned} \mathcal{G}_{13}&=-i[\delta\hat{\Omega}_p^{(-)}+\hat{\Omega}_s^{(-)}e^{-i\delta\tau}+\hat{\Omega}_4^{(-)}e^{i\delta\tau}]/2 , \\ \mathcal{G}_{23}&=\mathcal{G}_{13}^\dagger , \\ \mathcal{G}_{31}&=-2\mathcal{G}_{13}^\dagger , \\ \mathcal{G}_{32}&=-2\mathcal{G}_{13} , \end{aligned} \tag{4.6}$$

and

$$\hat{\mathbf{F}}=\begin{bmatrix} \hat{F}_2^\dagger \\ \hat{F}_2 \\ \hat{F}_1 \end{bmatrix} . \tag{4.7}$$

Essentially the same set of equations is used to describe the initiation of superfluorescence and Raman scattering [13–15], but then linearization about steady state corresponds to linearization with respect to the initial $\tau=0$ solutions. Thus in those cases, the initial conditions of the system are very important, because one is interested in short-time behavior (compared with the characteristic time of the system). Here, on the other hand, we are interested in the steady state, long-time limit with all the initial conditions damped away. This is the regime in which the dynamics dominate over the initial conditions.

We will now examine the long-time behavior of the atomic operators. They, of course, do not reach stationary values, since the terms proportional to the Langevin forces lead to fluctuations around their average values. If both the generated fields and the Langevin forces (vacuum fields) are small compared with the pump field, as in the case studied here, we may linearize our equations. Essentially the fluctuations in $\hat{\boldsymbol{\sigma}}$ and $\hat{w}$ due to the Langevin forces are small compared to their mean values.

First we can find the transformation matrix $\mathcal{T}$ that diagonalizes the coefficient matrix $\mathcal{A}$,

$$\mathcal{T}^{-1}\mathcal{A}\mathcal{T}=\Lambda=\begin{bmatrix} \Lambda_1 & 0 & 0 \\ 0 & \Lambda_2 & 0 \\ 0 & 0 & \Lambda_3 \end{bmatrix} . \tag{4.8}$$

The generic solution is

$$\mathcal{T}=(\mathcal{Y}_1,\mathcal{Y}_2,\mathcal{Y}_3) , \tag{4.9}$$

with $\mathcal{Y}_1,\mathcal{Y}_2,\mathcal{Y}_3$ the corresponding eigenvectors of matrix $\mathcal{A}$. That is,

$$\mathcal{A}\mathcal{Y}_j=\Lambda_j\mathcal{Y}_j , \quad j=1,2,3 . \tag{4.10}$$

The cubic equation (comparable to the "Mollow cubic") that determines the eigenvalues is

$$\left[\Lambda+\frac{\gamma}{2}\right]^3+\frac{\gamma}{2}\left[\Lambda+\frac{\gamma}{2}\right]^2+(|\Omega_p|^2+\Delta^2)\left[\Lambda+\frac{\gamma}{2}\right] +\frac{\gamma}{2}\Delta^2=0 . \tag{4.11}$$

This secular equation has all real coefficients, so the complex solutions must occur in conjugate pairs [27]. With $\Lambda_j=-\Gamma_j+i\delta_j$, for $j=p,s,4$ in the following order:

$$\begin{aligned} \Lambda_1&=\Lambda_s=-\Gamma-i\delta , \\ \Lambda_2&=\Lambda_4=-\Gamma+i\delta , \\ \Lambda_3&=\Lambda_p=-\Gamma_p . \end{aligned} \tag{4.12}$$

All the eigenvalues contain a negative real part, which ensures that the atomic operators will damp to around some stationary value.

We now can solve for the pump field to all orders, which reduces to the solution of

$$\frac{d\hat{\boldsymbol{\sigma}}_s}{d\tau}=\mathcal{A}\hat{\boldsymbol{\sigma}}_s+\hat{\boldsymbol{\sigma}}_0=0 , \tag{4.13}$$

where $\hat{\boldsymbol{\sigma}}_s$ is the steady-state solution of the above equation, and as given in Appendix C,

$$\hat{\boldsymbol{\sigma}}_s=-\mathcal{A}^{-1}\hat{\boldsymbol{\sigma}} = \frac{w_0}{\Delta^2+\frac{\gamma^2}{4}+\frac{|\Omega_p|^2}{2}}\begin{bmatrix} -\left[\Delta+i\frac{\gamma}{2}\right]\frac{\Omega_p}{2} \\ -\left[\Delta-i\frac{\gamma}{2}\right]\frac{\Omega_p^*}{2} \\ \Delta^2+\frac{\gamma}{4^2} \end{bmatrix} . \tag{4.14}$$

Since we want solutions to first order both in weak fields and in the Langevin forces (which act as the source of the weak fields), we can linearize the $\hat{\boldsymbol{\sigma}}$ equations around the $\hat{\boldsymbol{\sigma}}_s$ as

$$\delta\hat{\boldsymbol{\sigma}}=\hat{\sigma}-\hat{\boldsymbol{\sigma}}_s=\begin{bmatrix} \hat{\sigma}_{21}-\hat{\sigma}_{21}^s \\ \hat{\sigma}_{12}-\sigma_{12}^s \\ \hat{w}-\hat{w}^s \end{bmatrix}=\begin{bmatrix} \delta\hat{\sigma}_{21} \\ \delta\hat{\sigma}_{12} \\ \delta\hat{w} \end{bmatrix} . \tag{4.15}$$

Then we have

$$\frac{d\delta\hat{\boldsymbol{\sigma}}}{d\tau}=\mathcal{A}\delta\hat{\boldsymbol{\sigma}}+\hat{\mathbf{F}}+\mathcal{G}\hat{\boldsymbol{\sigma}}_s , \tag{4.16}$$

with

$$\begin{aligned} \hat{F}_1(\mathbf{r},\tau)&=-2(\hat{\sigma}_{21}^s\hat{\Omega}_{\rm vac}^{(+)}e^{i\omega_p\tau}+\hat{\Omega}_{\rm vac}^{(-)}\hat{\sigma}_{12}^s e^{-i\omega_p\tau}) , \\ \hat{F}_2(\mathbf{r},\tau)&=\hat{w}^s(\mathbf{r},\tau)\hat{\Omega}_{\rm vac}^{(+)}(\mathbf{r},\tau)e^{i\omega_p\tau} . \end{aligned} \tag{4.17}$$

After the linearization approximation, the normal ordering in expressions $\mathcal{G}\hat{\boldsymbol{\sigma}}$ and $\hat{\mathbf{F}}$ is not important any more, as $\mathcal{G}\hat{\sigma}_s$ and $\hat{\mathbf{F}}(\hat{\sigma}_s)$ are linear in the field operators. Transforming into the eigenvector representation

$$\hat{\mathcal{X}}=\mathcal{T}^{-1}\hat{\sigma} \;,\quad \hat{\mathcal{X}}=\mathcal{T}^{-1}\hat{\sigma}_s \;,\quad \delta\hat{\mathcal{X}}=\hat{\mathcal{X}}-\hat{\mathcal{X}}_s \;, \tag{4.18}$$

we get

$$\frac{d\delta\hat{\mathcal{X}}}{d\tau}=\Lambda\delta\hat{\mathcal{X}}+\mathcal{T}^{-1}\hat{\mathbf{F}}+\mathcal{T}^{-1}\mathcal{G}\hat{\sigma}_s \;. \tag{4.19}$$

The solution is

$$\delta\hat{\mathcal{X}}_j(\tau)=\delta\hat{\mathcal{X}}_j(0)e^{\Lambda_j\tau}+\int_0^{\tau}e^{\Lambda_j(\tau-\tau')}[\mathcal{T}^{-1}\hat{\mathbf{F}}(\tau')]_j d\tau' + \int_0^{\tau}e^{\Lambda_j(\tau-\tau')}[\mathcal{T}^{-1}\mathcal{G}\hat{\sigma}_s(\tau')]_j d\tau' \;. \tag{4.20}$$

The first term is due to the nonzero initial conditions, which will of course vanish in the steady-state limit. The remaining terms are the linear responses due to the Langevin and weak-field terms. The part due to the Langevin terms depends on $\hat{\mathbf{F}}$, which describes the fluctuations of the electromagnetic vacuum. It is very important to realize that the atomic raising operator $\delta\hat{\sigma}_{21}$ as written here depends not only on the creation operator $\hat{\Omega}_{\rm vac}^{(-)}$ but also on the annihilation operator $\hat{\Omega}_{\rm vac}^{(+)}$. Similarly the lowering operator $\delta\hat{\sigma}_{12}$ depends on both the creation and annihilation operators $\hat{\Omega}_{\rm vac}^{(\pm)}$. These results were obtained in the framework of the local-field dipole approximation and RWA. The mechanism for the coupling between the positive and negative-frequency parts of the field is provided by the strong pump field. Observe that the atomic operators depend on the electromagnetic vacuum field via a time integral over times prior to τ. Therefore not all the frequencies that contribute to the electromagnetic vacuum will significantly contribute to the atomic operators. The electromagnetic field is effectively "filtered" by the atomic system (see Appendix E). It is only those frequencies of the vacuum spectrum which are centered around one of the resonances, given by imaginary parts of Λ_j, and which have a bandwidth comparable to the real part of Λ_j, that significantly contribute to the fluctuations of the atomic operators. This filtering is crucial to understanding the long correlation lengths that give rise to the Cherenkov-type emission. The part of $\delta\hat{\sigma}_{21}$ that depends on the weak fields is essentially proportional to the $\chi^{(3)}$, the third-order susceptibility, which has a form familiar from the theory of 4WM. In the long-time limit the $(\mathcal{G}\sigma_s)$ reaches a steady state and the integrals in Eq. (4.20) are easily performed (see Appendix D). The solution of $\delta\hat{\sigma}_{21}$ is then

$$\delta\hat{\sigma}_{21}(\tau)=\delta\hat{\sigma}_{21}^{1}+\delta\hat{\sigma}_{21}^{2} \;. \tag{4.21}$$

$\delta\sigma_{21}^{1}$ will be discussed in Sec. V, where we will detail its interesting long correlation length properties. The linear-response part (stimulated part) of the polarization due to the weak fields is given by

$$\delta\hat{\sigma}_{21}^{2}(\tau)=\tilde{\alpha}_p\delta\hat{\Omega}_p^{(-)}+\tilde{\kappa}_p\delta\hat{\Omega}_p^{(+)}+\tilde{\alpha}_s\hat{\Omega}_s^{(-)}e^{-i\delta\tau} +\tilde{\kappa}_4\hat{\Omega}_4^{(+)}e^{-i\delta\tau}+\tilde{\alpha}_4\hat{\Omega}_4^{(-)}e^{i\delta\tau}+\tilde{\kappa}_s\hat{\Omega}_s^{(+)}e^{i\delta\tau} \tag{4.22}$$

where the $\tilde{\alpha}$'s and $\tilde{\kappa}$'s are given in Appendix D.

V. LONG COHERENCE LENGTH QUANTUM NOISE

In the above we have solved the atomic operator equations by keeping all orders in the strong pump field, and perturbatively to first order in the noise and weak-field terms. We have found that the solution is just the sum of the linear responses of both small signals. As detailed in Appendix D, the linear response due to the Langevin noise is

$$\delta\hat{\sigma}_{21}^{1}=\hat{\hat{V}}_p+\hat{\hat{V}}_s+\hat{\hat{V}}_4 \;, \tag{5.1}$$

where

$$\hat{\hat{V}}_p=\mathcal{D}_p^{(-)}\hat{\hat{\Omega}}_{p\rm vac}^{(-)}+\mathcal{D}_p^{(+)}\hat{\hat{\Omega}}_{p\rm vac}^{(+)} \;,\quad \hat{\hat{V}}_s=\mathcal{D}_s^{(-)}\hat{\hat{\Omega}}_{s\rm vac}^{(-)}+\mathcal{D}_s^{(+)}\hat{\hat{\Omega}}_{s\rm vac}^{(+)} \;,\quad \hat{\hat{V}}_4=\mathcal{D}_4^{(-)}\hat{\hat{\Omega}}_{4\rm vac}^{(-)}+\mathcal{D}_4^{(+)}\hat{\hat{\Omega}}_{4\rm vac}^{(+)} \;, \tag{5.2}$$

with

$$\hat{\hat{\Omega}}_{j\rm vac}^{(\pm)}=\int_0^{\tau}e^{\Lambda_j(\tau-\tau')}\hat{\Omega}_{\rm vac}^{(\pm)}e^{\pm i\omega_p\tau'}d\tau' \;. \tag{5.3}$$

The expressions for the $\mathcal{D}_j^{(\pm)}$ are given in Appendix D. As we show in Appendix E, Eq. (5.3) with the damped kernel is equivalent to a frequency filtering in the spectrum. The effective filter is centered at the central frequencies of the three fields we are describing and has a bandwidth determined by the damping rate. In general we will have problems of convergence due to large frequencies when we try to evaluate the correlation functions of the above-filtered vacuum. One standard way of getting rid of the divergence is the flat spectrum or pole approximation, which amounts to replacing the density of continuous modes by a constant evaluated at the center frequency. This is equivalent to saying that the filter is sufficiently strong so that it allows only the passband modes to go through.

