

Quantum theory of atomic four-wave mixing in Bose-Einstein condensates

Elena V. Goldstein and Pierre Meystre

Optical Sciences Center, University of Arizona, Tucson, Arizona 85721

(Received 12 November 1998)

We present an exact quantum-mechanical analysis of collinear four-wave mixing in a multicomponent Bose-Einstein condensate consisting of sodium atoms in the $F=1$ ground state. Technically, this is achieved by taking advantage of the conservation laws of the system to cast its Hamiltonian in terms of angular momentum operators. We discuss explicitly the build-up of matter-wave side modes from quantum fluctuations, as well as the correlations between these modes. We show the appearance of a strong quantum entanglement between hyperfine states. We also demonstrate that for finite atomic numbers, the system exhibits periodic collapses and revivals in the exchange of atoms between different spin states.

[S1050-2947(99)09705-X]

PACS number(s): 03.75.Fi, 05.30.Jp, 42.65.Hw

I. INTRODUCTION

Low-density Bose-Einstein condensates of alkali-metal atoms are described to an excellent degree of approximation by a Gross-Pitaevskii nonlinear Schrödinger equation. Such equations are ubiquitous in many fields of physics, including nonlinear optics, hence it is not surprising that many of the concepts first developed in optics can readily be extended to condensates. A number of theoretical investigations along these lines have already been presented, including the study of matter-wave solitons [1], phase conjugation [2,3], four-wave mixing [4], spin-mixing [5], etc. The experimental verification of these predictions is also underway, with the first announcement of four-wave mixing in a sodium condensate [6].

A recent experimental development of considerable importance in this context is the demonstration of multicomponent condensates, in particular of ^{23}Na condensates in far-off-resonance optical dipole traps [7]. These systems permit us to distinguish condensate modes in ways other than by their center-of-mass quantum numbers. This opens up new directions of research, such as the study of the stability and miscibility of quantum fluids, the analysis of pattern formation, the generation of ferromagnetic and antiferromagnetic states, etc. [8–10,5,6]. In the context of nonlinear atom optics, the coexistence of condensates with different magnetic quantum numbers is attractive in that it provides a way to perform four-wave mixing experiments in collinear geometries, with the considerable advantage of eliminating phase-matching limitations [3].

It is now well understood that the matter-wave analog of nonlinear optical interactions is provided by collisions. In particular, in the s -wave scattering regime (“shapeless” approximation) two-body collisions are mathematically equivalent to a medium with third-order nonlinear polarization (Kerr medium) and characterized by local and instantaneous response. Hence, the kinds of wave-mixing phenomena that can take place in a condensate are largely dictated by the properties of these collisions. For example, recent experimental studies of spin population dynamics in a ^{23}Na condensate have shown that as a result of spin changing collisions, a sample initially in the $m_F=0$ state eventually winds

up with almost equal populations of all hyperfine ground states [10]. The present paper exploits these collisions to generate four-wave mixing in ^{23}Na . It is organized as follows. Section II describes our physical model, which considers degenerate backward phase conjugation in (small) condensates. Section III derives a representation of the problem in terms of angular momentum algebra. This leads to an exact diagonalization of the four-wave mixing Hamiltonian and the determination of all eigenstates and eigenenergies. These results are applied in Sec. IV to the analysis of the dynamics of exchange of population between different spin states. The quantum correlations between these states are also discussed. Finally, Sec. V is a summary and outlook.

The phase conjugation of atomic waves has previously been discussed in a situation where the matter-wave modes involve the same electronic state but different center-of-mass components [2], and using the undepleted pump approximation [11]. Mode separation was achieved via Bragg scattering off an optical field. A full three-dimensional numerical solution of a closely related situation was recently given [4] in connection with experimental work at NIST [6]. Both of these analyses rely on the Gross-Pitaevskii equation, which effectively assumes a Hartree ansatz with a fixed number of particles, or spontaneous symmetry breaking. In addition, a recent analysis of matter-wave phase conjugation treats the central mode to all orders, but describes the side modes in a linearized fashion [3]. In contrast, the present theory, which also holds for small condensates, does not make any of these assumptions. It handles all modes on equal footing, to all orders, and makes no assumption about their statistical properties. We note that our approach is related to a recent analysis of spin mixing in spinor Bose-Einstein condensates [5], where the ground state of the system is determined by diagonalizing the system Hamiltonian expressed in terms of angular momentum operators.

