Quantum field theory of cooperative atom response: Low light intensity

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We study the interactions of a possibly dense and/or quantum degenerate gas with driving light. Both the atoms and the electromagnetic fields are represented by quantum fields throughout the analysis. We introduce a field-theory version of Markov and Born approximations for the interactions of light with matter, and devise a procedure whereby certain types of products of atom and light fields may be put to a desired, essentially normal, order. In the limit of low light intensity we find a hierarchy of equations of motion for correlation functions that contain one excited-atom field and one, three, five, etc., ground-state atom fields. It is conjectured that the entire linear hierarchy may be solved by solving numerically the classical equations for a coupled system of electromagnetic fields and charged harmonic oscillators. We discuss the emergence of resonant dipole-dipole interactions and collective linewidths, and delineate the limits of validity of the column density approach in terms of noncooperative atoms by presenting a mathematical example in which this approach is exact. [S1050-2947(97)03901-2]

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I. INTRODUCTION

Indications of Bose-Einstein condensation (BEC) in trapped alkali-metal vapors have been reported recently [1–3]. At this point all direct probing of such condensates has been carried out optically. Correspondingly, in anticipation of BEC and the role of light in the experiments, the optical response of degenerate atomic gases has been the subject of active theoretical research already for quite some time [4–12]. Aside from the interest in BEC, in an evaporatively cooled gas of alkali atoms one may have a homogeneously broadened, weakly interacting system at such a high density that there are many atoms in a cubic wavelength, $\rho\lambda^3 \gg 1$. This kind of a sample would in its own right serve to further our understanding of the interactions of light with matter.

Nonetheless, in spite of all the theoretical work, there still are quite basic unsettled issues in the theory of the optical properties of dense and/or degenerate gases. Under the condition $\rho\lambda^3 \gg 1$ the atoms no longer respond to the electromagnetic fields individually, but their properties are modified by the presence of nearby atoms. For instance, the atoms exhibit collective linewidths and line shifts. Inasmuch as it comes to the near-resonant response in the regime $\rho\lambda^3 \gg 1$, all treatments of the optical properties of degenerate gases known to the present authors (including ours) contain uncontrolled approximations that bear on linewidths and line shifts. As a result, the regions of validity and the relations between different treatments tend to be somewhat ill defined.

A rigorous study of atom-field interactions valid regardless of atom density, atom statistics, optical detuning, and so forth, is clearly called for. The paper of Morice, Castin, and Dalibard [9] is a step in this direction. They start from a full quantum-mechanical Hamiltonian, including quantized light and internal degrees of freedom and c.m. motion of the atoms. However, at an early stage these authors go over to a classical treatment of the c.m. motion of the atoms. They then derive equations of motion for a few correlation functions involving polarization and atom density, and solve the optical response including all photon exchange between any pair of atoms.

The program carried out in Sec. II of the present paper is similar to the agenda of Ref. [9]. We start in Sec. II A from our field-theory version of the Hamiltonian as in Ref. [10], amended with the atom-atom contact interaction [13,9] that derives from the Power-Zienau approach. The point of departure from Ref. [9] is that we describe the atoms with quantum fields throughout. The mathematical techniques introduced in the process are analogous to the time-honored tools in quantum optics: the field-theory version of the Born-Markov approximation (Sec. II B), and procedures to move noncommuting operators to a certain advantageous order (Sec. II C). In the present paper we complete the derivation by assuming the limit of low intensity for driving light. The end result in Sec. II D is a hierarchy of equations of motion for correlation functions that involve atomic polarization at one point and densities at 0,1, . . . , points in space. The lowest two equations coincide with those given in Ref. [9].

In the present paper the emphasis is on the structure of the theory. To gain more insight, we examine a few simple special cases in Sec. III. We demonstrate the exquisite subtlety of the propagation of radiation through an atomic sample by presenting one particular set of assumptions that yields the standard column density results of (optically) noncooperating atoms (Sec. III A), and by reviewing the density expansion of Ref. [9] (Sec. III B). In the case of only two atoms, what we call cooperative linewidth and line shift emerge as manifestations of the dipole-dipole interaction. This is discussed in Sec. III C. Here we also point out that in our limit of low intensity of the driving light, the collective linewidth and line shift could perfectly well have been derived from classical electrodynamics of classical atoms (charged harmonic oscillators). In this paper we do not attempt to derive any new results from the hierarchy of correlation functions, but the connection to classical physics points to a possible future method for exact solution of the hierarchy: classical simulations of a system of classical atoms. A few comments to this effect are made in Sec. III D.

Concluding remarks about possible solutions and extensions of our hierarchy are made in the final Sec. IV. Certain mathematical details concerning the divergence of the dipolar field and a summary of dipole matrix elements are deferred to the Appendix.

II. MOTION OF ATOM FIELDS

A. Basic dynamics of the fields

We begin by recapping, reformulating, and extending the salient results of [10]. The main items of this section are the coupled evolution equations for the light and matter fields exemplified by Eqs. (4) and (15). Overall, we emphasize the similarities of the theory to the classical electrodynamics of polarizable media.

1. Hamiltonian

For better or worse, in this paper we regard atoms as point dipoles. A mathematically rigorous treatment produces a δ-function term in the field of a dipole at the position of the dipole, which results in a contact interaction between dipoles. For mathematical consistency, this time around we therefore also keep the contact interaction generated in the Power-Zienau transformation from the $\mathbf{p}\cdot\mathbf{A}$ to the $\mathbf{d}\cdot\mathbf{E}$ gauge [13,9]. This interaction was ignored as presumably inconsequential in the limit of large detuning considered in Ref. [10], but for an arbitrary detuning it may become an issue.

The atoms have two internal energy levels, which we label g for ''ground'' and e for ''excited.'' We allow for the angular momentum degeneracy of the energy levels, so the complete specification of the internal state of an atom αm includes the level label $\alpha=e$ or g and the z component of angular momentum m. We assume dipole coupling of each atom to light.

In first quantization, we add to the Hamiltonian of [10] the contact interaction, the polarization energy

$$H_{\mathrm{P}}=\frac{1}{2\epsilon_0}\sum_{i\neq j}\mathbf{d}_i\cdot\mathbf{d}_j\,\delta(\mathbf{r}_i-\mathbf{r}_j). \tag{1}$$

Here $\mathbf{d}_i$ and $\mathbf{r}_i$ are the dipole operator and the center-of-mass position operator for the ith atom. There are also divergent self-energies with $i=j$, but we ignore these as we do not attempt a quantitative calculation of the Lamb shift. Equation (1) displays a standard two-body interaction, which is immediately converted to second quantization. As before, the many-atom system is described by Heisenberg picture quantum fields $\psi_{\alpha m}(\mathbf{r}t)$, which obey the proper commutator relations. While much of our development applies to fermions as well, in this paper we consider only bosons explicitly. In terms of the atom fields, the additional polarization energy is the integral of the Hamiltonian density

$$\begin{aligned}\mathcal{H}_{\mathrm{P}}=\frac{1}{\epsilon_0}\sum_{\substack{m_1m_2\\M_1M_2}}&[\mathbf{d}_{m_2M_1}\cdot\mathbf{d}_{M_2m_1}\psi^\dagger_{gm_2}\psi^\dagger_{eM_2}\psi_{eM_1}\psi_{gm_1}\\&+\tfrac{1}{2}\mathbf{d}_{M_2m_2}\cdot\mathbf{d}_{M_1m_1}\psi^\dagger_{eM_2}\psi^\dagger_{eM_1}\psi_{gm_2}\psi_{gm_1}\\&+\tfrac{1}{2}\mathbf{d}_{m_2M_2}\cdot\mathbf{d}_{m_1M_1}\psi^\dagger_{gm_2}\psi^\dagger_{gm_1}\psi_{eM_2}\psi_{eM_1}].\end{aligned} \tag{2}$$

The notation $\mathbf{d}_{mM}$ stands for the matrix element $\langle gm|\mathbf{d}|eM\rangle$ of the dipole operator of one atom, $\mathbf{d}$. We denote the energy level implicitly in such a way that a label of a Zeeman state with a lower case m refers to the ground state, and an upper case M to the excited state.

However, to simplify the notation as far as possible, we are going to adopt yet another convention that is in force unless we explicitly state otherwise. We do not write the magnetic quantum numbers explicitly. For instance, we write ψ_{e_2} in lieu of ψ_{eM_2}. Also, we write the matrix elements $\mathbf{d}_{mM}$ as $\mathbf{d}_{ge}$. Finally, if the same level index appears twice in a product, a sum over the magnetic substates of the level is implied. With these conventions, we write Eq. (2) anew as

$$\begin{aligned}\mathcal{H}_{\mathrm{P}}=\frac{1}{\epsilon_0}&[\mathbf{d}_{g_2e_1}\cdot\mathbf{d}_{e_2g_1}\psi^\dagger_{g_2}\psi^\dagger_{e_2}\psi_{e_1}\psi_{g_1}\\&+\tfrac{1}{2}\mathbf{d}_{e_2g_2}\cdot\mathbf{d}_{e_1g_1}\psi^\dagger_{e_2}\psi^\dagger_{e_1}\psi_{g_2}\psi_{g_1}\\&+\tfrac{1}{2}\mathbf{d}_{g_2e_2}\cdot\mathbf{d}_{g_1e_1}\psi^\dagger_{g_2}\psi^\dagger_{g_1}\psi_{e_2}\psi_{e_1}].\end{aligned} \tag{3}$$

2. Electromagnetic fields

Unlike in [10], and similarly to [9], we assume that there is a cutoff in the wave numbers q of the photons; we multiply the density of the states of the electromagnetic fields by $e^{-q^2\alpha^2/4}$, with $\alpha>0$ being a length scale. The cutoff removes all mathematical problems concerning, e.g., the exchange of the order of derivatives and integrals, which are abundant in the theory without the cutoff. At the end of the calculations we ultimately take the limit $\alpha\to 0$.

