Nonlinear-optical properties of a noninteracting Bose gas

M. A. Kasevich

Department of Physics, Stanford University, Stanford, California 94305

S. E. Harris

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

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We show that the characteristic Rabi frequency and nonlinear susceptibility of atoms in a dilute Bose gas remain unchanged by the process of condensation. We neglect the effects of atomic dipole–dipole interactions and spontaneous emission. © 1996 Optical Society of America

The recent field-opening experiments of Anderson *et al.*[1] and Bradley *et al.*[2] have demonstrated the formation of a Bose–Einstein condensate of atoms whose mean spacing is small compared with their de Broglie wavelength and large compared with the optical wavelength of their resonance transition. Because the de Broglie wavelength is larger than the average distance between the atoms, individual atoms cannot be distinguished, and the condensate must be considered a single, macroscopic, quantum state. When one does so, one finds that the matrix element for optical excitation between the symmetrized Bose product states varies as the square root of the number of particles in the condensate.

One might therefore be led to anticipate such extraordinary macroscopic properties as (i) a Rabi frequency that itself varies as the square root of the number of particles or, because third-order susceptibilities vary as the fourth power of the matrix element, (ii) a third-order susceptibility that is proportional to the square of the number of atoms in the sample.

In this Letter we show that, if all interactions between particles are neglected, these properties do not exist and, in fact, that the Rabi frequency and the nonlinear susceptibility of the sample are unchanged by condensation. In our model we assume N degenerate two-state atoms that interact with a semiclassical radiation field. We ignore spontaneous emission and atomic dipole–dipole interactions and consider the spatial extent of the condensate to be much larger than the optical wavelength. Hiroshima and Yamamoto[3] recently calculated $\chi^{(3)}$ for ensembles of tightly confined degenerate Bose and Fermi atoms. You and coworkers[4] considered the optical response of confined Bose atoms under the conditions of short-pulse excitation. Other related studies have focused on the linear-optical response of a Bose gas.[5–8]

To show the interesting and perhaps peculiar way in which the condensate properties reduce to those of single atoms, we work the problem in the atomic number state basis. As illustrated in Fig. 1, we let state $|q\rangle$ denote the excitation of q of these atoms from their internal ground state $|a\rangle$ to their excited state $|b\rangle$. For simplicity we take the center-of-mass state for atoms in $|a\rangle$ to be the momentum $\mathbf{p} = 0$ eigenstate and that of atoms in state $|b\rangle$ to be the $\mathbf{p} = \hbar\mathbf{k}$ momentum eigenstate ($\mathbf{k}$ is the wave vector of the incident electromagnetic field). The states $|q\rangle$ are then eigenstates of the free particle Hamiltonian $\mathcal{H}_0$ with eigenenergies

$$\mathcal{H}_0|q\rangle = q\left(\hbar\omega_0 + \frac{\hbar^2k^2}{2m}\right)|q\rangle, \tag{1}$$

where $\hbar\omega_0$ is the internal electronic excitation energy and $\hbar^2k^2/2m$ is the single-photon recoil energy.

The ith particle in the condensate interacts with an electric field $\mathbf{E}(\mathbf{R}_i) = \boldsymbol{\mathcal{E}}/2 \exp[j(\omega t - \mathbf{k}\cdot\mathbf{R}_i)] + \text{c.c.}$ through the electric–dipole interaction $V = -e\sum_{i=1}^{N} \mathbf{r}_i \cdot \mathbf{E}(\mathbf{R}_i)$. The interaction matrix element is

$$\langle q|V|q+1\rangle = -\frac{\hbar\Omega}{2}(q+1)^{1/2}(N-q)^{1/2}\exp(j\omega t) + \text{c.c.}, \tag{2}$$

where the single-atom Rabi frequency $\Omega \equiv \boldsymbol{\mu}_{ab}\cdot\boldsymbol{\mathcal{E}}/\hbar$ ($\boldsymbol{\mu}_{ab}$ is the single-atom dipole matrix element). The scaling of the matrix element with N and q follows directly from the required exchange symmetry of the condensate wave functions. Note that in the limit $N \gg q$ the strength of the interaction matrix element scales as $N^{1/2}$.