II. PHYSICAL MODEL

We consider a Bose-Einstein condensate of ^{23}Na atoms in the $F=1$ hyperfine ground state, with the three internal atomic states $|F=1, m_F=-1\rangle$, $|F=1, m_F=0\rangle$, and $|F=1, m_F=1\rangle$ of degenerate energies in the absence of external

magnetic fields. The condensate is confined by a far-off-resonance optical dipole trap. It is described by the three-component vector Schrödinger field

$$\mathbf{\Psi}(\mathbf{r}, t) = \{\Psi_{-1}(\mathbf{r}, t), \Psi_0(\mathbf{r}, t), \Psi_1(\mathbf{r}, t)\}, \quad (1)$$

which satisfies the bosonic commutation relations

$$[\Psi_i(\mathbf{r}, t), \Psi_j^\dagger(\mathbf{r}', t)] = \delta_{ij} \delta(\mathbf{r} - \mathbf{r}'). \quad (2)$$

Accounting for the possibility of two-body collisions, its dynamics is described by the second-quantized Hamiltonian

$$\begin{aligned} \mathcal{H} = & \int d\mathbf{r} \mathbf{\Psi}^\dagger(\mathbf{r}, t) H_0 \mathbf{\Psi}(\mathbf{r}, t) \\ & + \frac{1}{2} \int d\mathbf{r}_1 d\mathbf{r}_2 \mathbf{\Psi}^\dagger(\mathbf{r}_1, t) \mathbf{\Psi}^\dagger(\mathbf{r}_2, t) \\ & \times V(\mathbf{r}_1 - \mathbf{r}_2) \mathbf{\Psi}(\mathbf{r}_2, t) \mathbf{\Psi}(\mathbf{r}_1, t), \end{aligned} \quad (3)$$

where the single-particle Hamiltonian is given by

$$H_0 = \mathbf{p}^2/2M + V_{\text{trap}} \quad (4)$$

and the trap potential is of the general form

$$V_{\text{trap}} = \sum_{m=-1}^{+1} U(\mathbf{r}) |F=1, m\rangle \langle F=1, m|. \quad (5)$$

Here \mathbf{p} is the center-of-mass momentum of the atoms of mass M , and $U(\mathbf{r})$, the effective dipole trap potential for atoms in the $|1, m\rangle$ hyperfine state, is independent of m for a nonmagnetic trap.

The general form of the two-body interaction $V(\mathbf{r}_1 - \mathbf{r}_2)$ has been discussed in detail in Refs. [8,9,12]. We reproduce its main features for the sake of clarity. Considering exclusively situations where the hyperfine spin $F_i=1$ of the individual atoms is preserved, we label the hyperfine states of the combined system of two collision partners with total hyperfine spin $\mathbf{F} = \mathbf{F}_1 + \mathbf{F}_2$ by $|f, m\rangle$, where $f=0, 1, 2$ and $m = -f, \dots, f$. In the shapeless approximation, it can be shown that the two-body interaction is of the general form [9]

$$V(\mathbf{r}_1 - \mathbf{r}_2) = \delta(\mathbf{r}_1 - \mathbf{r}_2) \sum_{f=0}^2 \hbar g_f \mathcal{P}_f, \quad (6)$$

where

$$g_f = 4\pi\hbar a_f/M \quad (7)$$

and

$$\mathcal{P}_f \equiv \sum_m |f, m\rangle \langle f, m| \quad (8)$$

is the operator which projects the state of the atomic pair onto a state of total hyperfine quantum number f . Here a_f is the s -wave scattering length for the channel of total hyperfine spin f . As a result of the symmetry requirement for bosonic atoms, it can be shown that only states with even f contribute to $V(\mathbf{r}_1 - \mathbf{r}_2)$, so that

$$\begin{aligned} V(\mathbf{r}_1 - \mathbf{r}_2) &= \hbar \delta(\mathbf{r}_1 - \mathbf{r}_2) (g_2 \mathcal{P}_2 + g_0 \mathcal{P}_0) \\ &= \hbar \delta(\mathbf{r}_1 - \mathbf{r}_2) (c_0 + c_2 \mathbf{F}_1 \cdot \mathbf{F}_2). \end{aligned} \quad (9)$$

In this expression,

$$c_0 = (g_0 + 2g_2)/3, \quad (10)$$

$$c_2 = (g_2 - g_0)/3,$$

which follows from the identities $\mathcal{P}_1 + \mathcal{P}_2 = \hat{I}$ and $\mathbf{F}_1 \cdot \mathbf{F}_2 = \mathcal{P}_2 - 2\mathcal{P}_0$. Substituting this form of the two-body potential $V(\mathbf{r}_1 - \mathbf{r}_2)$ into the second-quantized Hamiltonian (3) leads to

$$\begin{aligned} \mathcal{H} = & \sum_m \int d\mathbf{r} \mathbf{\Psi}_m^\dagger(\mathbf{r}, t) \left[\frac{\mathbf{p}^2}{2M} + U(\mathbf{r}) \right] \mathbf{\Psi}_m(\mathbf{r}, t) \\ & + \frac{\hbar}{2} \int d\mathbf{r} \{ (c_0 + c_2) [\Psi_1^\dagger \Psi_1^\dagger \Psi_1 \Psi_1 \\ & + \Psi_{-1}^\dagger \Psi_{-1}^\dagger \Psi_{-1} \Psi_{-1} + 2\Psi_0^\dagger \Psi_0 (\Psi_1^\dagger \Psi_1 + \Psi_{-1}^\dagger \Psi_{-1})] \\ & + c_0 \Psi_0^\dagger \Psi_0^\dagger \Psi_0 \Psi_0 + 2(c_0 - c_2) \Psi_1^\dagger \Psi_1 \Psi_{-1}^\dagger \Psi_{-1} \\ & + 2c_2 (\Psi_1^\dagger \Psi_{-1}^\dagger \Psi_0 \Psi_0 + \text{H.c.}) \}. \end{aligned} \quad (11)$$

