Role of quantum statistics in the photoassociation of Bose-Einstein condensates

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We show that the photoassociation of an atomic Bose-Einstein condensate to form condensed molecules is a chemical process which not only does not obey the Arrhenius rules for chemical reactions, but that it can also depend on the quantum statistics of the reactants. Comparing the predictions of a truncated Wigner representation for different initial quantum states, we find that, even when the quantum prediction for an initial coherent state is close to the Gross-Pitaevskii prediction, other quantum states may result in very different dynamics.

I. INTRODUCTION

The production of a molecular Bose-Einstein condensate (BEC) via Raman photoassociation of an atomic condensate has attracted much theoretical and experimental interest in the past few years. That an atom-optical analog of the optical processes of frequency conversion should exist with condensates was first stated by Drummond *et al.* [1], who developed an effective quantum field theory to describe coupled atomic and molecular BECs. An early suggestion that a molecular condensate could be produced via photoassociation came from Javanainen and Mackie [2], who proposed a two-mode, phenomenological Hamiltonian to model the process. A more complete proposal, using an atomic and two molecular fields with spatial dependence, coupled via a two-color Raman transition so as to minimize spontaneous emission losses, was developed by Heinzen *et al.* [3]. This model utilized the Gross-Pitaevskii equation (GPE) approach and demonstrated that the molecular formation would not obey the usual Arrhenius rules for chemical reactions. Góral *et al.* [4] predicted that interactions with a thermal component would tend to destroy the oscillations predicted by Heinzen *et al.* after a short interaction time. As shown by Hope and Olsen in one dimension [5], and Hope in three dimensions [6], full quantum zero-temperature treatments using the positive-P representation [7,8] may not always agree with mean-field predictions, even for the mean-fields.

Experimental efforts to form molecular condensates by photoassociation have been at least partially successful, with ^{87}Rb molecules having been formed by Raman photoassociation techniques [9], although the atom-molecule oscillations predicted theoretically [3] were not observed. Using a similar technique, Gerton *et al.* [10] formed ^{7}Li molecules as a means to vary the atom number in a condensate with attractive interactions and hence study the growth and collapse dynamics. Molecules have also been formed from a sodium condensate by a single-photon transition, although this method does have problems with their subsequent spontaneous breakup [11]. It may not be long before experiments advance to the stage where the predicted superchemistry-type oscillations can be achieved and detected.

Another intriguing question has been the issue of the quantum state of a trapped condensate with repulsive interatomic interactions. Perhaps the two most natural choices are the coherent state and the Fock state, so useful in quantum optics. The coherent state appeals because of the coherence properties exhibited [12–14], but has the problem of a largish uncertainty in number, which is conceptually difficult to understand as atoms are not created or destroyed at typical temperatures. The number state is superficially an appealing choice, but as the condensate is in contact with an environment, particles can be added or removed. This state also has the problem that it has no defined phase. As the nonlinearity due to *s*-wave collisions between condensed atoms is equivalent to a Kerr interaction, we may expect to find that the actual state is none of the above. An early calculation [15] predicted an amplitude eigenstate, while a subsequent, more rigorous calculation [16] predicted a sheared Wigner function which approximated a number squeezed state. A more recent attempt, using the Hartree approximation, found a Q function which suggests both amplitude quadrature and number squeezing [17].

In this work we combine these issues, considering the effects of different possible initial states on the dynamics of Raman photoassociation, without actually solving the problem of which may be the most likely ground state of the trapped condensate. As the mathematics of photoassociation is essentially a more complex form of that of second-harmonic generation, and both quantum statistics [18,19] and Kerr nonlinearities [20] affect the dynamics of this process, it is of interest to investigate their effect in the present situation. As we investigate only the dynamics of the mean fields rather than quantum correlations, we stochastically integrate the appropriate equations in the truncated Wigner representation [8,21,22], which we expect to give reliable results for the numbers of particles involved.