In spite of the change in the Hamiltonian and the added cutoff of photon frequencies, the analysis of the electromagnetic fields proceeds almost as in [10]. In accordance with Ref. [13], it emerges from our results that what was called the electric field in [10] should more properly be interpreted as the electric displacement divided by the permittivity of the vacuum ϵ_0. We henceforth adopt this interpretation. The positive frequency part of the electric displacement is expressed in terms of the matter fields as

$$\begin{aligned}\mathbf{D}^+(\mathbf{r}t)=\mathbf{D}^+_F(\mathbf{r}t)+\epsilon_0\int_{-\infty}^{t}dt'\int d^3r'\,\mathbf{S}(\mathbf{d}_{ge};\mathbf{r}-\mathbf{r}',t-t')\\\times\psi^\dagger_g(\mathbf{r}'t')\,\psi_e(\mathbf{r}'t').\end{aligned} \tag{4}$$

In this approach electric displacement and matter fields are the primary degrees of freedom; $\mathbf{D}^+_F(\mathbf{r}t)$ is the free electric displacement that would apply if there were no coupling between matter and electromagnetic fields. The propagator that takes the radiation from the dipole source to the field point is

$$\begin{aligned}\mathbf{S}(\boldsymbol{\mathcal{D}};\mathbf{r}t)&=\frac{ic}{16\pi^3\epsilon_0}\int d^3q\,e^{-q^2\alpha^2/4}\,q\,\frac{\mathbf{q}}{q}\times\left(\frac{\mathbf{q}}{q}\times\boldsymbol{\mathcal{D}}\right)\\&\quad\times e^{i\mathbf{q}\cdot\mathbf{r}}(e^{icqt}-e^{-icqt})\qquad\text{(5a)}\\&=\frac{c}{4\pi\epsilon_0}(\boldsymbol{\mathcal{D}}\times\boldsymbol{\nabla})\times\boldsymbol{\nabla}\frac{\delta_\alpha[r-ct]-\delta_\alpha[r+ct]}{|\mathbf{r}-\mathbf{r}'|},\qquad\text{(5b)}\end{aligned}$$

where the δ function has acquired a finite width as a result of the cutoff in the photon energy spectrum,

$$\delta_\alpha(x)=\frac{1}{\sqrt{\pi}\alpha}\exp\left[-\left(\frac{x}{\alpha}\right)^2\right]. \tag{6}$$

As before, we assume that there is a dominant frequency Ω in the problem. In fact, we generally assume that a field such as $\psi_e(\mathbf{r}t)e^{i\Omega t}$, and similarly for the electromagnetic quantities, varies ''slowly'' in time in comparison with $e^{-i\Omega t}$. From now on a notation such as $\psi_e(\mathbf{r}t)$ refers to the slowly varying field $\psi_e(\mathbf{r}t)e^{i\Omega t}$, unless explicitly stated otherwise.

Based on an implicit cutoff such as α, we argued in [10] that δ functions with plus and minus signs in Eq. (5b) conspire to remove a term $\propto\delta(\mathbf{r}-\mathbf{r}')$ that results when the position derivatives act on $|\mathbf{r}-\mathbf{r}'|^{-1}$. What we did not realize is that this δ function does not outright vanish. Instead, it is smeared to a function whose integral over $\mathbf{r}$ is still unity but which has a finite width of the order α; see the Appendix, Sec. 1. From now on, as long as the integral operator with the kernel $\mathbf{S}$ acts on any smooth function $\phi(\mathbf{r}t)e^{-i\Omega t}$ of $\mathbf{r}$ and t in which the exponential is the dominant time dependence, we write

$$\int_{-\infty}^{t}dt'\int d^3r'\,\mathbf{S}(\boldsymbol{\mathcal{D}};\mathbf{r}-\mathbf{r}',t-t')\phi(\mathbf{r}'t')e^{-i\Omega t'}$$

$$=e^{-i\Omega t}\int d^3r'\,\mathbf{S}'(\boldsymbol{\mathcal{D}};\mathbf{r}-\mathbf{r}')\phi(\mathbf{r}'t_d). \tag{7}$$

Here

$$t_d=t-\frac{|\mathbf{r}-\mathbf{r}'|}{c} \tag{8}$$

is the retarded time. The monochromatic version of the propagator $\mathbf{S}$, $\mathbf{S}'$ may be written alternatively as

$$\mathbf{S}'(\boldsymbol{\mathcal{D}};\mathbf{r})=\frac{1}{4\pi\epsilon_0}(\boldsymbol{\mathcal{D}}\times\boldsymbol{\nabla})\times\boldsymbol{\nabla}\frac{e^{ikr}}{r} \tag{9a}$$

or

$$\mathbf{S}'(\boldsymbol{\mathcal{D}};\mathbf{r})=\mathbf{K}(\boldsymbol{\mathcal{D}};\mathbf{r})+\frac{2}{3\epsilon_0}\boldsymbol{\mathcal{D}}\delta(\mathbf{r}). \tag{9b}$$

The final kernel $\mathbf{K}(\boldsymbol{\mathcal{D}};\mathbf{r})$ is equal to the positive-frequency component of the electric field from a monochromatic dipole with the complex amplitude $\boldsymbol{\mathcal{D}}$, given that the dipole resides at the origin and the field is observed at $r\neq0$. The explicit expression is, of course [14]

$$\mathbf{K}(\boldsymbol{\mathcal{D}};\mathbf{r})=\frac{1}{4\pi\epsilon_0}\left\{k^2(\hat{\mathbf{n}}\times\boldsymbol{\mathcal{D}})\times\hat{\mathbf{n}}\frac{e^{ikr}}{r}+[3\hat{\mathbf{n}}(\mathbf{n}\cdot\boldsymbol{\mathcal{D}})-\boldsymbol{\mathcal{D}}]\right.$$

$$\left.\times\left(\frac{1}{r^3}-\frac{ik}{r^2}\right)e^{ikr}\right\}, \tag{10}$$

with

$$\hat{\mathbf{n}}=\frac{\mathbf{r}}{r},\quad k=\frac{\Omega}{c}. \tag{11}$$

It should be noted that, as the dipole radiation has a $1/r^3$ singularity, integrals such as Eq. (7) are generally not absolutely convergent. According to Sec. 1 of the Appendix, we resolve this problem by the rule that at least in the immediate vicinity of the divergence $\mathbf{r}'=\mathbf{r}$ the integral is to be performed in spherical polar coordinates, and the angles are first to be integrated over. It should also be borne in mind that the form (7) does not apply if the function ϕ is singular in $\mathbf{r}$.

We finally consider the quantum expectation value of Eq. (4). As is always done in this paper, we take the free field to be in a coherent state. We also assume that the expectation value $\langle\mathbf{D}_F^+\rangle$ is effectively monochromatic. It is physically evident that, at least in steady state, the expectation value of the product $\psi_g^\dagger\psi_e$ will then be monochromatic, and a smooth function of $\mathbf{r}$ as well. Inasmuch as the quantum expectation value of Eq. (4) is concerned, the transformation from kernel $\mathbf{S}$ to kernel $\mathbf{S}'$ is thus allowed. Moreover, the expectation value of the free field is a solution to the Helmholtz equation, and the function e^{ikr}/r is essentially the Green's function of the Helmholtz differential operator:

$$(\boldsymbol{\nabla}^2+k^2)\langle\mathbf{D}_F^+\rangle=0,\quad(\boldsymbol{\nabla}^2+k^2)\frac{e^{ikr}}{r}=-4\pi\delta(\mathbf{r}). \tag{12}$$

In view of Eqs. (7) and (9a), from Eq. (4) we thus have

$$(\boldsymbol{\nabla}^2+k^2)\langle\mathbf{D}^+\rangle=-\boldsymbol{\nabla}\times(\boldsymbol{\nabla}\times\langle\mathbf{P}^+\rangle). \tag{13}$$

Classically, the polarization of the medium is defined as the dipole moment per atom times the density of atoms. It is therefore clear that

$$\mathbf{P}^+(\mathbf{r})=\mathbf{d}_{ge}\psi_g^\dagger(\mathbf{r})\psi_e(\mathbf{r}) \tag{14}$$

should be identified as the (positive frequency part of the) quantum-mechanical polarization operator.

The value of Eq. (13) is twofold. First, it is a local differential equation, as opposed to the integral equation (4). Second, it has a well-known counterpart in the classical electrodynamics of polarizable media. This reinforces the interpretations of $\mathbf{D}$ and $\mathbf{P}$ as electric displacement and polarization operators.

3. Matter fields

We now turn to the equations of motion of the matter fields. Under the assumptions that the density of excited atoms is low and that an atom moves much less than a wavelength of resonant light during the time it remains excited, we have the equations of motion for the fields describing excited- and ground-state atoms:

$$\dot{\psi}_e(\mathbf{r}t)=i\,\delta\psi_e(\mathbf{r}t)+\frac{i}{\hbar}\mathbf{d}_{eg}\cdot\mathbf{E}^+(\mathbf{r}t)\psi_g(\mathbf{r}t), \tag{15a}$$

$$\dot{\psi}_g(\mathbf{r}t)=\frac{i}{\hbar}\mathbf{E}^-(\mathbf{r}t)\cdot\mathbf{d}_{ge}\psi_e(\mathbf{r}t)$$

$$-i\frac{H_{\text{c.m.}}(\mathbf{r})}{\hbar}\psi_g(\mathbf{r}t)+\left.\frac{d}{dt}\right|_C\psi_g(\mathbf{r}t). \tag{15b}$$

As the notation implies,

$$\mathbf{E}^+(\mathbf{r}t)=\frac{1}{\epsilon_0}[\mathbf{D}^+(\mathbf{r}t)-\mathbf{P}^+(\mathbf{r}t)] \tag{16}$$

is to be interpreted as the electric field. Furthermore, $\delta=\Omega-\omega_0$ is the detuning of the characteristic frequency of the light Ω from the atomic resonance frequency ω_0. We have carried out the rotating-wave approximation that takes into account the dominant field frequency Ω. Finally, $H_{\text{c.m.}}$ is the one-particle Hamiltonian governing the c.m. motion of the atoms, and the time derivative with the subscript C represents collisions.

4. Summary remarks

The two-state model of quantum optics is immediately seen to underlie Eqs. (15), and Eq. (4) describes the total field as the incident field plus the fields radiated by the dipoles, complete with propagation delays. In spite of the familiar appearances, though, it should be noted that the only real approximations so far have been to ignore the c.m. motion and collisions of the excited atoms. The formulation still fully accounts for quantum statistics of the many-atom system, and for the quantized electromagnetic fields. The effect of the dipole-dipole interactions on the transition frequencies and linewidths of the atoms is still included. On the other hand, as we have ignored the c.m. Hamiltonian of excited atoms, collisions between ground-state atoms and excited atoms can no longer be discussed.

B. Eliminating the vacuum field

Even in the absence of applied electromagnetic fields, the atoms bathe in vacuum fields that cause spontaneous emission and Lamb shifts. The purpose of the present section is to account for the vacuum fields. While pursuing this goal, we need to be prepared for singular functions with rapid spatial and temporal variations. We therefore start with the general field equation (4). Moreover, for the time being we argue in terms of the original atomic and electromagnetic fields untempered by the exponential $e^{i\Omega t}$.

To begin with, we insert Eq. (4) into Eq. (15a), and obtain

$$\begin{aligned}\dot{\psi}_e(\mathbf{r}t)=&-i\omega_0\psi_e(\mathbf{r}t)+\frac{i}{\hbar\epsilon_0}\mathbf{d}_{eg}\cdot\Big\{\mathbf{D}_F^+(\mathbf{r}t)\psi_g(\mathbf{r}t)\\&-\mathbf{d}_{g'e'}\psi_{g'}^\dagger(\mathbf{r}t)\psi_g(\mathbf{r}t)\psi_{e'}(\mathbf{r}t)\\&+\epsilon_0\int_\infty^t dt'\int d^3r'\,\mathbf{S}(\mathbf{d}_{g'e'};\mathbf{r}-\mathbf{r}',t-t')\\&\times\psi_{g'}^\dagger(\mathbf{r}'t')\psi_{e'}(\mathbf{r}'t')\psi_g(\mathbf{r}t)\Big\}.\end{aligned} \tag{17}$$

We are ultimately interested in quantum expectation values of atomic and electromagnetic field operators, and thus wish to be able to take expectation values of expressions such as Eq. (17) easily. It would be especially valuable to have the free-field operator $\mathbf{D}_F^+(\mathbf{r}t)$ farthest to the right. Because the (initial) free electromagnetic field is assumed to be in the coherent state, in an expectation value this operator would then reduce to a multiplicative classical field: the relation

$$\langle\mathcal{O}\mathbf{D}_F^+(\mathbf{r}t)\rangle=\langle\mathcal{O}\rangle\langle\mathbf{D}_F^+(\mathbf{r}t)\rangle \tag{18}$$

applies to any operator $\mathcal{O}$. Evidently we need commutators between atom operators and free-field operators, so that we may move the latter to the desired positions.