The physical interpretation of the various terms of this Hamiltonian has been discussed previously: The three terms quartic in one of the field operators, i.e., those of the form $\Psi_i^\dagger \Psi_i^\dagger \Psi_i \Psi_i$, are self-(de)focusing terms, the terms involving two hyperfine states conserve the populations of the individual spin states and merely lead to phase shifts, and the terms involving the central mode Ψ_0 and *both* side modes correspond to spin-exchange collisions. This ‘‘four-wave mixing’’ interaction, involving, e.g., the annihilation of a pair of atoms with $m_F=0$ and the creation of two atoms in the states $m_F = \pm 1$, leads to phase conjugation in quantum optics and to matter-wave phase conjugation in the present case [3].

The analogy with optical phase conjugation via four-wave mixing becomes even more apparent when we consider a situation where atoms in the $m_F=0$ state are placed in a linear superposition of two counterpropagating center-of-mass modes of momenta $\pm \hbar k_0$, that is, the field operator $\Psi_0(x)$ can be expanded as

$$\Psi_0(x) = \frac{1}{\sqrt{V}} (e^{ik_0x} a_{01} + e^{-ik_0x} a_{02}), \quad (12)$$

while the atoms of spin $m_F = \pm 1$ are taken to be at rest,

$$\Psi_{\pm 1}(x) = \frac{1}{\sqrt{V}} a_{\pm 1}. \quad (13)$$

Here a_{01} and a_{02} are the annihilation operators of the two counterpropagating $m_F=0$ modes, with $[a_{0i}, a_{0j}^\dagger] = \delta_{ij}$, $i, j = 1$ or 2 , and a_{\pm} are the annihilation operators for the modes associated with $m_F = \pm 1$. Finally, V is the confinement volume of the condensate. Inserting this mode expansion into

the Hamiltonian (11) and ignoring all non-phase-matched contributions finally yields the four-wave mixing Hamiltonian

$$\begin{aligned} \mathcal{H} = & \frac{\hbar^2 k_0^2}{2M} \hat{N} + \frac{\hbar c_0}{2} \hat{N}(\hat{N}-1) \\ & + \frac{\hbar c_2}{2} [a_1^\dagger a_1^\dagger a_1 a_1 + a_{-1}^\dagger a_{-1}^\dagger a_{-1} a_{-1} - 2a_1^\dagger a_1 a_{-1}^\dagger a_{-1} \\ & + 2(a_1^\dagger a_1 + a_{-1}^\dagger a_{-1})(a_{01}^\dagger a_{01} + a_{02}^\dagger a_{02}) \\ & + 4a_1^\dagger a_{-1}^\dagger a_{01} a_{02} + 4a_{1-1} a_{01}^\dagger a_{02}^\dagger], \end{aligned} \quad (14)$$

where we have introduced the total number of atoms

$$\hat{N} = a_1^\dagger a_1 + a_{-1}^\dagger a_{-1} + a_{01}^\dagger a_{01} + a_{02}^\dagger a_{02}. \quad (15)$$

Note that the coupling between the central and side modes of the condensate ($a_1^\dagger a_{-1}^\dagger a_{01} a_{02}$ and its Hermitian conjugate) requires both energy and momentum conservation. Energy conservation can be achieved, e.g., by introducing an external dc magnetic field.¹

III. ANGULAR MOMENTUM REPRESENTATION

Despite the formal analogy between the Hamiltonians describing optical and atomic four-wave mixing, there are important details in which these processes differ. One of them is phase matching, which is normally more difficult to achieve with de Broglie waves due to the quadratic dispersion relation of atoms. However, the use of multicomponent condensates permits us to avoid this difficulty, as we have just seen. More important perhaps is the fact that nonlinear optics experiments usually use coherent laser light as pumps. It is well known that the number of photons in these fields is not well determined. In contrast, the number of atoms in a condensate is a fixed (and integer) quantity in the absence of losses. This leads to important conservation laws, which we exploit in the following to reexpress the Hamiltonian (14) in terms of angular momentum operators. This leads in turn to an exact solution of the problem.