We study the dynamics of the light–condensate interaction in the interaction picture. With $|\psi(t)\rangle = \sum_{q=0}^{N} a_q(t)\exp(-jq\omega t)|q\rangle$ and the rotating-wave

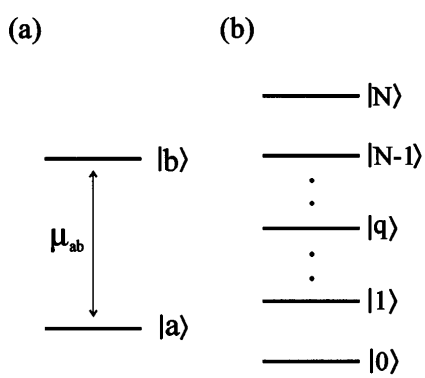

Fig. 1. (a) For a total of N atoms, in the basis of bare, independent atoms there are 2^N possible basis states; (b) in the symmetrized Bose basis there are $(N+1)$ states. The qth state is that where q atoms are excited.

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approximation, we arrive at the following coupled equations describing the evolution of the probability amplitudes $a_q(t)$:

$$\frac{\partial a_q}{\partial t} + jq\Delta\omega a_q = j\frac{\Omega^*}{2}\xi_{q-1}a_{q-1} + j\frac{\Omega}{2}\xi_q a_{q+1} \qquad (0 \le q \le N), \quad (3)$$

where $\xi_q = (q+1)^{1/2}(N-q)^{1/2}$, the detuning $\Delta\omega = \omega_0 + \hbar k^2/2m - \omega$, and the spurious amplitudes a_{-1} and a_{N+1} are zero for all t. Taking $N = 1$, Eq. (3) reduces to the standard pair of coupled equations for a two-level atom interacting with a driving radiation field. On resonance, these equations lead to the well-known sinusoidal oscillation of population between the ground and excited states at the Rabi frequency Ω.

We first investigate the limit $N \gg 1$ by numerically integrating Eq. (3) for large numbers of atoms.[9] Figure 2(a) shows the on-resonance time evolution of the probability of finding an ensemble of $N = 100$ atoms in the $|0\rangle$ state subject to the initial condition that, at $t = 0$, all the atoms are in state $|a\rangle$. We find what at first is strange behavior: the population is rapidly depleted at a rate proportional to $N^{1/2}\Omega$ but then returns on a time scale set by the single-atom Rabi frequency Ω.

To gain insight into the collective evolution of the entire set of probability amplitudes we show in Fig. 2(b) the time evolution of the mean number of excited atoms, $\langle q\rangle = \sum q|a_q|^2$. Here we find that the observable $\langle q\rangle$ oscillates at frequency Ω in exactly the same manner as a collection of N nondegenerate atoms.

As an example of a second observable for this system, we consider the induced polarization operator $\mathbf{P}(\mathbf{R})$:

$$\mathbf{P}(\mathbf{R}) = e\sum_i \mathbf{r}_i\delta(\mathbf{R} - \mathbf{R}_i). \quad (4a)$$

Assuming box normalization to volume $\mathcal{V}$ of the momentum eigenstates, the matrix elements of the dipole operator are

$$\langle q|\mathbf{P}(\mathbf{R})|q+1\rangle = \frac{\xi_q}{\mathcal{V}}\boldsymbol{\mu}_{ab}\exp(j\mathbf{k}\cdot\mathbf{R}), \quad (4b)$$

and the expected value of the induced polarization $\langle\mathbf{P}(\mathbf{R})\rangle$ is

$$\langle\psi(t)|\mathbf{P}(\mathbf{R})|\psi(t)\rangle = \frac{1}{\mathcal{V}}\boldsymbol{\mu}_{ab}\exp[j(\omega t - \mathbf{k}\cdot\mathbf{R})] \times \sum_{q=0}^{N-1}\xi_q a_q a_{q+1}^* + \text{c.c.} \quad (5)$$

In the limit where the detuning $\Delta\omega$ is much larger than the Rabi frequency $|\Omega|$, one can use perturbation theory to calculate $\langle\mathbf{P}\rangle$ to arbitrary order in the small parameter $|\Omega|/\Delta\omega$. The third-order term in this expansion, $\langle\mathbf{P}^{(3)}\rangle$, determines the third-order susceptibility $\chi^{(3)}$ and sets the magnitude of such effects as self-focusing and self-phase modulation. With the standard assumption that the condensate adiabatically evolves from the lowest-lying state $|0\rangle$ to state $|\psi(t)\rangle$ as the optical field is turned on we find that

$$\langle\mathbf{P}^{(3)}(\mathbf{R})\rangle = -\frac{1}{\mathcal{V}}\frac{\boldsymbol{\mu}_{ab}|\Omega|^2\Omega}{4\Delta\omega^3}\exp[j(\omega t - \mathbf{k}\cdot\mathbf{R})] \times (\xi_0{}^4 - \xi_0{}^2\xi_1{}^2/2) + \text{c.c.} \quad (6a)$$

$$= -\frac{N}{\mathcal{V}}\frac{\boldsymbol{\mu}_{ab}|\Omega|^2\Omega}{4\Delta\omega^3}\exp[j(\omega t - \mathbf{k}\cdot\mathbf{R})] + \text{c.c.} \quad (6b)$$

