Coherent two-color photoassociation in a spinor-1 Bose-Einstein condensate: Single-spin versus mixed-spin cases

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We investigate the coherent two-color photoassociation (PA) process in a spinor-1 atomic Bose-Einstein condensate by comparing the single-spin and mixed-spin cases. We find that, by tuning the PA light applied on different spin components, one can observe quite different features for the atomic spin-mixing dynamics (with weak PA field) or coherent atom-molecule conversion (with strong PA field), indicating a research field of so-called superchemistry or coherent PA in a spinor gas.

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Coherent atom-molecule conversion in ultracold degenerate matter-wave gases, sometimes also termed as superchemistry [1], describes the collective assembly of homonuclear or heteronuclear molecules with bosonic or fermionic atoms or some Bose-Fermi mixture by using the magnetic Feshbach resonance (FR) or the optical photoassociation (PA) technique [2]. The FR drives a colliding atom pair to jump from the free atomic state to a bound molecular state, while the PA works when two atoms are combined with one photon to create a bound-state molecule. The underlying physics for these non-Arrhenius superchemistry reactions can be understood by laserlike Bose enhancement or complex many-body effect with fermionic components [3] and a variety of novel quantum effects have been predicted such as the molecular Rabi oscillations [4], the superselectively rule in triatomic molecular dissociation [5], or the confinement-induced dissociation [6]. In very recent experiments, the atom-molecule dark state [7,8] and the rovibrational molecular ground state have also been realized. Most of these previous studies are focused on the single-component atomic condensate [9] and thus we are strongly motivated to generalize to study the superchemistry process in a spinor-1 Bose condensate.

The spin degrees of freedom of the polarized ultracold particles become accessible when a far-off-resonant optical trap is used to provide the confinement for all Zeeman states instead of the magnetic traps [10]. The spinor Bose condensate or the superfluid with internal spins has been observed for, e.g., $F=1$ [11] or $F=2$ [12] ^{23}Na or ^{87}Rb [13] atoms, together with the novel effects of spin domains, interdomain tunneling [14], coreless vortex states [15], or spin-mixing dynamics [16]. Very recently, we proposed to study the spin-mixing dynamics in the presence of an attenuated PA light applied on a single-spin component [17]. Almost at the same time, Hamley *et al.* [18] experimentally studied the single-spin and mixed-spin PA spectroscopy. However, the different roles of single-spin and mixed-spin PA on the atomic spin-mixing dynamics and even the atom-dimer conversion are still not probed.

In this paper, we study these interesting problems by considering a coherent two-color PA process in a spinor-1 condensate. As Fig. 1 illustrates, three hyperfine states in the lowest energy manifold of spinor-1 atoms are initially prepared in an optical trap and then we consider the free-quasibound-bound coupling with the so-called counterintuitive order of pulses for the single-spin (a) or mixed-spin (b) component. Obviously, each case contains three different possibilities of associating atom pairs, such as the case of $|0\rangle+|0\rangle$ or $|+\rangle+|-\rangle$ as Fig. 1(a) or 1(b) shows.

Single-spin PA case. As Fig. 1(a) shows, we first consider the single-spin PA case for the ferromagnetic ^{87}Rb atoms initially prepared in spin state $|F=1,m_F=0\rangle$. Two atoms in this state can be reversibly scattered into other two states $|m_F=1\rangle$ and $|m_F=-1\rangle$ [19] due to spin-exchange collisions or combined into the molecular ground state $|g\rangle$ due to the external PA couplings with pumping or dumping Rabi frequencies Ω_p or Ω_d. This system is described by the interacting Hamiltonian

