

Laser-catalyzed spin-exchange process in a Bose-Einstein condensate

H. Jing,^{1,2} Y. Jiang,¹ Weiping Zhang,² and P. Meystre³

¹*Department of Physics, Henan Normal University, Xinxiang 453007, People's Republic of China*

²*State Key Laboratory of Precision Spectroscopy, Department of Physics, East China Normal University, Shanghai 200062, People's Republic of China*

³*B2 Institute and Department of Physics, The University of Arizona, Tucson, Arizona 85721, USA*

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We show theoretically that it is possible to optically control collective spin-exchange processes in spinor Bose condensates through virtual photoassociation. The interplay between optically induced spin exchange and spin-dependent collisions provides a flexible tool for the control of atomic spin dynamics, including enhanced or inhibited quantum spin oscillations, the optically induced ferromagnetic-to-antiferromagnetic transition, and coherent matter-wave spin conversion.

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Spinor Bose-Einstein condensates, consisting of atoms with internal spin states, provide a promising bridge between atomic, molecular, and optical physics, matter-wave optics, many-body physics, and quantum information science [1]. Beside the study of their rich ground-state properties [2] and spatial spin structures [3–5], the magnetic control of quantum spin mixing has been a central topic of investigations for atomic spin systems [6–13]. Also, by steering the spin degrees of freedom in ultracold quantum gases, the creation of topological skyrmions, Dirac monopoles, dark solitons, and quantum entanglement has been investigated [14–16].

In parallel to these works, which concentrate largely on the role of external magnetic fields on spinor condensates, there have also been important developments on their magneto-optical manipulation. For example, Zhang *et al.* [17] studied the spin waves induced by light-induced dipole-dipole interactions in an atomic spin chain trapped in a lattice potential. More recently, advances in coherent photoassociation (PA) at ultracold temperatures [18] were exploited in theoretical and experimental studies of spin mixing and spin-dependent PA in spin-1 condensates [19,20]. In very recent work, Kobayashi *et al.* observed the spin-selective formation of spinor molecules in ferromagnetic atoms ⁸⁷Rb [21].

In this Rapid Communication, we demonstrate theoretically the optical control of atomic spin mixing in a ferromagnetic spin-1 Bose gas. In the proposed method the optical fields induce virtual PA processes in the atoms, for example, via a dark molecular state. We show that this step, which can be intuitively coined a laser-catalyzed spin-exchange process (LCSE), opens up an efficient and well-controlled optical channel for coherent atomic spin mixing. By tuning the strength ratio of these two channels of LCSE and spin-dependent collisions, three different regimes can be identified in the laser-controlled quantum spin dynamics, that is, going from the collision-dominated regime to the no-spin-mixing regime, and to the laser-induced antiferromagnetic regime, which is reminiscent of the prominent role of long-range dipole-dipole interaction in a dipolar spin gas (by changing the ratio of spin-dependent collision and dipolar interaction). As a result, and in contrast to the collision-dominated “single-channel” case, a wealth of important new effects arise in the spin dynamics of the atoms.

For concreteness we concentrate on two limiting situations, the adiabatic far off-resonant regime and the resonant case [22]. Our purpose here is to show that the interplay of two channels, the spin-dependent collisions and LCSE, provides a flexible tool for the control of the atomic spin dynamics, including enhanced and inhibited quantum spin oscillations, laser-induced ferromagnetic-to-antiferromagnetic (F-AF) transitions, and efficient coherent spin transfer triggered even by quantum vacuum noise. This method can be also extended to study, for example, the optical control of domain formation and of spin textures in a spinor gas [4]. As such, optical LCSE-controlled spinor condensates provide a promising new tool for the study of collective chemically driven quantum spin dynamics.

