PHYSICAL REVIEW A **70**, 033611 (2004)

Fock-state dynamics in Raman photoassociation of Bose-Einstein condensates

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(Received 18 December 2003; published 23 September 2004)

By stochastic modeling of the process of Raman photoassociation of Bose-Einstein condensates, we show that, the farther the initial quantum state is from a coherent state, the farther the one-dimensional predictions are from those of the commonly used zero-dimensional approach. We compare the dynamics of condensates, initially in different quantum states, finding that, even when the quantum prediction for an initial coherent state is relatively close to the Gross-Pitaevskii prediction, an initial Fock state gives qualitatively different predictions. We also show that this difference is not present in a single-mode type of model, but that the quantum statistics assume a more important role as the dimensionality of the model is increased. This contrasting behavior in different dimensions, well known with critical phenomena in statistical mechanics, makes itself plainly visible here in a mesoscopic system and is a strong demonstration of the need to consider physically realistic models of interacting condensates.

DOI: 10.1103/PhysRevA.70.033611 PACS number(s): 03.75.Kk, 03.75.Mn, 05.30.Jp

I. INTRODUCTION

In this work, we combine two issues which are of topical interest to the Bose-Einstein condensate (BEC) community and farther demonstrate that the dimensionality of models used to represent trapped condensates can be of crucial importance. The first issue is superchemistry, or Raman photoassociation of condensed atoms to form a molecular condensate [1]. The second issue is the question of the actual quantum state of a trapped condensate, as described by the Wigner function, and whether knowledge of this state may be important for theoretical analyses. The mathematics of photoassociation is essentially a more complex form of that of traveling-wave second-harmonic generation, in which both quantum statistics [2,3] and Kerr nonlinearities [4] are predicted to affect the dynamics of a zero-dimensional model in a quantitative manner. However, we will show that once we consider a one-dimensional model with spatial dependence for interacting condensates, the differences arising due to the quantum statistics are qualitative.

The first prediction of an atom-optical analog of the optical processes of frequency conversion with condensates was by Drummond *et al.* [5], who developed an effective quantum field theory to describe coupled atomic and molecular BECs. Javanainen and Mackie [6] later proposed a two-mode, phenomenological Hamiltonian to model the process of photoassociation. A more complete proposal, using an atomic and two molecular fields, coupled via a two-color Raman transition so as to minimize spontaneous-emission losses, was developed by Heinzen *et al.* [1]. This model, which utilized the Gross-Pitaevskii equation (GPE) approach, demonstrated that the molecular formation would not obey the usual Arrhenius rules for chemical reactions and introduced the name superchemistry. Hope and Olsen in one dimension [7], and Hope in three dimensions [8], subsequently showed that fully quantum treatments using the positive-P representation [9,10] do not always agree with mean-field predictions, even for the mean fields. Recent works by Olsen and Plimak [11] and Olsen [12] have shown that superchemistry not only does not obey the Arrhenius rules, but also depends on the quantum statistics of the reactants.

Experimental efforts to form molecular condensates by photoassociation of atomic condensates have been at least partially successful, although the most successful method for the production of condensed molecules to date has been the recent combination of pairs of fermions using Feshbach techniques [13–16]. Because of Pauli blocking of the dissociation channel, the dynamics of this process, even if it were to be carried out using photoassociative techniques, are expected to be different from those of superchemistry [17]. Photoassociation experiments have been performed using Raman techniques by Heinzen *et al.* [18] and Gerton *et al.* [19], although the atom-molecule oscillations predicted theoretically [1] were not observed. 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 [20]. A number of groups are continuing their efforts and we feel it may not be long before coupled atomic and molecular condensates are formed by photoassociation and can be controlled in the laboratory. This will open new areas for investigation, much as frequency conversion provided for in quantum and nonlinear optics. We note here that photoassociation is not the only possible method which can be used to form coupled atomic and molecular condensates, with Feshbach resonance techniques having been successfully used [21–24], although this technique would not normally be expected to produce the population oscillations predicted for photoassociation superchemistry.