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_{\text{col}} + \hat{H}^I_{\text{PA}}, \tag{1}$$

where $\hat{\mathcal{H}}_{\text{col}}$ and $\hat{H}^I_{\text{PA}}$ describe the parts of atomic spin mixing [16] and PA-induced atom-molecule conversion, respectively ($\hbar=1$),

$$\hat{\mathcal{H}}_{\text{col}} = \frac{c_0}{2}\sum \hat{a}_i^\dagger\hat{a}_j^\dagger\hat{a}_i\hat{a}_j + \frac{c_2}{2}(\hat{a}_+^{\dagger 2}\hat{a}_+^2 + \hat{a}_-^{\dagger 2}\hat{a}_-^2 - 2\hat{a}_+^\dagger\hat{a}_-^\dagger\hat{a}_-\hat{a}_+ + 2\hat{a}_+^\dagger\hat{a}_0^\dagger\hat{a}_0\hat{a}_+ + 2\hat{a}_-^\dagger\hat{a}_0^\dagger\hat{a}_0\hat{a}_- + 2\hat{a}_0^{\dagger 2}\hat{a}_+\hat{a}_- + 2\hat{a}_+^\dagger\hat{a}_-^\dagger\hat{a}_0^2), \tag{2}$$

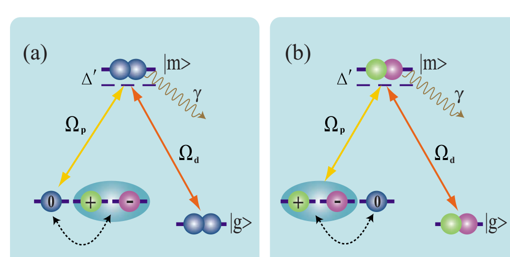

FIG. 1. (Color online) Schematic diagram of (a) single-spin or (b) mixed-spin superchemistry process in a spinor-1 Bose condensate. Besides the spin-exchange collisions, one can apply a free-quasibound-bound coupling, with the detuning Δ' and the decaying rate γ for the intermediate excited state $|m\rangle$.

$$\hat{H}^{I}_{\mathrm{PA}} = \left(\delta - \frac{1}{2}i\gamma\right)\hat{m}^\dagger\hat{m} - \Omega_p(\hat{a}_0^{\dagger 2}\hat{m} + \hat{m}^\dagger\hat{a}_0^2) - \Omega_d(\hat{m}^\dagger\hat{g} + \hat{g}^\dagger\hat{m}), \tag{3}$$

where $\hat{a}_{i,j}$ $(i,j=0,\pm)$, $\hat{m}$, and $\hat{g}$ are the annihilation operators of the atoms in state $|i\rangle$, the molecules in the excited state $|m\rangle$, and the ground state $|g\rangle$, respectively; the density-dependent and the spin-dependent parameters c_0 and c_2 are determined by the atomic s-wave scattering length, i.e., $c_0 = 4\pi\hbar^2(a_0+2a_2)/3M$, $c_2=4\pi\hbar^2(a_2-a_0)/3M$ (M is the atomic mass). We note that c_2, generally much smaller than c_0 [16], can be well tuned by using a PA light (see Ref. [18]). $\gamma=\gamma_s+\gamma_d$ denotes both the spontaneous and dissociative decaying of the intermediate state $|m\rangle$ with two-photon detuning δ. The collisions involving molecules are ignored since they are yet unknown in current experiments.

The intermediate excited state can be adiabatically eliminated by assuming δ as the largest parameter of this system [20]. Using $i\dot{\hat{m}}/\delta \approx 0$, we have

$$\hat{m} \simeq \left(\frac{\Omega_d\hat{g} + \Omega_p\hat{a}_0^2}{\delta}\right)\left(1 + i\frac{\gamma}{2\delta}\right),$$

and thus the effective Hamiltonian of PA coupling [20]

$$\hat{\mathcal{H}}^{I}_{\mathrm{PA}} = -\Delta\hat{g}^\dagger\hat{g} + \Omega(\hat{a}_0^{\dagger 2}\hat{g} + \hat{a}_0^2\hat{g}^\dagger) - \alpha\hat{a}_0^{\dagger 2}\hat{a}_0^2, \tag{4}$$

with

$$\Delta = (\Omega_d^2/\delta)(1 + i\gamma/2\delta),\ \Omega = \Omega_p\Omega_d/\delta,\ \alpha = \Omega_p^2/\delta.$$