Figure 1 illustrates the process under consideration, the dynamics of a spin-1 atomic condensate resulting from the virtual PA of two $m_F = 0$ atoms into a molecular state, followed by dissociation into a pair of $m_F = -1$ and $m_F = +1$ atoms. Accounting in addition for spin-dependent collisions between atoms, this system is described by the Hamiltonian ($\hbar = 1$)

$$H = H_{\text{coll}} + H_{\text{pa}}, \quad (1)$$

where H_{coll} and H_{pa} refer to spin-dependent collisions and the controllable light-assisted interactions, respectively, with

$$H_{\text{coll}} = \int d\mathbf{r} \left[\hat{\psi}_i^\dagger \left(-\frac{\hbar^2}{2m} \nabla^2 + V + E_i \right) \hat{\psi}_i + \frac{c'_0}{2} \hat{\psi}_i^\dagger \hat{\psi}_j^\dagger \hat{\psi}_j \hat{\psi}_i + \frac{c'_2}{2} \hat{\psi}_k^\dagger \hat{\psi}_i^\dagger (F_\gamma)_{ij} (F_\gamma)_{kl} \hat{\psi}_j \hat{\psi}_l \right], \quad (2)$$

$$H_{\text{pa}} = \int d\mathbf{r} \left[\Delta' \hat{\psi}_m^\dagger \hat{\psi}_m + \Omega_p (\hat{\psi}_0^{\dagger 2} \hat{\psi}_m + \hat{\psi}_m^\dagger \hat{\psi}_0^2) - \Omega_d (\hat{\psi}_m^\dagger \hat{\psi}_+ \hat{\psi}_- + \hat{\psi}_-^\dagger \hat{\psi}_+ \hat{\psi}_m) \right]. \quad (3)$$

Here $\hat{\psi}_i(\mathbf{r})$ is the annihilation operator of the i th component atom ($i = 0, \pm 1$) and $\hat{\psi}_m(\mathbf{r})$ the corresponding operator for the intermediate molecular field, V is the trap potential, E_i is the Zeeman shift, and $F_{\gamma=x,y,z}$ is the spin-1 matrix [3]. The coefficients $c'_0 = 4\pi\hbar^2(a_0 + 2a_2)/3m$ and $c'_2 = 4\pi\hbar^2(a_2 - a_0)/3m$ give the strength of the spin-conserving two-body collisions, with a_0, a_2 the scattering lengths of the accessible collision channels and m the atomic mass. The Rabi frequencies

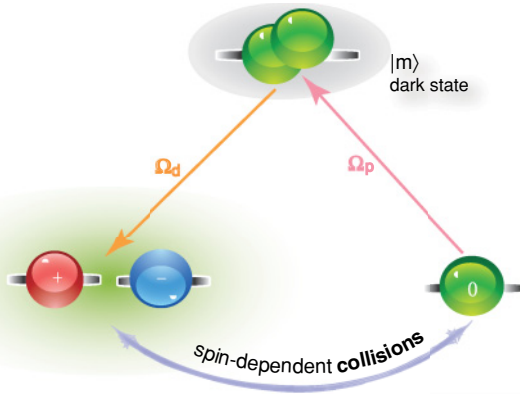


FIG. 1. (Color online) Schematic of coherent two-channel spin-exchange interactions in an optically controlled spin-1 Bose gas. In addition to the familiar collisional channel, the optical channel proceeds via the virtual PA of two $|m_F = 0\rangle$ atoms into an intermediate molecular state $|m\rangle$, which dissociates into a pair of $|m_F = +\rangle$ and $|m_F = -\rangle$ atoms.

$\Omega_{p,d}$ describe the strength of the PA of $m_F = 0$ atoms and dissociation into $m_F = \pm 1$ atoms, and Δ' is the detuning between the molecular and the atomic states.