The most obvious conserved quantity of the Hamiltonian (15) is the total number of atoms, as follows from the commutator

$$[\hat{N}, \mathcal{H}] = 0. \quad (16)$$

In addition, the population *differences*

$$\hat{D}_0 = a_{01}^\dagger a_{01} - a_{02}^\dagger a_{02}, \quad (17)$$

¹In this case, the energy of atoms in the $m_F=0$ state is not affected by the magnetic field and is determined solely by their kinetic energy. In contrast, the energy of atoms in the hyperfine states $m_F = \pm 1$ changes due to the quadratic Zeeman effect. For the fields used in Ref. [10] the $m_F = -1 \leftrightarrow m_F = 0$ and $m_F = +1 \leftrightarrow m_F = 0$ transitions differ in frequency by 0.9 MHz, which corresponds to an energy of 10^{-28} J. This leads to an energy excess in the side modes which can be used to compensate the mismatch in kinetic energies.

$$\hat{D} = a_1^\dagger a_1 - a_{-1}^\dagger a_{-1} \quad (18)$$

are also conserved. These two conservation laws follow directly from the fact that the Hamiltonian (14) describes the creation and annihilation of bosonic atoms in pairs. For example, the annihilation of two atoms in the center-of-mass modes 1 and 2 of the $m_F=0$ hyperfine state, described by the operator pair $a_{01} a_{02}$, results in the creation of a pair of atoms in the $m_F = \pm 1$ spin states via $a_1^\dagger a_{-1}^\dagger$.

Together with the conservation of the total number of atoms, these conservation laws yield the two additional conserved quantities

$$\begin{aligned} \hat{N}_1 & \equiv a_1^\dagger a_1 + a_{01}^\dagger a_{01}, \\ \hat{N}_2 & \equiv a_{-1}^\dagger a_{-1} + a_{02}^\dagger a_{02}. \end{aligned} \quad (19)$$

These conservation laws make it possible to define an angular momentum algebra for this four-wave mixing system analogous to the Schwinger coupled boson representation used in Ref. [13] for the description of two-mode condensates and in Ref. [5] for the three-mode coupling problem. However the present situation requires that these considerations be extended to a compound angular momentum representation [14,15].

We proceed by introducing spinor operators \mathbf{a}_1 and \mathbf{a}_2 ,

$$\mathbf{a}_1 \equiv \begin{pmatrix} a_{01} \\ a_1 \end{pmatrix} \quad \text{and} \quad \mathbf{a}_2 \equiv \begin{pmatrix} a_{-1} \\ a_{02} \end{pmatrix}, \quad (20)$$

as well as the two angular momentum operators \mathbf{S}_1 and \mathbf{S}_2 ,

$$\mathbf{S}_1 \equiv \mathbf{a}_1^\dagger \boldsymbol{\sigma} \mathbf{a}_1 = \frac{1}{2} \begin{pmatrix} a_{01}^\dagger a_1 + a_{01} a_1^\dagger \\ -i(a_{01}^\dagger a_1 - a_{01} a_1^\dagger) \\ a_{01}^\dagger a_{01} - a_1^\dagger a_1 \end{pmatrix} \quad (21)$$

and

$$\mathbf{S}_2 \equiv \mathbf{a}_2^\dagger \boldsymbol{\sigma} \mathbf{a}_2 = \frac{1}{2} \begin{pmatrix} a_{-1}^\dagger a_{02} + a_{02}^\dagger a_{-1} \\ -i(a_{02}^\dagger a_{-1} - a_{02} a_{-1}^\dagger) \\ a_{-1}^\dagger a_{-1} - a_{02}^\dagger a_{02} \end{pmatrix}, \quad (22)$$

where $\boldsymbol{\sigma}$ is the Pauli spin operator. The Casimir operators K_j associated with the angular momenta \mathbf{S}_j are also constants of motion since

$$K_j \equiv \mathbf{S}_j^2 = \frac{\hat{N}_j}{2} \left(\frac{\hat{N}_j}{2} + 1 \right). \quad (23)$$

Expressed in terms of the total spin operator

$$\mathbf{S} \equiv \mathbf{S}_1 + \mathbf{S}_2 \quad (24)$$

with components $S_j = S_{1j} + S_{2j}$, the Hamiltonian (14) becomes

$$\mathcal{H} = \frac{\hbar^2 k_0^2}{2M} \hat{N} + \frac{\hbar c_0}{2} \hat{N}(\hat{N}-1) + 2\hbar c_2 \left[\mathbf{S}^2 - \frac{\hat{N}}{2} - \left(\frac{\hat{D}_0}{2} \right)^2 \right]. \quad (25)$$

Together with the conserved Casimir operators, this form of the four-wave mixing Hamiltonian suggests expressing the state of the system in terms of the complete set of quantum numbers associated with the eigenstates of the operators \mathbf{S}_1^2 , \mathbf{S}_2^2 , \mathbf{S}^2 , and S_z . The Hamiltonian (25) can clearly be diagonalized in terms of these operators, yielding a complete set of eigenstates and eigenenergies.

