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Maxwell-Bloch equations: A unified view of nonlinear optics and nonlinear atom optics

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The linear and nonlinear properties of propagation of an electromagnetic field through a medium may be derived by the effective elimination of the medium from the coupled Maxwell-Bloch equations. In this paper we suggest, on the other hand, the elimination of the field from the Maxwell-Bloch equations as a means to derive a nonlinear master equation for, e.g., an atomic sample in a laser field. This presents a straightforward and physically intuitive derivation of equations for "nonlinear atom optics," obtained recently by other approaches.

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By laser cooling it is possible to produce atomic samples where the quantum nature of the center-of-mass motion becomes significant. At high densities collective effects are expected to be important, and recently a new domain of theoretical studies has emerged: nonlinear atom optics. A conventional starting point for the analysis of this situation is the construction of an equation for a system consisting of a number N of identical atoms. This equation may, by a Hartree-type approximation, be reduced to an effective nonlinear one-atom master equation or to a nonlinear Schrödinger equation in case of negligible dissipation [1-4].

The purpose of this Rapid Communication is to show that these nonlinear equations may be obtained easily from the well-known Maxwell-Bloch equations. The Maxwell-Bloch equations for the combined system of an electromagnetic field and a gas of polarizable atoms are often used to analyze the propagation of light through matter. By an elimination of the atomic degrees of freedom one gets an effective equation for the field only, involving the linear and nonlinear susceptibilities of the medium. As we are interested in the quantized motion of atoms in a laser field, e.g., in laser cooling and atom interferometry, we shall take the opposite viewpoint in this Rapid Communication and eliminate the field in the Maxwell-Bloch equations. Damping and dispersion of the field as it propagates through the medium depend on the spatial distribution of atomic dipoles, and hence the effective atomic master equation becomes nonlinear.

We start with the mean electric field $\vec{E}(\vec{r},t)$ in the medium, defined as the mean value of the total electric field operator, including both the transverse and the longitudinal components of the field. In the absence of free charges, free currents, and magnetization, the density of charge and current in the medium can be expressed in terms of the polarization density $\vec{P}(\vec{r},t)$. After elimination of the mean magnetic field, the Maxwell equations lead to the equation of propagation for the mean electric field:

$$\left(\Delta - \frac{1}{c^2} \partial_t^2\right) \vec{E}(\vec{r}, t) = \frac{1}{\varepsilon_0} \left(-\overrightarrow{\text{grad}} \operatorname{div} + \frac{1}{c^2} \partial_t^2\right) \vec{P}(\vec{r}, t),$$
(1)

where the polarization, appearing in Eq. (1), is calculated from the atomic density matrix, describing both internal and external states of the atom:

$$\vec{P}(\vec{r},t) = N \operatorname{Tr}[\rho(t)\,\delta(\vec{r}-\vec{R})\vec{D}] = N \operatorname{Tr}_{\operatorname{int}}[\langle \vec{r} | \rho(t) | \vec{r} \rangle \vec{D}].$$
(2)

 \vec{R} is the center-of-mass operator, \vec{D} is the dipole operator for the atom, and N denotes the total number of atoms. In the last term of Eq. (2) the trace over the external variables has been performed.

We approximate the atomic density matrix ρ by the solution to the usual master equation for an atom coupled, in the electric dipole approximation, to the classical electric field E(r,t), and to the quantized electromagnetic field in the vacuum state giving rise to energy shifts and radiative damping. Incorporating the effects of the atomic kinetic and internal energies and of spontaneous emission in the Liouville operator $\mathscr{L}[\rho]$, this equation can be written

$$\frac{d}{dt}\rho(t) = \mathscr{L}[\rho(t)] + \frac{1}{i\hbar} [-\vec{D} \cdot \vec{E}(\vec{R},t),\rho(t)].$$
(3)

Equations (1)-(3) constitute the coupled Maxwell-Bloch equations.

Truly, the electric field experienced by an atom contains a component due to incoherent scattering from the other atoms in the medium. This component has a vanishing mean value and does not contribute to $\vec{E}(\vec{r},t)$. The approximation made in order to obtain the Maxwell-Bloch equations is therefore to disregard this incoherent component of the electric field.