II. THE SYSTEM AND EQUATIONS OF MOTION

We consider that the initial atomic condensate is trapped such that one of the frequencies (ω_0) is much smaller than the other two, allowing use of a one-dimensional approximation. We consider here a Raman photoassociation scheme [3,5,6] with the excited molecular field adiabatically elimi-

nated. Photoassociation has previously been treated using a functional positive-P representation [5,6], which gives true stochastic differential equations for the coupled and molecular fields. However, numerical integration of these equations is very time consuming and can present serious stability problems [8]. Hence we use a truncated Wigner representation, which is much more stable and lends itself readily to the modeling of different initial quantum states of the atomic condensate. A full Wigner representation of this problem has derivatives of third order in the equation of motion for the pseudoprobability function, and, while it is possible to model these using stochastic difference equations [23], there are severe practical difficulties involved. However, as is common with the Wigner representation, we discard the third-order derivatives, which in this case leaves us with a Fokker-Planck equation with no diffusion matrix. This can be immediately mapped onto differential equations which have the appearance of coupled Gross-Pitaevskii-type equations. We stress here that there are, however, two important differences. The Wigner equations calculate symmetrically ordered operator products and averages must be taken over a large number of integrations, with initial conditions chosen so as to represent the Wigner function for the desired initial states. Only if the Wigner function were to be a Dirac δ, which is completely nonphysical, would we recover the Gross-Pitaevskii equation. We can now model the quantum fields (to a good level of approximation) via equations which are completely classical in appearance.

Using the usual oscillator units, with time measured in units of ω_0^{-1} and space in units of $\sqrt{\hbar/m\omega_0}$, we are then able to describe the process by two coupled equations for the complex atomic (ψ_a) and molecular (ψ_m) fields,

$$i\frac{\partial \psi_a}{\partial t} = -\frac{\partial^2 \psi_a}{\partial x^2} + V_a(x)\psi_a + (U_{aa}|\psi_a|^2 + U_{am}|\psi_m|^2)\psi_a$$

$$+ i\kappa\psi_a^*\psi_m,$$

$$i\frac{\partial \psi_m}{\partial t} = -\frac{1}{2}\frac{\partial^2 \psi_m}{\partial x^2} + V_m(x)\psi_m$$

$$+ (U_{mm}|\psi_m|^2 + U_{am}|\psi_a|^2 - \Delta)\psi_m - \frac{i}{2}\kappa\psi_a^2. \quad (1)$$

In the above, $V_a(x)[V_m(x)]$ represents the trapping potential for the atomic (molecular) condensate, U_{aa} is the atom-atom interaction strength, U_{mm} represents that between molecules, and U_{am} represents atom-molecule scattering, all in the s-wave δ-function approximation. The coupling strength κ, chosen as real here, represents the Raman laser coupling between atoms and molecules. Δ is the detuning from the Raman resonance. In this model we ignore spontaneous losses and interactions with the thermal cloud, which does not exist at zero temperature.

III. RESULTS

For the purposes of comparison, in all simulations we use as our starting point a ground-state solution of the GPE for a one-dimensional trapped atomic condensate with 2×10^4 atoms and a value of the nonlinear interaction, $U_{aa}=4\times10^{-3}$. This initial solution is obtained via propagation in imaginary time. In all our investigations we use $U_{am}=-1.5U_{aa}$, $U_{mm}=2U_{aa}$, $\kappa=1$, $\Delta=0$, and a molecular trapping potential twice that of the harmonic atomic potential, all consistent with Ref. [24]. The integrations begin with all particles in the atomic condensate. The equations are integrated over 10^4 trajectories, using a standard split-operator method, with momentum propagation in Fourier space and a three-step predictor-corrector method in position space. The accuracy and stability of the integration is checked by keeping track of the conserved quantity, N_a+2N_m, and by varying the time step. Over the times shown, $t=\pi/16$, results with a halved time step were virtually indistinguishable and number was conserved to within less than 0.05%.

To model the quantum states, each of the 1024 points in the spatial grid is given an initial value on each trajectory, chosen from the appropriate Wigner distribution. A coherent state is modeled by taking the (real) GPE solution for the nth spatial point and adding real and imaginary numbers drawn from a normal Gaussian distribution, giving $\psi(x_n)=\psi_{GP}(x_n)+0.5(\eta_1+i\eta_2)/\sqrt{\Delta x}$. It is easily verified that the trajectory average will be $|\psi_{GP}(x_n)|^2+1/(2\Delta x)$ at each point, with $1/(2\Delta x)$ needing to be subtracted at each point after the averaging. A minimum uncertainty squeezed state is modeled by adding $0.5(\eta_1 e^{-r}+i\eta_2 e^r)/\sqrt{\Delta x}$ at each point, where r is the squeezing parameter. A sheared state, typical of Kerr nonlinearities, as in Dunningham $et\ al.$ [16], is simulated by transforming the added squeezed state noise by a factor $\exp(iq\eta_3)$, where q is the shearing factor. The real noise terms have the correlations