To illustrate how this works, we consider for concreteness a condensate consisting of N atoms such that initially there are $N_{01} \equiv \langle a_{01}^\dagger a_{01} \rangle = N/2 - m$ atoms in center-of-mass mode 1 and $N_{02} \equiv \langle a_{02}^\dagger a_{02} \rangle = N/2$ atoms in center-of-mass mode 2 of the hyperfine state $m_F = 0$, $m \ll N/2$ atoms in the hyperfine state $m_F = 1$, and none in the state $m_F = -1$. In the ‘‘natural’’ eigenbasis of the operators $\{K_1, S_{1z}, K_2, S_{2z}\}$ the initial state of this system is described by the state vector

$$|\phi(0)\rangle = \left| \frac{N}{4}, \frac{N}{4} - m; \frac{N}{4}, -\frac{N}{4} \right\rangle. \quad (26)$$

Expanding it in terms of a complete set of eigenvectors $\{|N/4, N/4, S, S_z\rangle\}$ of the Hamiltonian (25), we find

$$|\phi(0)\rangle = \sum_S C \left(\frac{N}{4} - m, -\frac{N}{4}; S, -m \right) \left| \frac{N}{4}, \frac{N}{4}; S, -m \right\rangle, \quad (27)$$

where

$$C(S_{1z}, S_{2z}; S, -m) \equiv \left\langle \frac{N}{4}, \frac{N}{4}; S, -m \left| \frac{N}{4}, S_{1z}; \frac{N}{4}, S_{2z} \right\rangle \quad (28)$$

are Clebsch-Gordan coefficients [16], which are nonzero only for $S_z = S_{1z} + S_{2z} = -m$.

This conservation of S_z under the Hamiltonian (25) further implies that the state vector of the system is given at time t by

$$|\phi(t)\rangle = \sum_S \alpha_S(t) \left| \frac{N}{4}, \frac{N}{4}; S, -m \right\rangle \equiv \sum_S \alpha_S(t) |S, -m\rangle, \quad (29)$$

where in the second equality we have made explicit use of the value of the conserved quantities K_1 , K_2 , and S_z to simplify the notation via

$$\left| \frac{N}{4}, \frac{N}{4}; S, -m \right\rangle \rightarrow |S, -m\rangle. \quad (30)$$

Note that this simplification is not general, but is appropriate for the initial condition at hand.

The equations of motion for the probability amplitudes $\alpha_S(t)$ follow from the Schrödinger equation. For the condensate containing N atoms they read, in a frame rotating at the frequency $\hbar k_0^2 N/2M + c_0 N(N-1)/2 - 2c_2 [N/2 + (D_0/2)^2]$,

$$i\dot{\alpha}_S(t) = 2c_2 S(S+1) \alpha_S(t) \quad (31)$$

and can be integrated trivially with the initial condition

$$\alpha_S(t=0) = C \left(\frac{N}{4} - m, -\frac{N}{4}; S, -m \right).$$

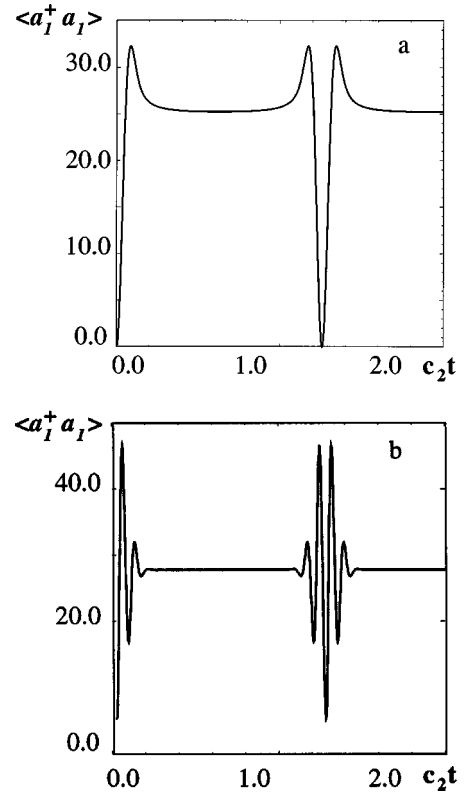


FIG. 1. Time evolution of the side-mode population $\langle a_1^\dagger a_1 \rangle$ for $N=100$ and initial populations (a) $\langle a_{01}^\dagger a_{01} \rangle = N/2$, $\langle a_{02}^\dagger a_{02} \rangle = N/2$, $\langle a_1^\dagger a_1 \rangle = 0$, and $\langle a_{-1}^\dagger a_{-1} \rangle = 0$; (b) $\langle a_{01}^\dagger a_{01} \rangle = N/2 - 5$, $\langle a_{02}^\dagger a_{02} \rangle = N/2$, $\langle a_1^\dagger a_1 \rangle = 5$, and $\langle a_{-1}^\dagger a_{-1} \rangle = 0$.