If we consider the case of an incident monochromatic field $\vec{E}_L(\vec{r},t)$ with a frequency ω_L , and we assume a steady state for the atomic dipole at this frequency, we may solve Eq. (1) by simply adding the field radiated by the individual dipoles. This leads to the simple relation between the positive frequency components of the total electric field, the incident field, and the polarization of the medium,

$$\vec{\mathscr{E}}(\vec{r}) = \vec{\mathscr{E}}_L(\vec{r}) + \frac{1}{\varepsilon_0} \int d^3 \vec{r}' [g(\vec{r} - \vec{r}')] \vec{\mathscr{P}}(\vec{r}'), \qquad (4)$$

with the expression for the 3×3 matrix $[g(\vec{r})]$ $(\alpha,\beta=x,y,z)$:

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$$g_{\alpha\beta}(\vec{r}) = -\int \frac{d^{3}\vec{k}}{(2\pi)^{3}} \sum_{\vec{e}\perp\vec{k}} ck\varepsilon_{\alpha}\varepsilon_{\beta}e^{i\vec{k}\cdot\vec{r}}\frac{1}{2} \left(\frac{1}{\omega_{L}-ck+i0^{+}} - \frac{1}{\omega_{L}+ck+i0^{+}}\right) - \delta_{\alpha\beta}\delta(\vec{r})$$
$$= -\frac{k_{L}^{3}}{4\pi} \frac{e^{ik_{L}r}}{k_{L}r} \left[\left(1 + \frac{3i}{k_{L}r} - \frac{3}{k_{L}^{2}r^{2}}\right)\frac{r_{\alpha}r_{\beta}}{r^{2}} - \left(1 + \frac{i}{k_{L}r} - \frac{1}{k_{L}^{2}r^{2}}\right)\delta_{\alpha\beta} \right] - \frac{1}{3}\delta_{\alpha\beta}\delta(\vec{r}).$$
(5)

 $g_{\alpha\beta}(\vec{r})$ is a Green's function for the positive frequency part of Eq. (1); the component with a common factor $e^{ik_L r}/k_L r$ provides the usual expression for the field emitted by a dipole [5], while the $\delta(\vec{r})$ term is required to determine the field at the location of the dipole, as shown in the static case in Ref. [5] when $\vec{P} \propto \delta(\vec{r})$. Equation (4) is readily generalized to the case of multichromatic incident light.

Before proceeding as outlined in the introduction, we would like to determine more carefully the field \vec{E}_d driving the atom. In the Maxwell-Bloch equations, which are the crudest mean-field approximation, \vec{E}_d is identical to \vec{E} . By noting that the driving field should contain only the contribution of the probe field \vec{E}_L and of the N-1 other atoms, it is possible to get a more accurate estimate of the positive frequency part $\vec{\mathcal{E}}_d$ of \vec{E}_d from Eq. (4), with $\vec{\mathcal{P}}$ replaced by $[(N-1)/N]\vec{\mathcal{P}}$. Also, the fact that our atoms are considered as individual entities suggests that a given atom should not be coupled to the field created by another atom at its own location [see the $\delta(\vec{r})$ term in Eq. (5)]. The effects of this physical requirement can be implemented by the introduction of a small fictitious excluded volume around the atoms. When put to a vanishing limiting value at the end of the calculation this excluded volume does not change the results obtained from an exact treatment of the problem, but it is supposed to lead to an improvement of our approximate mean-field approach. We therefore omit the $\delta(\vec{r})$ term of Eq. (5), which amounts to replacing $g_{\alpha\beta}(\vec{r})$ by $\tilde{g}_{\alpha\beta}(\vec{r})$ $=g_{\alpha\beta}(\vec{r})+(1/3)\delta_{\alpha\beta}\delta(\vec{r})$ in (4), and finally we get for the driving field:

$$\vec{\mathscr{E}}_{d}(\vec{R}) = \vec{\mathscr{E}}_{L}(\vec{R}) + \frac{N-1}{\varepsilon_{0}} \operatorname{Tr}'\{ [\tilde{g}(\vec{R} - \vec{R}')]\vec{D}'\rho(') \}.$$
(6)

We note that in a regime where $N-1 \approx N$ the replacement of g by \tilde{g} is equivalent to the addition of $\vec{P}(\vec{r})/3\varepsilon_0$ to the electric field amplitude in the optical Bloch equations, as is, e.g., done in [6] (local field correction).