Any atom operator, of course, commutes with the total electric displacement operator $\mathbf{D}^\pm$ at the same time. Thus from Eq. (4) we have

$$\begin{aligned}Q=&[\psi_g(\tilde{\mathbf{r}}t),\mathbf{D}_F^+(\mathbf{r}t)]\\=&-\epsilon_0\int_{-\infty}^t dt'\int d^3r'\,\mathbf{S}(\mathbf{d}_{g'e};\mathbf{r}-\mathbf{r}',t-t')\\&\times[\psi_g(\tilde{\mathbf{r}}t),\psi_{g'}^\dagger(\mathbf{r}'t')\psi_e(\mathbf{r}'t')].\end{aligned} \tag{19}$$

Here we are preparing for the eventuality that the commutator is required for two different field points.

The standard way of dealing with vacuum fields in quantum optics is the duo of Born and Markov approximations: the atom operators evolve as if no electromagnetic fields were present (Born) during the short vacuum correlation time (Markov) [15]. For implementations of this idea in the Heisenberg picture see, e.g., [16–18]. We evaluate the commutator Q under an approximation which, we think, is the field-theory equivalent of the standard Born and Markov approximations: We assume that during the time it takes radiation reaction effects to assert themselves, the atom fields evolve as if they were completely noninteracting. We temporarily restore the explicit notation for magnetic quantum numbers, and write the Born-Markov approximation for Eq. (19) as

$$\begin{aligned}\psi_{gm}(\mathbf{r}t')&=\frac{1}{\sqrt{V}}\sum_{\mathbf{k}}\,e^{i[\mathbf{k}\cdot\mathbf{r}-\epsilon_{\mathbf{k}}(t'-t)]}b_{gm\mathbf{k}}(t),\\\psi_{eM}(\mathbf{r}t')&=\frac{1}{\sqrt{V}}\sum_{\mathbf{k}}\,e^{i[\mathbf{k}\cdot\mathbf{r}-\omega_0(t'-t)]}b_{eM\mathbf{k}}(t).\end{aligned} \tag{20}$$

Here $\epsilon_{\mathbf{k}}=\hbar\mathbf{k}^2/2m$ gives the dispersion relation for an atom with mass m, b's are boson operators, and the sums run over the wave vectors $\mathbf{k}$ appropriate for the quantization volume V. In the standard continuum limit the relevant commutator becomes

$$[\psi_{gm}(\tilde{\mathbf{r}}t),\psi_{gm''}^\dagger(\mathbf{r}'t')]=\frac{\delta_{mm''}}{(2\pi)^3}\int d^3k\,e^{i\mathbf{k}\cdot(\tilde{\mathbf{r}}-\mathbf{r}')-i\epsilon_k(t-t')}. \tag{21}$$

We use $\mathbf{S}$ from Eq. (5a), the commutator from Eq. (21), $\psi_{gm'}^\dagger$ from (20), and add the conventional convergence factor $e^{-\eta t}$ to the time integral. Equation (19) is cast in the form

$$\begin{aligned}Q=&\frac{-ic}{16\pi^3\sqrt{V}}\sum_{\mathbf{K},M'}e^{i\mathbf{K}\cdot\tilde{\mathbf{r}}}b_{eM'\mathbf{K}}\int_0^\infty d\tau\,e^{-\eta\tau}\int d^3q\\&\times e^{i\mathbf{q}\cdot(\tilde{\mathbf{r}}-\mathbf{r})}e^{-\alpha^2q^2/4}\,q\frac{\mathbf{q}}{q}\times\left(\frac{\mathbf{q}}{q}\times\mathbf{d}_{mM'}\right)\\&\times(e^{icq\tau}-e^{-icq\tau})e^{-i\epsilon_{\mathbf{K}-\mathbf{q}}\tau+i\omega_0\tau}.\end{aligned} \tag{22}$$

Our final approximation is to ignore the c.m. energies in comparison with the energy of the internal transition of the atom; we write $\omega_0 - \epsilon_{\mathbf{K}-\mathbf{q}} \simeq \omega_0 \simeq \Omega$. The result is interesting:

$$Q = -\epsilon_0 \int_0^\infty d\tau\, \mathbf{S}(\mathbf{d}_{ge};\tilde{\mathbf{r}}-\mathbf{r},\tau) e^{i\Omega\tau} \psi_e(\tilde{\mathbf{r}}t). \tag{23}$$

The time integral is the same as the definition of the kernel $\mathbf{S}'(\mathbf{d}_{ge};\tilde{\mathbf{r}}-\mathbf{r})$ in Eq. (9), albeit still containing the cutoff parameter α. The cutoff is truly needed: in our immediate application to Eq. (17) we are to set $\tilde{\mathbf{r}}=\mathbf{r}$, and without the cutoff we would have to contend with a pernicious singularity of the type $\delta(\mathbf{r})/r^3$. For a small but nonzero α, the result is

$$[\psi_g(\mathbf{r}t), \mathbf{D}_F^+(\mathbf{r}t)] = \mathbf{d}_{ge}\left(\frac{2\pi^{-3/2}}{3\alpha^3} - i\frac{\omega_0^3}{6\pi c^3}\right)\psi_e(\mathbf{r}t). \tag{24}$$

Given the sum rule for the dipole moment matrix elements, Eq. (A15), the relevant terms in Eq. (17) become

$$\begin{aligned}\dot{\psi}_e(\mathbf{r}t) &= -i\omega_0\psi_e(\mathbf{r}t) + \frac{i}{\hbar\epsilon_0}\mathbf{d}_{eg}\cdot\mathbf{D}_F^+(\mathbf{r}t)\psi_g(\mathbf{r}t)\cdots \\ &= -i\omega_0\psi_e(\mathbf{r}t) - \left[\gamma + i\frac{2\mathcal{D}^2\sqrt{\pi}}{3\pi^2\epsilon_0\hbar\alpha^3}\right]\psi_e \\ &\quad + \frac{i}{\hbar\epsilon_0}\mathbf{d}_{eg}\cdot\psi_g(\mathbf{r}t)\mathbf{D}_F^+(\mathbf{r}t)\ldots,\end{aligned} \tag{25}$$

where $\mathcal{D}$ is the reduced dipole moment matrix element. The imaginary part in the second term on the right-hand side of Eq. (25) diverges as the photon momentum cutoff goes to infinity with $\alpha \to 0$. This part, after a proper renormalization, contributes to the Lamb shift. From now on we assume that the Lamb shift is already included in the transition frequency, and ignore the α^{-3} term in Eq. (25). What remains is the familiar spontaneous linewidth of the atomic transition,

$$\gamma = \frac{\mathcal{D}^2\omega_0^3}{6\pi\hbar\epsilon_0 c^3}. \tag{26}$$

There are no divergence problems with the commutator of ψ_g and $\mathbf{D}_F^+$ if the position arguments are different. We simply write

$$[\psi_g(\tilde{\mathbf{r}}t), \mathbf{D}_F^+(\mathbf{r}t)] = -\epsilon_0\mathbf{S}'(\mathbf{d}_{ge};\mathbf{r}-\tilde{\mathbf{r}})\psi_e(\tilde{\mathbf{r}}t). \tag{27a}$$

In fact, if the divergent in-phase part of the dipole field at $\tilde{\mathbf{r}}=\mathbf{r}$ is ignored (or incorporated into the Lamb shift), we may interpret Eq. (27a) to be valid even for $\tilde{\mathbf{r}}=\mathbf{r}$.

By the same token, we may carry out all commutators between atom fields and free electromagnetic fields. The two additional nonvanishing commutators that play some role in this paper are

$$[\psi_g^\dagger(\tilde{\mathbf{r}}t), \mathbf{D}_F^-(\mathbf{r}t)] = \epsilon_0\psi_e^\dagger(\tilde{\mathbf{r}}t)[\mathbf{S}'(\mathbf{d}_{ge};\mathbf{r}-\tilde{\mathbf{r}})]^*, \tag{27b}$$

$$[\psi_e(\tilde{\mathbf{r}}t), \mathbf{D}_F^-(\mathbf{r}t)] = -\epsilon_0\psi_g(\tilde{\mathbf{r}}t)[\mathbf{S}'(\mathbf{d}_{ge};\mathbf{r}-\tilde{\mathbf{r}})]^*. \tag{27c}$$

At this point we restore our convention of slowly varying fields. We also add another assumption to the effect that light has ample time to propagate across the atomic sample during the time that it takes the slowly varying fields to change appreciably. This permits us to ignore propagation delays in the time arguments of the slowly varying fields. The retarded time t_d is simply replaced by the external time t. We have thus obtained an equation of motion for the excited-state atom field that contains an explicit radiative damping,

$$\begin{aligned}\dot{\psi}_e(\mathbf{r}) &= (i\delta-\gamma)\psi_e(\mathbf{r}) + \frac{i}{\hbar\epsilon_0}\mathbf{d}_{eg}\cdot\Big\{\psi_g(\mathbf{r})\mathbf{D}_F^+(\mathbf{r}) \\ &\quad - \mathbf{d}_{g'e'}\psi_{g'}^\dagger(\mathbf{r})\psi_{e'}(\mathbf{r})\psi_g(\mathbf{r}) \\ &\quad + \epsilon_0\int d^3r'\,\mathbf{S}'(\mathbf{d}_{g'e'};\mathbf{r}-\mathbf{r}') \\ &\quad \times\psi_{g'}^\dagger(\mathbf{r}')\psi_{e'}(\mathbf{r}')\psi_g(\mathbf{r})\Big\}.\end{aligned} \tag{28}$$

Here, and in our subsequent expressions, the common time t is omitted in the notation.

Unlike in the ordinary treatments of spontaneous emission in the quantum optics of an isolated atom, no short vacuum correlation time suggests itself in our formulation. The use of the free-field evolution as in Eqs. (20) during the ''vacuum correlation time'' may thus seem like an *ad hoc* assumption. This approximation, however, did produce spontaneous damping and Lamb shift in accordance with the one-atom theory. Of course, even in standard quantum optics the atomic variables do not evolve completely freely during the vacuum correlation time. Spontaneous emission itself, as well as external driving electromagnetic fields and collisions between the atoms in principle affect spontaneous emission, but at ordinary conditions for laser spectroscopy these influences are negligible [15]. We conjecture that the same applies in our field-theory version of spontaneous emission. Finally, ignoring c.m. energies in comparison with the energy of the atomic transition is nothing new either. This is a standard approximation in the derivation of spontaneous emission in the theory of light pressure [19]. If such energies *were* included, a velocity-dependent spontaneous emission rate would emerge in manifest contradiction with special relativity [20].

