PHYSICAL REVIEW
LETTERS

VOLUME 81 | 14 DECEMBER 1998 | NUMBER 24

Quantum Spins Mixing in Spinor Bose-Einstein Condensates

C. K. Law,[1] H. Pu,[2] and N. P. Bigelow[2]
[1]*Rochester Theory Center for Optical Science and Engineering and Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627*
[2]*Laboratory for Laser Energetics, and Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627*
(Received 18 May 1998)

A set of collective spin states is derived for a trapped Bose-Einstein condensate in which atoms have three internal hyperfine spins. These collective states minimize the interaction energy among condensate atoms, and they are characterized by strong spin correlations. We also examine the internal dynamics of an initially spin-polarized condensate. The time scale of spin mixing is predicted. [S0031-9007(98)07921-6]

PACS numbers: 03.75.Fi

Bose-Einstein condensates (BEC) of atoms with internal degrees of freedom are new forms of macroscopically coherent matter which exhibit rich quantum structures. In the case of BEC with two internal spin states [1,2], theoretical studies have predicted interesting phenomena such as quantum entanglement of spins [3], suppression of quantum phase diffusion [4], and interference effects [5]. Recently, Stamper-Kurn *et al.* [6] have realized an optically trapped BEC in which all three hyperfine states in the lowest energy manifold of sodium atoms are involved. Such a three-component condensate raises new questions regarding the more complex ground state structure [7,8] and internal spin dynamics. One of the key features here is that there are spin exchange interactions which constantly mix different condensate spin components while the system as a whole remains in the ground state. For example, two atoms with respective hyperfine spins $+1$ and -1 interact and become two atoms with hyperfine spin 0. Therefore an important problem is to determine how atoms organize their spins in the ground state and how a spin-polarized BEC loses its polarization because of spin exchange interactions.

In this paper we approach the questions using an algebraic method found in quantum optics. By excluding effects of noncondensate atoms, we identify the fact that the interaction between spin components in a BEC is analogous to 4-wave mixing in nonlinear optics. However, since the trap is like a matter wave cavity, a more appropriate optical analogy is the 4-wave mixing in a high finesse cavity (i.e., a cavity QED system). With the help of the methods developed in a related cavity QED problem [9,10], we are able to study the organization of spins in the condensate ground state. We find that there exists a class of quantum superposition states which minimize the interaction energy. These quantum states are recognized as collective spin states which are characterized by strong correlations among different spin components, and in some cases we find that the number of atoms in an individual spin component shows large fluctuations. In this paper we also examine the *internal dynamics* of the spin-mixing process arising from the nonlinear interactions between condensate atoms [11]. For an initially spin-polarized BEC, we predict the time scale at which spins become strongly mixed.

To begin we consider a dilute gas of trapped bosonic atoms with hyperfine spin $f = 1$. The second quantized Hamiltonian of the system is given by ($\hbar = 1$)

$$\mathcal{H} = \sum_{\alpha} \int d^3x\, \hat{\Psi}_{\alpha}^{\dagger} \left(-\frac{\nabla^2}{2M} + V_T \right) \hat{\Psi}_{\alpha} + \sum_{\alpha,\beta,\mu,\nu} \Omega_{\alpha\beta\mu\nu} \int \hat{\Psi}_{\alpha}^{\dagger} \hat{\Psi}_{\beta}^{\dagger} \hat{\Psi}_{\mu} \hat{\Psi}_{\nu}\, d^3x\,, \quad (1)$$

where $\hat{\Psi}_{\kappa}$ ($\kappa = -1, 0, 1$) is the atomic field annihilation

operator associated with atoms in the hyperfine spin state $|f = 1, m_f = \kappa\rangle$. The summation indices in (1) run through the values $-1, 0, 1$. The mass of the atom is given by M, and the trapping potential V_T is assumed to be the same for all three components. The interactions between atoms are characterized by the coefficients $\Omega_{\alpha\beta\mu\nu}$ which are obtained from the two-body interaction model [8,11–13],

$$U(\vec{x}_1, \vec{x}_2) = \delta(\vec{x}_1 - \vec{x}_2) \sum_{F=0}^{2} g_F \sum_{M_F=-F}^{F} |F, M_F\rangle\langle F, M_F| . \tag{2}$$