$$\overline{\eta_j}=0, \quad \overline{\eta_i\eta_j}=\delta_{ij}. \quad (2)$$

Numerical checks of single-mode distributions produced using these methods show that they give the correct values for average numbers and quadrature variances. In our simulations for squeezed states, we use values of $r=\pm\ln 0.5$, while for the sheared state we use $q=0.005$, which give results similar to the Wigner function shown in Dunningham $et\ al.$ [16]. We also investigate a more extreme shearing, with $r=-\ln 0.2$ and $q=0.05$, as we are treating a larger condensate than those considered in Refs. [16,17], which will possess a larger effective Kerr nonlinearity, giving a more sheared Wigner distribution (which we call a $crescent$ state). The molecular field begins as a coherent vacuum. Although we would have also liked to investigate initial number states, this was not feasible as their Wigner function in terms of Laguerre polynomials [25] becomes incredibly difficult to treat for large numbers.

For purposes of comparison, we begin by numerically integrating the GPE-type equations, which give semiclassical results with the quantum statistics playing no part in the time evolution. What we find is that the spatial dependence of the trapped condensates plays an important role in the process, with the coupling rates at different densities being different. For the parameters used, this causes an interesting structure to emerge, with spatial sidebands forming in the distribu-
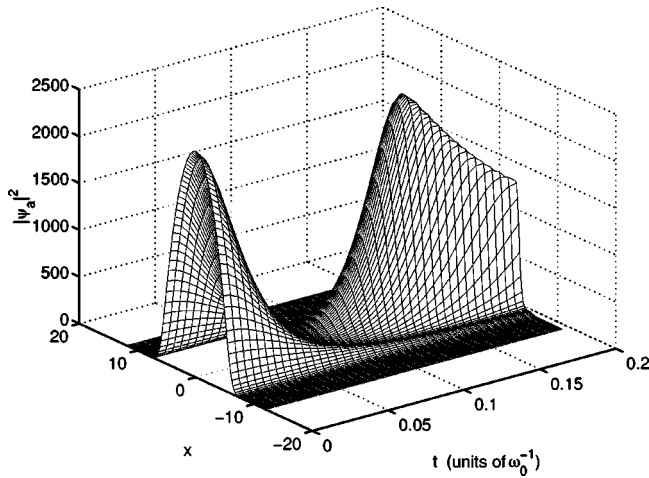
FIG. 1. GPE atomic field prediction up to $t = \pi/16$, showing the spatial dynamics of the atomic BEC, not calculable in a single-mode approach. The units of the spatial axis are $\sqrt{\hbar/m\omega_0}$.

tions, as shown in Figs. 1 and 2. Over the times shown here, the kinetic energy of the condensates plays a negligible role, with integration of spatially separate single-mode equations at each spatial point giving virtually identical predictions, both spatially and for the total particle numbers. This is not the case for longer interaction times.

Examining Figs. 3 and 4, which show the mean particle numbers, we see that when we use an initial coherent state in the Wigner equations, we do not see the dramatic differences from the GPE predictions reported previously [5,6]. The reason is simply that we are working with different parameters, with the ratio between κ and the strength of the nonlinear interactions being important. This was previously demonstrated to be the case in traveling-wave second-harmonic generation, with which, although it is not as rich a system as coupled condensates, an analogy can be made [20]. Initial states with the degree of amplitude squeezing and shearing as calculated in Refs. [16,17] also do not lead to vastly different predictions, the difference between the two being almost negligible. However, a dramatic difference occurs when we consider the initial crescent state, which is greatly
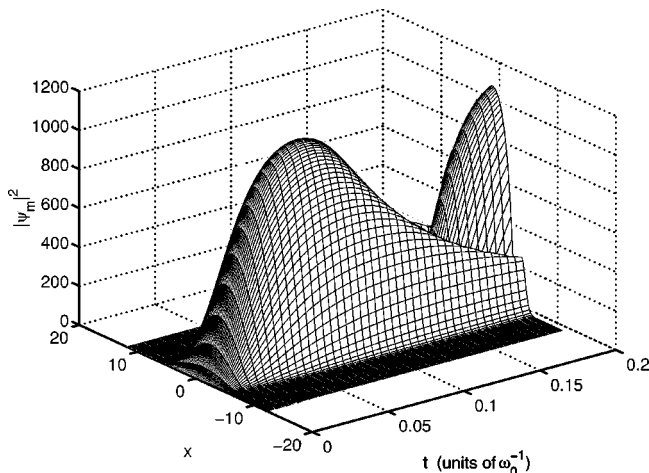