C. Hierarchy for operator products

A particularly relevant atomic variable is the polarization operator (14), which acts as the source for secondary radiation. Generalizing slightly, we now embark on a study of the time evolution of the operator product $\psi_g^\dagger(\mathbf{r})\psi_e(\mathbf{r})$.

We have in mind situations in which collisions and c.m. motion of the ground-state atoms have come to a steady state before the driving light is turned on. We regard the external field as a small perturbation, so that ground-state atoms remain materially unperturbed in the presence of the light. As the final item, we assume that collisions and c.m. motion of the ground-state atoms take place on a time scale much longer than the spontaneous emission time scale γ^{-1}. They are therefore not expected to interfere with spectroscopic probing of the atomic transition. Under these assumptions we will henceforth ignore the collision terms and the c.m. evolution of the ground-state atoms altogether. Nonetheless,

mathematical consistency dictates that in the intermediate steps of our calculations we take into account some light driven evolution even for the ground-state atoms.

We thus have the equation of motion from Eqs. (15b) and (28),

$$\frac{d}{dt}\psi_g^\dagger\psi_e=(i\delta-\gamma)\psi_g^\dagger\psi_e+\frac{i}{\hbar\epsilon_0}\mathbf{d}_{eg'}\cdot\Big\{\psi_g^\dagger\psi_{g'}\mathbf{D}_F^+-\psi_g^\dagger\mathbf{P}^+\psi_{g'}+\epsilon_0\int d^3r'\,\psi_g^\dagger\mathbf{S}'(\mathbf{P}^+(\mathbf{r}'),\mathbf{r}-\mathbf{r}')\psi_{g'}\Big\}-\frac{i}{\hbar\epsilon_0}\mathbf{d}_{e'g}\cdot\Big\{\psi_{e'}^\dagger\psi_e\mathbf{D}_F^+-\psi_{e'}^\dagger\mathbf{P}^+\psi_e+\epsilon_0\int d^3r'\,\psi_{e'}^\dagger\mathbf{S}'(\mathbf{P}^+(\mathbf{r}'),\mathbf{r}-\mathbf{r}')\psi_e\Big\},\tag{29}$$

where we have shown explicitly only the nonlocal position dependence. Generalizing, we are evidently about to derive a hierarchy of equations for operators of the type $\mathbf{P}^+$, $\psi_g^\dagger\mathbf{P}^+\psi_g$, $\psi_g^\dagger\psi_g^\dagger\mathbf{P}^+\psi_g\psi_g$, ..., with different position arguments for the different fields.

Now, light has to be present in order to produce excited atoms. Each excited-state field corresponds to one order in the strength of the driving light. To first order in $\mathbf{D}_F^\pm$ we might thus ignore the second term on the right-hand side of Eq. (29) altogether. This is, generally speaking, what we will do: only retain those products of operators that involve at most one of the operators $\mathbf{D}_F^+$, $\mathbf{D}_F^-$, ψ_e, or $\psi_e^\dagger$. However, caution must be exercised for two reasons. First, we will eventually arrange all atom fields to normal order; creation operators to the left, annihilation operators to the right. In addition, we move the free-field operators to prescribed positions. In the process commutators are generated that may be of different order in the strength of the driving field than the original terms. Second, some of the commutators are flat out divergent, analogous to the Lamb shift. It may be shown that such extra Lamb shifts cancel exactly, order by order in the strength of the driving field, but the cancellation of course fails if the calculations are not consistent in the orders. We will not dwell on the latter aspect anymore, but simply ignore all orders higher than the first immediately at the point when the operators have been brought to the desired order.

We illustrate the process of deriving the hierarchy of equations for operator products with a detailed treatment of the time evolution of a particular product,

$$\frac{d}{dt}[\psi_{g_1}^\dagger(\mathbf{r}')\psi_g^\dagger(\mathbf{r})\psi_e(\mathbf{r})\psi_{g_1'}(\mathbf{r}')]=\Big[\frac{d}{dt}\psi_{g_1}^\dagger(\mathbf{r}')\Big]\psi_g^\dagger(\mathbf{r})\psi_e(\mathbf{r})\psi_{g_1'}(\mathbf{r}')+\psi_{g_1}^\dagger(\mathbf{r}')\Big[\frac{d}{dt}\psi_g^\dagger(\mathbf{r})\psi_e(\mathbf{r})\Big]\psi_{g_1'}(\mathbf{r}')+\psi_{g_1}^\dagger(\mathbf{r}')\psi_g^\dagger(\mathbf{r})\psi_e(\mathbf{r})\frac{d}{dt}\psi_{g_1'}(\mathbf{r}').\tag{30}$$

In the way of preparation, let us first note from Eq. (4) and its Hermitian conjugate that the electric displacement may be written in the form

$$\mathbf{D}^\pm=\mathbf{D}_F^\pm+\mathbf{D}_S^\pm,\tag{31}$$

where the source term $\mathbf{D}_S^\pm$ is a normal-ordered combination of atom fields. We begin our analysis with the third term on the right-hand side of Eq. (30). By virtue of Eqs. (15b) and (16) we first have

$$\begin{aligned}\psi_{g_1}^\dagger(\mathbf{r}')\psi_g^\dagger(\mathbf{r})\psi_e(\mathbf{r})\frac{d}{dt}\psi_{g_1'}(\mathbf{r}')&=\frac{i}{\hbar}\psi_{g_1}^\dagger(\mathbf{r}')\psi_g^\dagger(\mathbf{r})\psi_e(\mathbf{r})\mathbf{d}_{g_1'e'}\cdot\mathbf{E}^-(\mathbf{r}')\psi_{e'}(\mathbf{r}')\\&=\frac{i}{\hbar\epsilon_0}\psi_{g_1}^\dagger(\mathbf{r}')\psi_g^\dagger(\mathbf{r})\psi_e(\mathbf{r})\mathbf{d}_{g_1'e'}\cdot[\mathbf{D}^-(\mathbf{r}')-\mathbf{P}^-(\mathbf{r}')]\psi_{e'}(\mathbf{r}')\\&=\frac{i}{\hbar\epsilon_0}[\psi_{g_1}^\dagger(\mathbf{r}')\psi_g^\dagger(\mathbf{r})\mathbf{d}_{g_1'e'}\cdot\mathbf{D}^-(\mathbf{r}')\psi_e(\mathbf{r})\psi_{e'}(\mathbf{r}')-\psi_{g_1}^\dagger(\mathbf{r}')\psi_g^\dagger(\mathbf{r})\psi_e(\mathbf{r})\mathbf{d}_{g_1'e'}\cdot\mathbf{P}^-(\mathbf{r}')\psi_{e'}(\mathbf{r}')]\\&=\frac{i}{\hbar\epsilon_0}[\psi_{g_1}^\dagger(\mathbf{r}')\psi_g^\dagger(\mathbf{r})\mathbf{d}_{g_1'e'}\cdot\mathbf{D}_S^-(\mathbf{r}')\psi_e(\mathbf{r})\psi_{e'}(\mathbf{r}')+\psi_{g_1}^\dagger(\mathbf{r}')\psi_g^\dagger(\mathbf{r})\mathbf{d}_{g_1'e'}\cdot\mathbf{D}_F^-(\mathbf{r}')\psi_e(\mathbf{r})\psi_{e'}(\mathbf{r}')\\&\quad-\psi_{g_1}^\dagger(\mathbf{r}')\psi_g^\dagger(\mathbf{r})\psi_e(\mathbf{r})\mathbf{d}_{g_1'e'}\cdot\mathbf{P}^-(\mathbf{r}')\psi_{e'}(\mathbf{r}')].\end{aligned}\tag{32}$$

Since the total displacement $\mathbf{D}^-$ commutes with all atom fields, we first moved it between atom creation and annihilation operators. The term involving the source field $\mathbf{D}_S^-$ is then readily in normal order. In addition, here the source field term is third order in the perturbation, so it may be omitted. However, we are not yet done with operator orderings. First, just as the free-field operator $\mathbf{D}_F^+$ is profitably moved to the right of atom operators, the free-field operator $\mathbf{D}_F^-$ should be transported all the way to the left. By virtue of Eq. (27b), this leaves behind two commutator terms. Nevertheless, both the term with $\mathbf{D}_F^-$ remaining and the commutators are formally third order in the strength of the driving field, and we ignore them all. Finally, the term involving $\mathbf{P}^-$ is not yet in normal order, but it may be made so easily by using the commutators of the atom fields. The rearranged term is third order and negligible, but the generated commutator is first order. We eventually have

$$\psi^\dagger_{g_1}(\mathbf{r}')\psi^\dagger_g(\mathbf{r})\psi_e(\mathbf{r})\frac{d}{dt}\psi_{g'_1}(\mathbf{r}') = -\frac{i}{\hbar\epsilon_0}\mathbf{d}_{g'_1e'}\cdot\mathbf{d}_{eg''}\delta(\mathbf{r}-\mathbf{r}')\psi^\dagger_{g_1}(\mathbf{r}')\psi^\dagger_g(\mathbf{r})\psi_{g''}(\mathbf{r}')\psi_{e'}(\mathbf{r}'). \tag{33}$$

The same analysis may be carried out with the other two terms in Eq. (30). The first term contributes nothing in the first order, while the second term gives a homogeneous term proportional to $i\delta-\gamma$, a driven term proportional to $\mathbf{D}^+_F$, and something of a two-atom analog of radiation reaction. The final result is

$$\begin{aligned}\frac{d}{dt}[\psi^\dagger_{g_1}(\mathbf{r}')\psi^\dagger_g(\mathbf{r})\psi_e(\mathbf{r})\psi_{g'_1}(\mathbf{r}')] &= (i\delta-\gamma)\psi^\dagger_{g_1}(\mathbf{r}')\psi^\dagger_g(\mathbf{r})\psi_e(\mathbf{r})\psi_{g'_1}(\mathbf{r}') + \frac{i}{\hbar\epsilon_0}\Big\{\psi^\dagger_{g_1}(\mathbf{r}')\psi^\dagger_g(\mathbf{r})\psi_{g'}(\mathbf{r})\psi_{g'_1}(\mathbf{r}')\mathbf{d}_{eg'}\cdot\mathbf{D}^+_F(\mathbf{r})\\ &+\psi^\dagger_{g_1}(\mathbf{r}')\psi^\dagger_g(\mathbf{r})\psi_{g'}(\mathbf{r})\psi_{e'}(\mathbf{r}')\mathbf{d}_{eg'}\cdot[\epsilon_0\mathbf{S}'(\mathbf{d}_{g'_1e'};\mathbf{r}-\mathbf{r}')-\mathbf{d}_{g'_1e'}\delta(\mathbf{r}-\mathbf{r}')]\\ &-\psi^\dagger_{g_1}(\mathbf{r}')\psi^\dagger_g(\mathbf{r})\mathbf{d}_{eg'}\cdot\mathbf{P}^+(\mathbf{r})\psi_{g'}(\mathbf{r})\psi_{g'_1}(\mathbf{r}')\\ &+\epsilon_0\psi^\dagger_{g_1}(\mathbf{r}')\psi^\dagger_g(\mathbf{r})\int d^3r''\mathbf{d}_{eg'}\cdot\mathbf{S}'(\mathbf{P}^+(\mathbf{r}'');\mathbf{r}''-\mathbf{r})\psi_{g'}(\mathbf{r})\psi_{g'_1}(\mathbf{r}')\Big\}.\end{aligned} \tag{34}$$