Here $|F, M_F\rangle$ is the total hyperfine spin state formed by two atoms each with spin $f = 1$, and $g_F \equiv 4\pi\hbar^2 a_F/M$ with a_F being the s-wave scattering length in the F channel. The interaction (2) is based on rather general symmetry assumptions of the system, because it preserves angular momentum and the rotation symmetry in hyperfine spin space [8]. The model also makes use of the δ potential approach which has been widely used in one-component dilute BEC.

By expanding the total spin state $|F, M_F\rangle$ in terms of basis vectors $|f = 1, m_f = \alpha\rangle \otimes |f = 1, m_f = \beta\rangle$, we obtain the Hamiltonian in the form $\mathcal{H} = \mathcal{H}_S + \mathcal{H}_A$, where

$$\mathcal{H}_S = \sum_\alpha \int d^3x\, \hat{\Psi}_\alpha^\dagger \left(-\frac{\nabla^2}{2M} + V_T\right)\hat{\Psi}_\alpha + \frac{\lambda_s}{2}\sum_{\alpha,\beta}\int \hat{\Psi}_\alpha^\dagger \hat{\Psi}_\beta^\dagger \hat{\Psi}_\alpha \hat{\Psi}_\beta\, d^3x , \tag{3}$$

$$\mathcal{H}_A = \frac{\lambda_a}{2}\int (\hat{\Psi}_1^\dagger\hat{\Psi}_1^\dagger\hat{\Psi}_1\hat{\Psi}_1 + \hat{\Psi}_{-1}^\dagger\hat{\Psi}_{-1}^\dagger\hat{\Psi}_{-1}\hat{\Psi}_{-1} + 2\hat{\Psi}_1^\dagger\hat{\Psi}_0^\dagger\hat{\Psi}_1\hat{\Psi}_0 + 2\hat{\Psi}_{-1}^\dagger\hat{\Psi}_0^\dagger\hat{\Psi}_{-1}\hat{\Psi}_0 - 2\hat{\Psi}_1^\dagger\hat{\Psi}_{-1}^\dagger\hat{\Psi}_1\hat{\Psi}_{-1} + 2\hat{\Psi}_0^\dagger\hat{\Psi}_0^\dagger\hat{\Psi}_1\hat{\Psi}_{-1} + 2\hat{\Psi}_1^\dagger\hat{\Psi}_{-1}^\dagger\hat{\Psi}_0\hat{\Psi}_0)\, d^3x. \tag{4}$$

Here $\lambda_s \equiv (g_0 + 2g_2)/3$ and $\lambda_a \equiv (g_2 - g_0)/3$ are defined. The Hamiltonian $\mathcal{H}$ is written as the sum of a symmetric part $\mathcal{H}_S$ and a nonsymmetric part $\mathcal{H}_A$, where $\mathcal{H}_S$ remains unchanged for any interchange of the spin component indices.

In this paper we assume that the symmetric interaction $\mathcal{H}_S$ is strong compared with $\mathcal{H}_A$. This occurs for atoms whose scattering lengths in different F channels have approximately the same values such that $|\lambda_s| \gg |\lambda_a|$. Recent estimations have indicated that ^{23}Na and ^{87}Rb atoms indeed have such a property [7,8,14]. With the symmetric $\mathcal{H}_S$ being the dominant Hamiltonian, the condensate wave functions $\phi_\kappa(\vec{x})$ ($\kappa = 0, \pm 1$) for each spin component are approximately described by the same wave function $\phi(\vec{x})$, i.e., $\phi_\kappa(\vec{x}) = \phi(\vec{x})$, which is defined by the Gross-Pitaevskii equation through $\mathcal{H}_S$,

$$\left(-\frac{\nabla^2}{2M} + V_T + \lambda_s N|\phi|^2\right)\phi = \mu\phi , \tag{5}$$

where μ is the mean field energy or the chemical potential.

