Spinor atom-molecule conversion via laser-induced three-body recombination

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Phys. Rev. A **83**, 043601 — Published 1 April 2011

DOI: 10.1103/PhysRevA.83.043601

Spinor atom-molecule conversion via laser-induced three-body recombination

H. Jing[1,2], Y. Deng[1], and P. Meystre[21]

[11] *Department of Physics, Henan Normal University, Xinxiang 453007, China*
[2] *B2 Institute, Department of Physics and College of Optical Sciences, The University of Arizona, Tucson, Arizona 85721*

We study theoretically several aspects of the dynamics of coherent atom-molecule conversion in spin-1 Bose-Einstein condensates. Specifically, we discuss how for a suitable dark-state condition the interplay of spin-exchange collisions and photoassociation leads to the stable creation of an atom-molecule pairs from three initial spin-zero atoms. This process involves *two* two-body interactions and can be intuitively viewed as an effective three-body recombination. We investigate the relative roles of photoassociation and of the initial magnetization in the "resonant" case where the dark state condition is perfectly satisfied. We also consider the "non-resonant" regime, where that condition is satisfied either approximately – the so-called adiabatic case – or not at all. In the adiabatic case, we derive an effective non-rigid pendulum model that allows one to conveniently discuss the onset of an antiferromagnetic instability of an "atom-molecule pendulum," as well as large-amplitude pair oscillations and atom-molecule entanglement.

PACS numbers: 42.50.-p, 03.75.Pp, 03.70.+k

I. INTRODUCTION

Recent years have witnessed rapid advances in the manipulation of the spin degrees of freedom of ultracold atoms [1–6]. By magnetically steering two-body collisions, a broad range of effects has been observed, including atomic magnetism [7–10], coherent spin mixing [5, 11], topological excitations [12], and an atomic analog of the Einstein-de Haas effect [13]. The *optical* control of atomic spin dynamics has also attracted much experimental interest [14–16]. For example, Dumke *et al.* [14] and Hamley *et al.* [15] have investigated the photoassociation (PA) diagnosis [14] and PA spectroscopy [15] of spin-1 atoms, opening the way to studies of PA-controlled regular [17] or chaotic [18] spin dynamics.

In a very recent experiment, the ro-vibrational ground-state molecules were successfully prepared via the all-optical association of laser-cooled atoms [19], which has triggered the investigation of coherent PA of a wide variety of ultracold atomic and molecular systems [20]. A result of particular relevance for the present study is an experiment by Kobayashi *et al.*, who used a coherent two-color PA technique to create spinor molecules in a spin-1 atomic Bose condensate [16]. In particular, these authors found that for strong PA couplings the atomic spin oscillations are significantly suppressed and the dominant process is scalar-like atom-molecule conversion. That is, only the populations of the spin components that are associated into molecules are observed to decrease, while the other spin component remains almost unchanged on the experimentally relevant timescale [16].

In this paper we show that under appropriate two-photon resonance conditions quantum interferences between optical PA and atomic spin mixing can lead to the existence of a dark state of the spin-down atoms, which can in turn be exploited in the stable formation of a spinor atom-molecule pair from three initial spin-zero atoms. This process, which involves *two* two-body interactions, can be thought of as an effective three-body spin-exchange effect. The important role of the initial magnetization in creating the atom-molecule pairs is also analyzed. We also analyze dynamical features that occur in the "non-resonant" regime where no dark state is formed, including large-amplitude coherent oscillations of the atom-molecule pairs population and an antiferromagnetic instability. As such, these manifestations of the interplay between two-color PA and spin-exchange collisions sheds significant new insight into the study of quantum spin gases and ultracold chemistry [20].

The article is organized as follows. Section II discusses the "resonant" situation where the dynamics of the system is characterized by the existence of a dark state. We first introduce our model, which we then apply to the description of scalar-like photoassociation [16]. We then derive a dark-state condition for the spin-down atoms and show that when satisfied, it results in the stable resonant creation of atom-molecule pairs. The role of the initial atomic magnetization is also discussed. Section III then turns to the non-resonant regime. We show that in that case the system can be described in terms of a non-rigid pendulum model. Two important dynamical manifestations of this regime, large-amplitude atom-molecule oscillations, and a regime of antiferromagnetic instability are explicitly discussed. Finally Section IV is a summary and conclusion.