Continuing in this manner, we obtain the equations of motion for an entire hierarchy of products of atomic operators. As before, we put the positive-frequency free-field operators to the right, all atom operators to normal order, and then keep only the terms that are first order in the perturbation strength. The full result is

$$\begin{aligned}&\frac{d}{dt}\psi^\dagger_{g_n}(\mathbf{r}_n)\ldots\psi^\dagger_{g_1}(\mathbf{r}_1)\psi_e(\mathbf{r}_1)\psi_{g'_2}(\mathbf{r}_2)\ldots\psi_{g'_n}(\mathbf{r}_n)\\ &=(i\delta-\gamma)\psi^\dagger_{g_n}(\mathbf{r}_n)\ldots\psi^\dagger_{g_1}(\mathbf{r}_1)\psi_e(\mathbf{r}_1)\psi_{g'_2}(\mathbf{r}_2)\ldots\psi_{g'_n}(\mathbf{r}_n)\\ &+\frac{i}{\hbar\epsilon_0}\Big\{\psi^\dagger_{g_n}(\mathbf{r}_n)\ldots\psi^\dagger_{g_1}(\mathbf{r}_1)\psi_g(\mathbf{r}_1)\psi_{g'_2}(\mathbf{r}_2)\ldots\psi_{g'_n}(\mathbf{r}_n)\mathbf{d}_{eg}\cdot\mathbf{D}^+_F(\mathbf{r}_1)\\ &-\psi^\dagger_{g_n}(\mathbf{r}_n)\ldots\psi^\dagger_{g_1}(\mathbf{r}_1)\mathbf{d}_{eg}\cdot\mathbf{P}^+(\mathbf{r}_1)\psi_g(\mathbf{r}_1)\psi_{g'_2}(\mathbf{r}_2)\ldots\psi_{g'_n}(\mathbf{r}_n)\\ &+\epsilon_0\psi^\dagger_{g_n}(\mathbf{r}_n)\ldots\psi^\dagger_{g_1}(\mathbf{r}_1)\int d^3r'\,\mathbf{d}_{eg}\cdot\mathbf{S}'(\mathbf{P}^+(\mathbf{r}');\mathbf{r}_1-\mathbf{r}')\psi_g(\mathbf{r}_1)\psi_{g'_2}(\mathbf{r}_2)\ldots\psi_{g'_n}(\mathbf{r}_n)\\ &+\epsilon_0\psi^\dagger_{g_n}(\mathbf{r}_n)\ldots\psi^\dagger_{g_1}(\mathbf{r}_1)\psi_g(\mathbf{r}_1)\sum_{k=2}^{n}\psi_{g'_2}(\mathbf{r}_2)\ldots\psi_{g'_{k-1}}(\mathbf{r}_{k-1})\psi_{e'}(\mathbf{r}_k)\psi_{g'_{k+1}}(\mathbf{r}_{k+1})\ldots\psi_{g'_n}(\mathbf{r}_n)\mathbf{d}_{eg}\cdot\mathbf{W}(\mathbf{d}_{g'_ke'};\mathbf{r}_1-\mathbf{r}_k)\Big\},\end{aligned} \tag{35}$$

where the notation in the last term implies that $\psi_{g'_k}(\mathbf{r}_k)$ is missing from the k term of the sum. We have defined

$$\mathbf{W}(\boldsymbol{\mathcal{D}};\mathbf{r}) = \mathbf{S}'(\boldsymbol{\mathcal{D}};\mathbf{r}) - \frac{1}{\epsilon_0}\boldsymbol{\mathcal{D}}\delta(\mathbf{r}) = \mathbf{K}(\boldsymbol{\mathcal{D}};\mathbf{r}) - \frac{1}{3\epsilon_0}\boldsymbol{\mathcal{D}}\delta(\mathbf{r}). \tag{36}$$

This is precisely the classical expression of the electric field (not displacement) of a dipole $\boldsymbol{\mathcal{D}}$ residing at the origin, as measured at the point $\mathbf{r}$. Even the peculiar δ-function divergence of the dipolar field at the origin [14] is there.

D. Hierarchy for correlation functions

By taking expectation values of the operator hierarchy (35), we obtain a hierarchy of equations of motion for correlation functions. In order to simplify, in the rest of the paper we only consider a $J_g=0\rightarrow J_e=1$ transition. Then there are no Zeeman substates in the ground level, and a single g suffices in all of the Eqs. (35). The three excited Zeeman states are also handled easily; cf. Sec. 2 of the Appendix.

We define a succession of correlation functions

$$\begin{aligned}\mathbf{P}_1(;\mathbf{r}_1) &= \langle\psi^\dagger_g(\mathbf{r}_1)\mathbf{d}_{ge}\psi_e(\mathbf{r}_1)\rangle \equiv \langle\mathbf{P}^+(\mathbf{r}_1)\rangle,\\ \mathbf{P}_2(\mathbf{r}_1;\mathbf{r}_2) &= \langle\psi^\dagger_g(\mathbf{r}_1)\mathbf{P}^+(\mathbf{r}_2)\psi_g(\mathbf{r}_1)\rangle,\\ \mathbf{P}_3(\mathbf{r}_1,\mathbf{r}_2;\mathbf{r}_3) &= \langle\psi^\dagger_g(\mathbf{r}_1)\psi^\dagger_g(\mathbf{r}_2)\mathbf{P}^+(\mathbf{r}_3)\psi_g(\mathbf{r}_2)\psi_g(\mathbf{r}_1)\rangle,\\ &\ldots,\end{aligned} \tag{37}$$

and similarly

$$\begin{aligned}\rho_1(\mathbf{r}_1) &= \langle\psi^\dagger_g(\mathbf{r}_1)\psi_g(\mathbf{r}_1)\rangle,\\ \rho_2(\mathbf{r}_1,\mathbf{r}_2) &= \langle\psi^\dagger_g(\mathbf{r}_1)\psi^\dagger_g(\mathbf{r}_2)\psi_g(\mathbf{r}_2)\psi_g(\mathbf{r}_1)\rangle,\\ &\ldots.\end{aligned} \tag{38}$$

$\mathbf{P}_k(\mathbf{r}_1,\ldots,\mathbf{r}_{k-1};\mathbf{r}_k)$ is the correlation function of the polarization at $\mathbf{r}_k$ and the atom density at $k-1$ positions $\mathbf{r}_1,\ldots,\mathbf{r}_{k-1}$, and ρ_k is a k-point density correlation function. All of these are normally ordered.

We finally reiterate that the driving field is in a coherent state, so that the factorization (18) is warranted. In fact, with-

out further ado, we let $\mathbf{D}_F^+$ stand for the *expectation value* of the coherent free field, or, equally well, for a classical incident field. It is now a simple matter to derive a hierarchy of equations for the correlation functions from the operator equations (35). We consolidate the results into the form

$$\dot{\mathbf{P}}_1(;\mathbf{r}_1)=(i\delta-\gamma)\mathbf{P}_1(;\mathbf{r}_1)+i\kappa\rho_1(\mathbf{r}_1)\mathbf{D}_F^+(\mathbf{r}_1)+\int d^3r_2\mathsf{G}(\mathbf{r}_1-\mathbf{r}_2)\mathbf{P}_2(\mathbf{r}_1;\mathbf{r}_2), \tag{39a}$$

$$\dot{\mathbf{P}}_2(\mathbf{r}_1;\mathbf{r}_2)=(i\delta-\gamma)\mathbf{P}_2(\mathbf{r}_1;\mathbf{r}_2)+\mathsf{G}(\mathbf{r}_2-\mathbf{r}_1)\mathbf{P}_2(\mathbf{r}_2;\mathbf{r}_1)+i\kappa\rho_2(\mathbf{r}_1,\mathbf{r}_2)\mathbf{D}_F^+(\mathbf{r}_2)+\int d^3r_3\mathsf{G}(\mathbf{r}_2-\mathbf{r}_3)\mathbf{P}_3(\mathbf{r}_1,\mathbf{r}_2;\mathbf{r}_3), \tag{39b}$$

$$\dot{\mathbf{P}}_3(\mathbf{r}_1,\mathbf{r}_2;\mathbf{r}_3)=(i\delta-\gamma)\mathbf{P}_3(\mathbf{r}_1,\mathbf{r}_2;\mathbf{r}_3)+\mathsf{G}(\mathbf{r}_3-\mathbf{r}_1)\mathbf{P}_3(\mathbf{r}_3,\mathbf{r}_2;\mathbf{r}_1)+\mathsf{G}(\mathbf{r}_3-\mathbf{r}_2)\mathbf{P}_3(\mathbf{r}_1,\mathbf{r}_3;\mathbf{r}_2)+i\kappa\rho_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)\mathbf{D}_F^+(\mathbf{r}_3)+\int d^3r_4\mathsf{G}(\mathbf{r}_3-\mathbf{r}_4)\mathbf{P}_4(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3;\mathbf{r}_4),\quad\ldots\ . \tag{39c}$$

We have defined the scalar constant

$$\kappa=\frac{\mathcal{D}^2}{\hbar\epsilon_0}, \tag{40a}$$

and the 3×3 tensor

$$\mathsf{G}_{ij}(\mathbf{r})=i\kappa\left\{\left[\frac{\partial}{\partial r_i}\frac{\partial}{\partial r_j}-\delta_{ij}\boldsymbol{\nabla}^2\right]\frac{e^{ikr}}{4\pi r}-\delta_{ij}\delta(\mathbf{r})\right\}. \tag{40b}$$

The first two equations of the hierarchy (39) coincide exactly with the results of Morice, Castin, and Dalibard [9]. These authors do not proceed any further, but their method, which at this point had become tantamount to classical electrodynamics, undoubtedly could have yielded the entire hierarchy.

The terms in Eqs. (39) with $i\delta-\gamma$ obviously come from the damped free evolution of the polarization in each correlation function, and the term $\propto\mathbf{D}_F^+$ corresponds to excitation of a ground-state atom by the driving light to make polarization. To grasp the dual role of the tensor G, let us consider the equation of motion for $\mathbf{P}_2(\mathbf{r}_1;\mathbf{r}_2)$ as an example, the correlation function of polarization at $\mathbf{r}_2$ and density at $\mathbf{r}_1$. The integral term obviously characterizes processes in which a dipole at yet another position $\mathbf{r}_3$ radiates and thereby promotes a ground-state atom at $\mathbf{r}_2$, so that density becomes dipole density. On the other hand, the term with $\mathsf{G}(\mathbf{r}_2-\mathbf{r}_1)$ describes photon exchange between the two sites $\mathbf{r}_1$ and $\mathbf{r}_2$; an excited atom radiates at $\mathbf{r}_1$ and falls to the ground state, while the emitted radiation promotes an atom at $\mathbf{r}_2$ to the excited state.