Under the condition that atoms in different spin states are described by the same wave function, we can approximate field operators at the zero temperature by

$$\hat{\Psi}_\kappa \approx \hat{a}_\kappa \phi(\vec{x}) \qquad \kappa = 0, \pm 1 . \tag{6}$$

Here $\hat{a}_\kappa$ is the annihilation operator associated with the condensate mode, and it satisfies the usual commutation relation $[\hat{a}_\kappa, \hat{a}_\gamma] = 0$ and $[\hat{a}_\kappa, \hat{a}_\gamma^\dagger] = \delta_{\kappa\gamma}$. Using (5) and (6), $\mathcal{H}_S$ and $\mathcal{H}_A$ have leading parts H_s and H_a, respectively,

$$\mathcal{H}_S \approx \mu\hat{N} - \lambda_s'\hat{N}(\hat{N} - 1) \equiv H_s , \tag{7}$$

$$\mathcal{H}_A \approx \lambda_a'(\hat{a}_1^\dagger\hat{a}_1^\dagger\hat{a}_1\hat{a}_1 + \hat{a}_{-1}^\dagger\hat{a}_{-1}^\dagger\hat{a}_{-1}\hat{a}_{-1} - 2\hat{a}_1^\dagger\hat{a}_{-1}^\dagger\hat{a}_1\hat{a}_{-1} + 2\hat{a}_1^\dagger\hat{a}_0^\dagger\hat{a}_1\hat{a}_0 + 2\hat{a}_{-1}^\dagger\hat{a}_0^\dagger\hat{a}_{-1}\hat{a}_0 + 2\hat{a}_0^\dagger\hat{a}_0^\dagger\hat{a}_1\hat{a}_{-1} + 2\hat{a}_1^\dagger\hat{a}_{-1}^\dagger\hat{a}_0\hat{a}_0) \equiv H_a . \tag{8}$$

Here $2\lambda_i' \equiv \lambda_i \int |\phi(\vec{x})^4|\, d^3x$ ($i = s, a$), and $\hat{N} \equiv \hat{a}_1^\dagger\hat{a}_1 + \hat{a}_0^\dagger\hat{a}_0 + \hat{a}_{-1}^\dagger\hat{a}_{-1}$ is the total number of atoms in the condensate.

Our goal is to find the quantum states that minimize the energy $H_s + H_a$. Since H_s is a function of $\hat{N}$ only, H_s is a constant operator for a fixed number of atoms. Therefore it is sufficient to look for the ground state of H_a. It is quite remarkable that a similar structure of H_a also appeared in nonlinear wave-mixing processes in cavity QED [9]. We follow Refs. [9,10] and identify the algebraic structure of the system. We notice that the operators $\hat{L}_- \equiv \sqrt{2}\,(\hat{a}_1^\dagger\hat{a}_0 + \hat{a}_0^\dagger\hat{a}_{-1})$, $\hat{L}_+ \equiv \sqrt{2}\,(\hat{a}_0^\dagger\hat{a}_1 + \hat{a}_{-1}^\dagger\hat{a}_0)$, and $\hat{L}_z \equiv (\hat{a}_{-1}^\dagger\hat{a}_{-1} - \hat{a}_1^\dagger\hat{a}_1)$ obey angular momentum commutation relations: $[\hat{L}_+, \hat{L}_-] = 2\hat{L}_z$ and $[\hat{L}_z, \hat{L}_\pm] = \pm\hat{L}_\pm$. In other words, the operators $\hat{L}_+$, $\hat{L}_-$ can be interpreted as raising and lowering operators of a kind of "orbital angular momentum," and $\hat{L}_z$ is the "z component" in the standard notations. From the theory of angular momentum, $\hat{L}^2$ and $\hat{L}_z$ have a complete set of common eigenvectors $|l, m_l\rangle$ defined by