By inserting the expression for the driving field into the atomic master equation (3), we obtain the equation for the atomic density matrix:

$$\frac{d}{dt} \rho = \mathscr{L}[\rho] - \frac{1}{i\hbar} [\vec{D}^{+} \cdot \vec{\mathscr{E}}_{L}(\vec{R}) + \text{H.c.}, \rho]
- \frac{N-1}{i\hbar\varepsilon_{0}} \text{Tr}' \left(\left[\sum_{\alpha\beta} \tilde{g}_{\alpha\beta}(\vec{R} - \vec{R}') D_{\alpha}^{+} D_{\beta}^{+-} + \tilde{g}_{\alpha\beta}^{*}(\vec{R} - \vec{R}') D_{\alpha}^{-} D_{\beta}^{++}, \rho \otimes \rho(') \right] \right).$$
(7)

 $D_{\alpha}^{+}, D_{\alpha}^{-}$ are the α components of the raising and lowering parts of the atomic dipole. As the same density matrix ρ is applied for primed and unprimed variables, we obtain a non-linear master equation.

We can interpret the tensor product in the last term of Eq. (7) as a Hartree approximation to a two-atom density matrix. It is then natural to include the bosonic ($\eta = 1$) or fermionic ($\eta = -1$) exchange symmetry of two-particle states by using properly symmetrized density matrices, so that in Eq. (7) we make the replacement,

$$\rho \otimes \rho(\prime) \to (1 + \eta P_{12}) \rho \otimes \rho(\prime) / [1 + \eta \operatorname{Tr}(\rho^2)].$$
(8)

The exchange operator P_{12} is defined by its action on twoparticle state vectors, $P_{12}|\xi\rangle \otimes |\xi'\rangle = |\xi'\rangle \otimes |\xi\rangle$. If we are far from having a macroscopic population of any quantum state, i.e., $\langle \psi | \rho | \psi \rangle \ll 1$ for any $| \psi \rangle$, the normalization factor can be replaced by unity. The resulting equation, in the case of a two-level atom, turns out to be identical to the one obtained in Ref. [2], except for the contact term, which is different from the one in our Eq. (5), and which is kept in the further treatment of that paper. In view of the special treatment, suggested for this term above Eq. (6), we do not consider this difference to be of physical significance. Our master equation (7) reduces to the nonlinear Schrödinger equation obtained in a specific situation, neglecting dissipation, in Ref. [4], when the definition of ξ in Refs. [3,4] is corrected to $\xi = k_L |\vec{r} - \vec{r'}|$. Here the contact term is not included.

We believe that the present derivation is more straightforward and that it can bring more insight because the nonlinearity of the master equation is related directly to the propagation of the electromagnetic field in the medium. A special example illustrating this aspect is that of laser cooling of an optically thick gas, Ref. [7]: attenuation of the laser beams as they propagate through the gas results in imbalanced intensities and hence a net force on atoms pointing from the edges

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towards the center of the sample; an interpretation of this effect in terms of nonlocal interactions is possible but clearly less appropriate.

As we obtain exactly the same equations, the validity of the calculations for a specific problem is not improved, but we may have a better feeling for the approximations made than what is offered by the approach in Refs. [1-4]. One should expect that, e.g., the incoherent spontaneously emitted light, disregarded in our equations, influences the atomic dynamics, and, indeed, the corresponding contribution to radiation pressure is considered to be an important ingredient in the understanding of the dynamics of a trapped atomic cloud [8,9].