We have implemented several approximations. The most relevant physical assumption is the perturbative limit with respect to the strength of the driving light, the most conspicuous technical assumption is the $J_g=0\rightarrow J_e=1$ transition. However, Eqs. (39) do not contain any assumptions concerning spontaneous emission except for our field-theory version of the Born and Markov approximations. Moreover, there are no assumptions, *ad hoc* or otherwise, concerning multiple scattering of light or resonant dipole-dipole interactions; these are included exactly. Hence so are collective linewidths and line shifts.

As far as the interactions of atoms with electromagnetic fields are concerned, we have regarded the atoms as point dipoles. For real atoms at short distances, when higher multipoles and electron exchange become relevant, this assumption evidently fails. The relevance of contact interactions and δ-function contributions to the dipolar field is questionable, as both operate at *zero* distance between the atoms only. Now, real atoms cannot overlap because of the hard core of the interatomic potential. A reader troubled by the δ functions may therefore want to consider cutting off and setting to zero all correlation functions at distances between the atoms shorter than the typical length scale of a molecular bond. The effect is that all δ-function contributions to the field propagator G of Eq. (40b) should be omitted. As the derivatives of e^{ikr}/r also produce δ functions, such an omission is tantamount to replacing the term $-\delta_{ij}\delta(\mathbf{r})$ by $-\frac{2}{3}\delta_{ij}\delta(\mathbf{r})$. However, in the present paper we use the propagator exactly as given in Eq. (40b).

III. EXAMPLES

In this section we illustrate the correlation function hierarchy with a few simple examples. At this time we have made little progress toward a full, exact solution of the hierarchy (39) in any nontrivial situation. Evidently, radically different approaches are needed. We hope that either ourselves or our readers will in the end be inspired to come up with a successful solution of, say, the optical response of a Bose-Einstein condensate (BEC) in the limit of truly dense sample, $\rho\lambda^3\gg1$.

A. Semi-infinite BEC without collective coupling

1. Nature of condensate

An ideal Bose condensate of noninteracting particles is made of a macroscopic number of particles in the same one-particle quantum state. Traditionally, the condensate is described by a macroscopic wave function, whose absolute square gives the particle density, and which also has a phase. More in the vein of quantum optics, one could assume that the condensate is in a coherent state, albeit with an unknown phase. In normally ordered operator expressions the field operator ψ_g then behaves as a c number. We write

$$\rho_k(\mathbf{r}_1,\ldots,\mathbf{r}_k)=\langle\psi_g^\dagger(\mathbf{r}_1)\ldots\psi_g(\mathbf{r}_1)\rangle \tag{41}$$

$$\simeq\psi_g^*(\mathbf{r}_1)\ldots\psi_g^*(\mathbf{r}_k)\psi_g(\mathbf{r}_k)\ldots\psi_g(\mathbf{r}_1)$$

$$=\psi_g^*(\mathbf{r}_1)\psi_g(\mathbf{r}_1)\ldots\psi_g^*(\mathbf{r}_k)\psi_g(\mathbf{r}_k)$$

$$=\langle\psi_g^\dagger(\mathbf{r}_1)\psi_g(\mathbf{r}_1)\rangle\ldots\langle\psi_g^\dagger(\mathbf{r}_k)\psi_g(\mathbf{r}_k)\rangle$$

$$=\rho_1(\mathbf{r}_1)\ldots\rho_1(\mathbf{r}_k). \tag{42}$$

In other words, density correlation functions factorize. For the purposes of the present paper, we take the factorization of normally ordered density correlation functions as the hallmark of the condensate even for a weakly interacting condensate. We ignore noncondensate atoms altogether.

Contrary to the experimental realities, we ignore the finite dimensions of the condensate. We cannot outright declare $\rho_1(\mathbf{r})$ as a constant all over space, because propagation through an infinite medium would cause extinction of all light before it reaches any position with finite $|\mathbf{r}|$. Instead, we assume that a homogeneous condensate with density ρ fills the half-space $z \geqslant 0$. We assume that the incident and induced radiations as well as the induced polarizations all propagate in the z direction. Finally, as we have the $m_g = 0$ spherically symmetric ground state that cannot exhibit any directional preferences, we take all fields to have the same transverse polarization $\hat{\mathbf{e}}$. In particular, the initial free field is written

$$\mathbf{D}_F^+(\mathbf{r}) = D_F\, \hat{\mathbf{e}}\, e^{ikz}. \tag{43}$$

This has the dispersion relation of light in vacuum, an oddity in the presence of matter. In fact, in accordance with the Ewald-Oseen extinction theorem [21], it will turn out that the matter responds with a field that exactly cancels the applied field.

2. Optical response without collective coupling

We solve the response by ignoring the collective line shifts and dampings, i.e., those G terms in Eqs. (39) that do not appear inside integrals. It turns out that the simplest conceivable ansatz, a fully factorized, damped plane-wave solution of the form

$$\mathbf{P}_n(\mathbf{r}_1, \ldots; \mathbf{r}_n) = \begin{cases} P\rho^{n-1}\hat{\mathbf{e}}\, e^{ik' z_n}, & z_1 \geqslant 0, \ldots, z_n \geqslant 0 \\ 0 \quad \text{otherwise} \end{cases} \tag{44}$$

succeeds for suitable choices of P and k', with $\mathrm{Im}(k') > 0$.

To see this, we need the integral

$$\int_{z_2 \geqslant 0} d^3 r_2\, \hat{\mathbf{e}}^* \cdot \mathsf{G}(\mathbf{r}_1 - \mathbf{r}_2) \cdot \hat{\mathbf{e}}\, e^{ik' z_2} = i\kappa \left[\frac{k^2}{k'^2 - k^2} e^{ik' z_1} + \frac{k'^2}{2k(k-k')} e^{ikz_1} \right], \tag{45}$$

where the vector $\hat{\mathbf{e}}$ takes care of the polarizations. The integral is valid for $z_1 > 0$. For $z_1 < 0$ the integral yields a reflected wave $\propto e^{-ikz}$ instead, but we do not consider this case any further. With the ansatz (44), the integrals on the right hand sides of Eqs. (39) produce sums of two exponentials, one with the wave number k appropriate for light in vacuum and the other with the wave number k' for light in the medium. In steady state of Eqs. (39) the vacuum component $\propto e^{ikz}$ must cancel the corresponding free-field terms, and the remaining $e^{ik'z}$ term must pair up with the polarization correlation functions $\mathbf{P}_n$. All Eqs. (39) then reduce to the following two conditions:

$$\left(i\delta - \gamma + \frac{i\rho\kappa k^2}{k'^2 - k^2} \right) P = 0, \tag{46a}$$

$$D_F + \frac{k'^2}{2k(k-k')} P = 0\,. \tag{46b}$$

The first one gives the wave number k', and the second one may then be read as a condition for the polarization amplitude P.

One expects that the total displacement from Eq. (4) should be of the form $D\hat{\mathbf{e}}\, e^{ik'z}$ as well. With the choice (46b), the vacuum type contributions e^{ikz} indeed cancel. We have the condition for the polarization amplitude and the amplitude of electric displacement,

$$D = \frac{k'^2}{k'^2 - k^2} P. \tag{47}$$

This implies that an electric field of the form $E\hat{\mathbf{e}}\, e^{ik'z}$ also propagates in the medium, with the amplitude given by

$$E = \frac{D - P}{\epsilon_0} = \frac{1}{\epsilon_0} \frac{k^2}{k'^2 - k^2} P. \tag{48}$$

As is customary, we define the refractive index n in such a way that $k' = nk$, and susceptibility χ such that $P = \epsilon_0 \chi E$. Equations (46)–(48) immediately give the explicit expressions

$$n^2 - 1 = \chi = -\frac{\rho\kappa}{\delta + i\gamma} \tag{49}$$

for these quantities.

The remarkable feature of the result (49) is that it is so unremarkable: susceptibility is obtained as atomic polarizability times atom density. This is precisely the conventional column density approach that the experimenters routinely use to analyze their BEC results. In other words, we have proven that, for a plausible model of the BEC and within a precisely formulated approximation that ignores collective linewidths and line shifts, the column density approach is *exact*. We regard this as an important result, in that it displays precisely and explicitly the underlying assumptions of the column density arguments. The flip side is that, at high density, simply ignoring the collective effects is another uncontrolled approximation.

B. Density expansion to second order

In the present section we review the density expansion of Morice, Castin, and Dalibard [9] from the point of view of our development. In effect, they truncate the hierarchy for correlation functions by writing

$$\mathbf{P}_3(\mathbf{r}_1, \mathbf{r}_2; \mathbf{r}_3) \simeq \frac{\rho_2(\mathbf{r}_1, \mathbf{r}_2)}{\rho(\mathbf{r}_2)} \mathbf{P}_2(\mathbf{r}_2; \mathbf{r}_3). \tag{50}$$

If (and probably in some fairly strong sense only if) one inserts this particular factorization into Eq. (39b), one may at the same time eliminate both the free-field term and the integral term from the steady-state versions of Eqs. (39a) and (39b). This gives an algebraic relation between $\mathbf{P}_1$ and $\mathbf{P}_2$,

$$0=\mathbf{P}_2(\mathbf{r}_1;\mathbf{r}_2)-\frac{\mathsf{G}(\mathbf{r}_2-\mathbf{r}_1)}{(i\delta-\gamma)}\mathbf{P}_2(\mathbf{r}_2;\mathbf{r}_1)+\frac{\rho_2(\mathbf{r}_1,\mathbf{r}_2)}{\rho_1(\mathbf{r}_2)}\mathbf{P}_1(;\mathbf{r}_2). \tag{51}$$

Now assume a constant density of atoms ρ in the half-space $z\geqslant 0$, and write the physically justifiable ansatz

$$\rho_2(\mathbf{r}_1,\mathbf{r}_2)=\rho^2[1+\varphi(\mathbf{r}_1-\mathbf{r}_2)]. \tag{52}$$