$$\hat{L}^2|l, m_l\rangle = l(l + 1)\,|l, m_l\rangle , \tag{9}$$

$$\hat{L}_z|l, m_l\rangle = m_l|l, m_l\rangle , \tag{10}$$

where $m_l = 0, \pm 1, \pm 2, \ldots, \pm l$. For a given total number

of atoms N, the allowable values of l are $l = 0, 2, 4, \ldots, N$ if N is even, and $l = 1, 3, 5, \ldots, N$ if N is odd.

With the help of the angular momentum operators, H_a takes a very simple form,

$$H_a = \lambda_a'(\hat{L}^2 - 2\hat{N}). \tag{11}$$

This is the main result of the paper because the energy spectrum of H_a is now solved. Equation (11) indicates that $|l, m_l\rangle$ are eigenstates of H_a with the energy E_l^a

$$E_l^a = \lambda_a'[l(l+1) - 2N]. \tag{12}$$

The lowest energy state of H_a depends on the sign of λ_a'. In the following we discuss two cases: (I) $\lambda_a' > 0$ and (II) $\lambda_a' < 0$.

(I) $\lambda_a' > 0$.—In this case $|l = 0, m_l = 0\rangle$ is the ground state of H_a. Using the Fock states $|n_1, n_0, n_{-1}\rangle$ defined by the number operators $\hat{n}_j \equiv \hat{a}_j^\dagger \hat{a}_j$ for the three spin components (i.e., $\hat{n}_j|n_1, n_0, n_{-1}\rangle = n_j|n_1, n_0, n_{-1}\rangle$), $|l = 0, m_l = 0\rangle$ has the form

$$|l = 0, m_l = 0\rangle = \sum_{k=0}^{[N/2]} A_k |k, N - 2k, k\rangle, \tag{13}$$

where the amplitudes A_k obey the recursion relation

$$A_k = -\sqrt{\frac{N - 2k + 2}{N - 2k + 1}}\, A_{k-1}. \tag{14}$$

We see that the state $|l = 0, m_l = 0\rangle$ is a quantum superposition of a chain of Fock states $|k, N - 2k, k\rangle$ in which the numbers of atoms in the spins 1 and -1 are equal. We stress that such a quantum state is a *collective* spin state which cannot be expressed as product states of individual atoms. The amplitudes A_k are arranged in such a way that the interaction energy H_a is almost completely canceled. This can be seen from the disappearance of N^2 dependence in the energy of H_a. It is not difficult to show that for the state (13), the average numbers of atoms in each component are all equal, i.e., $\langle\hat{n}_0\rangle = \langle\hat{n}_1\rangle = \langle\hat{n}_{-1}\rangle = N/3$. Since A_k are almost uniformly distributed (see Fig. 1a), there are large fluctuations of particle numbers in individual components although the total particle number N is fixed. More precisely, we find that $\langle\Delta\hat{n}_0\rangle \approx 2N/\sqrt{5}$ for $N \gg 1$, i.e., a super-Poisson distribution. Our further calculations indicate that a super-Poisson distribution of particle numbers is a common feature for low energy eigenstates of H_a when $\lambda_a' > 0$.