IV. DYNAMICS

First we apply the results of the preceding section to the study of the dynamics of population exchange between the different modes of the condensate. For instance, the population of the $m_F = 1$ hyperfine spin state can readily be determined from the expectation value of S_{1z} . With Eqs. (19) and the definition of S_{1z} we find

$$\langle a_1^\dagger a_1 \rangle = \frac{N}{4} - \langle S_{1z} \rangle. \quad (32)$$

From Eq. (29) we have

$$\langle S_{1z} \rangle = \sum_p p \left| \sum_S \alpha_S(t) C(p, -(p+m); S, -m) \right|^2, \quad (33)$$

where we inserted the identity operator

$$\hat{I} = \sum_{p_1, p_2} \left| \frac{N}{4}, p_1; \frac{N}{4}, p_2 \right\rangle \left\langle \frac{N}{4}, p_1; \frac{N}{4}, p_2 \right| \quad (34)$$

and used the simplified notation (30) as well as the property that the Clebsch-Gordan coefficients are nonzero only for $S_z = S_{1z} + S_{2z}$. Equations (33) can be evaluated numerically.

The evolution of the population of the $m_F = 1$ side mode is shown in Fig. 1 for $N=100$ atoms in the system. In case (b) the initial mode populations are $\langle a_{01}^\dagger a_{01} \rangle = N/2 - m$, $\langle a_{02}^\dagger a_{02} \rangle = N/2$, $\langle a_1^\dagger a_1 \rangle = m = 5$, $\langle a_{-1}^\dagger a_{-1} \rangle = 0$ while case (a) illustrates the build-up from quantum fluctuations, $m = 0$. In

both cases, the side-mode population exhibits an initial growth to the point where it contains about 1/3 of the atoms in the first case and about half of the atoms in the second. This is followed by a collapse to a quasi-steady-state population, as well as a subsequent revival at $2c_2t_1 = \pi$. This dynamics then repeats itself periodically, with revivals at $2c_2t_n = \pi n$, independently of N . This is similar to the periodic revivals which occur in the two-photon Jaynes-Cummings model discussed in [17].

During the periods of collapse, one has $S_{jz} = -m/2$, so that all modes are almost equally macroscopically populated with $\langle a_{01}^\dagger a_{01} \rangle = N/4 - m/2$, $\langle a_1^\dagger a_1 \rangle = N/4 + m/2$, $\langle a_{02}^\dagger a_{02} \rangle = N/4 + m/2$, and $\langle a_2^\dagger a_2 \rangle = N/4 - m/2$.

A particularly interesting aspect of the present study is that it allows one to obtain the quantum correlations between side modes. In optics, for example, four-wave mixing provides a method to study purely quantum-mechanical effects such as squeezing and nonclassical states of the radiation field, and also to prepare states of composite systems exhibiting strong quantum-mechanical entanglement [18]. These states are of considerable interest in tests of the foundations of physics as well as quantum information processing such as quantum cryptography [19,20] and quantum computing [21]. Macroscopic quantum states of massive particles present an interesting alternative to all-optical systems, hence it is of considerable interest to determine to which extent quantum entanglement between side modes can be achieved in Bose-Einstein condensation.

In analogy with the optical case, one can quantify the amount of quantum entanglement between condensate modes by determining the extent to which the Cauchy-Schwartz inequality is violated by the second-order cross-correlation functions between modes [18]. In particular, for a ‘‘classically looking’’ optical system with positive Glauber \mathcal{P} representation, the single-time second-order cross-correlation function is bound by

$$G_{i,j}^{(2)}(t) \leq [G_i^{(2)}(t)G_j^{(2)}(t)]^{1/2}. \quad (35)$$

In case the \mathcal{P} representation is not positive or does not exist, in contrast, the upper bound is higher, namely

$$G_{i,j}^{(2)}(t) \leq \{[G_i^{(2)}(t) + G_i^{(1)}(t)][G_j^{(2)}(t) + G_j^{(1)}(t)]\}^{1/2}. \quad (36)$$

In these inequalities, we have introduced the single-time and single-mode first-order correlation functions

$$G_j^{(1)}(t) \equiv \langle \phi(t) | a_j^\dagger a_j | \phi(t) \rangle \quad (37)$$

as well as the single-time two-mode second-order correlation functions

$$G_{ij}^{(2)}(t) \equiv \langle \phi(t) | a_i^\dagger a_j^\dagger a_j a_i | \phi(t) \rangle \quad (38)$$

and the single-time, second-order correlation functions

$$G_j^{(2)}(t) \equiv \langle \phi(t) | a_j^\dagger a_j^\dagger a_j a_j | \phi(t) \rangle. \quad (39)$$

As was the case for the side-mode populations, the single-time single-mode second-order cross correlation between the side modes $m_F = \pm 1$ can be expressed in terms of the z component of the individual pseudospins as