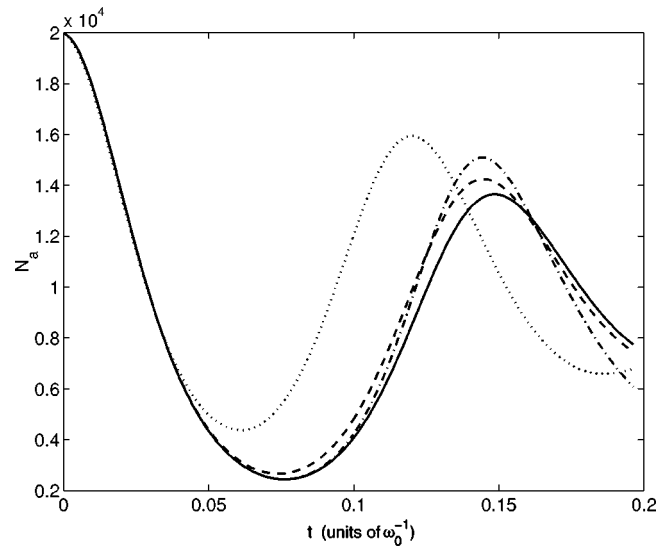FIG. 2. GPE molecular-field prediction up to $t = \pi/16$.

FIG. 3. Atomic population predictions. The dash-dotted line is from the GPE approach, the solid line is for an initial coherent state, the dashed line is the amplitude squeezed state, and the dotted line is the crescent state.

sheared in phase space with a large degree of number squeezing (the single-mode Fano factor for this distribution is ≈ 0.2), but being well above the minimum uncertainty product in the quadratures [single-mode $V(X) \approx 0.6, V(Y) \approx 15$].

The differences seen are not due to the initial spatial intensity correlation, defined using the field operators as

$$g^{(2)}(x,x) = \frac{\langle \psi_a^\dagger(x) \psi_a^\dagger(x) \psi_a(x) \psi_a(x) \rangle}{\langle \psi_a^\dagger(x) \psi_a(x) \rangle^2},\tag{3}$$

which is expected to affect the initial conversion rate [18,26]. This factor varies between 1 and 1.04 at the center for the states considered here, and the initial conversion rate is almost unchanged. The differences come in the first minimum of the atomic population and in subsequent revivals and are
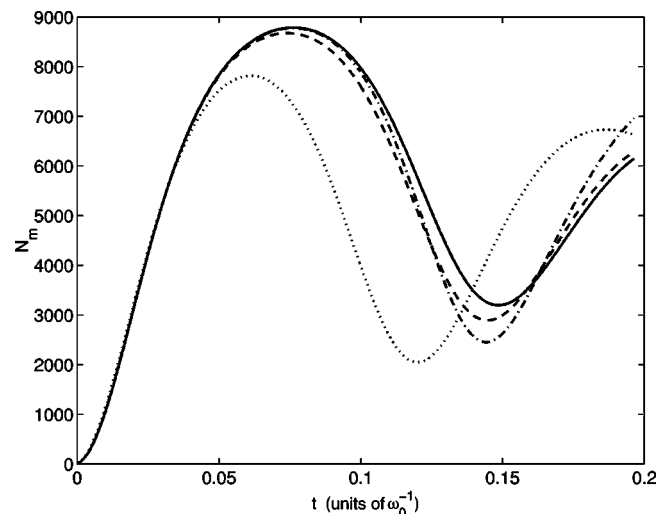

FIG. 4. Molecular population predictions, with lines as in Fig. 3.

more readily explained by the initial degree of phase uncertainty. It can be readily seen by examination of Eq. (1) that whether association or disassociation is predominant will depend on the phase of the products $\psi_a^* \psi_m$ and ψ_a^2. As the crescent state has a larger phase uncertainty than the others considered, the photodisassociation process begins to dominate and the mean number of atoms begins to revive at an earlier time than for the others.