(II) $\lambda_a' < 0$.—In this case H_a has $2N + 1$ degenerate ground states given by $|l = N, m_l\rangle$ where $m_l = 0, \pm 1, \pm 2, \ldots, \pm N$. The energy (12) of these states is $\lambda_a' N(N - 1)$, and the general form of $|l = N, m_l\rangle$ is given by

$$|l = N, m_l\rangle = \sum_k B_k^{(m_l)} |k, N - 2k - m_l, k + m_l\rangle. \tag{15}$$

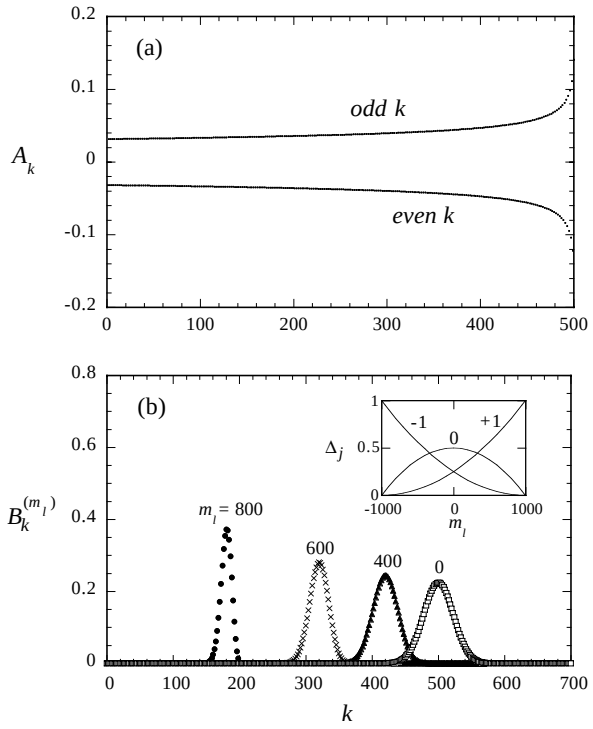

FIG. 1. Amplitudes of Fock states associated with the ground states of H_a for $N = 10^3$ atoms: (a) $\lambda_a' > 0$; (b) $\lambda_a' < 0$. The inset in (b) shows the normalized number fluctuations $\Delta_j \equiv \langle\Delta n_j\rangle^2/\langle\hat{n}_j\rangle$ in the three spin components ($j = 0, \pm 1$) as a function of m_l. Sub-Poisson distributions are defined by $\Delta_j < 1$.

Here the summation index k runs over all physical Fock states $|k, N - 2k - m_l, k + m_l\rangle$ (i.e., those with non-negative numbers in each component). The simplest case of (15) is $|l = N, m_l = -N\rangle = |N, 0, 0\rangle$, and with this we can construct the amplitudes $B_k^{(m_l)}$ by repeatedly applying the raising operator $\hat{L}_+$. To give an illustration, we plot in Fig. 1b the Fock state amplitudes $B_k^{(m_l)}$ for several m_l's. We see that $B_k^{(m_l)}$ has a narrow distribution which indicates well defined particle numbers in each spin component. It is interesting that all the degenerate states (15) have sub-Poisson number fluctuations in each spin component (see the inset of Fig. 1b). This feature is just the opposite of the previous case $\lambda_a' > 0$.

Finally, let us look at the spin-mixing dynamics of an initially spin-polarized condensate in which all atoms in the condensate are prepared in the spin-0 state at $t = 0$, i.e., $|\psi(0)\rangle = |0, N, 0\rangle$. In this case two atoms in the spin-0 state can be converted into one atom in the spin-1 state and the other in the spin-(-1) state. Assuming the spin-mixing process does not introduce appreciable changes of shape of the spatial condensate wave function, we can use $H_s + H_a$ as our approximate Hamiltonian to describe the internal dynamics [15]. The internal state at time t is given by