We have separated expression Eq. (5.1) for the polarization into three parts corresponding to the time variation part of the resulting polarization. $\hat{\hat{\Omega}}_{p\rm vac}^{(\pm)}$ is the field with center frequency at ω_p, and with resulting polarization at ω_p. $\hat{\hat{\Omega}}_{s\rm vac}^{(-)}$ is the field with center frequency at ω_s, and resulting polarization at ω_s. $\hat{\hat{\Omega}}_{s\rm vac}^{(+)}$ is the field with center frequency at ω_4, and resulting polarization at ω_s. $\hat{\hat{\Omega}}_{4\rm vac}^{(-)}$ is the field with center frequency at ω_4, and resulting polarization at ω_4. $\hat{\hat{\Omega}}_{4\rm vac}^{(+)}$ is the field with center frequency at ω_s, and resulting polarization at ω_4. Thus terms $\hat{\hat{\Omega}}_{s\rm vac}^{(-)}$ and $\hat{\hat{\Omega}}_{4\rm vac}^{(+)}$ behave like spontaneous four-wave mixing.

A very important comment is in order here. As mentioned earlier, the vacuum fluctuations will be partitioned among the three fields of interest, which are identified by the three eigenvalues listed above. However, the pump field will propagate in the medium as well. It is by no means a constant plane-wave field; rather, due to self-focusing, diffraction, etc., it will be a space-time-dependent quantity, so at different locations in the medium, the generated fields will center at different frequencies $\omega_p+\delta_j[\Omega_p(\mathbf{r})]$, and will have different widths $\Gamma_j[\Omega_p(\mathbf{r})]$. Thus the distribution of the pump field strength will cause the separation of the triplet spectrum to vary, but as the center of the triplet is always the same as the pump frequency, and the two sidebands are at least Δ apart, we can usually replace the center frequencies of the sidebands with that corresponding to the saturated field strength in the filaments [4]. We then simply assume that the sidebands are coherent irrespective of the exact position of the center frequency. These coherent fields are then additive. If, on the other hand, we also want to look at the spectrum of the generated fields, we have to be more careful.

Since we are performing our calculations in terms of normal ordering of the field operators, and we are only interested in the intensity distribution, the nonvanishing correlation functions that will contribute to the far-field normal-ordered intensity are of the following antinormal-ordered form:

$$D_{jj'}(\mathbf{r},\mathbf{r}';\tau)=\langle \hat{\hat{\Omega}}^{(+)}_{j\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(+)\dagger}_{j'\text{vac}}(\mathbf{r}',\tau)\rangle \ . \tag{5.4}$$

In the above, $\langle\ \rangle$ represents the quantum-mechanical expectation value over the state vector $|\Psi(t)\rangle$, which is

$$|\Psi(t)\rangle=|\Psi(t=0)\rangle=|\Psi_{\text{field}}\rangle\otimes|\Psi_{\text{atoms}}\rangle \ , \tag{5.5}$$

with

$$|\Psi_{\text{field}}\rangle=|\text{vacuum}\rangle \ , \quad |\Psi_{\text{atoms}}\rangle=|\text{all the atoms in the ground-state level 1}\rangle \ . \tag{5.6}$$

Before we proceed to calculate the correlation function of the quantum-noise fields, we briefly review the properties of the vacuum fluctuations, and point out some special cases of interest. For the free vacuum field,

$$\hat{a}_\lambda(t)=\hat{a}_\lambda(0)e^{-i\omega_\lambda t} \ . \tag{5.7}$$

Then according to the expansion Eq. (1.2), we get

$$\begin{aligned}\langle \hat{E}^{(+)}_{\text{vac}}(\mathbf{r},t)\hat{E}^{(-)}_{\text{vac}}(\mathbf{r}',t')\rangle &= \sum_\lambda (2\pi\hbar\omega_\lambda/V)e^{i\mathbf{k}_\lambda\cdot(\mathbf{r}-\mathbf{r}')-i\omega_\lambda(t-t')}\\ &=2\left[\frac{1}{2\pi}\right]^3\int d\mathbf{k}_\lambda(2\pi\hbar\omega)e^{i\mathbf{k}_\lambda\cdot(\mathbf{r}-\mathbf{r}')-i\omega(t-t')}\\ &=\frac{\hbar c}{\pi|\mathbf{r}-\mathbf{r}'|}\int_0^\infty(e^{ik_\lambda r_-}-e^{-ik_\lambda r_+})k_\lambda^2dk_\lambda \ ,\end{aligned} \tag{5.8}$$

where $r_\pm$ are defined by

$$r_-=|\mathbf{r}-\mathbf{r}'|-(t-t') \ , \quad r_+=|\mathbf{r}-\mathbf{r}'|+(t-t') \ . \tag{5.9}$$

The above integration has been discussed elsewhere [28,29]; it is singular at $r_\pm$, and can be expressed in terms of the δ function $\delta(r_\pm)$ and its derivatives under proper regularizations. The correlation properties of the vacuum field are such that two points of space-time which cannot be connected by light signals are not correlated at all. However, as is often the case for calculations in quantum optics with vacuum fields, we are interested in a limited frequency band, and therefore a flat spectrum may be assumed within that band. In the present case, a limited band around the atomic resonance is the most important part, so instead of going through rigorous algebra, we will investigate the properties under the flat spectrum approximation. Thus

$$\begin{aligned}\langle \hat{E}^{(+)}_{\text{vac}}(\mathbf{r},t)\hat{E}^{(-)}_{\text{vac}}(\mathbf{r}',t')\rangle &= \frac{\hbar c}{\pi|\mathbf{r}-\mathbf{r}'|}\int_0^\infty(e^{ik_\lambda r_-}-e^{-ik_\lambda r_+})k_\lambda^2dk_\lambda\\ &\approx \frac{\hbar c}{\pi|\mathbf{r}-\mathbf{r}'|}k_{21}^2\left[e^{ik_{21}r_-}\int_{-k_{21}}^\infty e^{ik'r_-}dk'-e^{-ik_{21}r_+}\int_{-k_{21}}^\infty e^{-ik'r_+}dk'\right]\\ &\approx \frac{\hbar c}{\pi|\mathbf{r}-\mathbf{r}'|}k_{21}^2[2\pi\delta(r_-)-2\pi\delta(r_+)] \ ,\end{aligned} \tag{5.10}$$

where $k'=k_\lambda-k_{21}$, and we can see that the essential physics is retained. In terms of the local time τ,

$$\langle \hat{E}^{(+)}_{\text{vac}}(\mathbf{r},\tau)\hat{E}^{(-)}_{\text{vac}}(\mathbf{r}',\tau')\rangle \approx \frac{\hbar c}{\pi|\mathbf{r}-\mathbf{r}'|}k_{21}^2[2\pi\delta(r_-')-2\pi\delta(r'_+)] \ , \tag{5.11}$$

where

$$\begin{aligned} r'_- &=|\mathbf{r}-\mathbf{r}'|-(\tau-\tau')-(z-z')/c \ ,\\ r'_+ &=|\mathbf{r}-\mathbf{r}'|+(\tau-\tau')+(z-z')/c \ .\end{aligned} \tag{5.12}$$

Under the same approximation, we can also discuss the following special cases.

At $t=t'$, Eq. (5.8) reduces to

$$\langle \hat{E}^{(+)}_{\rm vac}(\mathbf{r},t)\hat{E}^{(-)}_{\rm vac}(\mathbf{r}',t)\rangle = 4\pi\hbar\omega_{21}\delta(\mathbf{r}-\mathbf{r}') \ . \tag{5.13}$$

This implies that at the same time the vacuum fields are indeed white noise in space.

At $\mathbf{r}=\mathbf{r}'$, we obtain from Eq. (5.8),

$$\langle \hat{E}^{(+)}_{\rm vac}(\mathbf{r},t)\hat{E}^{(-)}_{\rm vac}(\mathbf{r},t')\rangle \approx 4\hbar k^3_{21}\delta(t-t') \ . \tag{5.14}$$

This approximation has been used in many cases. It implies that the vacuum field is indeed white noise in time if we are not considering any propagation, and imagine the interactions occurring at a point.

At $\tau=\tau'$, we will have from Eq. (5.11)

$$\langle \hat{E}^{(+)}_{\rm vac}(\mathbf{r},\tau)\hat{E}^{(-)}_{\rm vac}(\mathbf{r}',\tau)\rangle \approx \frac{\hbar c}{\pi|\mathbf{r}-\mathbf{r}'|}k^2_{21}[2\pi\delta(R_-)-2\pi\delta(R_+)] \ , \tag{5.15}$$

where $R_\pm$ are defined by

$$R_- = |\mathbf{r}-\mathbf{r}'|-(z-z') \ , \quad R_+ = |\mathbf{r}-\mathbf{r}'|+(z-z') \ . \tag{5.16}$$

All the above correlation functions are only functions of the difference variables $\mathbf{r}-\mathbf{r}'$, $t-t'$, or $\tau-\tau'$. They are preserved under the translation $\mathbf{r}\to\mathbf{r}+\delta\mathbf{r}$, $t\to t+\delta t$, or $\tau\to\tau+\delta\tau$ at all times. That is why we sometimes say that the vacuum field is stationary. But it is *not* of the following form:

$$\langle \hat{E}^{(+)}_{\rm vac}(\mathbf{r},\tau')\hat{E}^{(-)}_{\rm vac}(\mathbf{r}',\tau')\rangle \neq (\ \cdots\ )\delta(\mathbf{r}-\mathbf{r}')\delta(\tau-\tau') \ , \tag{5.17}$$

or any other factorizable form between time and space. Note, however, that Eq. (5.17) may be a good model for collisional Langevin-type noise [15], since no propagation is involved in the collisional process.

In our case, at $\tau=0$, before the interaction is turned on, the initial condition for the above-defined correlation function Eq. (5.4) is the same as for the vacuum field case $\tau=\tau'$ of Eq. (5.15). After the interaction is turned on, the atomic medium acts as a filter, and the spatial coherence develops in the polarization of the medium at the various frequencies due to the vacuum fields.

We note that in the transient situation, for example, SF initiation, it is the δ-correlated [Eq. (5.14)] incident vacuum at the entrance plane that is important. We are interested in the other limit, namely the stationary limit, when the interaction becomes the dominant dynamics and the initial conditions have all died away. We will find that in this case, the above correlation is not δ correlated, but has some typical coherence length δR. We will also find that if δk is the spread in wave vector passed by the filter, the correlation length δR is qualitatively given by $\delta k\delta R\sim 1$, which is the Heisenberg uncertainty relation.

In the long-time limit, we get the filtered vacuum

$$\begin{aligned}\hat{\Omega}^{(+)}_{j\rm vac}(\mathbf{r},\tau) &= \sum_\lambda \int_0^\tau e^{\Lambda_j(\tau-\tau')}g_\lambda e^{i\mathbf{k}_\lambda\cdot\mathbf{r}}\hat{a}_\lambda(0) \times e^{-i\omega_\lambda(\tau'+z/c)}e^{i\omega_p\tau'}d\tau' \\ &\sim \sum_\lambda g_\lambda e^{i\mathbf{k}_\lambda\cdot\mathbf{r}}\hat{a}_\lambda(0)e^{-i\omega_\lambda z/c}e^{i(\omega_p-\omega_\lambda)\tau} \times \frac{1}{-\Lambda_j-i\omega_\lambda+i\omega_p} \ .\end{aligned} \tag{5.18}$$

Since $\hat{\Omega}^{(+)}_{j\rm vac}(\mathbf{r},\tau)$ occurs in the dipole polarization $\delta\hat{\sigma}^1_{21}$, as we will see in Sec. VI, it will be used to determine the slowly varying envelope of the fields of interest. Thus we expect that important contributions to the polarization will also be slowly varying. Since the filtering is centered around one of the Mollow triplets, we should expect the contribution from the k_λ integral to come mainly from $k_\lambda\sim\omega_j/c$, with $\omega_j=\omega_p+\mathrm{Im}(\Lambda_j)$. This is of course consistent with our SVEA, RWA, and the flat spectrum (pole) approximations. This also means that for positive k_λ, it is the main source for the copropagating waves, and for negative k_λ, the source for the counterpropagating waves. In Appendix F we find in the steady-state limit,

$$D_{jj'}(\mathbf{r},\mathbf{r}';\tau) = -i\delta_{jj'}\frac{2}{\hbar}\frac{|\mathbf{d}_{21}|^2}{(2\pi c)^2}\frac{\pi}{\Gamma_j}\left[\omega_j^2+\frac{c^2\partial^2}{\partial(x-x')^2}\right]\frac{1}{|\mathbf{r}-\mathbf{r}'|}[e^{-\Gamma_j(|R_-|/c)}e^{i\omega_j(R_-/c)}-e^{-\Gamma_j(|R_+|/c)}e^{-i\omega_j(R_+/c)}] \ . \tag{5.19}$$

The long coherence length is obviously given by

$$L_c\sim\frac{c}{\Gamma_j} \ , \tag{5.20}$$

which is consistent with the Heisenberg uncertainty relation. This length is, in most cases of interest, longer than the cell length. Equation (5.18) is not, however, convenient for determining the slowly varying fields $\hat{E}_s$ and $\hat{E}_4$ since it has a singularity at $\mathbf{r}=\mathbf{r}'$. The SVEA implies that it is sufficient to consider as their source the quantities $\hat{\hat{V}}_j$, which are averaged over a volume element that is large compared to λ_p^3. Of course, to be consistent, we should ultimately make sure that our observable does not depend on the size of the volume element. Alternatively for angle θ, we require $a\sim 1/(k_j\theta)$.