II. THE MODEL

This section introduces our model and exploit it to describe the main features of scalar-like PA [16]. We also discuss a regime of stable atom-molecule pair formation, and analyze the role of initial magnetization in the system dynamics.

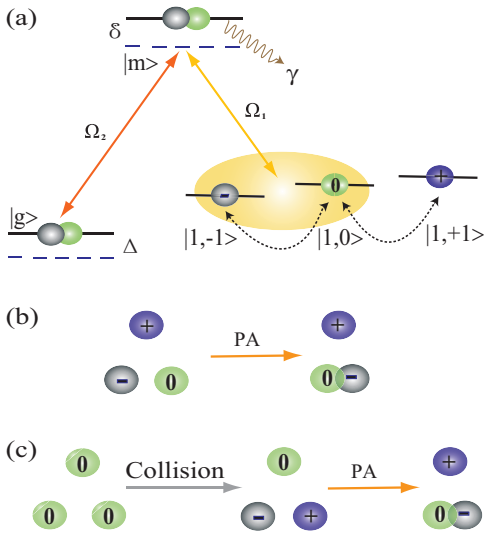

FIG. 1: (Color online). (a) Schematic of coherent two-color PA in a spin-1 atomic condensate. Here δ and Δ are the one- and two-photon detunings of the laser fields with Rabi frequencies $\Omega_{1,2}(t)$, and γ accounts for the spontaneous decay of the excited state $|m\rangle$. (b) Scalar-like atom-molecule conversion as observed in a recent experiment of Kobayashi *et al.* [16]. (c) Effective three-body recombination resulting from the interplay of *two* two-body interactions (see text).

A. Theoretical model

The system that we consider is illustrated in Fig. 1. It consists of a spin-1 atomic condensate undergoing spin-changing two-body collisions and coupled via 2-photon coherent PA to a ground-state diatomic molecular condensate.

Denoting by $\hat{\psi}_{i,j=0,\pm1}$ and $\hat{\psi}_{m,g}$ the annihilation operators of the three atomic components and of the excited or ground-state molecules, respectively, the Hamiltonian of the binary atomic and molecular condensate is ($\hbar = 1$)

$$\hat{H} = \hat{\mathcal{H}}_0 + \hat{\mathcal{H}}_{\rm coll} + \hat{\mathcal{H}}_{\rm PA}, \tag{1}$$

where

$$\begin{aligned}
\hat{\mathcal{H}}_0 &= \int d\mathbf{r} \left[\sum_{i=-1,0,1} \hat{\psi}_i^\dagger \left(V + E_i\right) \hat{\psi}_i \right. \\
&+ \left. \left(\delta - \frac{1}{2} i\gamma\right) \hat{\psi}_m^\dagger \hat{\psi}_m + (\Delta + \delta) \hat{\psi}_g^\dagger \hat{\psi}_g \right], \qquad (2) \\
\hat{\mathcal{H}}_{\rm coll} &= \frac{1}{2} \int d\mathbf{r} \left[\mathbf{c_0'} \hat{\psi}_\mathbf{i}^\dagger \hat{\psi}_\mathbf{j}^\dagger \hat{\psi}_\mathbf{j} \hat{\psi}_\mathbf{i} \right. \\
&+ \left. c_2' \hat{\psi}_i^\dagger (F_\kappa)_{ij} \hat{\psi}_j \hat{\psi}_k^\dagger (F_\kappa)_{kl} \hat{\psi}_l \right], \qquad (3) \\
\hat{\mathcal{H}}_{\rm PA} &= \int d\mathbf{r} \left[-\Omega_2 \hat{\psi}_g^\dagger \hat{\psi}_m + \Omega_1 \hat{\psi}_m^\dagger \hat{\psi}_0 \hat{\psi}_{-1} + H.c. \right]. \qquad (4)
\end{aligned}$$