An intriguing issue is raised by the quantum state of a trapped condensate with repulsive interatomic interactions, which is perhaps best described in terms of the Wigner function [25]. This function has long been a useful tool in quantum optics and has a simple definition in the single-mode case,

$$W(\alpha) = \frac{2}{\pi^2}\int_{-\infty}^{\infty} \mathrm{d}^2\beta \langle \alpha + \beta|\rho|\alpha - \beta\rangle \exp(\alpha^*\beta - \alpha\beta^*), \quad (1)$$

which is useful if the density matrix is known. A condensate, however, is not a single mode and, even in the case of a noninteracting condensate, the dimensionality of the Hilbert space required to describe the density matrix makes this approach intractable. Another approach is to solve for a steady-state solution of the appropriate Fokker-Planck equation, which can sometimes be done in terms of a potential solution [26]. In the case of a trapped condensate, the equation of motion for the Wigner function is a generalized Fokker-Planck equation with third-order derivatives and spatial dependence, so that the steady-state solution is not easily found [10]. In the absence of a complete solution for the Wigner function, obtained without making various approximations, researchers have often chosen to use either the coherent or Fock states. In previous works [11,12], the photoassociation dynamics for different initial states were stochastically simulated, but the Fock, or number state, was not considered due to difficulties in simulating its Wigner function numerically. (For a short discussion of what we consider to be the relative merits of different choices, see Refs. [11,12].) Here we adapt an approximation developed by Gardiner *et al.* [27], which allows us to approximately represent Fock states without having to deal with negative pseudoprobabilities. As we investigate only the dynamics of the mean fields rather than quantum correlations, we will stochastically integrate the appropriate equations in the truncated Wigner representation [10,28,29], which we expect to give reliable results for the numbers of particles involved. For the parameters used in Ref. [12], where it was shown that the superchemistry-type oscillations can eventually disappear due solely to the quantum noise, without any thermal component being present, we will show here that, for an initial Fock atomic state, the oscillations may not even appear.

II. QUANTUM STATES IN THE WIGNER REPRESENTATION

Due to the difficulty of solving the equations of motion analytically, all our solutions will be obtained numerically, as averages over a large number of stochastic trajectories. When we wish to calculate the time evolution for a particular initial quantum state, we must begin by representing the Wigner distribution for the chosen state. To model these, each of the 512 points on the spatial grid is given an initial value on each trajectory, chosen from the appropriate Wigner distribution. Coherent states and the crescent state, given this name because its Wigner contours are sheared in phase space due to the $\chi^{(3)}$ nonlinearity, are modeled as explained in the appendix of Ref. [12]. We note here that, unlike in the single-mode zero-dimensional models used in quantum optics, there is some freedom in the way these initial states can be modeled once spatial dependence is included. For the purposes of simple comparisons, we will use the same method as in Refs. [11,12]. The Wigner function for the Fock state $|N\rangle$ is

$$W_N(\alpha,\alpha^*) = 2\frac{(-1)^N}{\pi}\exp(-2|\alpha|^2)L_N(4|\alpha|^2), \quad (2)$$

where L_N is the Laguerre polynomial of order N. This distribution is oscillatory and can obviously be either positive or negative, so it cannot be easily simulated numerically. However, in the large-N regime Gardiner has made the observation [27] that the cumulative distribution behaves very much like a step function centered at $|\alpha|^2 = N$. This distribution can then be approximated by a Gaussian which gives the right moments for the mean and variance and approximates the higher moments well. The appropriate distribution is

$$P_N(n,\theta) = \sqrt{\frac{2}{\pi}}\exp\left(-\frac{(n-N-1/2)^2}{2(1/4)}\right), \quad (3)$$

where we have taken $\alpha = \sqrt{n}e^{i\theta}$, with θ uniform on $[0,2\pi)$. The first three moments of this distribution are

$$\overline{\alpha^*\alpha} = N + \frac{1}{2}, \quad (4)$$

$$\overline{\alpha^{*2}\alpha^2} = \left(N+\frac{1}{2}\right)^2 + \frac{1}{4}, \quad (5)$$

$$\overline{\alpha^{*3}\alpha^3} = \left(N+\frac{1}{2}\right)^3 + \frac{3}{4}\left(N+\frac{1}{2}\right), \quad (6)$$

so that mean and variance are in agreement with Eq. (2). We can now show that such an approximation in fact generates all moments of Eq. (2), up to a correction of order $1/N^2$, which is negligible for large N.