Some of these interesting results have already shown up in a pair of two-level atoms spontaneous-emission and resonance fluorescence theory, and they are also ultimately related to the time delay due to the propagation behavior of the field [30].

VI. FIELD-OPERATOR EQUATIONS AND PARAXIAL APPROXIMATION

We now derive the field-operator equations. The polarization

$$\hat{\mathbf{P}}(\mathbf{r},t)=\hat{\mathbf{P}}_B(\mathbf{r},t)+\hat{\mathbf{P}}_A(\mathbf{r},t) \tag{6.1}$$

consists of two parts, where

$$\hat{\mathbf{P}}_B(\mathbf{r},\tau)=\hat{\mathbf{P}}_{Bp}(\mathbf{r},\tau)+\hat{\mathbf{P}}_{Bs}(\mathbf{r},\tau)+\hat{\mathbf{P}}_{B4}(\mathbf{r},\tau) \tag{6.2}$$

is the induced (nonresonant) polarization of the background medium (e.g., buffer gases). The usual linear relation between the polarization and the field holds, and

$$\begin{aligned}\hat{\mathbf{P}}^{(-)}_{Bp}(\mathbf{r},\tau)&=\chi_0(\omega_p)\hat{\mathbf{E}}^{(-)}_p ,\\ \hat{\mathbf{P}}^{(-)}_{Bs}(\mathbf{r},\tau)&=\chi_0(\omega_s)\hat{\mathbf{E}}^{(-)}_s ,\\ \hat{\mathbf{P}}^{(-)}_{B4}(\mathbf{r},\tau)&=\chi_0(\omega_4)\hat{\mathbf{E}}^{(-)}_4 ,\end{aligned} \tag{6.3}$$

where $\chi_0(\omega)$ is the linear susceptibility at that frequency with the resonant term omitted, as mentioned in Sec. III, we will neglect this part. The resonant part of the polarization

$$\hat{\mathbf{P}}_A(\mathbf{r},\tau)=\sum_j \hat{\mathbf{P}}^j\delta^3(\mathbf{r}-\mathbf{r}^{(j)}) \tag{6.4}$$

is due to the active medium. We have treated this part in more detail by solving the atomic-operator equations in Secs. IV and V.

As remarked earlier, we have performed the collective variable description approximation for the atomic operators, and both SVEA and RWA have been made for all the quantities except the polarization terms due to the vacuum field. Thus to be consistent with the SVEA we have to perform a volume average of the source terms, i.e., those that we have shown in Sec. V to have a long coherence length. Hence

$$\hat{\mathbf{P}}^j=\mathbf{d}^{(j)}_{12}\hat{\sigma}^{(j)}_{12}+\mathbf{d}^{(j)}_{21}\hat{\sigma}^{(j)}_{21} , \tag{6.5}$$

$$\begin{aligned}\hat{\mathbf{P}}_A(\mathbf{r},\tau)&=[[\hat{\mathbf{P}}(\mathbf{r},\tau)^j]]_{\rm av}\\ &=N[[\mathbf{d}_{21}\hat{\sigma}_{21}e^{i\omega_p\tau}+\mathbf{d}_{12}\hat{\sigma}_{12}e^{-i\omega_p\tau}]]_{\rm av} ,\end{aligned} \tag{6.6}$$

where $[[\ ]]_{\rm av}$ represents the appropriate volume average over size larger than λ_p^3. We stress again that if the SVEA is valid, then the final result should not depend on the specific volume chosen. We choose the volume element to be such that it is a sphere centered at position $\mathbf{r}$ with the following weighting function [31]:

$$u(\mathbf{r})=(\pi a^2)^{-3/2}e^{-r^2/a^2} , \tag{6.7}$$

then

$$[[\hat{\Omega}^{(\pm)}_{j\rm vac}(\mathbf{r},\tau)]]_{\rm av}=\int_V d\mathbf{r}'\hat{\Omega}^{(\pm)}_{j\rm vac}(\mathbf{r}',\tau)u(\mathbf{r}-\mathbf{r}') . \tag{6.8}$$

It is easy to work out as

$$\int_{-\infty}^{\infty}dx'e^{i\mathbf{k}_{\lambda x}x'}e^{-(x-x')^2/a^2}=(\pi a^2)^{1/2}e^{i\mathbf{k}_{\lambda x}x-k^2_{\lambda x}a^2/4} , \tag{6.9}$$

so we have

$$(\pi a^2)^{-3/2}\int_V d\mathbf{r}'e^{i\mathbf{k}_\lambda\cdot\mathbf{r}'}e^{-ik_\lambda z'}e^{-|\mathbf{r}-\mathbf{r}'|^2/a^2} =e^{i\mathbf{k}_\lambda\cdot\mathbf{r}-k_\lambda^2a^2/2+k_\lambda k_{\lambda z}a^2/2-ik_\lambda z} . \tag{6.10}$$

Then we obtain the volume-averaged source due to the vacuum field, which is in principle slowly varying over a distance of the order of a,

$$\begin{aligned}[[\hat{\Omega}^{(+)}_{j\rm vac}(\mathbf{r},\tau)]]_{\rm av}&=\sum_\lambda g_\lambda\hat{a}_\lambda(0)\frac{e^{i\mathbf{k}_\lambda\cdot\mathbf{r}-k_\lambda^2a^2/2+k_\lambda k_{\lambda z}a^2/2-ik_\lambda z}e^{i(\omega_p-\omega_\lambda)\tau}}{-\Lambda_j-i\omega_\lambda+i\omega_p}\\ &=[[\hat{\Omega}^{(+)}_{j\rm vac+}(\mathbf{r},\tau)]]_{\rm av}+[[\hat{\Omega}^{(+)}_{j\rm vac-}(\mathbf{r},\tau)]]_{\rm av} ,\end{aligned} \tag{6.11}$$

where

$$\begin{aligned}[[\hat{\Omega}^{(+)}_{j\rm vac+}(\mathbf{r},\tau)]]_{\rm av}&=\sum_{k_{\lambda z}>0} g_\lambda\hat{a}_\lambda(0)\frac{e^{i\mathbf{k}_\lambda\cdot\mathbf{r}-k_\lambda^2a^2/2+k_\lambda k_{\lambda z}a^2/2-ik_\lambda z}e^{i(\omega_p-\omega_\lambda)\tau}}{-\Lambda_j-i\omega_\lambda+i\omega_p} ,\\ [[\hat{\Omega}^{(+)}_{j\rm vac-}(\mathbf{r},\tau)]]_{\rm av}&=\sum_{k_{\lambda z}<0} g_\lambda\hat{a}_\lambda(0)\frac{e^{i\mathbf{k}_\lambda\cdot\mathbf{r}-k_\lambda^2a^2/2+k_\lambda k_{\lambda z}a^2/2-ik_\lambda z}e^{i(\omega_p-\omega_\lambda)\tau}}{-\Lambda_j-i\omega_\lambda+i\omega_p} .\end{aligned} \tag{6.12}$$

We have used $\pm$ to index the copropagating and counterpropagating waves. As we can see, the above integral is not only restricted by the filtering function to the widths of each individual element of the Mollow triplet, but also the direction is restricted to be mainly along the z axis (copropagating) or opposite to the direction of the z axis (counterpropagating). Similar definitions for $[[\hat{\Omega}^{(-)}_{j\rm vac\pm}(\mathbf{r},\tau)]]_{\rm av}$ also hold. Thus in principle we can write

$$\begin{aligned}[[\hat{V}_p(\mathbf{r},\tau)]]_{\rm av}&=\hat{\beta}_{p+}+\hat{\beta}_{p-}e^{2ik_pz}+\hat{\beta}'_{p-}e^{-2ik_pz} ,\\ [[\hat{V}_s(\mathbf{r},\tau)]]_{\rm av}&=\hat{\beta}_{s+}e^{-i\delta\tau}+\hat{\beta}_{s-}e^{-i\delta\tau}e^{2ik_sz}\\ &\quad+\hat{\beta}'_{s-}e^{-i\delta\tau}e^{-2ik_4z} ,\\ [[\hat{V}_4(\mathbf{r},\tau)]]_{\rm av}&=\hat{\beta}_{4+}e^{i\delta\tau}+\hat{\beta}_{4-}e^{i\delta\tau}e^{2ik_4z}+\hat{\beta}'_{4-}e^{i\delta\tau}e^{-2ik_sz} .\end{aligned} \tag{6.13}$$

Written out explicitly, we have

$$\hat{\hat{\beta}}_{p+}(\mathbf{r},\tau)=\mathcal{D}_p^{(-)}[[\hat{\hat{\Omega}}^{(-)}_{p\text{vac}+}(\mathbf{r},\tau)]]_{\text{av}}+\mathcal{D}_p^{(+)}[[\hat{\hat{\Omega}}^{(+)}_{p\text{vac}+}(\mathbf{r},\tau)]]_{\text{av}}\,,$$
$$\hat{\hat{\beta}}_{p-}(\mathbf{r},\tau)e^{2ik_pz}=\mathcal{D}_p^{(-)}[[\hat{\hat{\Omega}}^{(-)}_{p\text{vac}-}(\mathbf{r},\tau)]]_{\text{av}}\,, \tag{6.14}$$
$$\hat{\hat{\beta}}'_{p-}(\mathbf{r},\tau)e^{-2ik_pz}=\mathcal{D}_p^{(+)}[[\hat{\hat{\Omega}}^{(+)}_{p\text{vac}-}(\mathbf{r},\tau)]]_{\text{av}}\,,$$

$$\hat{\hat{\beta}}_{s+}(\mathbf{r},\tau)e^{-i\delta\tau}=\mathcal{D}_s^{(-)}[[\hat{\hat{\Omega}}^{(-)}_{s\text{vac}+}(\mathbf{r},\tau)]]_{\text{av}}+\mathcal{D}_s^{(+)}[[\hat{\hat{\Omega}}^{(+)}_{s\text{vac}+}(\mathbf{r},\tau)]]_{\text{av}}\,,$$
$$\hat{\hat{\beta}}_{s-}(\mathbf{r},\tau)e^{-i\delta\tau}e^{2ik_sz}=\mathcal{D}_s^{(-)}[[\hat{\hat{\Omega}}^{(-)}_{s\text{vac}-}(\mathbf{r},\tau)]]_{\text{av}}\,, \tag{6.15}$$
$$\hat{\hat{\beta}}'_{s-}(\mathbf{r},\tau)e^{-i\delta\tau}e^{-2ik_4z}=\mathcal{D}_s^{(+)}[[\hat{\hat{\Omega}}^{(+)}_{s\text{vac}-}(\mathbf{r},\tau)]]_{\text{av}}\,,$$

$$\hat{\hat{\beta}}_{4+}(\mathbf{r},\tau)e^{i\delta\tau}=\mathcal{D}_4^{(-)}[[\hat{\hat{\Omega}}^{(-)}_{4\text{vac}+}(\mathbf{r},\tau)]]_{\text{av}}+\mathcal{D}_4^{(+)}[[\hat{\hat{\Omega}}^{(+)}_{4\text{vac}+}(\mathbf{r},\tau)]]_{\text{av}}\,,$$
$$\hat{\hat{\beta}}_{4-}(\mathbf{r},\tau)e^{i\delta\tau}e^{2ik_4z}=\mathcal{D}_4^{(-)}[[\hat{\hat{\Omega}}^{(-)}_{4\text{vac}-}(\mathbf{r},\tau)]]_{\text{av}}\,, \tag{6.16}$$
$$\hat{\hat{\beta}}'_{4-}(\mathbf{r},\tau)e^{i\delta\tau}e^{-2ik_sz}=\mathcal{D}_4^{(+)}[[\hat{\hat{\Omega}}^{(+)}_{4\text{vac}-}(\mathbf{r},\tau)]]_{\text{av}}\,.$$

$\hat{\hat{\beta}}_{j\pm}(\mathbf{r},\tau)$ and $\hat{\hat{\beta}}'_{j-}(\mathbf{r},\tau)$ are by construction the slowing varying amplitudes. However, we cannot get very compact analytical formulas for them. But we can get the correlation functions of various kinds, which will then relate them to the observable normal-ordered intensity for the fields. In general, the nonvanishing correlation function for the copropagating source reduces to

$$D_{j+}(\mathbf{r},\mathbf{r}';\tau)=[[\langle\hat{\hat{\Omega}}^{(+)}_{j\text{vac}+}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(+)\dagger}_{j\text{vac}+}(\mathbf{r}',\tau)\rangle]]_{\text{av}}\,. \tag{6.17}$$

The copropagating and counterpropagating sources are not correlated, because

$$\langle\hat{\hat{\Omega}}_{j\text{vac}+}(\mathbf{r},\tau)\hat{\hat{\Omega}}_{j'\text{vac}-}(\mathbf{r}',\tau)\rangle=\langle\hat{\hat{\Omega}}_{j\text{vac}-}(\mathbf{r},\tau)\hat{\hat{\Omega}}_{j'\text{vac}+}(\mathbf{r},\tau)\rangle=0\,. \tag{6.18}$$

In Sec. II we introduced the coherent copropagating fields $\hat{E}_s$, $\hat{E}_4$, and $\delta\hat{E}_p$, and did not introduce any counterpropagating ones. It is clear from the phase factors associated with the above source terms, at any position the spontaneous four-wave mixing is possible irrespective of the direction, as given by the correlation function

$$\langle\hat{\hat{\beta}}_{j-}(\mathbf{r},\tau)\hat{\hat{\beta}}^{\dagger}_{j'-}(\mathbf{r}',\tau)\rangle=0\,,\quad\langle\hat{\hat{\beta}}'_{j-}(\mathbf{r},\tau)\hat{\hat{\beta}}'^{\dagger}_{j-}(\mathbf{r},\tau)\rangle\neq0\,, \tag{6.19}$$

because the resonance fluorescence in the Rabi sidebands are correlated. However, only in the forward direction is spatial phase matching satisfied such that the noise sources from different atoms are still correlated as a spontaneous four-wave mixing. The sources of the counterpropagating waves, on the other hand, are only correlated at the same position, and the phase factors are not well matched in an extended medium. Thus no coherent components will be generated in the backward direction. This justifies the choice we made in Sec. II.