In order to clearly see the new features of the atomic spin mixing in the presence of the PA field, we first write the equations of motion from Eq. (1) in the framework of the standard mean-field approach by replacing the operators by the c numbers: $\hat{a}_i \to a_i\sqrt{n}$, $\hat{g} \to g\sqrt{n}$, where n is the density of the total particle number

$$i\frac{da_+}{d\tau} = [1 - 2|g|^2 + c(|a_+|^2 + |a_0|^2 - |a_-|^2)]a_+ + ca_0^2a_-^*,$$

$$i\frac{da_0}{d\tau} = [1 - \delta' - 2|g|^2 + c(|a_+|^2 + |a_-|^2)]a_0 + 2ca_+a_-a_0^* - 2\alpha|a_0|^2a_0 + 2\Omega a_0^*g,$$

$$i\frac{da_-}{d\tau} = [1 - 2|g|^2 + c(|a_-|^2 + |a_0|^2 - |a_+|^2)]a_- + ca_0^2a_+^*,$$

$$i\frac{dg}{d\tau} = \Omega a_0^2 - \Delta g, \tag{5}$$

where we have taken $\tau=c_0nt$ as the scaled time and thus the scaling transformations $c_2/c_0 \sim c$, $\Delta/c_0n \sim \Delta$, $\Omega/c_0\sqrt{n} \sim \Omega$, and $\alpha/c_0 \sim \alpha$.

Mixed-spin PA case. As Fig. 1(b) shows, the mixed-spin PA case can be similarly described by

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_{\mathrm{col}} + \hat{\mathcal{H}}^{II}_{\mathrm{PA}}, \tag{6}$$

where $\hat{\mathcal{H}}_{\mathrm{col}}$ is the same as Eq. (2) and $\hat{\mathcal{H}}^{II}_{\mathrm{PA}}$ denotes the molecular creation from two atomic components with opposite spins ($|+\rangle+|-\rangle \to |g\rangle$), i.e.,

$$\hat{\mathcal{H}}^{II}_{\mathrm{PA}} = -\Delta\hat{g}^\dagger\hat{g} + \Omega(\hat{a}_+^\dagger\hat{a}_-^\dagger\hat{g} + \hat{g}^\dagger\hat{a}_+\hat{a}_-) - \alpha\hat{a}_+^\dagger\hat{a}_-^\dagger\hat{a}_+\hat{a}_-. \tag{7}$$

The corresponding equations of motion in the framework of the standard mean-field approach can be written as

$$i\frac{da_+}{d\tau} = [1 - 2|g|^2 + c(|a_+|^2 + |a_0|^2 - |a_-|^2)]a_+ + ca_0^2a_-^* + \Omega a_-^*g - \alpha|a_+|^2a_-,$$

$$i\frac{da_0}{d\tau} = [1 - \delta' - 2|g|^2 + c(|a_+|^2 + |a_-|^2)]a_0 + 2ca_+a_-a_0^*,$$

$$i\frac{da_-}{d\tau} = [1 - 2|g|^2 + c(|a_-|^2 + |a_0|^2 - |a_+|^2)]a_- + ca_0^2a_+^* + \Omega a_+^*g - \alpha|a_-|^2a_+,$$

$$i\frac{dg}{d\tau} = \Omega a_+a_- - \Delta g. \tag{8}$$

The other two sets of equations for single-spin or mixed-spin PA cases ($|0\rangle+|-\rangle \to |g\rangle$, $|0\rangle+|+\rangle \to |g\rangle$) can be similarly derived and facilitate numerical simulations and full comparisons of all the possible cases. In the following, we focus on the dynamics of the system in two particularly interesting regimes: the atomic spin-mixing regime with weak PA fields and then the atom-dimer conversion regime with strong PA fields.