Using the differential recursion relation for the Laguerre polynomials,

$$x\frac{d}{dx}L_N(x) = N[L_N(x) - L_{N-1}(x)], \quad (7)$$

a recursion relation for arbitrary even moments can be found. Writing

$$(\overline{\alpha^{*m}\alpha^m})_N = 2\frac{(-1)^N}{\pi}\int d^2\alpha \exp(-2|\alpha|^2)L_N(4|\alpha|^2)|\alpha|^{2m}, \quad (8)$$

we can use Eq. (7) to find

$$(\overline{\alpha^{*m}\alpha^{m}})_N = \frac{N+m}{2}(\overline{\alpha^{*m-1}\alpha^{m-1}})_N + \frac{N}{2}(\overline{\alpha^{*m-1}\alpha^{m-1}})_{N-1}. \tag{9}$$

The first three moments are

$$(\overline{\alpha^{*}\alpha})_N = N + 1/2, \tag{10}$$

$$(\overline{\alpha^{*2}\alpha^{2}})_N = (N+1/2)^2 + 1/4, \tag{11}$$

$$(\overline{\alpha^{*3}\alpha^{3}})_N = (N+1/2)^3 + \frac{5}{4}(N+1/2), \tag{12}$$

and comparison with Eq. (4) shows that the Gaussian approximation is exact for the mean and variance, and accurate to $O(1/N^2)$ for $m=3$. It is easily shown by induction on m that the exact moments satisfy

$$(\overline{\alpha^{*m}\alpha^{m}})_N = (N+1/2)^m + O(N^{m-2}), \tag{13}$$

so that the correction is always of order $1/N^2$ relative to the leading term. Returning to the Gaussian approximation, we see that it gives

$$\begin{aligned}\overline{\alpha^{*m}\alpha^{m}} &= \sqrt{\frac{2}{\pi}}\int_{-\infty}^{\infty} dz(z+N+1/2)^m \exp(-2z^2)\\ &= \sqrt{\frac{2}{\pi}}\int_{-\infty}^{\infty} dz[(N+1/2)^m + m(N+1/2)^{m-1}z\\ &\quad + O(N^{m-2})]\exp(-2z^2)\\ &= (N+1/2)^m + O(N^{m-2}),\end{aligned} \tag{14}$$

which will be an adequate description of the number state statistics for large N.

To simulate this distribution numerically, consider the choice (using a single mode for simplicity)

$$\alpha_i = p + q\eta_i, \tag{15}$$

where η_i is a normal Gaussian random variable, and p and q are yet to be determined. As we are using a Gaussian approximation, it is sufficient to reproduce the first two moments of α^2. (Note that α is a real variable here, with the phase distribution to be added.) We need to reproduce $\overline{\alpha^2}=N+1/2$ and $\overline{\alpha^4}=(N+1/2)^2+1/4$. The choices

$$p = \frac{1}{2}(2N+1+2\sqrt{N^2+N})^{1/2} \tag{16}$$

and

$$q = \frac{1}{4p} \tag{17}$$

reproduce the required distribution to a high degree of accuracy. The α_i thus chosen is then multiplied by the factor $\exp(2i\pi\xi_i)$, where ξ is randomly chosen from the uniform distribution $[0,1)$. The molecular field is always chosen to be in a coherent vacuum state at the beginning of the photoassociation process.

After stochastic integration, the trajectory averages will be $|\psi_{\mathrm{GP}}(x_n)|^2 + 1/(2\Delta x)$ at each point, with $1/(2\Delta x)$ needing to be subtracted at each point after the averaging. $\psi_{\mathrm{GP}}(x_n)$ is the ground-state solution of the GPE at spatial point x_n, with the mean number of atoms at each spatial point being $\Delta x|\psi_{\mathrm{GP}}(x_n)|^2$, so that when the summation is performed, the Wigner average is this plus 1/2. In the continuous limit, with an infinite number of spatial points, this causes a divergence in the Wigner equations, as the "vacuum" noise becomes infinite. In this sense, only the normally ordered P representations truly exist for continuous fields. However, as the s-wave scattering length imposes a physical limit on the grid size which may be used, we are able to use the Wigner representation. This ultraviolet divergence does not exist for single-mode treatments, as commonly used in quantum optics. However, as we shall demonstrate, the effects of the quantum noise change with the dimensionality, meaning that single-mode-type approaches to the dynamics of interacting condensates may not always be valid.

