# Photon blockade and quantum dynamics in intracavity coherent photoassociation of Bose-Einstein condensates

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We demonstrate that a photon blockade effect exists in the intracavity coherent photoassociation of an atomic Bose-Einstein condensate and that the dynamics of the coupled atomic and molecular condensates can only be successfully described by a quantum treatment of all the interacting fields. We show that the usual mean-field calculational approaches give answers that are qualitatively wrong, even for the mean fields. The quantization of the fields gives a degree of freedom that is not present in analogous nonlinear optical processes. The difference between the semiclassical and quantum predictions can actually increase as the three fields increase in size so that there is no obvious classical limit for this process.

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Using a fully quantum analysis of an intracavity electromagnetic field resonantly coupling atomic and molecular Bose-Einstein condensates (BEC), we find that a photon blockade effect can be caused and that the dynamics of the three fields are not even qualitatively similar to those of mean-field predictions. The effects we describe are not present for a traveling-wave electromagnetic field interacting with the condensate, but occur because of correlations that build up between the matter fields and the confined electromagnetic field. Unlike many nonclassical systems, these effects do not necessarily scale inversely with system size, demonstrating that a BEC is indeed a macroscopic quantum object. Although there are parametric processes in nonlinear optics where the noise properties are also important in the dynamics, the system we describe here exhibits a richer range of behaviors because the quantization of the electromagnetic field means that we effectively have a quantized $\chi^{(2)}$ nonlinearity, which is not possible with optical parametric systems.

The Gross-Pitaevski equation (GPE) has been largely successful in describing the dynamical features of weakly interacting dilute gas Bose-Einstein condensates [1]. However, it has recently been shown that for resonant coupling between atomic and molecular condensates using a Feshbach resonance, it gives predictions that are in disagreement with those of the Hartree-Fock-Bogoliubov theory [2], although the authors assume classical behavior for the molecular field. What we show here is that there is another simple dynamical process that it cannot describe accurately and for which a full quantum treatment is necessary, namely, the intracavity coherent photoassociation of an atomic condensate to form a molecular condensate.

Photoassociation of an atomic condensate to form a molecular condensate has been investigated using a simplified mean-field model [3], while coherent, molecular soliton formation has also been predicted in a similar system [4]. The more robust method of stimulated Raman adiabatic passage (STIRAP) has been studied in a mean-field approximation [5,6]. The quantum statistical properties of single mode STIRAP with classical electromagnetic fields and the mean-field multimode behavior in one dimension has also been studied [7]. The mean-field dynamical behavior of atomic and molecular condensates coupled by a Raman transition has been investigated in three-dimensions, showing giant collective oscillations between the atoms and molecules [8].

Methods have been developed to study the interaction of quantized matter and electromagnetic fields [9–11], although these have only been applied so far to different electronic levels of the atoms and then after making various approximations, including linearization of the resulting equations of motion. As the system of photoassociation we are considering here has formal similarities to second-harmonic generation (SHG) and behavior has been predicted there that is not calculable in a mean-field or linearized approximation [12], we have chosen to use the phase-space methods of quantum optics. The disadvantage of this is that we have to proceed numerically, but the advantage is that we have more control over any approximations that we may choose to make. What none of the approaches to photoassociation have done is to quantize the electromagnetic field, all treating the interaction as having an effective $\chi^{(2)}$ strength that remains constant, as in the familiar approaches to SHG and parametric down conversion. While this is a good approximation for a traveling-wave electromagnetic field, it is not sustainable if we consider the three fields interacting in an electromagnetic cavity, as we will show below.

The system we consider is as shown schematically in Fig. 1. A trapped atomic condensate is held in an electomagnetic cavity. Our formalism is applicable to both microwave and optical transitions. The empty cavity is resonant at the frequency of the transition between atomic and molecular states of the condensate. Here we make the approximation that all three fields can be represented as single modes, which is reasonable as long as we are considering short interaction times where the kinetic energy may be ignored. We can also ignore other vibrational and rotational levels of the molecular state as long as the energy spacing between these is more than the laser linewidth. We also ignore spontaneous disso-

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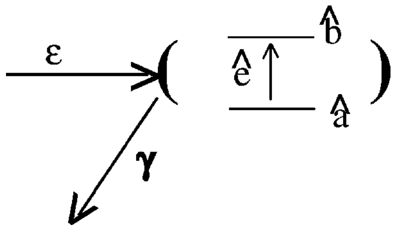


FIG. 1. Schematic of the condensate, represented by the operators $\hat{a}$ and $\hat{b}$, inside the electromagnetic cavity with field operator $\hat{e}$. The classical cavity pumping is represented by $\epsilon$ and the loss rate is represented by $\gamma$.

ciation of the molecules into noncondensed atomic states, again as a short-time approximation. We also make the normal zero-temperature approximation of quantum optics, as condensates exist at temperatures of the order of nanokelvins.