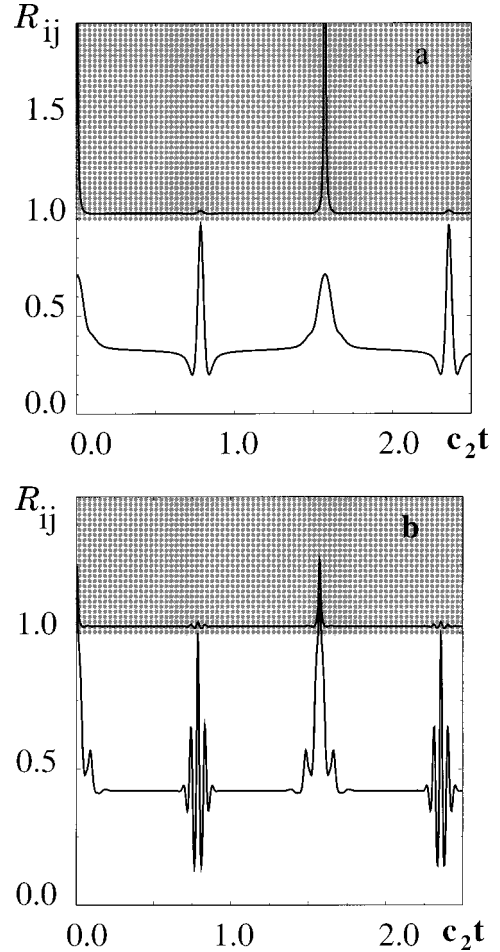


FIG. 2. Time evolution of the one-time normalized central-mode–side-mode correlation function (lower curve) $\mathcal{R}_{01,1}^{(2)}(t)$ and the one-time normalized side-mode–side-mode correlation function (upper curve) $\mathcal{R}_{-1,1}^{(2)}(t)$ for $N=100$ and (a) $\langle a_{01}^\dagger a_{01} \rangle = N/2$, $\langle a_{02}^\dagger a_{02} \rangle = N/2$, $\langle a_1^\dagger a_1 \rangle = 0$, and $\langle a_{-1}^\dagger a_{-1} \rangle = 0$; (b) $\langle a_{01}^\dagger a_{01} \rangle = N/2 - 5$, $\langle a_{02}^\dagger a_{02} \rangle = N/2$, $\langle a_1^\dagger a_1 \rangle = 5$, and $\langle a_{-1}^\dagger a_{-1} \rangle = 0$. The shaded area is the classically forbidden region of values of R_{ij} .

$$\begin{aligned} G_{-1,1}^{(2)}(t) &= \left\langle \left(\frac{N}{4} - S_{1z} \right) \left(\frac{N}{4} - S_{2z} \right) \right\rangle \\ &= \left(\frac{N}{4} \right)^2 - \frac{N}{4} (\langle S_{1z} \rangle + \langle S_{2z} \rangle) + \langle S_{1z} S_{2z} \rangle, \end{aligned} \quad (40)$$

where

$$\langle S_{1z} S_{2z} \rangle = \sum_p p(-p-m) \left| \sum_{s=m}^{[N/2]} \alpha_s(t) \mathcal{C}(p, -(p+m); S, m) \right|^2 \quad (41)$$

and the sum can easily be evaluated numerically.

Figure 2 compares the time dependence of the normalized central-mode–side-mode correlation function (lower curve)

$$\mathcal{R}_{01,1}^{(2)}(t) \equiv \frac{G_{01,1}^{(2)}(t)}{\sqrt{G_{01}^{(2)}(t)G_1^{(2)}(t)}} \quad (42)$$

and the side-mode–side-mode correlation (upper curve)

$$\mathcal{R}_{-1,1}^{(2)}(t) \equiv \frac{G_{-1,1}^{(2)}(t)}{\sqrt{G_1^{(2)}(t)G_{-1}^{(2)}(t)}} \quad (43)$$

for (a) the case $m=0$ where the side mode builds up from quantum fluctuations and (b) the case $m=5$ of an injected signal. These results illustrate how the correlations between central mode and side modes do satisfy the classical Cauchy-Schwartz inequality while the side-mode-side-mode cross correlations violate them. The violation is particularly strong in the case of build-up from quantum fluctuations, as should be intuitively expected. In that case the hyperfine side modes $m_F = \pm 1$ play symmetric roles, thus

$$G_{-1,1}^{(2)}(t) = G_1^{(2)}(t) + G_1^{(1)}(t), \quad (44)$$

i.e., Eq. (36) thus becomes an equality, corresponding to the maximum violation of the classical Cauchy-Schwartz inequality allowed by quantum mechanics.

The difference in the behavior of the two mode correlation functions between side modes and those involving one central and one side mode can be intuitively understood from the form of the wave-mixing term $a_1^\dagger a_{-1}^\dagger a_{01} a_{02}$ appearing in the Hamiltonian (14). Indeed, the coupling between side modes, involving two annihilation operators, is reminiscent of the interaction $a_1^\dagger a_2^\dagger$ in the Hamiltonian of parametric amplification leading to squeezing and quantum entanglement between two side modes. In contrast, the coupling between central and side modes involves both an annihilation and a creation operator.