In the following we calculate the above correlation functions under one-way propagation and paraxial approximations (PA). The D_{j+} are proportional to the correlation functions for the copropagating sources. They are similar to the correlation functions $D_{jj'}$ we have discussed above and which were found to have a long coherence length. Under the one-way approximation, the D_{j+} are of the same form, except that no volume average has been performed in $D_{jj'}$, and thus $D_{jj'}$ contains sources for all directions. To be consistent with the SVEA, we also have to perform an average over the volume as given in Eqs. (6.12) and (6.13). Then as in Appendix F

$$D_{j+}(\mathbf{r},\mathbf{r}';\tau)=\frac{2}{\hbar}\frac{|\mathbf{d}_{21}|^2}{(2\pi c)^3}\int_0^\infty\int_0^{\pi/2}\int_0^{2\pi}\omega_\lambda^3 d\omega_\lambda(1-\sin^2\theta\cos^2\phi)\sin\theta\,d\theta\,d\phi\times\frac{e^{[-ik_\lambda(z-z')-k_\lambda^2a^2](1-\cos\theta)+ik_\lambda(x-x')\sin\theta\cos\phi+ik_\lambda(y-y')\sin\theta\sin\phi}}{(\omega_\lambda-\omega_p-\delta_j)^2+\Gamma_j^2}\,. \tag{6.20}$$

As we have seen in Eq. (5.18), the poles are at $\omega_\lambda=\omega_j\pm i\Gamma_j$. With proper choice of contour the integrand vanishes on the semicircle at infinity; this leads to terms like $e^{-\Gamma_j(|R_-|/c)}$ and $e^{-\Gamma_j(|R_+|/c)}$ as in Eq. (5.19). Hence if the dimensions of the sample are less than c/Γ_j, which is generally the case (especially if we can ignore collisions), we can then ignore the Γ_j terms in the exponent. [This leads to Eq. (6.20) being a simple function of $x-x'$, $y-y'$, etc.] This paraxial approximation is equivalent to assuming an infinite correlation length in the z direction. Mathematically, it is the limiting case of $\Gamma_j\to0$, since

$$\lim_{\Gamma_j\to0}\frac{\Gamma_j}{(\omega-\omega_j)^2+\Gamma_j^2}=\pi\delta(\omega-\omega_j)\,. \tag{6.21}$$

Thus we are left with

$$D_{j+}(\mathbf{r},\mathbf{r}';\tau)=\frac{2}{\hbar}\frac{|\mathbf{d}_{21}|^2}{(2\pi c)^3}\frac{\pi}{\Gamma_j}\omega_j^3\int_0^{\pi/2}\int_0^{2\pi}(1-\sin^2\theta\cos^2\phi)\sin\theta\,d\theta\,d\phi\times e^{[-ik_j(z-z')-k_j^2a^2](1-\cos\theta)+ik_j(x-x')\sin\theta\cos\phi+ik_j(y-y')\sin\theta\sin\phi}\,. \tag{6.22}$$

Now it is easy to see that, due to the rapidly varying phase, the above integral is important for $\theta \sim 0$. We can then put

$$\cos\theta \approx 1-\frac{\theta^2}{2}, \quad \sin\theta \approx \theta . \tag{6.23}$$

Since $k_j^2 a^2 \gg 1$, the important angular integration is restricted to $\theta \ll 1$, so that we can extend the upper limit of the integration to infinity. This form shows a very important physical aspect of the paraxial approximation, namely if we are interested in angle θ with respect to the z axis, we must have a variation in the transverse direction a such that $k_j a\theta \sim 1$,

$$\begin{aligned} D_{j+}(\mathbf{r},\mathbf{r}';\tau) &\approx \frac{2}{\hbar}\frac{|\mathbf{d}_{21}|^2}{(2\pi c)^3}\frac{\pi}{\Gamma_j}\int_0^\infty\int_0^{2\pi}\theta\, d\theta\, d\phi\, e^{[-ik_j(z-z')-k_j^2a^2](\theta^2/2)+ik_j\theta|\delta\rho|\cos\phi} \\ &= \frac{2}{\hbar}\frac{|\mathbf{d}_{21}|^2}{(2\pi c)^3}\frac{\pi}{\Gamma_j}\int_0^\infty \theta\, d\theta\, e^{[-ik_j(z-z')-k_j^2a^2](\theta^2/2)(2\pi)J_0(k_j\theta|\delta\rho|)} \\ &= \frac{4}{\hbar}\frac{|\mathbf{d}_{21}|^2}{(2\pi c)^3}\frac{\pi^2}{\Gamma_j}\frac{1}{ik_j(z-z')+k_j^2a^2}\exp\left[-\frac{(k_j|\delta\rho|)^2}{2[ik_j(z-z')+k_j^2a^2]}\right], \end{aligned} \tag{6.24}$$

where $\delta\boldsymbol{\rho}=\boldsymbol{\rho}-\boldsymbol{\rho}'$. In Fig. 4 we plot the above correlation function. It is easy to see that the spatial average is similar to a limit in the high-frequency behavior, which effectively eliminates the singularity at the locations of individual atoms. It is straightforward to show that this approximation will give an intensity expectation value that is real and positive definite. Ultimately, any intensity calculated with Eq. (6.24) should not depend significantly on the quantity a in order to be self-consistent.

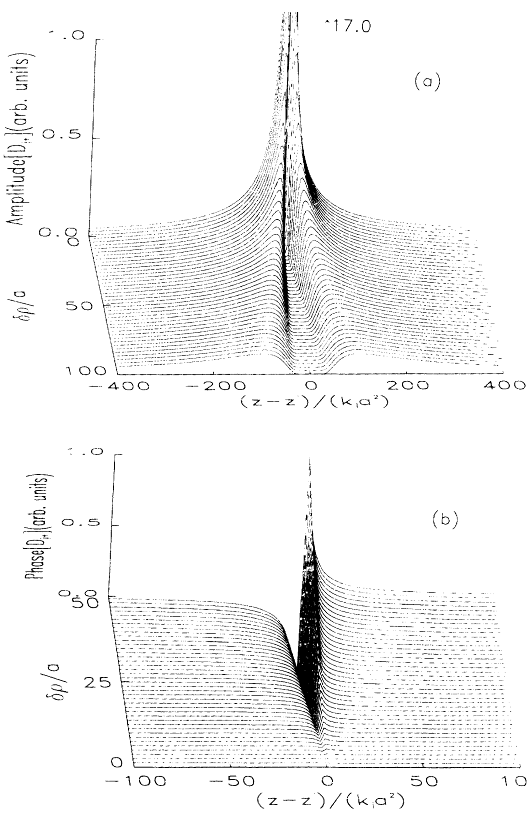

FIG. 4. (a) The amplitude of Eq. (6.24); (b) the phase of Eq. (6.24). Note the coarse-grained noise sources are not singular. We have scaled $\boldsymbol{\rho}$ (x and y) to a, and z to k_j/a^2.

We now have the SVEA polarization as follows:

$$[[\langle\hat\sigma_{21}\rangle]]_{\rm av}=[[\hat\sigma_{21}^s]]_{\rm av}+[[\langle\delta\hat\sigma_{21}\rangle]]_{\rm av}, \tag{6.25}$$

$$\begin{aligned}[][[\delta\hat\sigma_{21}]]_{\rm av}=&\tilde\alpha_p\delta\hat\Omega_p^{(-)}+\tilde\kappa_s\delta\hat\Omega_p^{(+)}+\tilde\alpha_s\hat\Omega_s^{(-)}e^{-i\delta r} \\ &+\tilde\kappa_4\hat\Omega_4^{(+)}e^{-i\delta\tau}+\tilde\kappa_s\hat\Omega_s^{(+)}e^{i\delta\tau}+\tilde\alpha_4\hat\Omega_4^{(-)}e^{i\delta\tau} \\ &+\hat{\tilde\beta}_{p+}(\tau)+\hat{\tilde\beta}_{s+}e^{-i\delta\tau}+\hat{\tilde\beta}_{4+}e^{i\delta\tau} . \end{aligned} \tag{6.26}$$

Then from Eq. (6.6)

$$\hat{\mathbf{P}}_A^{(-)}(\mathbf{r},\tau)=\hat{\mathbf{P}}_{Ap}^{(-)}(\mathbf{r},\tau)+\hat{\mathbf{P}}_{As}^{(-)}(\mathbf{r},\tau)+\hat{\mathbf{P}}_{A4}^{(-)}(\mathbf{r},\tau), \tag{6.27}$$

with

$$\begin{aligned} \hat{\mathbf{P}}_{Ap}^{(-)}(\mathbf{r},\tau)&=N\mathbf{d}_{21}[\hat\sigma_{21}^s+\tilde\alpha_p\delta\hat\Omega_p^{(-)}+\tilde\kappa_p\delta\hat\Omega_p^{(+)}+\hat{\tilde\beta}_{p+}]e^{i\omega_p\tau}, \\ \hat{\mathbf{P}}_{As}^{(-)}(\mathbf{r},\tau)&=N\mathbf{d}_{21}[\alpha_s\hat\Omega_s^{(-)}+\tilde\kappa_4\hat\Omega_4^{(+)}+\hat{\tilde\beta}_{s+}]e^{i(\omega_p-\delta)\tau}, \\ \hat{\mathbf{P}}_{A4}^{(-)}(\mathbf{r},\tau)&=N\mathbf{d}_{21}[\alpha_4\hat\Omega_4^{(-)}+\tilde\kappa_s\hat\Omega_s^{(+)}+\hat{\tilde\beta}_{4+}]e^{i(\omega_p+\delta)\tau} . \end{aligned} \tag{6.28}$$

Then according to Maxwell's equation for the total smoothed field $\hat{\mathbf{E}}$ with transverse Laplacian ∇_T^2,

$$\left[\nabla_T^2+\frac{\partial^2}{\partial z^2}-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right]\hat{\mathbf{E}}(\mathbf{r},t)=\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{P}(\mathbf{r},t) . \tag{6.29}$$

Now it is clear that the polarization of the medium oscillates at approximately three different frequencies (if δ is large compared to Γ_j's). We therefore assign a distinct source to each of the three different fields. Presumably this is a good approximation provided the spread Γ_j around each frequency is negligible compared to the spacing of the triplet components.