We first consider the adiabatic off-resonant regime by assuming that Δ' is the largest parameter in the system. From $\partial\hat{\psi}_m/\Delta't \simeq 0$, we have $\hat{\psi}_m \simeq \frac{\Omega_d\hat{\psi}_+\hat{\psi}_- - \Omega_p\hat{\psi}_0^2}{\Delta'}$, and substitute this form into the Heisenberg equations of motion derived from Eq. (1) [23]. It is easily seen that the resulting equations can also be derived from the effective Hamiltonian

$$\mathcal{H}_{\text{eff}} = H_{\text{coll}} + \int d\mathbf{r} [\Omega'(\hat{\psi}_+^\dagger\hat{\psi}_-^\dagger\hat{\psi}_0^2 + \hat{\psi}_0^{\dagger 2}\hat{\psi}_+\hat{\psi}_-) + \mathcal{O}], \quad (4)$$

with

$$\Omega' = \Omega_p\Omega_d/\Delta', \quad \mathcal{O} = -\frac{\Omega_p^2}{\Delta'}\hat{\psi}_0^{\dagger 2}\hat{\psi}_0^2 - \frac{\Omega_d^2}{\Delta'}\hat{\psi}_-^\dagger\hat{\psi}_+^\dagger\hat{\psi}_+\hat{\psi}_-.$$

In addition to the familiar spin-dependent collisions, spin coupling now also results from optical LCSE, resulting in the two-channel spin-exchange Hamiltonian

$$\mathcal{H} = \mathcal{C}' \int d\mathbf{r} [\hat{\psi}_+^\dagger(\mathbf{r}, t)\hat{\psi}_-^\dagger(\mathbf{r}, t)\hat{\psi}_0(\mathbf{r}, t)\hat{\psi}_0(\mathbf{r}, t) + \text{H.c.}].$$

Here $\mathcal{C}' = \Omega' + c_2'$ describes the combined effect of spin-dependent collisions and optical LCSE. Clearly, the effective spin-coupling strength \mathcal{C}' can be negative or positive with suitable optical parameters, leading to significantly different spin dynamics.

In the limit where the spatial degrees of freedom decouple from the spinor dynamics, that is, when the spin healing length is larger than the condensate size, it is possible to invoke the single-mode approximation [9,13] where $\hat{\psi}_i(\mathbf{r}, t) \rightarrow \phi(\mathbf{r})\hat{a}_i(t)$, $\hat{\psi}_m(\mathbf{r}, t) \rightarrow \phi(\mathbf{r})\hat{m}(t)$, where $\phi(r)$ is the spatial wave function of the condensate with \hat{a}_i and \hat{m} being the atomic or molecular annihilation operators. The rest of this article presents results obtained in this approximation. For convenience we also introduce the scaled parameters $c_{0,2} = c'_{0,2} \int d\mathbf{r} |\phi(\mathbf{r})|^4$, $\Omega = \Omega' \int d\mathbf{r} |\phi(\mathbf{r})|^4$, $\Delta = \frac{\Omega_p^2}{\Delta'} \int d\mathbf{r} |\phi(\mathbf{r})|^4$, and $\tau = c_0 n t$, where n is the initial atomic density.

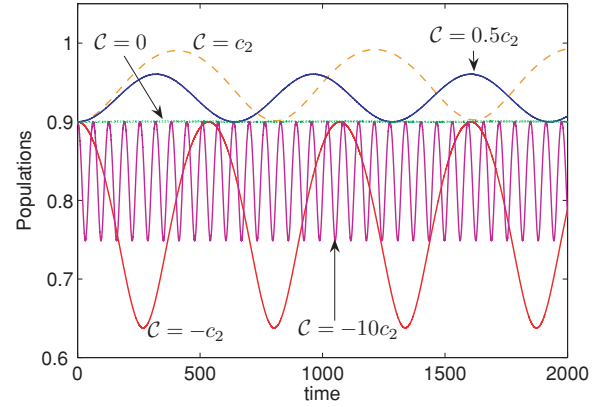


FIG. 2. (Color online) Population of spin-zero atoms with (solid lines) or without (dashed lines) laser fields. In all cases the atomic density is $2 \times 10^{14} \text{ cm}^{-3}$ [24], and the time is scaled to $\tau = c_0 n t$, corresponding to the time unit 0.1 ms.