The interaction Hamiltonian for this system in the rotating-wave approximation is

$$\mathcal{H}=\frac{i\hbar g}{2}[\hat{a}^{\dagger 2}\hat{b}\hat{e}^{\dagger}-\hat{a}^{2}\hat{b}^{\dagger}\hat{e}]+\hbar\chi_a\hat{a}^{\dagger 2}\hat{a}^{2}+\hbar\chi_b\hat{b}^{\dagger 2}\hat{b}^{2}+i\hbar(\epsilon\hat{e}^{\dagger}-\epsilon^{*}\hat{e})+\Gamma^{\dagger}\hat{e}+\Gamma\hat{e}^{\dagger}, \tag{1}$$

where $g$ represents the effective coupling strength between the condensates and the electromagnetic field, $\hat{a}(\hat{b})$ is the annihilation operator for the atomic (molecular) condensate and $\hat{e}$ is the annihilation operator for the intracavity electromagnetic field. The $\chi_j$ represent the self-interaction terms between the atoms or molecules, $\epsilon$ represents the classical pumping of the cavity, and $\Gamma$ is a bath operator for the electromagnetic field.

Following the standard methods [13], we find a partial differential equation for the $P$ distribution of this system

$$\begin{aligned}\frac{\partial P}{\partial t}=\Bigg\{&\frac{\partial}{\partial\alpha}[-ge^{*}\alpha^{*}\beta+2i\chi_a\alpha^{*}\alpha^{2}]+\frac{\partial}{\partial\alpha^{*}}[-ge\alpha\beta^{*}-2i\chi_a\alpha^{*2}\alpha]+\frac{\partial}{\partial\beta}\left[\frac{g}{2}\alpha^{2}e+2i\chi_b\beta^{*}\beta^{2}\right]\\&+\frac{\partial}{\partial\beta^{*}}\left[\frac{g}{2}\alpha^{*2}e^{*}-2i\chi_b\beta^{*2}\beta\right]+\frac{\partial}{\partial e}\left[-\frac{g}{2}\alpha^{*2}\beta+\gamma e-\epsilon\right]+\frac{\partial}{\partial e^{*}}\left[-\frac{g}{2}\alpha^{2}\beta^{*}+\gamma e^{*}-\epsilon^{*}\right]\\&+\frac{1}{2}\left[\frac{\partial^{2}}{\partial\alpha^{2}}(g\beta e^{*}-2i\chi_a\alpha^{2})+\frac{\partial^{2}}{\partial\alpha^{*2}}(g\beta^{*}e+2i\chi_a\alpha^{*2})+\frac{\partial^{2}}{\partial\beta^{2}}(-2i\chi_b\beta^{2})+\frac{\partial^{2}}{\partial\beta^{*2}}(2i\chi_b\beta^{*2})\right.\\&\left.+\frac{\partial^{2}}{\partial\alpha\,\partial e}(2g\alpha^{*}\beta)+\frac{\partial^{2}}{\partial\alpha^{*}\,\partial e^{*}}(2g\alpha\beta^{*})\right]-\frac{1}{6}\left[\frac{\partial^{3}}{\partial\alpha^{2}\,\partial e}(3g\beta)+\frac{\partial^{3}}{\partial\alpha^{*2}\,\partial e^{*}}(3g\beta^{*})\right]\Bigg\}P(\alpha,\beta,e,t),\end{aligned} \tag{2}$$

where $\gamma$ represents the loss rate of the optical field from the cavity.