V. SUMMARY AND CONCLUSIONS

In contrast to the optical case, where it is usually difficult to study quantum mechanically four-wave mixing in the strong field limit, this task can be achieved relatively easily for matter waves, a direct consequence of the conservation of the total number of atoms in a condensate at $T=0$ and in a

lossless trap. The resulting conservation laws permit us to develop an angular momentum algebra analysis that leads to an exact solution of the problem away from the linear regime where the side-mode populations remain small.

In this paper we have applied this technique to the study of the dynamics of the population exchange between hyperfine levels of a condensate. We found that this exchange is periodic and it is characterized by a sequence of ‘‘collapses’’ and ‘‘revivals’’ reminiscent of those appearing in the two-photon Jaynes-Cummings model of optical physics. In addition, we found that strong quantum correlations can develop between the central modes and the side modes, the general state of the system exhibiting a strong quantum entanglement between the modes $m_F = \pm 1$. Thus, it appears that multi-component condensates offer a fascinating method to create quantum entanglement at a truly macroscopic level, a possibility made even more attractive by the fact that these systems suffer very little from dissipation, since they consist of ground-state atoms. The finite lifetime of condensates is usually attributed to three-body collisions, which result in losses on a time scale of seconds.

We conclude by noting that the revivals in the population exchange occur at a time independent of the number of atoms in the condensate. Hence, they allow for a direct and absolute determination of the coefficient c_2 . In practice, however, c_2 seems to be too small to observe revivals on the time scales of the condensate lifetime.

ACKNOWLEDGMENTS

This work was supported in part by the U.S. Office of Naval Research under Contract No. 14-91-J1205, by the National Science Foundation under Grant No. PHY98-01099, by the U.S. Army Research Office, and by the Joint Services Optics Program. The valuable comments and suggestions by O. Zobay are gratefully acknowledged. E.V.G. also acknowledges stimulating discussions with J. H. Eberly.

-
- [1] S. A. Morgan, R. J. Ballagh, and K. Burnett, *Phys. Rev. A* **55**, 4338 (1997); W. P. Reinhardt and C. W. Clark, *J. Phys. B* **30**, L785 (1997); R. Dum, J. I. Cirac, M. Lewenstein, and P. Zoller, *Phys. Rev. Lett.* **80**, 2972 (1998); T. F. Scott, R. J. Ballagh, and K. Burnett, *J. Phys. B* **31**, L329 (1998); A. D. Jackson, G. M. Kavoulakis, and C. J. Pethick, *Phys. Rev. A* **58**, 2417 (1998); O. Zobay, S. Pötting, E. M. Wright, and P. Meystre, *ibid.* **59**, 643 (1999).
- [2] E. V. Goldstein, K. Plättner, and P. Meystre, *J. Res. Natl. Inst. Stand. Technol.* **101**, 583 (1996).
- [3] E. V. Goldstein and P. Meystre, *Phys. Rev. A* **59**, 1509 (1999).
- [4] M. Trippenbach, Y. B. Band, and P. S. Julienne, *Opt. Express* **3**, 530 (1998).
- [5] C. K. Law, H. Pu, and N. P. Bigelow, *Phys. Rev. Lett.* **81**, 5257 (1998).
- [6] L. Deng *et al.*, *Nature (London)* **398**, 218 (1999).
- [7] D. M. Stamper-Kurn *et al.*, *Phys. Rev. Lett.* **80**, 2027 (1998).
- [8] T. Ohmi and K. Machida, *J. Phys. Soc. Jpn.* **67**, 1822 (1998).
- [9] T.-L. Ho, *Phys. Rev. Lett.* **81**, 742 (1998).
- [10] J. Stenger *et al.*, *Nature (London)* **396**, 345 (1998).
- [11] See, e.g., Chap 2 in P. Meystre and M. Sargent III, *Elements of Quantum Optics*, 3rd ed. (Springer-Verlag, New York, 1998).
- [12] W. Zhang and D. F. Walls, *Phys. Rev. A* **57**, 1248 (1998).
- [13] G. J. Milburn, J. Corney, E. M. Wright, and D. F. Walls, *Phys. Rev. A* **55**, 4318 (1997); M. J. Steel and M. J. Collett, *ibid.* **57**, 2920 (1998).
- [14] D. C. Mattis, *The Theory of Magnetism I* (Springer-Verlag, New York, 1981).
- [15] L. Wang and J. H. Eberly, *Phys. Rev. A* **47**, 4248 (1993).
- [16] M. Abramowitz and I. A. Stegun, *Handbook of Mathematical Functions* (Dover, New York, 1972).
- [17] B. Buck and C. V. Sukumar, *Phys. Lett.* **81A**, 132 (1981).
- [18] D. F. Walls and G. J. Milburn, *Quantum Optics* (Springer-Verlag, Berlin, 1994).
- [19] C. H. Bennett, *Science* **257**, 752 (1992).
- [20] W. Tittel, G. Ribordy, and N. Gisin, *Phys. World* **11**, 41 (1998).
- [21] A. Ekert and R. Jozsa, *Rev. Mod. Phys.* **68**, 733 (1996).