The mean-field evolution of the spin-0 atomic population n_0 is illustrated in Fig. 2 for several values of $\mathcal{C} = \Omega + c_2$ and for the initial state $(n_+, n_0, n_-) = (0.05, 0.9, 0.05)$. The specific example of atoms ^{87}Rb is considered here, with $a_0 = (101.8 \pm 2)a_B$ and $a_2 = (100.4 \pm 1)a_B$ [25], where a_B is the Bohr radius. In our calculations, we assume the optical detuning $\Delta' = 100\Omega_p$ and $\Omega_d = 10\Omega_p$, which is reasonable for the adiabatic off-resonant case. The typical initial atomic density $n \sim 10^{14} \text{ cm}^{-3}$, corresponds to $c_0 n \sim 9700 \text{ Hz}$.

Significantly different regimes are reached by varying the optical detuning Δ' : (i) In the collision-dominated regime $|c_2| > |\Omega|$, the population of spin-0 atoms is always larger than its initial value 0.9. In this perturbed regime, the spin coupling is still ferromagnetic ($\mathcal{C} < 0$). (ii) For $\mathcal{C} = 0$, that is, $\Omega = -c_2$ (for $\Delta' > 0$), the two channels (spin-dependent collisions and LCSE) interfere destructively, leading to frozen or inhibited spin mixing. (iii) The sign of \mathcal{C} can be reversed by tuning the laser fields, resulting in an effective antiferromagnetic regime $\mathcal{C} > 0$. This is reminiscent of the F-AF transition induced by long-range dipolar interactions in a spinor gas [10]. In this reversed regime, the atomic spin oscillations ($|\Omega| > |c_2|$) turn out to be below the initial value 0.9.

The mean-field atomic spinor dynamics is well described by a nonrigid pendulum model [3]. By expressing the c -number amplitudes a_i in terms of real amplitudes and phases, that is, $a_{\pm,0} = \sqrt{n_{\pm,0}} e^{-i\theta_{\pm,0}}$, the energy functional of the optically controlled spinor system can be derived as

$$\mathcal{E} = q(1 - n_0) + \mathcal{C}\sqrt{(1 - n_0)^2 - m^2} \cos \theta + c_2 n_0(1 - n_0) + \frac{\Delta}{4} n_0(2 - n_0) - \frac{\Omega^2}{\Delta} n_0^2, \quad (5)$$

where q denotes the quadratic Zeeman effect, $\theta = \theta_+ + \theta_- - 2\theta_0$ is the relative phase of the spin components, and $m = n_+ - n_-$ is the atomic magnetization. The last two terms in Eq. (4) account for the optical energy shift.

The effective spin-coupling parameter \mathcal{C} has an important impact on the properties of the system, as already mentioned. Figure 3 plots equal-energy contours in the phase space (θ, n_0) for several values of \mathcal{C} for $m = 0$ and $q = 0.01$, corresponding to the fixed magnetic field of about 460 mG in our specific