The first term in Eq. (6a) is the contribution of perturbation path a [shown in Fig. 3(a)]; the second is that of path b [shown in Fig. 3(b)]. Note that either path, if alone, yields a contribution proportional to N^2. When both paths are included, the N^2 terms cancel and $\langle\mathbf{P}^{(3)}\rangle$ is the same as that of N nondegenerate atoms.

From these results one is led to wonder whether all stimulated optical properties of a dilute condensate reduce to those of a nondegenerate gas. Further insight can be gained by consideration of the class of Hamiltonians of the form $\mathcal{H} = \sum_i \mathcal{H}^{(i)}$, where the operators $\mathcal{H}^{(i)}$ depend on the coordinates of the ith particle alone. For example, the Hamiltonian for the noninteracting Bose gas considered above falls into this class, but one that includes atom–atom interactions does not. For this class of Hamiltonians the equations of motion are separable in the coordinates of the individual particles, and the condensate wave function $|\psi(t)\rangle = \prod_i |\varphi_i(t)\rangle$, where $|\varphi_i(t)\rangle$ are solutions to the single-particle equations $\mathcal{H}^{(i)}|\varphi_i(t)\rangle = j\hbar\partial/\partial t|\varphi_i(t)\rangle$. [Note that $|\psi(t)\rangle$ is automatically symmetric under particle interchange if all particles initially reside in the same quantum state.]

We now specialize this result to the example of Rabi oscillations considered above. The on-resonance,

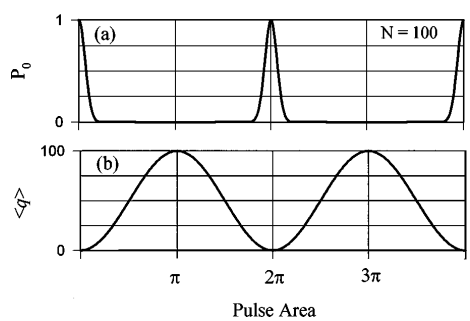

Fig. 2. Numerical integration of the coupled equations [Eq. (3)] for $N = 100$ atoms: (a) population in state $|0\rangle$, (b) mean number of excited atoms $\langle q\rangle$.

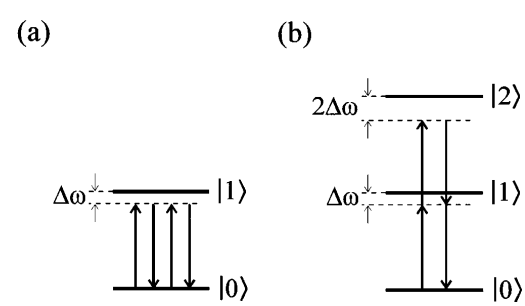

Fig. 3. Perturbation paths for calculation of $\chi^{(3)}$: (a) path a, (b) path b.

single-atom solutions are $|\psi_i\rangle = \cos(\Omega t/2)|a_i\rangle + \sin(\Omega t/2)|b_i\rangle$. Here the kets $|a_i\rangle$ and $|b_i\rangle$ correspond to the ground state (with momentum $\mathbf{p} = 0$) and the excited state (with momentum $\mathbf{p} = \hbar\mathbf{k}$), respectively, of the ith particle. In this notation the number state $|0\rangle$ of the N atom system is $\prod_i |a_i\rangle$. Thus the probability of finding an atom in state $|0\rangle$ [as shown in Fig. 2(a)] is $\cos^{2N}(\Omega t/2)$. We also find immediately that $\langle q\rangle$ [shown in Fig. 2(b)] oscillates at frequency Ω. Similar arguments apply to calculation of the induced dipole moment. It follows directly that $\langle\mathbf{P}\rangle = \sum_i \langle\mathbf{P}_i\rangle$, where $\langle\mathbf{P}_i\rangle$ are the single-atom contributions to the induced dipole moment.

In conclusion, we have shown that the Rabi frequency and susceptibility do not depend on the momentum degeneracy of dilute atomic samples and that these results follow directly from the separability of the collective Hamiltonian.

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