III. THE SYSTEM AND EQUATIONS OF MOTION

The system we treat is the same as in previous works [11,12], where we again treat the trapped zero-temperature atomic condensate as one-dimensional. We consider a Raman photoassociation scheme [1,7,8] with the excited molecular field adiabatically eliminated. Following the same procedure as detailed in Ref. [12], we map the Hamiltonian onto differential equations which have the appearance of coupled Gross-Pitaevskii-type equations. These 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 desired Wigner function. We can now model the quantum fields (to a good level of approximation) via equations which are completely classical in appearance. (Note that a full version of the derivation of these equations is given in Ref. [12].) Using the usual oscillator units, with time measured in units of ω_0^{-1} and space in units of $\sqrt{\hbar/m\omega_0}$, the two coupled equations for the complex atomic (ψ_a) and molecular (ψ_m) fields are

$$\begin{aligned}i\frac{\partial\psi_a}{\partial t} &= -\frac{\partial^2\psi_a}{\partial x^2} + V_a(x)\psi_a + (U_{aa}|\psi_a|^2 + U_{am}|\psi_m|^2)\psi_a\\ &\quad + i\kappa\psi_a^*\psi_m,\\ i\frac{\partial\psi_m}{\partial t} &= -\frac{1}{2}\frac{\partial^2\psi_m}{\partial x^2} + V_m(x)\psi_m + (U_{mm}|\psi_m|^2 + U_{am}|\psi_a|^2 - \Delta)\psi_m\\ &\quad - \frac{i}{2}\kappa\psi_a^2.\end{aligned} \tag{18}$$

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 be-

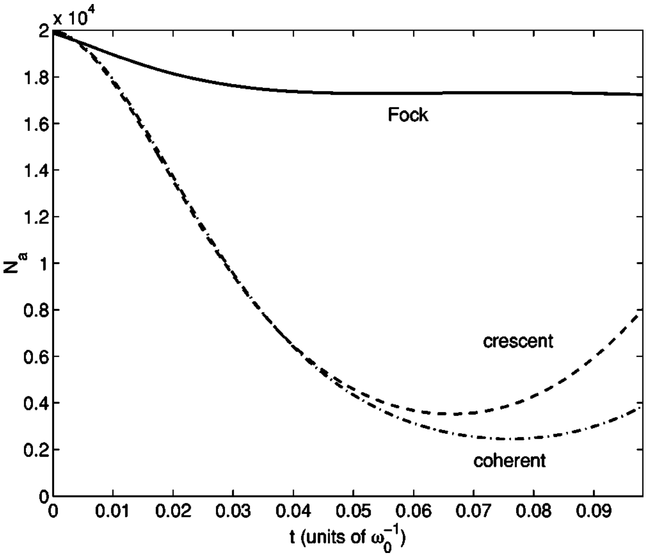

FIG. 1. Atom number evolution for Fock, crescent, and coherent initial states.

tween 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.

IV. RESULTS

In all simulations, we use as our starting point a ground-state solution of the GPE with 2×10^4 atoms and a value of the nonlinear interaction, $U_{aa}=4\times10^{-3}$. 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. Physically, the one-dimensional value chosen for U_{aa} is at the low end of the range, representing either a three-dimensional scattering length smaller than that of sodium, or a weak trap. We have chosen this value because, given the value of κ, the differences from the GPE predictions happen after a somewhat short integration time, even for initial coherent states. (For the importance of the ratio of $\chi^{(2)}$ to $\chi^{(3)}$ nonlinearities in the quantum dynamics, at least in the zero-dimensional case, see Ref. [4]). We note that scattering lengths can be tuned using Feshbach resonance techniques. The coupling parameter κ depends on both laser intensities and the one-photon detuning from the excited molecular levels, as well as on the overlap integrals of the states involved in the transition [1]. Using the same procedure as Kheruntsyan and Drummond [30], we find that the value of κ used here is comparable to the one used in that work, the value of the coupling equating to approximately 2.1×10^{-7} m$^{1/2}$ s^{-1}. Although we have not attempted to model an actual experiment, with all the complications involved in the process, we can have some confidence that our parameters are not physically unattainable.

The integrations begin with all particles in the atomic condensate. The equations are integrated over 10^4 trajectories, using a standard split-operator method, as described in Ref. [12]. In Fig. 1, we show the results for the atomic numbers, using initial Fock, crescent, and coherent states, all with the same initial mean numbers of atoms. Immediately obvious is that, for these parameters, the dynamics for the Fock state are completely different from the other two. Not only is the initial conversion rate less, but the degree of conversion to molecules is much less, and there are no oscillations seen between atoms and molecules. The different dynamics of the Fock state cannot be ascribed to the initial spatial intensity correlation, $g^{(2)}(x,x)$ (see Refs. [12,31]), as this between $1-1/N$ for a Fock state, 1 for a coherent state, and 1.04 at the center for the crescent state. As demonstrated previously [11,12], the more uncertainty in phase that a given state has by comparison with a coherent state, the more difference we see in the dynamics. A Fock state, which exhibits the maximum possible phase uncertainty of 2π, is therefore expected to differ most in its dynamics from the coherent state.