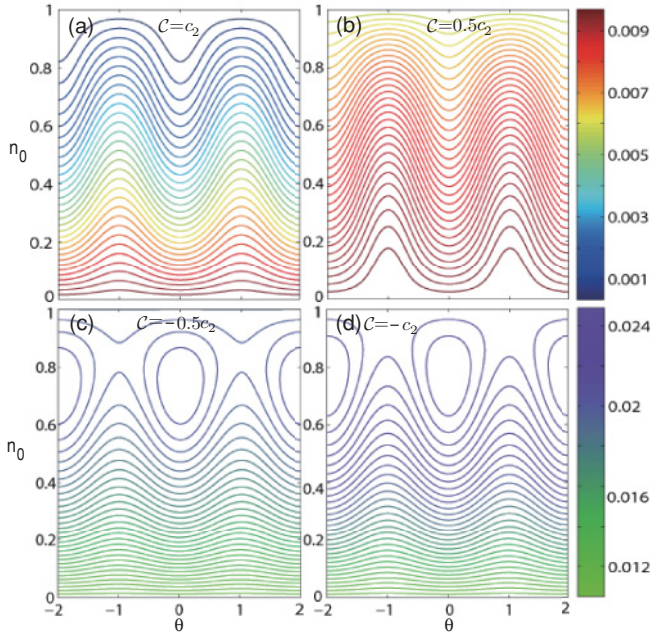


FIG. 3. (Color online) Equal-energy contours of \mathcal{E} without (a) ($C = c_2$) and with (b) ($C = 0.5c_2$), (c) ($C = -0.5c_2$), and (d) ($C = -c_2$) optically controlled LCSE for $q = 0.01$ and $m = 0$.

example, which is larger than the resonance magnetic field $B_{\text{res}} \sim 330$ mG (see [26]). We have also carried out numerical simulations for values of q corresponding to $B < B_{\text{res}}$ and found similar results for appropriate values of C . For our parameters only open trajectories exist in the absence of laser fields ($C = c_2$) as well as for perturbed case $C = 0.5c_2$. In contrast, the “reversed” cases $C = -0.5c_2$ and $C = -c_2$ are characterized by the coexistence of closed and open trajectories.

We remark that Eq. (5) also makes it possible to study the instabilities and domain formation in spin-1 atoms [3,5]. We actually have done this and found that the LCSE-dominated case is again different from the collision-dominated case [27].

We now turn to the resonant situation, where one can exploit the existence of an atom-molecule dark state to prevent the buildup of a significant molecular population throughout the process [18]. This two-photon resonant control of atomic spinor dynamics represents a promising step in developing the field of all-optical manipulation of matter-wave spins. In a mean-field approach where $\hat{\psi}_i \rightarrow \sqrt{n}\phi_i$ [19], we find

$$\begin{aligned}
 \frac{d\phi_+}{d\tau} &= -i[c_2(|\phi_+|^2 + |\phi_0|^2 - |\phi_-|^2)]\phi_+ \\
 &\quad - ic_2\phi_0^2\phi_-^* + i\Omega'_d\phi_m\phi_-^* - i(\Theta + \delta)\phi_+, \\
 \frac{d\phi_0}{d\tau} &= -i[c_2(|\phi_+|^2 + |\phi_-|^2)]\phi_0 - 2ic_2\phi_+\phi_-\phi_0^* \\
 &\quad - 2i\Omega'_p\phi_m\phi_0^*, \\
 \frac{d\phi_-}{d\tau} &= -i[c_2(|\phi_-|^2 + |\phi_0|^2 - |\phi_+|^2)]\phi_- \\
 &\quad + i\Omega'_d\phi_m\phi_+^*, \\
 \frac{d\phi_m}{d\tau} &= i\Omega'_d\phi_+\phi_- - i\Omega'_p\phi_0^2 - (i\delta + \gamma)\phi_m,
 \end{aligned} \tag{6}$$

where $\Omega'_p = \Omega_p/c_0\sqrt{n}$, $\Omega'_d = \Omega_d/c_0\sqrt{n}$, $\Theta = \Delta'/c_0n$, $\delta = \delta'/c_0n$, δ is the frequency difference between the molecular state and the atomic hyperfine states, and the phenomenological decay rate γ accounts for the loss of intermediate molecules. Following the method of Ref. [22], we find that Eqs. (7) admit a steady-state coherent population trapping (CPT) solution ($n_m = 0$),

$$n_{\pm,s} = \frac{1}{2 + \Omega'_d/\Omega'_p}, \quad n_{0,s} = \frac{1}{1 + 2\Omega'_p/\Omega'_d},$$

under the generalized two-photon resonance condition

$$\Theta(t) = -\delta + c_2[2(n_{+,s} + n_{-,s}) + 2\sqrt{n_{+,s}n_{0,s}} - 4n_{0,s}].$$

This time-dependent resonance condition is determined by the CPT steady-state values of the atomic density, which thereby can be tuned by choosing suitable pumping and (time-dependent) dumping laser fields.