We note here that Eq. (2) is not of the standard Fokker-Planck form, as it contains third-order derivatives. Although formal methods do exist for dealing with these [14], they are not easy to use. An approximation, which is commonly made especially in the Wigner representation, is to truncate the equation at second order. This has been shown to be accurate for the dynamics and quadrature variances of second-harmonic generation [12] and for calculating first-order correlation functions in trapped BEC [15], although it is not accurate for the calculation of higher-order correlations in traveling-wave SHG [16]. This truncation can be justified by claiming that the coefficients of the third-order terms are smaller than the other coefficients in the equation, which is certainly the case in our present example. After truncation, we can map Eq. (2) onto the following set of Ito stochastic differential equations in the positive-$P$ representation

$$\begin{aligned}\frac{d\alpha}{dt}&=-2i\chi_a\alpha^{\dagger}\alpha^{2}+ge^{\dagger}\alpha^{\dagger}\beta+\frac{\sqrt{g}}{2}(e^{\dagger}+\beta)\eta_1(t)\\&\quad+\frac{i\sqrt{g}}{2}(e^{\dagger}-\beta)\eta_3(t)+\sqrt{-2i\chi_a\alpha^{2}}\,\eta_5(t),\\\frac{d\alpha^{\dagger}}{dt}&=2i\chi_a\alpha^{\dagger 2}\alpha+ge\alpha\beta^{\dagger}+\frac{\sqrt{g}}{2}(e+\beta^{\dagger})\eta_2(t)\\&\quad-\frac{i\sqrt{g}}{2}(e-\beta^{\dagger})\eta_4(t)+\sqrt{2i\chi_a\alpha^{\dagger 2}}\,\eta_6(t),\\\frac{d\beta}{dt}&=-2i\chi_b\beta^{2}\beta^{\dagger}-\frac{g}{2}\alpha^{2}e+\sqrt{-2i\chi_b\beta^{2}}\,\eta_7(t),\\\frac{d\beta^{\dagger}}{dt}&=2i\chi_b\beta^{\dagger 2}\beta-\frac{g}{2}\alpha^{\dagger 2}e^{\dagger}+\sqrt{2i\chi_b\beta^{\dagger 2}}\,\eta_8(t),\end{aligned} \tag{3}$$

$$\frac{de}{dt}=\epsilon-\gamma e+\frac{g}{2}\alpha^{\dagger 2}\beta+\sqrt{g}\alpha^{\dagger}\eta_1(t)+i\sqrt{g}\alpha^{\dagger}\eta_3(t),$$

$$\frac{de^{\dagger}}{dt}=\epsilon^{*}-\gamma e^{\dagger}+\frac{g}{2}\alpha^{2}\beta^{\dagger}+\sqrt{g}\alpha\eta_2(t)-i\sqrt{g}\alpha\eta_4(t),$$

where the real-noise sources have the properties

$$\overline{\eta_i(t)}=0,\quad \overline{\eta_j(t)\,\eta_k(t')}=\delta_{jk}\delta(t-t'). \tag{4}$$

There is a correspondence between the $c$-number variables $[\alpha,\alpha^{\dagger},\beta,\beta^{\dagger},e,e^{\dagger}]$ and the operators $[\hat{a},\hat{a}^{\dagger},\hat{b},\hat{b}^{\dagger},\hat{e},\hat{e}^{\dagger}]$, although, as always with the positive-$P$, a variable such as $\alpha^{\dagger}$ is not complex conjugate to $\alpha$ except in the mean, due to the independence of the noise sources. We note here that it is possible to write the noise terms in many different ways, amounting to different factorizations of the diffusion matrix of Eq. (2). We should note here that the above equations, although having a formal similarity to those used to describe traveling-wave SHG with an additional $\chi^{(3)}$ nonlinearity [17], exhibit one important difference. Instead of a constant $\kappa$, the effective $\chi^{(2)}$ interaction used in [17], we now have the field-dependent $ge$. Another difference in our present case would be that we now have a term $(g/2)\alpha^{\dagger 2}\beta$ in the equation for the electromagnetic field.

We have solved Eq. (3) numerically for a range of parameters and found behavior of the mean fields that is strikingly different from that found in the usual mean-field approximation. This is completely different from many situations in quantum optics or in the study of condensates where the dynamics of the mean fields can be successfully described by considering only the drift terms in the appropriate Fokker-Planck equation. In these cases, as long as care is taken with the parameter regimes, the full stochasticity of the problem only becomes important when we wish to consider quantum-statistical properties.