Since the $\delta\hat\Omega_p^{(-)}$ is always at the same frequency as the pump field, we cannot distinguish them, so we have to introduce

$$\hat{\Omega}_p^{(-)}=\Omega_p+\delta\hat{\Omega}_p^{(-)} . \tag{6.30}$$

Namely we will also take the pump field as a dynamic variable and study its propagation, including noise. We then get the following in the SVEA [32]:

$$\left[\nabla_T^2-2ik_p\left[\frac{\partial}{\partial z}+\frac{1}{v_p}\frac{\partial}{\partial t}\right]\right]\hat{\Omega}_p^{(-)} = -\frac{4\pi N\omega_p^2|\mathbf{d}_{21}|^2}{c^2\hbar}(\hat{\sigma}_{21}^s+\tilde{\alpha}_p\delta\hat{\Omega}_p^{(-)}+\tilde{\kappa}_p\delta\hat{\Omega}_p^{(+)}+\hat{\tilde{\beta}}_{p+}) ,$$

$$\left[\nabla_T^2-2ik_s\left[\frac{\partial}{\partial z}+\frac{1}{v_s}\frac{\partial}{\partial t}\right]\right]\hat{\Omega}_s^{(-)} = -\frac{4\pi N\omega_s^2|\mathbf{d}_{21}|^2}{c^2\hbar}(\tilde{\alpha}_s\hat{\Omega}_s^{(-)}+\tilde{\kappa}_4\hat{\Omega}_4^{(+)}+\hat{\tilde{\beta}}_{s+}) , \tag{6.31}$$

$$\left[\nabla_T^2+2ik_4\left[\frac{\partial}{\partial z}+\frac{1}{v_4}\frac{\partial}{\partial t}\right]\right]\hat{\Omega}_4^{(+)} = -\frac{4\pi N\omega_4^2|\mathbf{d}_{21}|^2}{c^2\hbar}(\tilde{\alpha}_4^*\hat{\Omega}_4^{(+)}+\tilde{\kappa}_s^*\hat{\Omega}_s^{(-)}+\hat{\tilde{\beta}}_{4+}^\dagger) .$$

We have neglected the terms containing e^{-2ik_jz} in the negative-frequency part of the polarization because they lead to coupling with counterpropagating components. Note that the zeroth-order solution is determined by solving the equation for pump field coupled to Eq. (4.14) for steady-state atomic-operator solutions, i.e.,

$$\left[\nabla_T^2-2ik_p\left[\frac{\partial}{\partial z}+\frac{1}{v_p}\frac{\partial}{\partial t}\right]\right]\Omega_p=-\frac{4\pi N\omega_p^2|\mathbf{d}_{21}|^2}{c^2\hbar}\hat{\sigma}_{21}^s , \tag{6.32}$$

$$\hat{\sigma}_{21}^s=-\frac{w_0\left[\Delta+i\frac{\gamma}{2}\right]}{\Delta^2+\frac{\gamma^2}{4}+\frac{|\Omega_p|^2}{2}}\frac{\Omega_p}{2} .$$

We thus have the steady-state values for the atomic operators $\hat{\sigma}_s(\mathbf{r})$ according to Eq. (4.14), from which all the $\tilde{\alpha}_j$, $\tilde{\kappa}_j$, and $\hat{\tilde{\beta}}_{j+}$ can be obtained. However, Eq. (6.31) for $\hat{\Omega}_p^{(-)}$ tells us how this, locally strong, field varies with position, and gets modified by the coherent component of resonance fluorescence noise at the same frequency. According to our approximations, we have kept all terms to first order in the generated fields. Thus we can solve Eq. (6.32) for the final output of the three fields, although the component centered at the pump frequency is of course predominately (zeroth-order) pump field.

The linear polarization term $\chi_0(\omega_j)\hat{\Omega}_j^{(-)}$ causes a slight change in the propagation velocity, i.e., $v_j=c/[1+4\pi\chi_0(\omega_j)]^{1/2}\approx c$. (This is not the velocity of the wave, because the right-hand side also contains the nonlinear polarization, which also affects the velocity. In fact, the velocity of the wave will not be known before we solve the problem. In the SVEA we are only eliminating the part traveling at c and leave the deviation from c for the detailed treatment.) Since this nonresonant polarization is small, it and its effects on dispersion will be neglected.

For the copropagating waves in the comoving frame, we have

$$\left[\nabla_T^2-2ik_p\frac{\partial}{\partial z}\right]\hat{\Omega}_p^{(-)}=-\alpha_p\hat{\Omega}_p^{(-)}+\hat{\beta}_{p+} ,$$

$$\left[\nabla_T^2-2ik_s\frac{\partial}{\partial z}\right]\hat{\Omega}_s^{(-)}=-\alpha_s\hat{\Omega}_s^{(-)}+\kappa_4\hat{\Omega}_4^{(+)}+\hat{\beta}_{s+} , \tag{6.33}$$

$$\left[\nabla_T^2+2ik_4\frac{\partial}{\partial z}\right]\hat{\Omega}_4^{(+)}=-\alpha_4^*\hat{\Omega}_4^{(+)}+\kappa_s^*\hat{\Omega}_s^{(-)}+\hat{\beta}_{4+}^\dagger ,$$

where

$$\alpha_p=\zeta(\omega_p)\frac{\hat{\sigma}_{21}^s(\Omega_p\to\hat{\Omega}_p^{(-)})}{\hat{\Omega}_p^{(-)}}$$

$$\alpha_j=\zeta(\omega_j)\tilde{\alpha}_j \ \text{ for } j=s,4 ,$$

$$\kappa_j=-\zeta(\omega_j)\tilde{\kappa}_j \ \text{ for } j=s,4 , \tag{6.34}$$

$$\hat{\beta}_{j\pm}=-\zeta(\omega_j)\hat{\tilde{\beta}}_{j\pm} \ \text{ for } j=p,s,4 ,$$

$$\zeta(\omega_j)=\frac{4\pi N\omega_j^2|\mathbf{d}_{21}|^2}{c^2\hbar} .$$

$\sigma_{21}^s(\Omega_p\to\hat{\Omega}_p^{(-)})$ denotes the steady-state value of σ_{21}^s with Ω_p replaced by $\hat{\Omega}_p^{(-)}$. We can easily check that

$$\tilde{\alpha}_p\delta\hat{\Omega}_p^{(-)}+\tilde{\kappa}_p\delta\Omega_p^{(+)}=\delta\hat{\sigma}_{21}^s=\frac{\partial\hat{\sigma}_{21}^s}{\partial\Omega_p^*}\delta\hat{\Omega}_p^{(-)}+\frac{\partial\hat{\sigma}_{21}^s}{\partial\Omega_p}\delta\hat{\Omega}_p^{(+)} \tag{6.35}$$

(taking the variation with respect to the small change of the fields). Thus

$$\hat{\sigma}_{21}^s(\Omega_p)+\tilde{\alpha}_p\delta\hat{\Omega}_p^{(-)}+\tilde{\kappa}_p\delta\hat{\Omega}_p^{(+)}=\hat{\sigma}_{21}^s(\Omega_p\to\hat{\Omega}_p^{(-)}) . \tag{6.36}$$

Since in this paper, we are interested in the redshifted s field only, we are left with

$$\left[\nabla_T^2-2ik_s\frac{\partial}{\partial z}\right]\hat{\Omega}_s^{(-)}=-\alpha_s\hat{\Omega}_s^{(-)}+\kappa_4\hat{\Omega}_4^{(+)}+\hat{\beta}_{s+} ,$$
$$\left[\nabla_T^2+2ik_4\frac{\partial}{\partial z}\right]\hat{\Omega}_4^{(+)}=-\alpha_4^*\hat{\Omega}_4^{(+)}+\kappa_s^*\hat{\Omega}_s^{(-)}+\hat{\beta}_{4+}^\dagger , \tag{6.37}$$

and boundary conditions under the one-way approximation are

$$\hat{\Omega}_s^{(-)}(\boldsymbol{\rho}',0,\tau)=0 ,$$
$$\hat{\Omega}_4^{(+)}(\boldsymbol{\rho}',0,\tau)=0 . \tag{6.38}$$

This equation can be formally solved as follows:

$$\hat{\Omega}_s^{(-)}(\boldsymbol{\rho},z,\tau)=\int_s\int_0^z K_{ss}(\boldsymbol{\rho},z;\boldsymbol{\rho}',z')\hat{\beta}_{s+}(\boldsymbol{\rho}',z',\tau)d\boldsymbol{\rho}'dz' + \int_s\int_0^z K_{s4}(\boldsymbol{\rho},z;\boldsymbol{\rho}',z')\hat{\beta}_{4+}^\dagger(\boldsymbol{\rho}',z',\tau)d\boldsymbol{\rho}'dz' , \tag{6.39}$$

where the $K_{ss}(\rho,z;\boldsymbol{\rho}',z')$ and $K_{s4}(\rho,z;\boldsymbol{\rho}',z')$ are the corresponding propagators (Green's functions).

The observable normal-ordered intensity for the $\hat{E}_s$

field is then related to the following correlations:

$$\langle \hat{\beta}_{s+}(\mathbf{r},\tau)\hat{\beta}^{\dagger}_{s+}(\mathbf{r}',\tau)\rangle = \zeta^2(\omega_s)|\mathcal{D}_s^{(+)}|^2[[\langle \hat{\hat{\Omega}}^{(+)}_{s\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(+)\dagger}_{s\text{vac}}(\mathbf{r}',\tau)\rangle]]_{\text{av}} ,$$

$$\langle \hat{\beta}^{\dagger}_{4+}(\mathbf{r},\tau)\hat{\beta}_{4+}(\mathbf{r}',\tau)\rangle = \zeta^2(\omega_4)|\mathcal{D}_4^{(-)}|^2[[\langle \hat{\hat{\Omega}}^{(-)\dagger}_{4\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(-)}_{4\text{vac}}(\mathbf{r}',\tau)\rangle]]_{\text{av}} , \tag{6.40}$$

$$\langle \hat{\beta}_{s+}(\mathbf{r},\tau)\hat{\beta}_{4+}(\mathbf{r}',\tau)\rangle = \zeta(\omega_s)\zeta(\omega_4)\mathcal{D}_s^{(+)}\mathcal{D}_4^{(-)}[[\langle \hat{\hat{\Omega}}^{(+)}_{s\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(-)}_{4\text{vac}}(\mathbf{r}',\tau)\rangle]]_{\text{av}} ,$$

$$\langle \hat{\beta}^{\dagger}_{4+}(\mathbf{r},\tau)\hat{\beta}^{\dagger}_{s+}(\mathbf{r}',\tau)\rangle = \zeta(\omega_s)\zeta(\omega_4)(\mathcal{D}_s^{(+)}\mathcal{D}_4^{(-)})^* \times[[\langle \hat{\Omega}^{(-)\dagger}_{4\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(+)\dagger}_{s\text{vac}}(\mathbf{r}',\tau)\rangle]]_{\text{av}} .$$

The so-called spontaneous four-wave mixing is very clear here, as the normal-ordered intensity for the E_s field is only connected to the correlation function of the vacuum modes centered around the ω_s. (The vacuum modes centered around the ω_s are also part of the expression for $\hat{\beta}_{j+}$. They do not contribute to the normal-ordered intensity, but they play an important role in preserving the commutation relations for the fields. In contrast, if an antinormal ordering were used the modes around ω_s would determine the intensity.) The $\mathcal{D}_j^{(\pm)}$ terms in the above prefactors are functions of the steady-state atomic operators $\hat{w}^s$ and $\hat{\sigma}^s_{21}$ (see Appendix D). It is easy to check that

$$\begin{aligned}
&[[\langle \hat{\hat{\Omega}}^{(+)}_{s\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(+)\dagger}_{s\text{vac}}(\mathbf{r}',\tau)\rangle]]_{\text{av}} = D_{s+}(\mathbf{r},\mathbf{r}';\tau) ,\\
&[[\langle \hat{\hat{\Omega}}^{(-)\dagger}_{4\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(-)}_{4\text{vac}}(\mathbf{r}',\tau)\rangle]]_{\text{av}} = D_{s+}(\mathbf{r},\mathbf{r}';\tau) ,\\
&[[\langle \hat{\hat{\Omega}}^{(+)}_{s\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(-)}_{4\text{vac}}(\mathbf{r}',\tau)\rangle]]_{\text{av}} = D_{s+}(\mathbf{r},\mathbf{r}';\tau) ,\\
&[[\langle \hat{\hat{\Omega}}^{(-)\dagger}_{4\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(+)\dagger}_{s\text{vac}}(\mathbf{r}',\tau)\rangle]]_{\text{av}} = D_{s+}(\mathbf{r},\mathbf{r}';\tau) .
\end{aligned} \tag{6.41}$$

A similar solution exists for the $\hat{E}_4$ field, and its intensity is related to the following correlation functions:

$$\langle \hat{\beta}_{4+}(\mathbf{r},\tau)\hat{\beta}^{\dagger}_{4+}(\mathbf{r}',\tau)\rangle = \zeta^2(\omega_4)|\mathcal{D}_4^{(+)}|^2[[\langle \hat{\hat{\Omega}}^{(+)}_{4\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(+)\dagger}_{4\text{vac}}(\mathbf{r}',\tau)\rangle]]_{\text{av}} ,$$

$$\langle \hat{\beta}^{\dagger}_{s+}(\mathbf{r},\tau)\hat{\beta}_{s+}(\mathbf{r}',\tau)\rangle = \zeta^2(\omega_s)|\mathcal{D}_s^{(-)}|^2[[\langle \hat{\hat{\Omega}}^{(-)\dagger}_{s\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(-)}_{s\text{vac}}(\mathbf{r}',\tau)\rangle]]_{\text{av}} , \tag{6.42}$$

$$\langle \hat{\beta}_{4+}(\mathbf{r},\tau)\hat{\beta}_{s+}(\mathbf{r}',\tau)\rangle = \zeta(\omega_s)\zeta(\omega_4)\mathcal{D}_s^{(-)}\mathcal{D}_4^{(+)}[[\langle \hat{\hat{\Omega}}^{(+)}_{4\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(-)}_{s\text{vac}}(\mathbf{r}',\tau)\rangle]]_{\text{av}} ,$$

$$\langle \hat{\beta}^{\dagger}_{s+}(\mathbf{r},\tau)\hat{\beta}^{\dagger}_{4+}(\mathbf{r}',\tau)\rangle = \zeta(\omega_s)\zeta(\omega_4)(\mathcal{D}_s^{(-)}\mathcal{D}_4^{(+)})^* \times[[\langle \hat{\hat{\Omega}}^{(-)\dagger}_{s\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(+)\dagger}_{4\text{vac}}(\mathbf{r}',\tau)\rangle]]_{\text{av}} ,$$

and

$$\begin{aligned}
&[[\langle \hat{\hat{\Omega}}^{(+)}_{4\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(+)\dagger}_{4\text{vac}}(\mathbf{r}',\tau)\rangle]]_{\text{av}} = D_{4+}(\mathbf{r},\mathbf{r}',\tau) ,\\
&[[\langle \hat{\hat{\Omega}}^{(-)\dagger}_{s\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(-)}_{s\text{vac}}(\mathbf{r}',\tau)\rangle]]_{\text{av}} = D_{4+}(\mathbf{r},\mathbf{r}';\tau) ,\\
&[[\langle \hat{\hat{\Omega}}^{(+)}_{4\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(-)}_{s\text{vac}}(\mathbf{r}',\tau)\rangle]]_{\text{av}} = D_{4+}(\mathbf{r},\mathbf{r}';\tau) ,\\
&[[\langle \hat{\hat{\Omega}}^{(-)\dagger}_{s\text{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(+)\dagger}_{4\text{vac}}(\mathbf{r}',\tau)\rangle]]_{\text{av}} = D_{4+}(\mathbf{r},\mathbf{r}';\tau) .
\end{aligned} \tag{6.43}$$

We note here that we are describing the fields emitted by the atoms only. The total field at any position $\mathbf{r}$ and time t in the medium free region is in fact

$$\hat{E}(\mathbf{r},t) = E_p(\mathbf{r},t) + \hat{E}_s(\mathbf{r},t) + \hat{E}_4(\mathbf{r},t) + \hat{E}_{\text{vac}}(\mathbf{r},t) , \tag{6.44}$$

which will preserve the appropriate commutator relations for field operators.