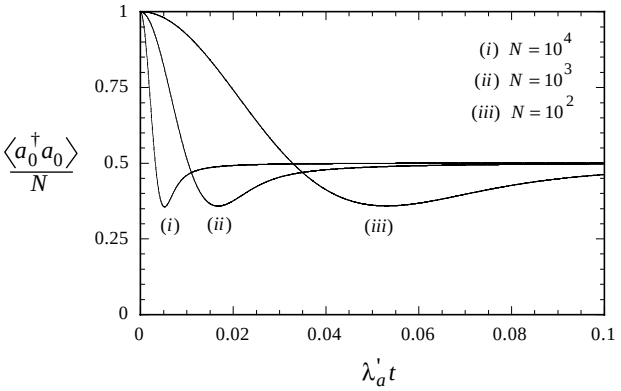

FIG. 2. Time dependence of average number of atoms in the spin-0 state normalized by the total number of atoms N. The initial state of the system is $|\psi(0)\rangle = |0, N, 0\rangle$. We show three cases with $N = 10^2, 10^3, 10^4$.

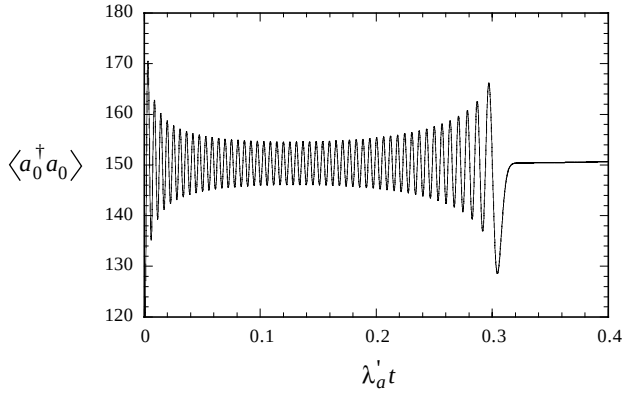

FIG. 3. Time dependence of average number of atoms in the spin-0 state. The initial state of the system is $|\psi(0)\rangle = |N/3, N/3, N/3\rangle$, where $N = 300$.

$$|\psi(t)\rangle = e^{-i\theta_N(t)} \sum_{l=0}^{N} C_l e^{-i\lambda_a' l(l+1)t} |l, m_l = 0\rangle , \quad (16)$$

where $C_l = \langle l, m_l = 0 \mid 0, N, 0\rangle$ and $\theta_N(t) = [\mu N - \lambda_s' N(N-1) - 2\lambda_a' N]t$. In Fig. 2 we present the time dependence of the particle number in the spin-0 component for $N = 10^2, 10^3, 10^4$ cases. We see that the number of atoms in the spin-0 component becomes steady at $\langle \hat{n}_0 \rangle = N/2$ after a time t_c,

$$t_c \approx \frac{1}{2|\lambda_a'|\sqrt{N}} . \quad (17)$$

This is the time scale for the spin-mixing process purely due to the nonlinear interaction between condensate atoms [16]. In the Thomas-Fermi (large N) limit, we find that for a spherical harmonic trap, $(g_2 - g_0)t_c \approx 5.1 N^{1/10}[(g_0 + 2g_2)/M\omega^2]^{3/5}$ where ω is trap frequency. Therefore t_c becomes quite insensitive to N in the Thomas-Fermi limit. To give a realistic example, for a sodium condensate with $N = 10^4$ and $\omega = 2\pi \times 370$ Hz, we find that t_c is about 0.5 sec, using the recent experimental value of the difference of scattering lengths $a_2 - a_0 = 0.2$ nm [7]. In Fig. 2 we also see that an apparent damping of the individual spin populations can be observed, even without dissipation, purely as a result of the nonlinear spin-mixing process.

We remark that the spin-mixing dynamics can be quite different for different initial conditions. In Fig. 3, we give an example for the case when all three components initially have the same atom numbers, i.e., $|\psi(0)\rangle = |N/3, N/3, N/3\rangle$. It is quite surprising that the particle number executes fast oscillations with a frequency of the order of $\lambda_a' N$, and then the system suddenly becomes steady. This interesting behavior indicates that there are complex quantum dynamics governed by the nonlinear interaction H_a. In fact, since H_a has a discrete spectrum, quantum recurrence or revival is expected in a much longer time scale (which is typically of the order of π/λ_a'). It is worth further exploring the quantum dynamics in the context of either BEC or cavity QED.