Atomic spin-mixing regime with weak PA fields. Figure 2 shows the typical numerical results for single-spin and mixed-spin cases with weak PA fields. The main feature of this regime is that while the extremely small number of created molecules can be safely neglected, the atomic spin-mixing dynamics are largely influenced by the external PA light. To be specific, we found that for the single-spin PA $|0\rangle+|0\rangle \to |g\rangle$ (with $\Delta=-2$ and $\Omega=0.05$), rapid oscillations can be observed for the atomic populations in the state $|0\rangle$ (in comparison to the well-known case without any PA field, see Fig. 2(a) [19]); however, for the mixed-spin cases ($|+\rangle+|-\rangle \to |g\rangle$ or $|0\rangle+|-\rangle \to |g\rangle$), these oscillations occur in the two components: $|+\rangle$, $|-\rangle$ [Fig. 2(b)] or $|0\rangle$, $|+\rangle$ [Fig. 2(c)]. In contrast, no rapid oscillations can happen if one chooses the positive detuning $\Delta=2$ [Fig. 2(d)].

Similar features are also found for other PA cases. In fact, we have carried out our numerical simulations for a large set of other values of Ω and Δ and found the essentially same features as Fig. 2 (more rapid oscillations for stronger PA light with the same conditions).

Atom-dimer conversion regime for strong PA field. In the presence of a strong PA field, the coherent PA process is dominating over the collision-induced atomic spin mixing. For the specific examples, we plotted the populations of dimers for the cases of single-spin and mixed-spin PAs in

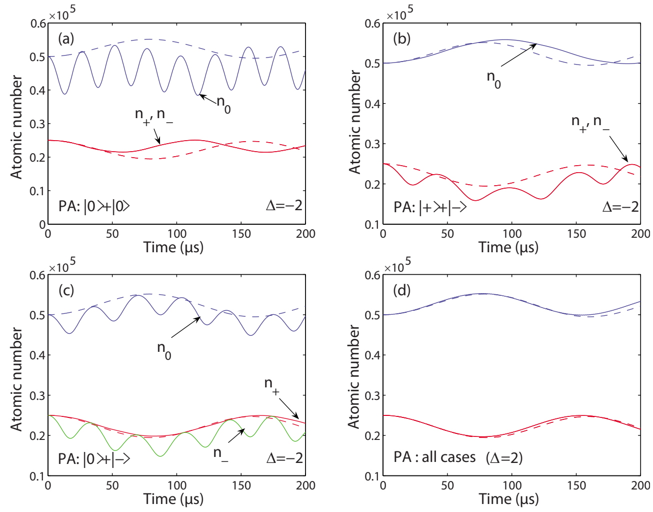

FIG. 2. (Color online) Population of atoms (solid lines) with a weak PA field ($\Omega=0.05$). Dashed lines denote the case without the PA field. The initial atomic states are $f_0=[\sqrt{0.25},\sqrt{0.5},\sqrt{0.25},0]$. (a)–(c) The weak PA field can be applied to two atoms in the same or different spin states. (d) A positive detuning Δ, however, can lead to a suppression of the weak PA effect.

Fig. 3. The PA process $|+\rangle+|-\rangle\rightarrow|g\rangle$ is very similar to that of $|0\rangle+|0\rangle\rightarrow|g\rangle$ [Fig. 3(a)], but quite different from the cases of $|0\rangle+|\pm\rangle\rightarrow|g\rangle$ [Fig. 3(b)]. We have also calculated the single-spin cases $|\pm\rangle+|\pm\rangle\rightarrow|g\rangle$ and found quite different features in comparison with that of $|0\rangle+|0\rangle\rightarrow|g\rangle$. This suggests that the atom-dimer conversion in a spinor gas strongly depends on the total spin of the closed-channel dimers and the two-photon detuning Δ [Figs. 3(c) and 3(d)]. Another feature in Fig. 3 is that for each certain value of Δ, an optimal PA field strength Ω can exist for a largest conversion rate $\eta=N_g/N_{\rm tol}$ (N_g is the number of dimers and $N_{\rm tol}$ is the total number of the initial atoms). Clearly, new features can be observed in the creation of molecules from a spinor gas with respect to the familiar scalar gas case.