Here V is the trap potential, E_i is the energy of the spin state i with a static magnetic field lifting their degeneracy, $F_{\kappa=x,y,z}$ are spin-1 matrices, and

$$c_0' = 4\pi(a_0 + 2a_2)/3M$$

and

$$c_2' = 4\pi(a_2 - a_0)/3M$$

where $a_{0,2}$ are s-wave scattering lengths [8], $\Omega_i, i = \{1, 2\}$ are the Rabi frequencies of the PA fields, and γ is a phenomenological decay factor. The detunings δ and Δ between the PA fields and the atomic and molecular levels are defined in Fig. 1. We have ignored the kinetic energy of the particles by assuming a dilute ensemble. Note also that this model ignores molecular collisions since there is currently no knowledge of their strength.

To extract the main aspects of the system dynamics we invoke a single-mode approximation, a simplification that has proven successful in describing key aspects of related systems in the past [3, 9, 10]. It amounts to approximating the fields operators of the three spin components of the atomic condensate as

$$\hat{\psi}_i(\vec{r}, t) = \sqrt{N} \hat{a}_i(t) \phi(\vec{r}) \exp(-i\mu t/\hbar),$$

where N is the initial atomic number, μ the chemical potential, $\phi(\vec{r})$ is the normalized condensate wave function for each spin component, satisfying $\int d\vec{r} |\phi(\vec{r})|^2 = 1$ [9, 10], and $\hat{a}_i(t)$ are bosonic annihilation operators. The molecular field is described likewise in a single-mode approximation, with the annihilation operators $\hat{m}$ and $\hat{g}$ describing excited and ground-sate molecules.

For large enough detunings δ the intermediate molecular state $|m\rangle$ can be adiabatically eliminated [21], simplifying the Heisenberg equations of motion of the atom-molecule system to

$$\begin{aligned}
i\frac{d\hat{a}_+}{d\tau} &= \chi_2(\rho_+ + \rho_0 - \rho_-)\hat{a}_+ + \chi_2 \hat{a}_0^2 \hat{a}_-^\dagger, \\
i\frac{d\hat{a}_0}{d\tau} &= \chi_2(\rho_+ + \rho_-)\hat{a}_0 - \omega\rho_- \hat{a}_0 + 2\chi_2 \hat{a}_+ \hat{a}_- \hat{a}_0^\dagger + \Omega \hat{g} \hat{a}_-^\dagger, \\
i\frac{d\hat{a}_-}{d\tau} &= -\Gamma \hat{a}_- + \chi_2 \hat{a}_0^2 \hat{a}_+^\dagger + \Omega \hat{g} \hat{a}_0^\dagger, \\
i\frac{d\hat{g}}{d\tau} &= \Omega \hat{a}_0 \hat{a}_- + (\Delta + \delta - \delta')\hat{g}. \qquad (5)
\end{aligned}$$

These equations will be treated in the following by using the mean-field replacement $\hat{a}_i,\ \hat{g} \to \sqrt{\rho_{i,g}}$, with $\rho_i = |a_i|^2$ or $\rho_g = |g|^2$ denoting the atomic or molecular density [3]. The rescaled parameters in Eqs. (5) are

$$c_{0,2} = c_{0,2}' \int d\mathbf{r} |\phi(\mathbf{r})|^4, \tag{6}$$

$$\delta' = \frac{\Omega_2^2}{c_0 N \delta} \left(1 + \frac{i\gamma}{2\delta}\right) \tag{7}$$

and we have introduced the dimensionless variables $\tau = c_0 N t$, $\chi_2 = c_2/c_0$, $\omega = \Omega_1^2/(c_0 N \delta)$, $\Gamma = \omega\rho_0 - \chi_2(\rho_- + \rho_0 - \rho_+)$, and