VII. CONCLUSION

In this paper we have investigated quantum mechanically the resonant light propagation in an active medium based on a two-level atom formulation. In the steady-state limit, the slowly varying field envelopes for the copropagating fields obey the following equations:

$$\begin{aligned}
&\left[\nabla_T^2 - 2ik_p\frac{\partial}{\partial z}\right]\hat{\Omega}_p^{(-)} = -\alpha_p\hat{\Omega}_p^{(-)} + \hat{\beta}_{p+} ,\\
&\left[\nabla_T^2 - 2ik_s\frac{\partial}{\partial z}\right]\hat{\Omega}_s^{(-)} = -\alpha_s\hat{\Omega}_s^{(-)} + \kappa_4\hat{\Omega}_4^{(+)} + \hat{\beta}_{s+} ,\\
&\left[\nabla_T^2 + 2ik_4\frac{\partial}{\partial z}\right]\hat{\Omega}_4^{(+)} = -\alpha_4^*\hat{\Omega}_4^{(+)} + \kappa_s^*\hat{\Omega}_s^{(-)} + \hat{\beta}^{\dagger}_{4+} .
\end{aligned} \tag{7.1}$$

On the right-hand side of the field equations are the polarizations of the medium. In addition to the terms due to the linear response of the weak fields, there are also quantum-noise terms, which we have proven to be of long coherence length. These long-coherence-length noise terms are coming from spontaneous-emission noise, the statistics of which we have determined from the dynamics of the atomic medium and the vacuum fluctuations.

Of particular interest are the following second-order correlations under the SVEA and one-way approximation, which are related to the observable normal-order intensity of the two generated quantum fields $\hat{E}_s$ and $\hat{E}_4$:

$$\begin{aligned}
&\langle \hat{\beta}_{s+}(\mathbf{r},\tau)\hat{\beta}^{\dagger}_{s+}(\mathbf{r}',\tau)\rangle = \zeta^2(\omega_s)|\mathcal{D}_s^{(+)}|^2 D_{s+}(\mathbf{r},\mathbf{r}';\tau) ,\\
&\langle \hat{\beta}^{\dagger}_{4+}(\mathbf{r},\tau)\hat{\beta}_{4+}(\mathbf{r}',\tau)\rangle = \zeta^2(\omega_4)|\mathcal{D}_4^{(-)}|^2 D_{s+}(\mathbf{r},\mathbf{r}';\tau) ,
\end{aligned} \tag{7.2}$$

$$\langle \hat{\beta}_{s+}(\mathbf{r},\tau)\hat{\beta}_{4+}(\mathbf{r}',\tau)\rangle = \zeta(\omega_s)\zeta(\omega_4)\mathcal{D}_s^{(+)}\mathcal{D}_4^{(-)} D_{s+}(\mathbf{r},\mathbf{r}';\tau) ,$$

$$\langle \hat{\beta}^{\dagger}_{4+}(\mathbf{r},\tau)\hat{\beta}^{\dagger}_{s+}(\mathbf{r}',\tau)\rangle = \zeta(\omega_s)\zeta(\omega_4)(\mathcal{D}_s^{(+)}\mathcal{D}_4^{(-)})^* D_{s+}(\mathbf{r},\mathbf{r}';\tau) ,$$

$$\langle \hat{\beta}_{4+}(\mathbf{r},\tau)\hat{\beta}^\dagger_{4+}(\mathbf{r}',\tau)\rangle = \zeta^2(\omega_4)|\mathcal{D}_4^{(+)}|^2 D_{4+}(\mathbf{r},\mathbf{r}';\tau) ,$$
$$\langle \hat{\beta}^\dagger_{s+}(\mathbf{r},\tau)\hat{\beta}_{s+}(\mathbf{r}',\tau)\rangle = \zeta^2(\omega_s)|\mathcal{D}_s^{(-)}|^2 D_{4+}(\mathbf{r},\mathbf{r}';\tau) ,$$
$$\langle \hat{\beta}_{4+}(\mathbf{r},\tau)\hat{\beta}_{s+}(\mathbf{r}',\tau)\rangle = \zeta(\omega_s)\zeta(\omega_4)\mathcal{D}_s^{(-)}\mathcal{D}_4^{(+)} D_{4+}(\mathbf{r},\mathbf{r}';\tau) , \quad (7.3)$$
$$\langle \hat{\beta}^\dagger_{s+}(\mathbf{r},\tau)\hat{\beta}^\dagger_{4+}(\mathbf{r}',\tau)\rangle = \zeta(\omega_s)\zeta(\omega_4)(\mathcal{D}_s^{(-)}\mathcal{D}_4^{(+)})^* D_{4+}(\mathbf{r},\mathbf{r}';\tau) ,$$

where

$$D_{j+}(\mathbf{r},\mathbf{r}';\tau) = \frac{4}{\hbar}\frac{|\mathbf{d}_{21}|^2}{(2\pi c)^3}\frac{\pi^2}{\Gamma_j}\frac{1}{ik_j(z-z')+k_j^2a^2} \times \exp\left[-\frac{(k_j|\delta\boldsymbol{\rho}|)^2}{2[ik_j(z-z')+k_j^2a^2]}\right] . \quad (7.4)$$

We note that in this formulation, collisional effects have been neglected, which might prevent us from making rigorous comparisons with experiments, since most experiments to date have been performed in the buffer-gas broadened cells. The extension via a collisional Langevin term is straightforward. Also neglected in the formulation are the Doppler distribution averages, which in principle could easily be included. However, since we are basically interested in regimes where the detuning and on-axis Rabi frequency of the pump are larger than the Doppler width, we expect this average will not be crucial.

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APPENDIX A

In order to evaluate $\hat{s}_{21}^{(l)}$ of Eq. (3.6) we first of all make the Markov approximation of Eq. (3.7). This is valid because the sum $\sum_\lambda$ infers a correlation time τ_c of order ω_{21}^{-1} which is short compared to the time scale (of order the spontaneous decay time γ^{-1}) over which $\hat{\sigma}_{21}^{(l)}(t)$ changes. A Born approximation is also implied by Eq. (3.7) since only free evolution during this correlation time is considered, which again will be valid if $\gamma\tau_c$ and $|\Omega_p|\tau_c$ are small.

From Eq. (3.7) we have

$$\begin{aligned}\tilde{\gamma} &= \sum_\lambda |g_\lambda|^2 \int_0^t e^{i(\omega_\lambda-\omega_{21})(t-t')}dt' \\ &\approx \sum_\lambda |g_\lambda|^2 \int_0^\infty e^{i(\omega_\lambda-\omega_{21})}\tau' d\tau' \\ &= \sum_\lambda |g_\lambda|^2 \left[\pi\delta(\omega_\lambda-\omega_{21}) + iP\frac{1}{\omega_\lambda-\omega_{21}}\right] \\ &= \frac{\gamma}{2} + i\tilde{\Delta}(\omega_{21}) ,\end{aligned} \quad (A1)$$

where

$$\begin{aligned}\gamma &= 2\pi \sum_\lambda |g_\lambda|^2 \delta(\omega_\lambda-\omega_{21}) \\ &= \frac{4\pi^2}{\hbar V}\sum_{\mathbf{k}_\lambda}\sum_{\epsilon_\lambda}(\mathbf{d}_{21}\cdot\boldsymbol{\epsilon}_\lambda)(\mathbf{d}_{12}\cdot\boldsymbol{\epsilon}^*_\lambda)\omega_\lambda\delta(\omega_\lambda-\omega_{21}) .\end{aligned} \quad (A2)$$

The summation can be easily done as

$$\sum_{\epsilon_\lambda}(\boldsymbol{\epsilon}_\lambda\cdot\mathbf{a})(\boldsymbol{\epsilon}^*_\lambda\cdot\mathbf{b}) = \mathbf{a}\cdot\mathbf{b} - \frac{(\mathbf{k}_\lambda\cdot\mathbf{a})(\mathbf{k}_\lambda\cdot\mathbf{b})}{k_\lambda^2} , \quad (A3)$$

and with

$$\frac{1}{V}\sum_{\mathbf{k}_\lambda}\cdots \to \left[\frac{1}{2\pi}\right]^3 \int d\mathbf{k}_\lambda \cdots , \quad (A4)$$

$$\begin{aligned}\gamma &= \frac{4\pi}{\hbar}\frac{|\mathbf{d}_{21}|^2}{(2\pi c)^3}\int_0^\infty\int_0^{2\pi}\int_0^\pi \omega_\lambda^3 d\omega_\lambda(1-\cos^2\theta) \\ &\quad\times \sin\theta\, d\theta\, d\phi\, \delta(\omega_\lambda-\omega_{21}) \\ &= \frac{4|\mathbf{d}_{21}|^2\omega_{21}^3}{3\hbar c^3} ,\end{aligned} \quad (A5)$$

$$\tilde{\Delta}(\omega_{21}) = \sum_\lambda |g_\lambda|^2 \mathrm{P}\left[\frac{1}{\omega_\lambda-\omega_{21}}\right] , \quad (A6)$$

where P$(1/x)$ represents the principal part of the expression.

APPENDIX B

From Eq. (3.10) we have

$$\sum_\lambda 2(g_\lambda\hat{\sigma}_{21}^{(l)}\hat{a}_\lambda e^{i\mathbf{k}_\lambda\cdot\mathbf{r}_l} + g_\lambda^*\hat{a}_\lambda^\dagger\hat{\sigma}_{12}^{(l)}e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}_l}) = \hat{s}^{(l)} + \hat{m}^{(l)} + \hat{f}^{(l)} , \quad (B1)$$

with

$$\hat{s}^{(l)} = \sum_\lambda 2|g_\lambda|^2\left[\hat{\sigma}_{21}^{(l)}\int_0^t \hat{\sigma}_{12}^{(l)}(t')e^{-i\omega_\lambda(t-t')}dt' + \int_0^t \hat{\sigma}_{21}^{(l)}(t')e^{i\omega_\lambda(t-t')}dt'\hat{\sigma}_{12}^{(l)}\right] ,$$

$$\hat{m}^{(l)} = \sum_\lambda\sum_{j(\neq l)} 2|g_\lambda|^2\left[\hat{\sigma}_{21}^{(l)}e^{i\mathbf{k}_\lambda\cdot\mathbf{r}_l}e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}_j}\int_0^t \hat{\sigma}_{12}^{(j)}(t')e^{-i\omega_\lambda(t-t')}dt' + e^{i\mathbf{k}_\lambda\cdot\mathbf{r}_j}\int_0^t \hat{\sigma}_{21}^{(j)}(t')e^{i\omega_\lambda(t-t')}dt'\hat{\sigma}_{12}^{(l)}e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}_l}\right] , \quad (B2)$$

$$\hat{f}^{(l)} = \sum_\lambda 2[g_\lambda\hat{\sigma}_{21}^{(l)}\hat{a}_\lambda(0)e^{-i\omega_\lambda t}e^{i\mathbf{k}_\lambda\cdot\mathbf{r}_l} + g_\lambda^*\hat{a}_\lambda^\dagger(0)e^{i\omega_\lambda t}\hat{\sigma}_{12}^{(l)}e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}_l}] .$$