In our simulations, we begin with an atomic condensate inside a cavity that begins to be pumped at $t=0$. Initially neither molecules nor electromagnetic field are present, with the atomic field being treated as being initially in a coherent state.

Through numerical investigations, we have found that this system exhibits at least three regimes of behavior, only one of which we describe in detail here. The behavior shown in our plots comes from what we may consider the strong-interaction regime and always exhibits short-time oscillations and photon blockade. In the weak-interaction regime, which may be reached by decreasing the strength of $g$ or the number of atoms, the solutions approach those found by treating all fields semiclassically. The solutions for atom and molecule number are similar to those found in superchemistry [8]. There are almost total oscillations between the two states and the photon blockade is not seen. There is also a regime between these two in which there are no oscillations, but partial conversion between atoms and molecules with the photon blockade effect being seen as the conversion stops.

In Fig. 2(a) we show the time development of the atomic and molecular fields as the cavity is turned on for the parameters $g=10^{-5}$, $|\epsilon|^2=10^6$, $\chi_{a,b}=10^{-9}$, and $|\alpha(0)|^2=10^6$, which are all scaled in terms of the cavity loss rate. We have taken the means over $5\times10^5$ stochastic trajectories, which were sufficient to ensure excellent convergence. What we see is that the atoms begin to associate to form molecules, but that only a small fraction are converted before the system undergoes transient oscillations between its atomic and molecular components. After a few cavity lifetimes, both components reach a steady state, with over 90% of the population still being in the atomic state.

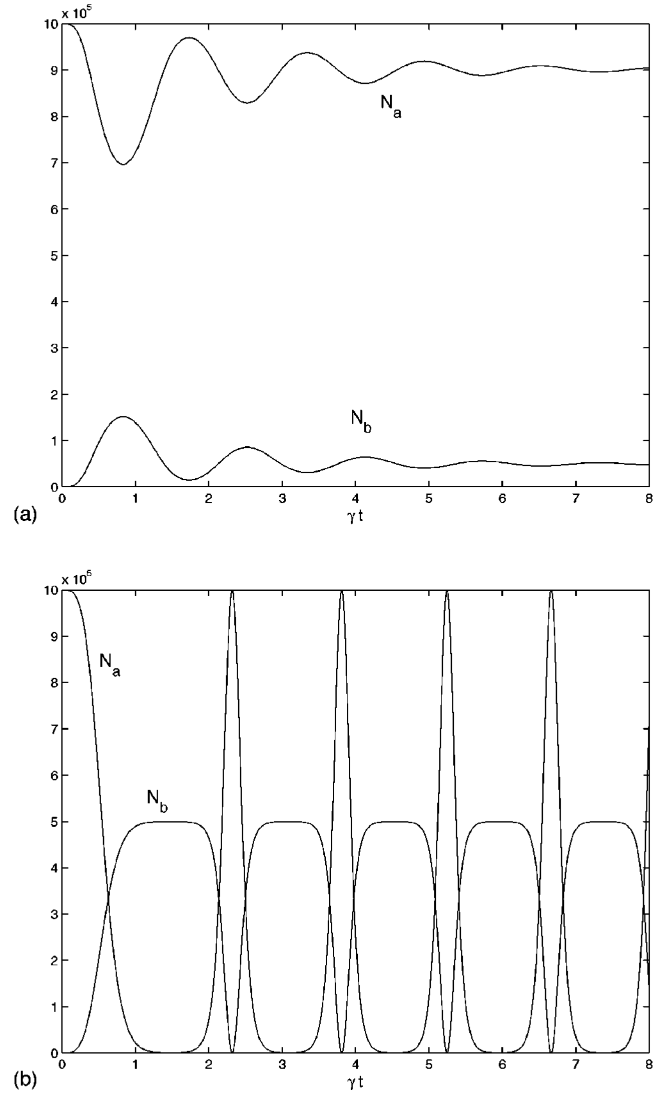


FIG. 2. (a) Occupation numbers of the atomic and molecular condensates as a function of time according to $5\times10^5$ quantum trajectories. The parameters are $g=10^{-5}$,$|\epsilon|^2=10^6$, $\chi_{a,b}=10^{-9}$, and $|\alpha(0)|^2=10^6$. In all graphs, the quantities are dimensionless. (b) Linearized solutions for the occupation numbers of the atomic and molecular condensates as a function of time.