To conclude, we have examined the spin-mixing interaction of a Bose-Einstein condensate with three internal spin components. It is quite remarkable that the model interaction (2) (which is based on general symmetry assumptions) can lead to a simple algebraic representation, and from which we can construct the collective spin states which minimize the interaction energy among condensate atoms. These collective states exhibit spin correlations and characteristic particle number fluctuations which depend crucially on the sign of λ_a'. We have also investigated the spin-mixing dynamics due to the nonlinear interaction between condensate atoms. The time scale of mixing for an initially spin-polarized system is identified. This study provides a theoretical treatment of the structure and dynamics of spinor BEC. However, our analysis is limited to interactions between condensate atoms, and it remains to be answered how noncondensate atoms will decohere the condensate structure. In the future we hope to address this question as well as to generalize our approach to systems with other hyperfine spin values.

We thank Professor J. H. Eberly and Professor G. Shlyapnikov for discussions. This research was supported by NSF Grants No. PHY-9415583 and No. PHY-9457897, and by the David and Lucile Packard Foundation.

[1] C. J. Myatt, E. A. Burt, R. W. Ghrist, E. A. Cornell, and C. E. Wieman, Phys. Rev. Lett. **78**, 586 (1997).
[2] D. S. Hall, M. R. Matthews, J. R. Ensher, C. E. Wieman, and E. A. Cornell, Phys. Rev. Lett. **81**, 1539 (1998).
[3] J. I. Cirac, M. Lewenstein, K. Molmer, and P. Zoller, Phys. Rev. A **57**, 1208 (1998).
[4] C. K. Law, H. Pu, N. P. Bigelow, and J. H. Eberly, Phys. Rev. A **58**, 531 (1998).

[5] C. M. Savage, Janne Ruostekoski, and D. F. Walls, Phys. Rev. A **57**, 3805 (1998).
[6] D. M. Stamper-Kurn, M. R. Andrews, A. P. Chikkatur, S. Inouye, H.-J. Miesner, J. Stenger, and W. Ketterle, Phys. Rev. Lett. **80**, 2027 (1998).
[7] J. Stenger, S. Inouye, D. M. Stamper-Kurn, H. J. Miesner, A. P. Chikkatur, and W. Ketterle, Nature (London) (to be published).
[8] T. L. Ho, Phys. Rev. Lett. **81**, 742 (1998).
[9] Liwei Wang, R. R. Puri, and J. H. Eberly, Phys. Rev. A **46**, 7192 (1992).
[10] Ying Wu, Phys. Rev. A **54**, 4534 (1996).
[11] A different aspect of spin-mixing dynamics in the context of atomic phase conjugation was reported by E. V. Goldstein and P. Meystre, cond-mat/9806165.
[12] T. Ohmi and K. Machida, cond-mat/9803160.
[13] W. Zhang and D. F. Walls, Phys. Rev. A **57**, 1248 (1998).
[14] J. P. Burke, Jr., C. H. Greene, and J. L. Bohn, Phys. Rev. Lett. **81**, 3355 (1998).
[15] For an initially spin-polarized sodium condensate, our numerical tests indicate that the single mode approximation is good for particle number N up to 10^5 in a spherical trap with a trap frequency 100 Hz. However, appreciable higher modes excitations may occur for larger N or for condensates with negative λ_a'. Detailed calculations including the time evolution of the spatial wave functions will be presented elsewhere.
[16] Our further calculations indicate that $\langle \Delta \hat{n}_j \rangle$ also become steady for $t > t_c$. These particle number fluctuations (for $t > t_c$) are found to be quite significant: $\langle \Delta \hat{n}_1 \rangle = \langle \Delta \hat{n}_1 \rangle = \langle \Delta \hat{n}_0 \rangle / 2 \approx 0.18N$.