In the above numerical calculations, we have used the parameters of ^{87}Rb atoms: $a_0=101.8(2)a_B$, $a_2=100.4(1)a_B$, where $a_B\approx0.529$ Å is the Bohr atomic radius. The initial atomic density n is taken as 2×10^{14} cm^{-3}, leading to $c_0n\approx1$ MHz, $c=c_2/c_0\sim-0.005$. The other parameters are $\delta=150\times2\pi$ MHz and $\gamma/\delta=0.08$ [21].

We note that, according to a very recent experiment [18], the value of c_2 can also be widely tuned by the PA light (even from the ferromagnetic to the antiferromagnetic regimes). We thus carried out our numerical procedures for $c\in[-0.02,0.02]$ and found that the atomic spin-mixing dynamics can be largely changed (with weak PA field) but no obvious change for coherent atom-dimer conversion (with strong PA field).

In summary, we have investigated the two-color coherent PA process in a spinor-1 atomic Bose gas, focusing on the different effects of single-spin and mixed-spin PA on the dynamics of atomic spin mixing (for the regime of weak PA field) or atom-dimer conversion (for the regime of strong PA field). In comparison to the familiar cases of atomic spin mixing without PA or the atom-dimer conversion in a scalar gas, the PA light in different spin components, leads to some features indicating a research field of so-called superchemistry or coherent PA in a spinor gas. In our future work we plan

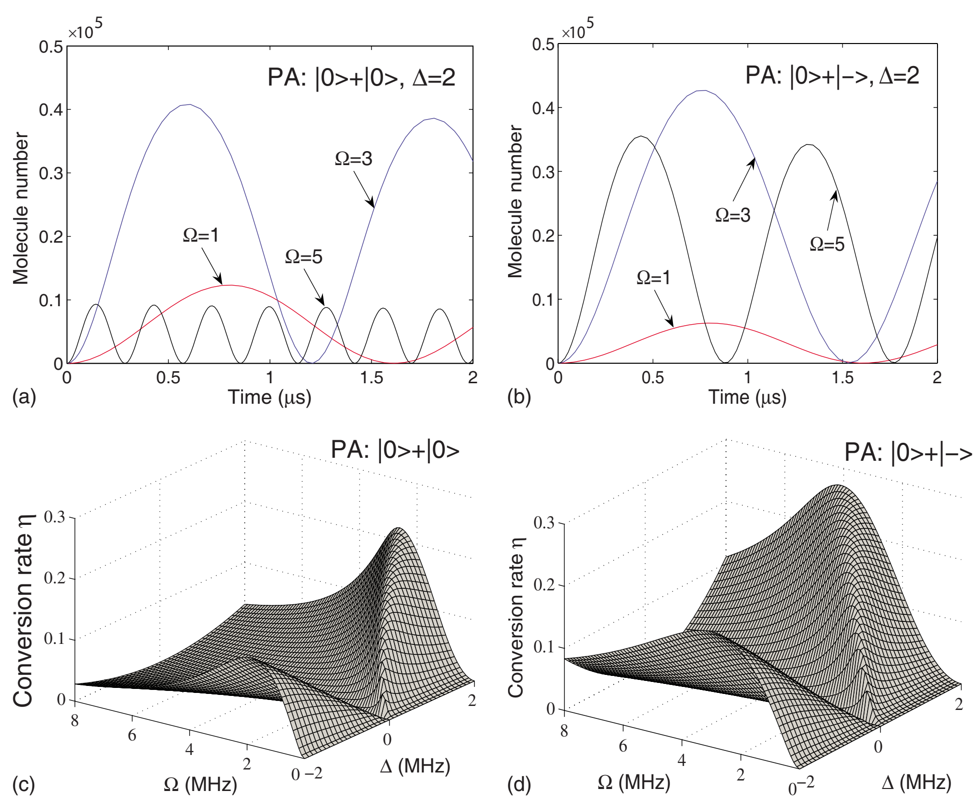

FIG. 3. (Color online) The molecule number and conversion rate with a strong PA field for single-spin or mixed-spin case.

to study the atomic spin mixing with the aid of coherent abstraction reaction [22], the coherent PA in a spinor-2 gas, and the possible effects of rogue photodissociation [23].

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