IV. CONCLUSION

We have shown that the quantum state of the initial atomic condensate can play an important role in the mean-field dynamics of photoassociation, even when the GPE predicts the dynamics for an initial coherent state accurately. All the quantum states considered give some difference from the GPE predictions, but the crescent state, possibly the most likely for BEC, is the most dramatic, showing a markedly less complete conversion to molecules. Over the time scales we considered, the quantum statistics are much more important than the spatial dependence of the condensate. These results suggest that, for the purposes of Raman photoassociation, a careful preparation of the initial atomic condensate may be important for the resulting dynamics.

[1] P.D. Drummond, K.V. Kheruntsyan and H. He, Phys. Rev. Lett. **81**, 3055 (1998).

[2] J. Javanainen and M. Mackie, Phys. Rev. A **59**, R3186 (1999).

[3] D.J. Heinzen, R. Wynar, P.D. Drummond, and K.V. Kheruntsyan, Phys. Rev. Lett. **84**, 5029 (2000).

[4] K. Góral, M. Gajda, and K. Rzążewski, Phys. Rev. Lett. **86**, 1397 (2001).

[5] J.J. Hope and M.K. Olsen, Phys. Rev. Lett. **86**, 3220 (2001).

[6] J.J. Hope, Phys. Rev. A **64**, 053608 (2001).

[7] P.D. Drummond and C.W. Gardiner, J. Phys. A **13**, 2353 (1980).

[8] M.J. Steel, M.K. Olsen, L.I. Plimak, P.D. Drummond, S.M. Tan, M.J. Collett, D.F. Walls, and R. Graham, Phys. Rev. A **58**, 4824 (1998).

[9] R. Wynar, R.S. Freeland, D.J. Han, C. Ryu, and D.J. Heinzen, Science **287**, 1016 (2000).

[10] J.M. Gerton, D. Strekalov, I. Prodin, and R.G. Hulet, Nature (London) **408**, 692 (2000).

[11] C. McKenzie *et al.*, Phys. Rev. Lett. **88**, 120403 (2002).

[12] M.R. Andrews, C.G. Townsend, H.J. Meisner, D.S. Durfee, D.M. Kurn, and W. Ketterle, Science **275**, 637 (1997).

[13] W. Ketterle and H.J. Miesner, Phys. Rev. A **56**, 3291 (1997).

[14] J.M. Vogels, J.K. Chin, and W. Ketterle, Phys. Rev. Lett. **90**, 030403 (2003).

[15] M. Lewenstein and L. You, Phys. Rev. Lett. **77**, 3489 (1996).

[16] J.A. Dunningham, M.J. Collett, and D.F. Walls, Phys. Lett. A **245**, 49 (1998).

[17] J. Rogel-Salazar, S. Choi, G.H.C. New, and K. Burnett, Phys. Lett. A **299**, 476 (2002).

[18] Y.R. Shen, Phys. Rev. **155**, 921 (1967).

[19] M.K. Olsen, L.I. Plimak, and A.Z. Khoury, Opt. Commun. **201**, 373 (2002).

[20] V.I. Kruglov and M.K. Olsen, Phys. Rev. A **64**, 053802 (2001).

[21] A. Sinatra, C. Lobo, and Y. Castin, Phys. Rev. Lett. **87**, 210404 (2001).

[22] A. Sinatra, C. Lobo, and Y. Castin, J. Phys. B **35**, 3599 (2002).

[23] L.I. Plimak, M.K. Olsen, M. Fleischhauer, and M.J. Collett, Europhys. Lett. **56**, 372 (2001).

[24] B.J. Cusack, T.J. Alexander, E.A. Ostrovskaya, and Y.S. Kivshar, Phys. Rev. A **65**, 013609 (2002).

[25] C.W. Gardiner, *Quantum Noise* (Springer-Verlag, Berlin, 1991).

[26] K.V. Kheruntsyan, D.M. Gangardt, P.D. Drummond, and G.V. Shlyapnikov, Phys. Rev. Lett. **91**, 040403 (2003).