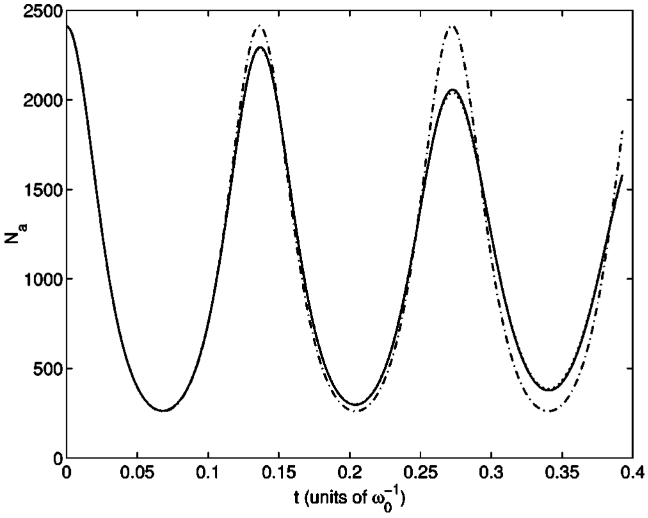

FIG. 2. Zero-dimensional predictions for the atomic evolution. The dash-dotted line represents the classical mean-field prediction, the solid line is for an initial Fock state (8.25×10^5 trajectories), and the dotted line is for an initial coherent state, as previously shown in Ref. [12] (3.45×10^5 trajectories), which is almost indistinguishable from the Fock state.

V. ZERO-DIMENSIONAL APPROACH

By comparison with a zero-dimensional quantum-optics-type approach, sometimes used to represent Raman photoassociation of atomic condensates [32], we can show that the quantum statistics become more important as the dimension increases. We can investigate the zero-dimensional system with the coupled equations given in Ref. [12]. We show the results for the atomic dynamics in Fig. 2, comparing the predictions of the truncated Wigner with initial coherent and Fock states to those of the classical approach, both for an initial atom number equal to $|\psi_a|^2$ at the center of the densities used for the harmonic trap. Note that this is not the same as the atomic number at the center of the one-dimensional grid, which is $\Delta x|\psi_a|^2$, but is the number which enters into the one-dimensional equations. We find that the classical approach, which predicts regular periodic behavior in this case, is reasonably accurate up to the second oscillation, but then begins to differ from the quantum prediction. The quantum result shows a damping of the oscillations, due solely to the quantum noise. However, when compared with the one-dimensional predictions, the results for the Fock state are completely different. Although the difference increases in one dimension for the coherent state, it is only quantitative. For a Fock state, the difference with dimension is qualitative and the zero-dimensional predictions, as used in Refs. [33,34] with initial Fock states, are far from the 1D results. We note here that, while there are certain physical conditions to be fulfilled so that a trapped BEC may be effectively considered as one-dimensional (see, for example, Ref. [31]), we are not aware of any physical conditions which would allow for a zero-dimensional model of the photoassociation process as treated here.

VI. CONCLUSION

Using the truncated Wigner representation and a Gaussian approximation, we have shown that the photoassociation dynamics for an initial Fock state of a trapped condensate can be completely different from those of other commonly considered quantum states. We have demonstrated that, to see this difference theoretically, it is necessary to use at least a one-dimensional model, with the differences not being apparent in a zero-dimensional treatment. In zero dimensions, for the parameters used in our simulations, we still find giant oscillations between the atomic and molecular populations for all three quantum states considered here. In one dimension, we do not see a single oscillation for the Fock state, with the dynamics being noticeably different from the time the interaction begins. This is a clear example of the importance of the underlying dimensionality, which has long been appreciated in critical phenomena and is now shown to play a role in the quantum dynamics of interacting atomic and molecular condensates.

ACKNOWLEDGMENTS

This research was supported by the New Zealand Foundation for Research, Science and Technology (Contracts No. UFRJ0001 and No. VUWX03014), the Australian Research Council, the Instituto do Milênio de Informação Quântica, CNPq (Conselho Nacional de Desenvolvimento Científico e Tecnológico), PRONEX (Programa de Apoio a Núcleos de Excelência), and FAPEAL (Fundação de Amparo à Pesquisa do Estado de Alagoas).