Under the same Markov approximation,

$$\hat{s}^{(l)}=\sum_{\lambda}2|g_{\lambda}|^2\left[\hat{\sigma}_{21}^{(l)}\int_0^t\hat{\sigma}_{12}^{(l)}(t')e^{-i\omega_\lambda(t-t')}dt'+\int_0^t\hat{\sigma}_{21}^{(l)}(t')e^{i\omega_\lambda(t-t')}dt'\hat{\sigma}_{12}^{(l)}\right]$$

$$\approx 4\bar{\gamma}\hat{\sigma}_{21}^{(l)}(t)\hat{\sigma}_{12}^{(l)}(t)$$

$$=2\gamma\hat{\sigma}_{22}^{(l)}(t)$$

$$=-\gamma(\hat{w}^{(l)}-\hat{w}_0)\ . \tag{B3}$$

Then from $\hat{\sigma}_{11}^{(l)}-\hat{\sigma}_{22}^{(l)}=\hat{w}^{(l)}$, and $\hat{\sigma}_{11}^{(l)}+\hat{\sigma}_{22}^{(l)}=\hat{w}_0$, we find

$$\hat{\sigma}_{22}^{(l)}=\tfrac{1}{2}(\hat{w}_0-\hat{w}^{(l)})\ . \tag{B4}$$

APPENDIX C

From Eq. (4.9) we have

$$\mathcal{T}=(\boldsymbol{\mathcal{Y}}_1,\boldsymbol{\mathcal{Y}}_2,\mathcal{Y}_3)\ ,$$

with

$$\hat{\boldsymbol{\mathcal{Y}}}_j=\begin{bmatrix} i\dfrac{\Omega_p}{2}\left[i\Delta-\dfrac{\gamma}{2}-\Lambda_j\right] \\ -i\dfrac{\Omega_p^*}{2}\left[-i\Delta-\dfrac{\gamma}{2}-\Lambda_j\right] \\ \left[i\Delta-\dfrac{\gamma}{2}-\Lambda_j\right]\left[-i\Delta-\dfrac{\gamma}{2}-\Lambda_j\right]\end{bmatrix},$$

then

$$\mathcal{T}=\begin{bmatrix} i\left[i\Delta-\dfrac{\gamma}{2}-\Lambda_1\right]\dfrac{\Omega_p}{p} & i\left[i\Delta-\dfrac{\gamma}{2}-\Lambda_2\right]\dfrac{\Omega_p}{2} & i\left[i\Delta-\dfrac{\gamma}{2}-\Lambda_3\right]\dfrac{\Omega_p}{2} \\ -i\dfrac{\Omega_p^*}{2}\left[-i\Delta-\dfrac{\gamma}{2}-\Lambda_1\right] & -i\dfrac{\Omega_p^*}{2}\left[-i\Delta-\dfrac{\gamma}{2}-\Lambda_2\right] & -i\dfrac{\Omega_p^*}{2}\left[-i\Delta-\dfrac{\gamma}{2}-\Lambda_3\right] \\ \Delta^2+\left[\dfrac{\gamma}{2}+\Lambda_1\right]^2 & \Delta^2+\left[\dfrac{\gamma}{2}+\Lambda_2\right]^2 & \Delta^2+\left[\dfrac{\gamma}{2}+\Lambda_3\right]^2\end{bmatrix},$$

and

$$\mathcal{T}^{-1}=\begin{bmatrix} \dfrac{\left[i\Delta+\frac{\gamma}{2}\right]^2+\left[i\Delta+\frac{\gamma}{2}\right]\Lambda_2+\left[i\Delta+\frac{\gamma}{2}\right]\Lambda_3+\Lambda_2\Lambda_3}{-\Delta(\Lambda_1-\Lambda_2)(\Lambda_1-\Lambda_3)\Omega_p} & \dfrac{\left[i\Delta-\frac{\gamma}{2}\right]^2-\left[i\Delta-\frac{\gamma}{2}\right]\Lambda_2-\left[i\Delta-\frac{\gamma}{2}\right]\Lambda_3+\Lambda_2\Lambda_3}{-\Omega_p^*\Delta(\Lambda_1-\Lambda_2)(\Lambda_1-\Lambda_3)} & \dfrac{1}{(\Lambda_1-\Lambda_2)(\Lambda_1-\Lambda_3)} \\ \dfrac{\left[i\Delta+\frac{\gamma}{2}\right]^2+\left[i\Delta+\frac{\gamma}{2}\right]\Lambda_1+\left[i\Delta+\frac{\gamma}{2}\right]\Lambda_3+\Lambda_1\Lambda_3}{-\Delta(\Lambda_2-\Lambda_1)(\Lambda_2-\Lambda_3)\Omega_p} & \dfrac{\left[i\Delta-\frac{\gamma}{2}\right]^2-\left[i\Delta-\frac{\gamma}{2}\right]\Lambda_1-\left[i\Delta-\frac{\gamma}{2}\right]\Lambda_3+\Lambda_1\Lambda_3}{-\Omega_p^*\Delta(\Lambda_2-\Lambda_1)(\Lambda_2-\Lambda_3)} & \dfrac{1}{(\Lambda_2-\Lambda_1)(\Lambda_2-\Lambda_3)} \\ \dfrac{\left[i\Delta+\frac{\gamma}{2}\right]^2+\left[i\Delta+\frac{\gamma}{2}\right]\Lambda_1+\left[i\Delta+\frac{\gamma}{2}\right]\Lambda_2+\Lambda_1\Lambda_2}{-\Delta(\Lambda_3-\Lambda_1)(\Lambda_3-\Lambda_2)\Omega_p} & \dfrac{\left[i\Delta-\frac{\gamma}{2}\right]^2-\left[i\Delta-\frac{\gamma}{2}\right]\Lambda_1-\left[i\Delta-\frac{\gamma}{2}\right]\Lambda_2+\Lambda_1\Lambda_2}{-\Omega_p^*\Delta(\Lambda_3-\Lambda_1)(\Lambda_3-\Lambda_2)} & \dfrac{1}{(\Lambda_3-\Lambda_1)(\Lambda_3-\Lambda_2)}\end{bmatrix},$$

$$\mathcal{A}^{-1}=\begin{bmatrix} \frac{-i\Delta\gamma+\gamma\frac{\gamma}{2}+\frac{|\Omega_p|^2}{2}}{-\Delta^2\gamma-\frac{\gamma^3}{4}-|\Omega_p|^2\frac{\gamma}{2}} & \frac{\frac{\Omega_p^2}{2}}{-\Delta^2\gamma-\frac{\gamma^3}{4}-|\Omega_p|^2\frac{\gamma}{2}} & \frac{-\Delta\frac{\Omega_p}{2}-i\frac{\gamma}{2}\frac{\Omega_p}{2}}{-\Delta^2\gamma-\frac{\gamma^3}{4}-|\Omega_p|^2\frac{\gamma}{2}} \\ \frac{\frac{\Omega_p^{*2}}{2}}{-\Delta^2\gamma-\frac{\gamma^3}{4}-|\Omega_p|^2\frac{\gamma}{2}} & \frac{i\Delta\gamma+\gamma\frac{\gamma}{2}+\frac{|\Omega_p|^2}{2}}{-\Delta^2\gamma-\frac{\gamma^3}{4}-|\Omega_p|^2\frac{\gamma}{2}} & \frac{-\frac{\Omega_p^*}{2}\Delta+i\frac{\Omega_p^*}{2}\frac{\gamma}{2}}{-\Delta^2\gamma-\frac{\gamma^3}{4}-|\Omega_p|^2\frac{\gamma}{2}} \\ \frac{-\Omega_p^*\Delta-i\Omega_p^*\frac{\gamma}{2}}{-\Delta^2\gamma-\frac{\gamma^3}{4}-|\Omega_p|^2\frac{\gamma}{2}} & \frac{-\Delta\Omega_p+i\frac{\gamma}{2}\Omega_p}{-\Delta^2\gamma-\frac{\gamma^3}{4}-|\Omega_p|^2\frac{\gamma}{2}} & \frac{\Delta^2+\frac{\gamma}{4^2}}{-\Delta^2\gamma-\frac{\gamma^3}{4}-|\Omega_p|^2\frac{\gamma}{2}} \end{bmatrix} .$$

APPENDIX D

From Eq. (4.20) we have

$$\delta\hat{\mathcal{X}}_i(\tau)=\delta\hat{\mathcal{X}}_i(0)e^{\Lambda_i\tau}+\int_0^\tau e^{\Lambda_i(\tau-\tau')}[\mathcal{T}^{-1}\hat{\mathbf{F}}(\tau')]_i d\tau' + \int_0^\tau e^{\Lambda_i(\tau-\tau')}[\mathcal{T}^{-1}\mathcal{G}\hat{\sigma}_s(\tau')]_i d\tau' , \tag{D1}$$

$$\delta\hat{\sigma}_{21}(\tau)=\mathcal{T}_{11}\delta\hat{\mathcal{X}}_1(\tau)+\mathcal{T}_{12}\delta\mathcal{X}_2(\tau)+\mathcal{T}_{13}\delta\mathcal{X}_3(\tau) =\delta\hat{\sigma}_{21}^1+\delta\hat{\sigma}_{21}^2 , \tag{D2}$$

where

$$\delta\hat{\sigma}_{21}^1=\sum_{j=1}^{3}\mathcal{T}_{1j}\mathcal{T}_{j1}^{-}\int_0^\tau e^{\Lambda_j(\tau-\tau')}\hat{F}_2^\dagger(\tau')d\tau' + \sum_{j=1}^{3}\mathcal{T}_{1j}\mathcal{T}_{j2}^{-}\int_0^\tau e^{\Lambda_j(\tau-\tau')}\hat{F}_2(\tau')d\tau' + \sum_{j=1}^{3}\mathcal{T}_{1j}\mathcal{T}_{j3}^{-}\int_0^\tau e^{\Lambda_j(\tau-\tau')}\hat{F}_1(\tau')d\tau' =\hat{\hat{V}}_p+\hat{\hat{V}}_s+\hat{\hat{V}}_4 , \tag{D3}$$

with

$$\hat{\hat{V}}_j=\mathcal{D}_j^{(-)}\hat{\hat{\Omega}}_{j\text{vac}}^{(-)}+\mathcal{D}_j^{(+)}\hat{\hat{\Omega}}_{j\text{vac}}^{(+)} ,$$

$$\mathcal{D}_j^{(-)}=\mathcal{T}_{1j}\mathcal{T}_{j1}^{-}\hat{w}^s-2\mathcal{T}_{1j}\mathcal{T}_{j3}^{-}\hat{\sigma}_{12}^s , \tag{D4}$$

$$\mathcal{D}_j^{(+)}=\mathcal{T}_{1j}\mathcal{T}_{j2}^{-}\hat{w}^s-2\mathcal{T}_{1j}\mathcal{T}_{j3}^{-}\hat{\sigma}_{21}^s ,$$

$$\hat{\hat{\Omega}}_{j\text{vac}}^{(-)}=\int_0^\tau e^{\Lambda_j(\tau-\tau')}\hat{\Omega}_{\text{vac}}^{(-)}e^{-i\omega_p\tau'}d\tau' , \quad \hat{\hat{\Omega}}_{j\text{vac}}^{(+)}=\int_0^\tau e^{\Lambda_j(\tau-\tau')}\hat{\Omega}_{\text{vac}}^{(+)}e^{i\omega_p\tau'}d\tau' . \tag{D5}$$

Then we obtain

$$\delta\hat{\sigma}_{21}^2=\sum_{j=1}^{3}\mathcal{T}_{1j}\mathcal{T}_{j1}^{-}\int_0^\tau e^{\Lambda_j(\tau-\tau')}(\mathcal{G}\hat{\sigma}_s)_1(\tau')d\tau' + \sum_{j=1}^{3}\mathcal{T}_{1j}\mathcal{T}_{j2}^{-}\int_0^\tau e^{\Lambda_j(\tau-\tau')}(\mathcal{G}\hat{\sigma}_s)_2(\tau')d\tau' + \sum_{j=1}^{3}\mathcal{T}_{1j}\mathcal{T}_{j3}^{-}\int_0^\tau e^{\Lambda_j(\tau-\tau')}(\mathcal{G}\hat{\sigma}_s)_3(\tau')d\tau' , \tag{D6}$$

with

$$(\mathcal{G}\hat{\sigma}_s)_1=-i\hat{w}^s[\delta\hat{\Omega}_p^{(-)}+\hat{\Omega}_s^{(-)}e^{-i\delta\tau}+\hat{\Omega}_4^{(-)}e^{i\delta\tau}]/2 ,$$
$$(\mathcal{G}\hat{\sigma}_s)_2=i\hat{w}^s[\delta\hat{\Omega}_p^{(+)}+\hat{\Omega}_s^{(+)}e^{i\delta\tau}+\hat{\Omega}_4^{(+)}e^{-i\delta\tau}]/2 ,$$
$$(\mathcal{G}\hat{\sigma}_s)_3=-i\hat{\sigma}_{21}^s[\delta\hat{\Omega}_p^{(+)}+\hat{\Omega}_s^{(+)}e^{i\delta\tau}+\hat{\Omega}_4^{(+)}e^{-i\delta\tau}] +i\hat{\sigma}_{12}^s[\delta\hat{\Omega}_p^{(-)}+\hat{\Omega}_s^{(-)}e^{-i\delta\tau}+\hat{\Omega}_4^{(-)}e^{i\delta\tau}] . \tag{D7}$$