To estimate the extent to which atomic interactions are taken into account in Eq. (7), we consider a gas $(N \ge 1)$ of ground-state atoms, illuminated by a weak laser beam $\vec{E}_L(\vec{r},t) = \tilde{\mathscr{E}}_L(\vec{r})e^{-i\omega_L t} + \text{c.c.}, \text{ on a transition between a}$ ground state of angular momentum $j_g = 0$ and an excited state of angular momentum $j_e = 1$. In a regime where the Doppler shift due to the atomic motion and the recoil shift $\hbar k_L^2/2M$ are much smaller than the rate Γ of spontaneous emission, the atomic kinetic energy $\vec{p}^2/2M$ can be neglected during the time required (a few Γ^{-1}) for the mean atomic dipole to reach a steady state. In the absence of quantum statistical effects and to first order in the probe field amplitude, we get from Eq. (3) (with \vec{E} replaced by \vec{E}_d) and from Eq. (2) the positive frequency part of the polarization density, $\hat{\mathcal{P}}(\vec{r}) = \varepsilon_0 \alpha \rho(\vec{r}) \hat{\mathcal{E}}_d(\vec{r})$, where $\rho(\vec{r}) = N \langle g, \vec{r} | \rho | g, \vec{r} \rangle$ is the density of ground-state atoms and α is the atomic polarizability. We eliminate the driving field using Eq. (6) $(N \ge 1)$, and from Eq. (1) we derive in the usual manner the (complex) refractive index of the probe field for a uniform density $\rho(\vec{r}) = \rho_0$, $n = \sqrt{1 + \alpha \rho_0}/(1 - \alpha \rho_0/3)$, which is the well-known Lorentz-Lorenz formula [10]. (For a transition with a dipole moment d and a resonance frequency ω_A , the polarizability has the value $\alpha = -d^2/[\hbar \varepsilon_0(\omega_L - \omega_A)]$ $+i\Gamma/2$)].)

This result for the refractive index does not include neardipole effects such as the shift of energy levels by resonant van der Waals interactions [11-13]. Therefore these high density corrections are not taken into account in the Maxwell-Bloch equations. In the further discussions of the nonlinear equations in Refs. [2,4] severe approximations are made, so as, e.g., to replace a term equivalent to our $g_{\alpha\beta}(\vec{r})$ by $A\delta(\vec{r})\delta_{\alpha\beta}$. The validity of this approximation may be difficult to assess from the derivations in the cited papers. Here, we associate the spatial structure of the nonlinear term directly with the propagation of electromagnetic fields, and the replacement corresponds to a simplified radiation pattern, probably oversimplified for most applications. As readily seen from Eq. (4), the total field \mathcal{E} emerging from a medium would in this case be identical to the probe field \mathcal{E}_L , which is not correct when the field dephasing or absorption is not negligible over the atomic sample.

Our treatment of the coupled Maxwell-Bloch equations has a classical analog in the self-consistent field approach to ionized plasmas. The electric and magnetic fields, appearing through the usual Lorentz force in the transport equation for the electron and ion distribution functions, may be expressed in terms of the charge densities and currents. This procedure establishes the Vlasov equations [14], and it is, indeed, very similar to the one adopted in the present paper. The Vlasov equations are derived under the assumption of a collisionfree plasma, disregarding an extra collision integral from the equations. This approximation may be related to the neglect of incoherently scattered light in our calculations.

Coupled equations for electromagnetic fields and charged particles also appear in the theory of semiconductors. Here it is established that a Hartree-Fock approximation of the electronic state, disregarding collisions, yields the "coherent contribution" to the dynamics only [15]. The fact that effects of incoherently scattered light are not treated exactly by the Hartree approach in [2] is thus no surprise, but the reported agreement with our approach probably reveals this feature more clearly.

Finally, it should be mentioned that in contrast to the usual Maxwell-Bloch equations, the center-of-mass motion of the atoms in the medium is treated here fully quantum mechanically. It is of course possible to also extract the solution for the electromagnetic field from our equations without neglecting the atomic motion, and one may anticipate a number of new phenomena in field generation from such considerations.

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