It is instructive to consider the linearized solutions for the mean fields, i.e., the solutions of Eq. (3) with the noise terms removed. This means that we are now treating all three fields semiclassically. Because of the dependence of the noise terms on all three fields, it is not sensible here to treat, for

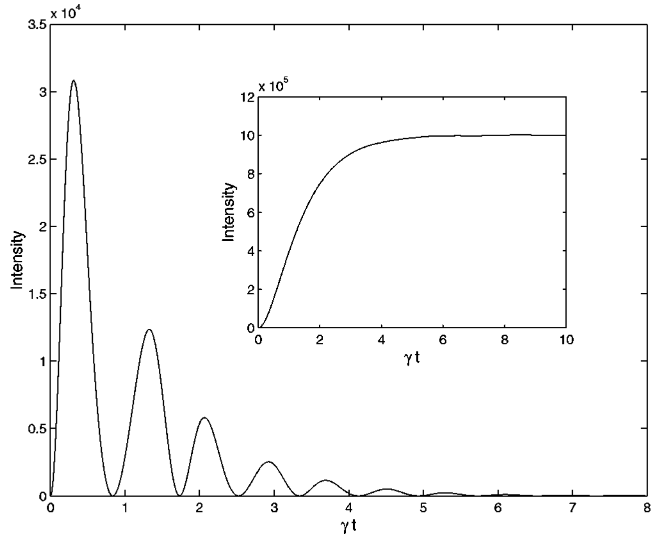


FIG. 3. The intracavity intensity of the electromagnetic field as calculated quantum mechanically, showing the photon blockade. The semiclassical solution is shown in the inset and rises to a value of almost $10^6$.

example, the matter fields quantum mechanically and the electromagnetic field semiclassically.

However, when we look at these solutions for our present problem, we see from Fig. 2(b) that after approximately the first third of a cavity lifetime, they do not even approximate the quantum solutions. This disagreement is even more striking than that previously found for pure traveling-wave SHG [12] and can be qualitatively explained as the result of correlations that build up between the three fields. We obtain some insight into the reason for this unexpected behavior when we examine the dynamics of the intracavity electromagnetic field as shown in Fig. 3. We find an initial build up of intensity in the cavity, with this field also becoming oscillatory and eventually almost vanishing completely. As the cavity continues to be pumped at the same rate, what we see is that it has become opaque. That is, a photon blockade effect is operating due to correlations that build up between the electromagnetic and matter fields [18,19]. This effect has been seen previously in systems that develop an effective giant $\chi^{(3)}$ nonlinearity. In the linearized approach, the electromagnetic field rises monotonically to a steady-state value very close to $|\epsilon|^2/\gamma$, as shown in Fig. 3, in stark contrast to the effectively empty cavity of the quantum solutions. What we can see here is that even going one step past the usual approach, which has treated the field-matter coupling as constant, and linearizing the quantum equations, which maintains to some degree the dynamics of the effective interaction, is not enough to give the correct solutions.

What we cannot say about this system is whether, in the situations where photon blockade is achieved, there will be later revivals of the oscillations. This type of effect has been predicted in parametric downconversion and for ultracold atoms in a driven microwave cavity [9], but would need a prohibitive amount of computer time to calculate using stochastic integration. The other open questions are about correlations between the three fields and the quantum statistics due to the interactions. As we have truncated the third-order terms in order to be able to calculate mean-field dynamics, we cannot have faith that our approach would adequately describe these correlations, which remain as a topic for further investigation. It is also important to ask whether the generalization to multimode atomic and molecular fields could cause the quantum and semiclassical solutions to converge drastically enough to allow a mean-field approach to describe the dynamics of this system. Results we have obtained for traveling-wave photoassociation [20] suggest that this is highly unlikely to be the case.

In conclusion, we have described a situation in which the Gross-Pitaevski approach does not describe adequately the dynamics of a Bose-Einstein condensate. The differences are not of the order of the inverse of the system size, but are qualitative. In contrast to the usual wisdom in which quantum effects become less important as system sizes increase, we have seen from numerical investigations that our solutions become closer to the semiclassical solutions as the number of atoms becomes smaller. This is a sign of the nonlinearity of the quantum dynamics, where correlations are built up between the three fields.

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