In the long-time limit the field amplitudes reach a steady state and the integral can be performed adiabatically as

$$\delta\hat{\sigma}_{21}^2(\tau)=\tilde{\alpha}_p\delta\hat{\Omega}_p^{(-)}+\tilde{\kappa}_p\delta\hat{\Omega}_p^{(+)}+\tilde{\alpha}_s\hat{\Omega}_s^{(-)}e^{-i\delta\tau} +\tilde{\kappa}_4\hat{\Omega}_4^{(+)}e^{-i\delta\tau}+\tilde{\alpha}_4\hat{\Omega}_4^{(-)}e^{i\delta\tau}+\tilde{\kappa}_s\hat{\Omega}_s^{(+)}e^{i\delta\tau} , \tag{D8}$$

where $\tilde{\alpha}$'s and $\tilde{\kappa}$'s are defined as

$$\tilde{\alpha}_p=\xi_1(0) , \quad \tilde{\alpha}_s=\xi_1(\delta) , \quad \tilde{\alpha}_4=\xi_1(-\delta) ,$$
$$\xi_1(\epsilon)=i\frac{\hat{w}^s}{2}\sum_{j=1}^{3}\mathcal{T}_{1j}\mathcal{T}_{j1}^{-}(\Lambda_j+i\epsilon)^{-1} -i\hat{\sigma}_{12}^s\sum_{j=1}^{3}\mathcal{T}_{1j}\mathcal{T}_{j3}^{-}(\Lambda_j+i\epsilon)^{-1} , \tag{D9}$$

$$\tilde{\kappa}_p=\xi_2(0) , \quad \tilde{\kappa}_s=\xi_2(-\delta) , \quad \tilde{\kappa}_4=\xi_2(\delta) ,$$
$$\xi_2(\epsilon)=-i\frac{\hat{w}^s}{2}\sum_{j=1}^{3}\mathcal{T}_{1j}\mathcal{T}_{j2}^{-}(\Lambda_j+i\epsilon)^{-1} +i\hat{\sigma}_{21}^s\sum_{j=1}^{3}\mathcal{T}_{1j}\mathcal{T}_{j3}^{-}(\Lambda_j+i\epsilon)^{-1} . \tag{D10}$$

APPENDIX E

Generically, if

$$\hat{\hat{U}}_j(\tau)=\int_0^{\tau} e^{\Lambda_j(\tau-\tau')}\hat{U}(\tau')d\tau' = \int_{-\infty}^{\infty}\Theta(\tau-\tau')\hat{H}(\tau')d\tau' , \quad \text{(E1)}$$

where

$$\Theta(\tau-\tau')=e^{\Lambda_j(\tau-\tau')}\Theta(\tau-\tau') , \quad \hat{H}(\tau')=\hat{U}(\tau')\Theta(\tau') , \quad \text{(E2)}$$

$$\Theta(\tau)=\begin{cases}1 & \text{if } \tau>0\\ 0 & \text{if } \tau<0 ,\end{cases} \quad \text{(E3)}$$

the Fourier transform of $\hat{\hat{U}}_j$ is

$$\hat{\hat{U}}_j(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty} e^{i\omega\tau}\hat{\hat{U}}_j(\tau)d\tau=\Theta(\omega)\hat{H}(\omega) , \quad \text{(E4)}$$

where

$$\Theta(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty} e^{i\omega\tau}\Theta(\tau)d\tau , \quad \hat{H}(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty} e^{i\omega\tau}\hat{H}(\tau)d\tau . \quad \text{(E5)}$$

We see that the damped oscillation function $\Theta(\tau)$ is an effective filtering function

$$\Theta(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty} e^{i\omega\tau}\Theta(\tau)d\tau=\frac{1}{2\pi}\int_0^{\infty} e^{(i\omega+\Lambda)\tau}d\tau = -\frac{1}{2\pi}\frac{1}{i\omega+\Lambda_j} , \quad \text{(E6)}$$

which is centered around $-\mathrm{Im}(\Lambda_j)$.

APPENDIX F

From Eq. (5.4) we have

$$\begin{aligned} D_{jj'}(\mathbf{r},\mathbf{r}';\tau) &= \langle \hat{\hat{\Omega}}^{(+)}_{j\mathrm{vac}}(\mathbf{r},\tau)\hat{\hat{\Omega}}^{(+)\dagger}_{j\mathrm{vac}}(\mathbf{r}',\tau)\rangle \\ &= \int_0^{\tau} e^{\Lambda_j(\tau-\tau')}d\tau' \int_0^{\tau} e^{\Lambda_j^*(\tau-\tau'')}d\tau'' \langle \hat{\Omega}^{(+)}_{\mathrm{vac}}(\mathbf{r},\tau')\hat{\Omega}^{(-)}_{\mathrm{vac}}(\mathbf{r}',\tau'')e^{i\omega_p(\tau'-\tau'')}\rangle \\ &\approx \delta_{jj'}\sum_{\lambda'}\sum_{\lambda}\langle \hat{a}_{\lambda'}(0)\hat{a}^\dagger_{\lambda}(0)\rangle g_{\lambda'} e^{i\mathbf{k}_{\lambda'}\cdot\mathbf{r}} e^{-i\omega_{\lambda'}z/c} e^{i(\omega_p-\omega_{\lambda'}\tau)} \frac{1}{-\Lambda_j-i\omega_{\lambda'}+i\omega_p} \\ &\quad \times g^*_{\lambda} e^{-i\mathbf{k}_{\lambda}\cdot\mathbf{r}'} e^{i\omega_{\lambda}z'/c} e^{-i(\omega_p-\omega_{\lambda})\tau} \frac{1}{-\Lambda_j^*+i\omega_{\lambda}-i\omega_p} \\ &= \delta_{jj'}\sum_{\lambda}|g_{\lambda}|^2 \frac{e^{i\mathbf{k}_{\lambda}\cdot(\mathbf{r}-\mathbf{r}')}e^{-ik_{\lambda}(z-z')}}{(\omega_{\lambda}-\omega_j)^2+\Gamma_j^2} . \end{aligned} \quad \text{(F1)}$$

In writing down the $\delta_{jj'}$ in the above expression, we have neglected the overlapping of the wings of the Mollow triplet. We choose the dipole moment to be perpendicular to the direction of propagation (z axis), say in the x direction for simplicity, and then

$$\sum_{\epsilon}|\boldsymbol{\epsilon}\cdot\mathbf{d}_{21}|^2=|\mathbf{d}_{21}|^2(1-|\mathbf{k}_{\lambda 1}\cdot\hat{x}|^2)=|\mathbf{d}_{21}|^2(1-\sin^2\theta\cos^2\phi) , \quad \text{(F2)}$$

$$D_{jj}(\mathbf{r},\mathbf{r}';\tau)=\frac{2}{\hbar}\frac{|\mathbf{d}_{21}|^2}{(2\pi c)^3}\int_0^{\infty}\int_0^{\pi}\int_0^{2\pi}\omega_{\lambda}^3 d\omega_{\lambda}(1-\sin^2\theta\cos^2\phi)\sin\theta\, d\theta\, d\theta \times \frac{e^{ik_{\lambda}(z-z')\cos\theta+ik_{\lambda}(x-x')\sin\theta\cos\phi+ik_{\lambda}(y-y')\sin\theta\sin\phi}}{(\omega_{\lambda}-\omega_j)^2+\Gamma_j^2} e^{-ik_{\lambda}(z-z')} . \quad \text{(F3)}$$

The integration over azimuth angle ϕ is

$$\int_0^{2\pi}(1-\sin^2\theta\cos^2\phi)d\phi\, e^{ik_{\lambda}(x-x')\sin\theta\cos\phi+ik_{\lambda}(y-y')\sin\theta\sin\phi} = \int_0^{2\pi}\left[1+\frac{\partial^2}{k_{\lambda}^2\partial(x-x')^2}\right]d\phi\, e^{ik_{\lambda}|\rho-\rho'|\sin\theta\cos(\phi-\phi_0)} = 2\pi\left[1+\frac{\partial^2}{k_{\lambda}^2\partial(x-x')^2}\right]J_0(k_{\lambda}|\rho-\rho'|\sin\theta) , \quad \text{(F4)}$$

where

$$\phi_0=\tan^{-1}\left[\frac{y-y'}{x-x'}\right] . \quad \text{(F5)}$$

The integration over polar angle θ is

$$g_r=\int_0^{\pi}\sin\theta\, d\theta\, J_0(k_\lambda|\boldsymbol{\rho}-\boldsymbol{\rho}'|\sin\theta)e^{ik_\lambda(z-z')\cos\theta}=\frac{2\sin(k_\lambda|\mathbf{r}-\mathbf{r}'|)}{k_\lambda|\mathbf{r}-\mathbf{r}'|} . \tag{F6}$$

We then get

$$D_{jj}(\mathbf{r},\mathbf{r}';\tau)=\frac{2}{\hbar}\frac{|\mathbf{d}_{21}|^2}{(2\pi c)^3}\int_0^{\infty}\omega_\lambda^3 d\omega_\lambda(2\pi)\left[1+\frac{c^2\partial^2}{\omega_\lambda^2\partial(x-x')^2}\right]g_r\frac{e^{-ik_\lambda(z-z')}}{(\omega_\lambda-\omega_j)^2+\Gamma_j^2} . \tag{F7}$$

Now it is easy to see that this can be integrated by making the change of variable $\omega=\omega_\lambda-\omega_j$,

$$\int_0^{\infty}\omega_\lambda^3 d\omega_\lambda\left[1+\frac{c^2\partial^2}{\omega_\lambda^2\partial(x-x')^2}\right]g_r\frac{e^{-ik_\lambda(z-z')}}{(\omega_\lambda-\omega_j)^2+\Gamma_j^2}$$

$$\approx\frac{c}{i}\left[\omega_j^2+\frac{c^2\partial^2}{\partial(x-x')^2}\right]\frac{1}{|\mathbf{r}-\mathbf{r}'|}\int_{-\infty}^{\infty}d\omega\frac{e^{i(R_-/c)(\omega+\omega_j)}-e^{-i(R_+/c)(\omega+\omega_j)}}{\omega^2+\Gamma_j^2}$$

$$=\frac{c}{i}\left[\omega_j^2+\frac{c^2\partial^2}{\partial(x-x')^2}\right]\frac{1}{|\mathbf{r}-\mathbf{r}'|}$$

$$\times\int_{-\infty}^{\infty}d\omega\left[e^{i\omega_j(R_-/c)}\frac{\cos\left[\frac{R_-}{c}\omega\right]+i\sin\left[\frac{R_-}{c}\omega\right]}{\omega^2+\Gamma_j^2}-e^{-i\omega_j(R_+/c)}\frac{\cos\left[\frac{R_+}{c}\omega\right]-i\sin\left[\frac{R_+}{c}\omega\right]}{\omega^2+\Gamma_j^2}\right]$$

$$=\frac{c}{i}\left[\omega_j^2+\frac{c^2\partial^2}{\partial(x-x')^2}\right]\frac{1}{|\mathbf{r}-\mathbf{r}'|}\left[\frac{\pi}{\Gamma_j}e^{-\Gamma_j(|R_-|/c)}e^{i\omega_j(R_-/c)}-\frac{\pi}{\Gamma_j}e^{-\Gamma_j(|R_+|/c)}e^{-i\omega_j(R_+/c)}\right] . \tag{F8}$$

Finally we have

$$D_{jj'}(\mathbf{r},\mathbf{r}';\tau)=-i\delta_{jj'}\frac{2}{\hbar}\frac{|\mathbf{d}_{21}|^2}{(2\pi c)^2}\frac{\pi}{\Gamma_j}\left[\omega_j^2+\frac{c^2\partial 2}{\partial(x-x')^2}\right]\frac{1}{|\mathbf{r}-\mathbf{r}'|}[e^{-\Gamma_j(|R_-|/c)}e^{i\omega_j(R_-/c)}-e^{-\Gamma_j(|R_+|/c)}e^{-i\omega_j(R_+/c)}] . \tag{F9}$$

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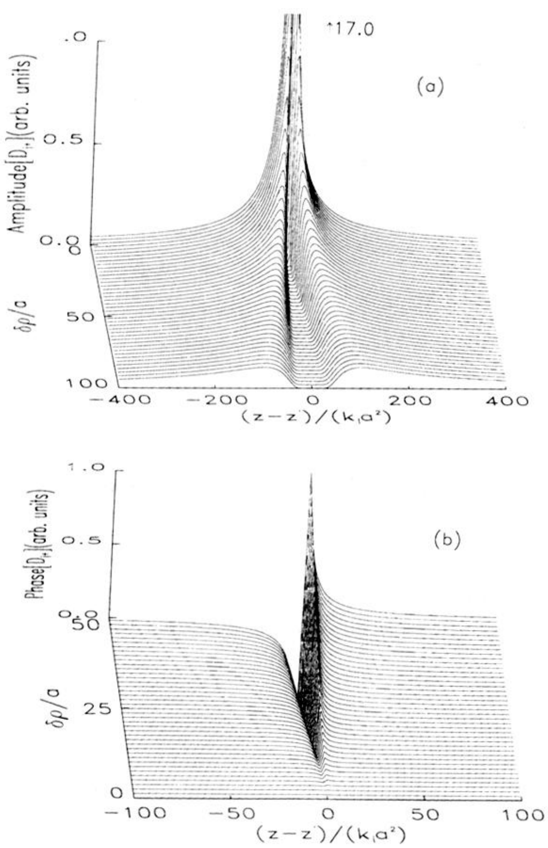

FIG. 4. (a) The amplitude of Eq. (6.24); (b) the phase of Eq. (6.24). Note the coarse-grained noise sources are not singular. We have scaled $\boldsymbol{\rho}$ (x and y) to a, and z to k_j/a^2.