Figure 4 shows the populations of the atomic sublevels for $\Omega'_p = 1$ (corresponding to 9700 Hz) and

$$\Omega'_d(t) = \Omega'_{d,0}\text{sech}(t/t_0),$$

with $\Omega'_{d,0} = 40$, $t_0 = 20$, the other parameters being as in Fig. 2. In contrast to the off-resonant spin oscillations, we have now a full transfer of population from the initial state, say $|m_F = 0\rangle$ to a final coherent superposition of $|m_F = \pm 1\rangle$ hyperfine states. We have carried out numerical simulations for a large set of initial seeds of spin- \pm atoms and found that the stable spin conversion is always possible for, for example, $\delta = 3$. The departure of the spin- \pm populations from the ideal CPT value is due to the fact that only an approximate adiabatic condition exists for the CPT state [22].

Following the preparation of all atoms in the spin-0 state, Klempt *et al.* [28] have recently studied the magnetic field dependence of the fraction of atoms transferred to the spin- \pm states. This transfer process is of a quantum nature as the mean-field equations break down for an initial vacuum of spin- \pm atoms. Our system also makes it possible to study quantum spin transfer with a quantum-noise-triggered seed. Following

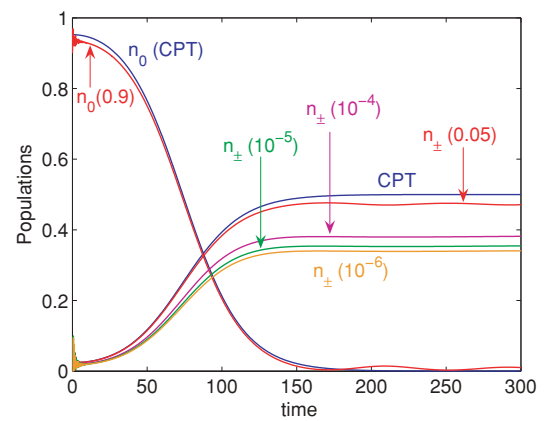


FIG. 4. (Color online) Atomic populations in the resonant regime for ^{87}Rb , with detuning $\delta = 3$ and $\gamma = 1$. To illustrate the role of small seeds of atoms n_{\pm} , several values of the initial populations of the corresponding spin states have been considered, as denoted in the parentheses. The line labeled “CPT” shows the ideal population of the target states $|m_F = \pm 1\rangle$.

the strategy familiar from quantum optics (see, e.g., [1,29,30]), we separate the problem into an initial stage dominated by quantum noise, followed by a classical stage that arises once the target state has acquired a macroscopic population [31]. We have performed an analysis of the present system along these lines, and it confirms that its evolution from quantum noise is also efficient, with dynamics similar to those with the classical seed $n_{\pm} = 10^{-5}$ in Fig. 4 [27,31].

In conclusion, we have demonstrated theoretically that the interplay of LCSE and spin-dependent collisions leads to a variety of remarkable effects such as inhibited spin oscillations, laser-induced F-AF transition, and quantum spin transfer between different components. The optical tuning or even elimination of spin-exchange interactions may be used to, for example, probe the weak signal of dipolar interactions, which is generally far smaller than the spin-dependent collisions [10]. The optical control of atomic spin interactions is somewhat reminiscent of the role of magnetic Feshbach resonances in the study of ultracold gases. This work hints at promising possibilities to carry out an all-optical control of

atomic spins [32]. When compared to the magnetic control of spinor atoms [1–16], LCSE offers an exciting new route to the study of atomic spin coupling independent of the collisions. Ultimately, magneto-optical methods will likely combine the best of both optical and purely magnetic approaches. Future work will study the magneto-optical control of spatial spin structures and explore possibilities of spin-dependent ultracold chemistry or atom-molecule hybrid spin mixing.

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