Relation (51) and its counterpart with $\mathbf{r}_1$ and $\mathbf{r}_2$ interchanged may then be solved to give

$$\begin{aligned}\mathbf{P}_2(\mathbf{r}_1;\mathbf{r}_2)&=\frac{\rho[1+\varphi(\mathbf{r}_1-\mathbf{r}_2)]}{1-[\mathsf{G}(\mathbf{r}_1-\mathbf{r}_2)/(i\delta-\gamma)]^2}\\&\quad\times\left[\mathbf{P}_1(;\mathbf{r}_2)-\frac{\mathsf{G}(\mathbf{r}_1-\mathbf{r}_2)}{(i\delta-\gamma)}\mathbf{P}_1(;\mathbf{r}_1)\right]\\&=\rho\mathbf{P}_1(;\mathbf{r}_2)+\frac{\rho}{1-g^2}[(\varphi+g^2)\mathbf{P}_1(;\mathbf{r}_2)\\&\quad-g\mathbf{P}_1(;\mathbf{r}_1)],\end{aligned} \tag{53}$$

where we have introduced an obvious temporary notation. The reason for the split in the second form of Eq. (53) is the following. Suppose we attempt an ansatz of the form (43), together with $\mathbf{P}_1(;\mathbf{r})=P\hat{\mathbf{e}}\,e^{ik'z}$. Then, when Eq. (53) is inserted into the integral in the right-hand side of Eq. (39a), in accordance with Eq. (45) the term $\rho\mathbf{P}_1$ will produce a term with the spatial dependence of the free field e^{ikz}. All other contributions to the integral will behave as $e^{ik'z}$, at least for $z\to\infty$. Now the requirement that the free-field contributions cancel (the Ewald-Oseen extinction theorem [21] again) gives one relation between D_F, P, and k', and Eq. (13) furnishes another. One then obtains an equation out of which one may solve k', and therefore ultimately the entire response of (a thick slab of) the gas. This equation reads

$$\frac{k'^2}{k^2}=1-\frac{\kappa\rho}{\delta+i\gamma}\frac{1}{1+C}, \tag{54a}$$

with

$$C=\frac{\rho}{i\delta-\gamma}\int d^3r\,\varphi(\mathbf{r})e^{-ik'z}\,\hat{\mathbf{e}}^*\cdot\mathsf{G}(\mathbf{r})\cdot\hat{\mathbf{e}}+\rho\int d^3r[1+\varphi(\mathbf{r})]\hat{\mathbf{e}}^*\cdot\left[\frac{e^{-ik'z}[\mathsf{G}(\mathbf{r})/(i\delta-\gamma)]^3-[\mathsf{G}(\mathbf{r})/(i\delta-\gamma)]^2}{1-[\mathsf{G}(\mathbf{r})/(i\delta-\gamma)]^2}\right]\cdot\hat{\mathbf{e}}. \tag{54b}$$

Ingenuous as the analysis of Ref. [9] is, it elicits two questions. First, the factorization (50) is the way it is to facilitate the mathematics, not because Eq. (50) would be a particularly apt physics assumption. There is no guarantee that correlations between the ground-state atoms are treated adequately. Second, while Morice, Castin, and Dalibard argued that the result is the correct expansion in the parameter $\rho\lambda^3$ up to the order $(\rho\lambda^3)^2$ and, in fact, includes all multiple-scattering events between any *pair* of atoms, the mathematical structure of the hierarchy (39) does not directly bear this out. The difficulty is that each integral of the form $\int d^3r_k\mathsf{G}\mathbf{P}_k$ must produce a component that cancels the associated free-field term. In other words, the integral does not automatically signal an increasing power in $\rho\lambda^3$. Within the present approach, a rigorous mathematical counting of the powers of $\rho\lambda^3$ seems elusive.

C. Resonant dipole-dipole interaction of two atoms

Let us take two atoms with small nonoverlapping c.m. wave packets $\phi_\pm(\mathbf{r})$ centered around $\mathbf{r}_\pm$. Inasmuch as the sizes of the wave packets are much smaller than the wavelength of the exciting light, for the purposes of the analysis of optical response we may write the atom density and the density correlation function as

$$\begin{aligned}\rho_1(\mathbf{r}_1)&=|\phi_+(\mathbf{r}_1)|^2+|\phi_-(\mathbf{r}_1)|^2\simeq\delta(\mathbf{r}_1-\mathbf{r}_+)+\delta(\mathbf{r}_1-\mathbf{r}_-),\\\rho_2(\mathbf{r}_1,\mathbf{r}_2)&=|\phi_+(\mathbf{r}_1)|^2|\phi_-(\mathbf{r}_2)|^2+|\phi_+(\mathbf{r}_2)|^2|\phi_-(\mathbf{r}_1)|^2\\&\simeq\delta(\mathbf{r}_1-\mathbf{r}_+)\delta(\mathbf{r}_2-\mathbf{r}_-)+\delta(\mathbf{r}_2-\mathbf{r}_+)\delta(\mathbf{r}_1-\mathbf{r}_-).\end{aligned} \tag{55}$$

To simplify the results further, we assume that the geometry of the situation is such that for the driving light at the positions of the atoms we have $\mathbf{D}_F^+(\mathbf{r}_+)=\mathbf{D}_F^+(\mathbf{r}_-)\equiv\mathbf{D}_F^+$. In particular, this is justified if the atoms are well within a wavelength of one another. In such a case the two-atom system may be discussed in terms of Dicke states, some superradiant and some subradiant. The field configuration we have chosen does not excite the subradiant states at all, so these will not come up in our analysis.

Since we have two atoms present, all correlation functions referring to more than two atoms vanish: $\rho_n=0$ and $\mathbf{P}_n=0$ for $n=3,4,\ldots$. The steady state of the hierarchy (39) is then found trivially:

$$\mathbf{P}_1(;\mathbf{r}_1)=\mathbf{p}[\,\delta(\mathbf{r}_1-\mathbf{r}_+)+\delta(\mathbf{r}_1-\mathbf{r}_-)], \tag{56}$$

with

$$\mathbf{p}=-\frac{i\kappa}{(i\delta-\gamma)+\mathsf{G}}\mathbf{D}_F^+. \tag{57}$$

Unraveling Eq. (57) (with the 3×3 tensor G in the denominator) gives a complicated expression that depends on the polarization of the driving light $\mathbf{D}_F^+$, the direction between the atoms, and the distance between the atoms, r. The main features, however, are intuitively obvious. For $r\ll\lambda$ the optical response shows a split resonance at

$$\delta=\frac{\kappa}{4\pi r^3},\quad \delta=-\frac{\kappa}{2\pi r^3}. \tag{58}$$

The configurations that produce only one or the other of these resonance are such that the polarization of the driving field (and, hence, of the induced dipoles) is perpendicular or parallel to the vector joining the dipoles, respectively. These correspond to the doubly degenerate π_u and the nondegenerate σ_u configurations of the molecule consisting of the two atoms. The cooperative width of the resonances is 2γ instead of the one-atom linewidth γ. Incidentally, the subradiant states that are invisible within our approximations correspond to the molecular configurations π_g and σ_g, which appropriately do not have dipole coupling to the molecular ground state.

Because our theory is linear in the external field, it should not come as a surprise that exactly the same dipole-dipole response ensues for two classical, isotropic, charged harmonic oscillators. In such a calculation one may want to put the one-oscillator damping γ in by hand in order to dispense with an explicit treatment of the divergent radiation reaction. Nonetheless, the collective linewidth and dipole-dipole interactions are easily derived from the classical analysis of the radiation that the oscillators exchange among themselves. On the other hand, the molecular analogy also hints at some aspects of physics that are missing from our formulation. First, once more, at close enough distances two atoms may no longer be regarded as point dipoles. Second, since we have ignored all c.m. evolution, the possibility of quantized vibrational states of the molecules and the ensuing optical resonances have fallen by the wayside.

As the connection of our hierarchy to dipole-dipole interactions has now come up explicitly, we should point out that there are numerous discussions of two- and n-atom responses in the literature in which (in effect) the dipole-dipole interaction is derived (in effect) by eliminating vacuum electromagnetic fields with the aid of Born and Markov approximations; see, e.g., [16]. As it comes to dense and/or degenerate gases, it seems that dipole-dipole interactions are going to occupy an increasingly prominent position in theory and eventually perhaps in experiments as well. ''Nonlinear atom optics'' [22–25], in which the self-interaction of the atom wave derives from dipole-dipole interactions, is a prominent example. In fact, the analysis of Zhang and Walls [23] in terms of Heisenberg picture field operators is in spirit quite close to our treatment, and it seems plausible that our correlation function hierarchy could also be derived from the Schrödinger picture master equation as presented by Lenz, Meystre, and Wright [24].

D. Prospects of exact solution

Dipole radiation presents mathematical difficulties both at short and long distances. One has $1/r^3$, and indeed a δ-function divergence at short distances, which means that the results are sensitive to short-range correlations between the atoms. Also, the dipole interaction falls off as $1/r$ at large distances. Integrals involving the dipole interaction are not absolutely convergent on the falloff of the interaction alone, and local approximations of the type

$$\int d^3r' \mathsf{G}(\mathbf{r}-\mathbf{r}')f(\mathbf{r}') \simeq f(\mathbf{r}) \int d^3r' \mathsf{G}(\mathbf{r}-\mathbf{r}')$$

cannot be made. There may be a global coupling between the electromagnetic fields and the polarization reaching across the entire sample. In the face of such mathematical hazards, any uncontrolled approximation should be viewed with suspicion. An essentially exact, most likely numerical, solution of the hierarchy (39) appears highly desirable.

The idea of attempting a direct numerical solution of the hierarchy of integral equations (39) in any conceivable future is clearly stillborn. Instead, on several occasions we have seen hints of an alternative. We noted in our treatment of two atoms that in the limit of low light intensity the atoms act like classical charged harmonic oscillators. Moreover, even though the derivation was in terms of commutator properties of various fields, both the shift and the damping of the atoms associated with the dipole-dipole interaction could be viewed simply as manifestations of the classical radiation transmitted from one atom to the other. It is conceivable that a proper (stochastic) spatial distribution of classical radiators could share the correlation function hierarchy (39). The hierarchy might thus be solved by simulating a system of classical atoms and classical electromagnetic fields numerically. However, so far we have no mathematically rigorous prescription for such a simulation, let alone a practical implementation.

IV. CONCLUDING REMARKS

We have presented a fully quantum-mechanical, careful analysis of the response of a gas, possibly degenerate, to electromagnetic fields. The main technical ingredients are the field-theory version of Born and Markov approximations, and procedures to move atom fields and electromagnetic fields to a certain (basically normal) order. The outcome is a hierarchy of equations of motion for atomic correlation functions, specifically developed in this paper for the limit of low light intensity.

Under our assumptions, notably low intensity, the hierarchy is in a sense obvious. In retrospect, it could have been outright guessed on the basis of classical electrodynamics. We note, though, that our methods would work in many generalizations that go beyond classical physics. Given that even the simplest low-intensity limit has not yet been solved satisfactorily for the near-resonance response of a dense gas, we do not address more complicated cases at any length. Nonetheless, a few possible generalizations should be mentioned.

The price of an arbitrary intensity would be a more complicated hierarchy containing atomic correlation functions with more than one excited-state field ψ_e. In the case of an arbitrary intensity, a two-level atom no longer behaves identically to a charged harmonic oscillator, so that scattered light may be nonclassical, and statistics of the atoms may play a nontrivial role. The hierarchy for an arbitrary intensity most likely will not admit a classical simulation. Solutions for a finite number of atoms and density expansions might be extracted, but the full hierarchy would present a truly daunting problem.