[1] D. J. Heinzen, R. Wynar, P. D. Drummond, and K. V. Kheruntsyan, Phys. Rev. Lett. **84**, 5029 (2000).
[2] Y. R. Shen, Phys. Rev. **155**, 921 (1967).
[3] M. K. Olsen, L. I. Plimak, and A. Z. Khoury, Opt. Commun. **201**, 373 (2002).
[4] V. I. Kruglov and M. K. Olsen, Phys. Rev. A **64**, 053802 (2001).
[5] P. D. Drummond, K. V. Kheruntsyan, and H. He, Phys. Rev. Lett. **81**, 3055 (1998).
[6] J. Javanainen and M. Mackie, Phys. Rev. A **59**, R3186 (1999).
[7] J. J. Hope and M. K. Olsen, Phys. Rev. Lett. **86**, 3220 (2001).
[8] J. J. Hope, Phys. Rev. A **64**, 053608 (2001).
[9] P. D. Drummond and C. W. Gardiner, J. Phys. A **13**, 2353 (1980).
[10] 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).
[11] M. K. Olsen and L. I. Plimak, Phys. Rev. A **68**, 031603 (2003).
[12] M. K. Olsen, Phys. Rev. A **69**, 013601 (2004).
[13] M. Greiner, C. A. Regal, and D. S. Gin, Nature (London) **426**, 537 (2003).
[14] J. Cubizolles, T. Bourdel, S. J. J. M. F. Kokkelmans, G. V. Shlyapnikov, and C. Salomon, Phys. Rev. Lett. **91**, 240401 (2003).
[15] M. W. Zwierlein, C. A. Stan, C. H. Schunck, S. M. F. Raupach, S. Gupta, Z. Hadzibabic, and W. Ketterle, Phys. Rev. Lett. **91**, 250401 (2003).
[16] K. E. Strecker, G. B. Partridge, and R. G. Hulet, Phys. Rev. Lett. **91**, 080406 (2003).
[17] J. F. Corney and P. D. Drummond (unpublished).
[18] R. Wynar, R. S. Freeland, D. J. Han, C. Ryu, and D. J. Heinzen, Science **287**, 1016 (2000).
[19] J. M. Gerton, D. Strekalov, I. Prodin, and R. G. Hulet, Nature (London) **408**, 692 (2000).
[20] C. McKenzie *et al.*, Phys. Rev. Lett. **88**, 120403 (2002).
[21] E. A. Donley, N. R. Claussen, S. T. Thompson, and C. E. Wieman, Nature (London) **417**, 529 (2002).
[22] J. Herbig, T. Kraemer, M. Mark, T. Weber, C. Chin, H.-C. Nägerl, and R. Grimm, Science **301**, 1510 (2003).
[23] K. Xu, T. Mukaiyama, J. R. Abo-Shaeer, J. K. Chin, D. E. Miller, and W. Ketterle, Phys. Rev. Lett. **91**, 210402 (2003).
[24] S. Dürr, T. Volz, A. Marte, and G. Rempe, Phys. Rev. Lett. **92**, 020406 (2004).
[25] E. P. Wigner, Phys. Rev. **40**, 749 (1932).
[26] D. F. Walls and G. J. Milburn, *Quantum Optics* (Springer-Verlag, Berlin, 1995).
[27] C. W. Gardiner, J. R. Anglin, and T. I. A. Fudge, J. Phys. B **35**, 1555 (2002).
[28] A. Sinatra, C. Lobo, and Y. Castin, Phys. Rev. Lett. **87**, 210404 (2001).
[29] A. Sinatra, C. Lobo, and Y. Castin, J. Phys. B **35**, 3599 (2002).
[30] K. V. Kheruntsyan and P. D. Drummond, Phys. Rev. A **66**, 031602 (2002).
[31] K. V. Kheruntsyan, D. M. Gangardt, P. D. Drummond, and G. V. Shlyapnikov, Phys. Rev. Lett. **91**, 040403 (2003).
[32] M. Koštrun, M. Mackie, R. Côté, and J. Javanainen, Phys. Rev. A **62**, 063616 (2000).
[33] J. Calsamiglia, M. Mackie, and K.-A. Suominen, Phys. Rev. Lett. **87**, 160403 (2001).
[34] O. Dannenberg, M. Mackie, and K.-A. Suominen, Phys. Rev. Lett. **91**, 210404 (2003).