While we have treated the c.m. degrees of freedom quantum mechanically, and at all times properly retained the quantum statistics of the atoms, for the most part we have ignored the c.m. Hamiltonian. We have effectively consigned the atoms to immobility. This is by no means necessary. We could add the kinetic energy of the atoms, a con-

fining potential, and even molecular potential curves to the theory. Of course, this again entails complications: the entire physics of molecules made of a single atomic species, photon recoil, cold collisions, etc., become special cases of our approach.

The ultimate objective of our formulation is to find the expectation value of the polarization, $\langle \mathbf{P}^+ \rangle$. Out of $\langle \mathbf{P}^+ \rangle$ one may deduce the expectation value of the scattered field, and hence, the expectation value of the total electric field $\langle \mathbf{E}^+ \rangle$. The flaw here is that a typical detector of light does not measure $\langle \mathbf{E}^+ \rangle$, but rather expectation values of quadratic quantities such as $\langle \mathbf{E}^- \mathbf{E}^+ \rangle$. One may approximate, say, the measured intensity as

$$I(\mathbf{r}t) = \langle \mathbf{E}^-(\mathbf{r}t)\mathbf{E}^+(\mathbf{r}t) \rangle \simeq \langle \mathbf{E}^-(\mathbf{r}t) \rangle \langle \mathbf{E}^+(\mathbf{r}t) \rangle, \quad (59)$$

but it is known in quantum optics that this type of an approximation broadly speaking misses the intensity of *inelastically* scattered light. Our hierarchy is tantamount to a collection of classical linear harmonic oscillators interacting with light, a system in which one expects elastic scattering only. However, when one goes beyond the low-intensity limit, even a single two-level atom scatters inelastically; and if the c.m. Hamiltonian is fully included, photon recoil gives additional inelastic scattering. To include inelastic scattering properly, one needs to develop a hierarchy starting from $\langle \mathbf{P}^-(\mathbf{r}t)\mathbf{P}^+(\mathbf{r}'t) \rangle$, and proceed consistently at least in the *second* order in the strength of the driving electric field.

Finally, to calculate the spectrum of scattered radiation, one employs the two-time correlation function of the electric field $\langle \mathbf{E}^-(\mathbf{r}t)\mathbf{E}^+(\mathbf{r}t') \rangle$, which is obtainable from the two-time polarization correlation function $\langle \mathbf{P}^-(\mathbf{r}t)\mathbf{P}^+(\mathbf{r}'t') \rangle$. To compute the latter, one needs not only the field-theory version of Born and Markov approximations, but also further considerations that essentially amount to the regression theorem [26].

The modifications of the spectrum of the scattered light in the presence of the condensate discussed in Ref. [10] are due to inelastic scattering associated with photon recoil. Ironically, to analyze even this seemingly simple case within our present framework, we would have to generalize so that the c.m. motion is included, and also develop a quantum regression theorem. Our linear hierarchy is no panacea; major generalizations are needed in many relevant problems. Nonetheless, we hope that the eventual solutions of the hierarchy, by means of classical simulations or otherwise, will shed light on the near-resonance optical response of dense atomic samples.

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APPENDIX: MATHEMATICAL DETAILS

1. Dipole radiation with wave-number cutoff

We first consider the following integral involving the propagator that governs dipole radiation from matter fields:

$$I(\rho) = \int_0^\infty dt \int_{r<\rho} d^3r\, \mathbf{S}(\boldsymbol{\mathcal{D}};\mathbf{r},t). \quad (A1)$$

Here the $\mathbf{r}$ integral runs over a sphere of radius ρ. Given the finite-width δ-function (6), we have

$$I(\rho) = -\frac{1}{4\pi\epsilon_0} \int_{r<\rho} d^3r\, \boldsymbol{\nabla} \times (\boldsymbol{\mathcal{D}} \times \boldsymbol{\nabla}) \frac{\mathrm{erf}(r/\alpha)}{r}$$
$$= -\frac{1}{4\pi\epsilon_0} \int_S dS\, \hat{\mathbf{n}} \times \left[\boldsymbol{\mathcal{D}} \times \boldsymbol{\nabla} \frac{\mathrm{erf}(r/\alpha)}{r} \right], \quad (A2)$$

where the integral now runs over the surface S of the sphere.

Two obvious limiting cases emerge depending on the radius of the sphere. For $\rho \gg \alpha$ the error function inside the integral may be regarded as a constant equal to 1, and the integral gives

$$I(\rho) = \frac{2}{3\epsilon_0} \boldsymbol{\mathcal{D}}, \quad \rho \gg \alpha. \quad (A3)$$

In the contrary case $\rho \ll \alpha$ we may expand the error function as a power series in r. This immediately yields

$$\lim_{\rho \to 0} I(\rho) = 0\,. \quad (A4)$$

We have, in effect, shown that there is something akin to a δ-function, albeit with a finite width $\sim \alpha$, in the immediate proximity of the origin $\mathbf{r} = 0$ in the expression $\int dt\, \mathbf{S}$.

Next consider an integral of the form

$$I = \int_0^\infty dt' \int d^3r'\, \mathbf{S}(\boldsymbol{\mathcal{D}}, \mathbf{r} - \mathbf{r}', t - t')\, \phi(\mathbf{r}'t') e^{-i\Omega(t-t')}, \quad (A5)$$

where the characteristic spatial and temporal scales of the function ϕ satisfy

$$\Delta r \gg \alpha, \quad \Delta t \gg \alpha/c, \quad \Delta t \gg \Omega^{-1}. \quad (A6)$$

Moreover, we assume that the cutoff of electromagnetic frequencies is much higher than Ω, $c/\alpha \gg \Omega$. With these conditions, it is possible to choose a length ρ such that simultaneously $\alpha \ll \rho \ll \Delta r$ and $\rho \ll c\Omega^{-1} = \lambda/2\pi$. We divide the spatial integral into two regions: a sphere of radius ρ, and the complement of the sphere. In the outer region the function δ_α acts effectively as a δ function, so we have

$$I_{r \geq \rho} = \frac{1}{4\pi\epsilon_0} \int_{r \geq \rho} d^3r'\, \boldsymbol{\nabla} \times (\boldsymbol{\mathcal{D}} \times \boldsymbol{\nabla})$$
$$\times \left[\frac{\phi(\mathbf{r}', t - |\mathbf{r} - \mathbf{r}'|/c) e^{ik|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r} - \mathbf{r}'|} \right]$$
$$\simeq \int_{r \geq \rho} d^3r'\, \mathbf{K}(\boldsymbol{\mathcal{D}}; \mathbf{r} - \mathbf{r}')\, \phi(\mathbf{r}', t - |\mathbf{r} - \mathbf{r}'|/c), \quad (A7)$$

where $\mathbf{K}$ is the standard dipole radiation formula (10). On the other hand, in analogy with the previous expression (A3), the integral over the inner region gives

$$I_{r<\rho}=\frac{2}{3\epsilon_0}\mathcal{D}\phi(\mathbf{r}t). \tag{A8}$$

Summarizing, when the result is used in conjunction with a smooth function of $\mathbf{r}$ and t that also varies slowly in comparison with $e^{-i\Omega t}$, we may write

$$\int_0^\infty dt\,\mathbf{S}(\mathcal{D};\mathbf{r},t)e^{-i\Omega t}=\frac{1}{4\pi\epsilon_0}((\mathcal{D}\times\nabla)\times\nabla)\frac{e^{ikr}}{r}$$
$$=\frac{1}{4\pi\epsilon_0}\left[\mathbf{K}(\mathcal{D},\mathbf{r})+\frac{8\pi}{3}\mathcal{D}\delta(\mathbf{r})\right]. \tag{A9}$$

In addition, this prescription comes with explicit directions about how to handle integrals involving the dipole radiation. Because of the $1/r^3$ divergence of the dipole radiation $\mathbf{K}$, such integrals are generally not absolutely convergent, and their values depend on how they are performed. It is clear from our development that the proper way to carry out such integrals is to remove a sphere of a finite radius ρ around the divergence, calculate the integral, and then let $\rho\to 0$. In practice, this is the same as performing the integral in spherical coordinates with the origin at the divergence, and integrating over the angles first. This is the prescription adopted in the present paper.

One may wonder where precisely our rule for handling the $1/r^3$ singularity came from, and whether there are plausible alternatives. Mathematically, our rule originates from the assumption that the photon modes were truncated in a manner that preserves the isotropy of photon phase space. We surmise, albeit without proof, that our prescription, indeed, is essentially unique if there is to be no intrinsically favored directions for photons.

2. Vectors and matrix elements

Alongside with the Cartesian unit vectors, we introduce the conventional circular unit vectors as

$$\hat{\mathbf{e}}_+=-\frac{1}{\sqrt{2}}(\hat{\mathbf{e}}_1+\hat{\mathbf{e}}_2),\quad \hat{\mathbf{e}}_0=\hat{\mathbf{e}}_3,\quad \hat{\mathbf{e}}_-=\frac{1}{\sqrt{2}}(\hat{\mathbf{e}}_1-i\hat{\mathbf{e}}_2), \tag{A10}$$

These are orthonormal, in that

$$\hat{\mathbf{e}}_\sigma^*\cdot\hat{\mathbf{e}}_{\sigma'}=\delta_{\sigma\sigma'},\quad \sum_\sigma \hat{\mathbf{e}}_\sigma\hat{\mathbf{e}}_\sigma^*=1\,. \tag{A11}$$

We also define a shorthand for the Clebsch-Gordan coefficients,

$$\langle J_e M;1J_g|1\sigma J_g m\rangle\equiv\langle M|m\sigma\rangle\equiv\langle m\sigma|M\rangle. \tag{A12}$$

The dipole operator is defined as

$$\mathbf{d}=\mathcal{D}\sum_{mM\sigma}|J_eM\rangle\langle M|m\sigma\rangle\langle J_gm|\hat{\mathbf{e}}_\sigma^*+\text{H.c.}, \tag{A13}$$

where $\mathcal{D}$ is the reduced dipole moment matrix element that would pertain to a transition with unit Clebsch-Gordan coefficient. In this paper we have chosen $\mathcal{D}$ to be real. The reason for the complex conjugate in Eq. (A13) is that we want a light field with the polarization $\hat{\mathbf{e}}_+$ to drive transitions with $M-m=1$. The dipole matrix elements are explicitly

$$\mathbf{d}_{Mm}=\mathcal{D}\sum_\sigma\langle M|m\sigma\rangle\hat{\mathbf{e}}_\sigma^*,\quad \mathbf{d}_{mM}=\mathbf{d}_{Mm}^*. \tag{A14}$$

In particular, it may be verified from the orthonormality of the Clebsch-Gordan coefficients that these matrix elements satisfy

$$\sum_m \mathbf{d}_{Mm}\cdot\mathbf{d}_{mM'}=\mathcal{D}^2\delta_{MM'}. \tag{A15}$$

In the special case $J_g=0\to J_e=1$, we have

$$\langle -1|0-1\rangle=\langle 0|00\rangle=\langle +1|0+1\rangle=1\,. \tag{A16}$$

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