$$\Omega = \frac{\Omega_1 \Omega_2}{c_0 N \delta}.$$

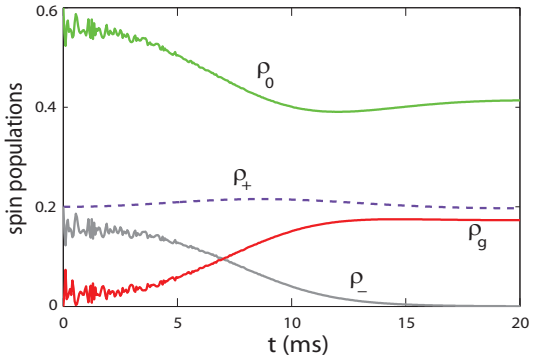

FIG. 2: (Color online) Scalar-like atom-molecule conversion of ^{87}Rb atoms, with essentially unchanged population of the spin-up state [16]. The initial condition is $f = [\sqrt{0.2}, \sqrt{0.6}, \sqrt{0.2}]$, and $\Omega = \Omega_m \mathrm{sech}(t/4)$ with $|\Omega_m/\chi_2| = 1.44 \times 10^4$ [16]. The other parameters are $\chi_2 = -0.01$, $\delta = -100\chi_2$, $\gamma = 10|\chi_2|$, and $c_0 N = 10^5 s^{-1}$.

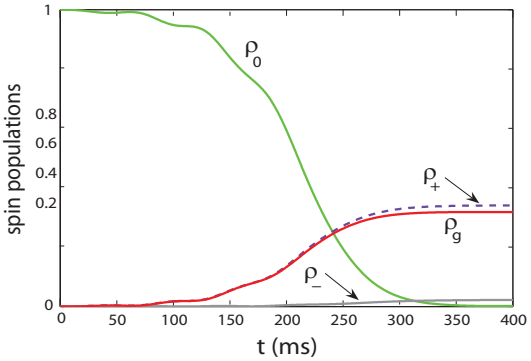

FIG. 3: (Color online) Atom-molecule pairs formation as a function of time for ^{87}Rb atoms, under the dark state condition for spin-down atoms. The initial state is $f = [0, 1, 0]$ and the other parameters are the same as Fig. 2.

B. Scalar-like photoassociation

In their recent experiment on two-color PA of the spinor atoms ^{87}Rb [16], Kobayashi *et al.* observed the spin-selective formation of the molecular state $|2, -1\rangle$ from reactant atoms in the state $|1, -1\rangle$ and $|1, 0\rangle$. One important feature of their experimental results is that while the populations of the reactant atoms decreased, the population of the state $|1, 1\rangle$ remained almost unchanged. This is the situation illustrated in Fig. 1(b) [16].

To test our model against that experiment we assume that the energy degeneracy of the atomic magnetic sublevels is lifted by a static magnetic field and that the atomic condensate is initially prepared in the state $f = [\sqrt{0.2}, \sqrt{0.6}, \sqrt{0.2}]$ [16]. The experiment used two lasers of maximum powers $I_1 = I_2/2 = 10W$, detuning $\delta = 2\pi \times 300$MHz and $\Omega/\sqrt{I} = 7\mathrm{MHz}(\mathrm{Wcm}^{-2})^{-\frac{1}{2}}$, which yields in our case $\Omega_1 = 139$ MHz and $\Omega_2 = 197$ MHz. As we will see in the following these values are well beyond the regime of atom-molecule pair formation, and as illustrated in Fig. 2 our model does confirm that the two-color PA of atoms into molecules is scalar-like in this case.

C. Stable atom-molecule pair formation

The scalar-like photoassociation sketched in the previous subsection results from the binding of a pair of Rb atoms of spin-0 and spin-down. We now consider the case of PA from spin-0, but in the presence of spin-changing collisions, the situation sketched in Fig. 1(c). Specifically, we assume that the atomic condensate is initially prepared in the spin-0 state $|1, 0\rangle$. Spin-exchange collisions couple then a pair of spin-0 atoms to a pair of atoms with opposite spins, $2A_0 \rightarrow A_\downarrow + A_\uparrow$ [15], while PA fields of appropriate wavelengths selectively combine a spin-down atom and a spin-0 atom into the molecular ground state $|g\rangle$ via a virtual transition to an excited molecular $|m\rangle$, $A_0 + A_\downarrow \rightarrow A_0 A_\downarrow$ [16]. The outcome of these combined mechanisms is the creation of an atom-molecule pair from three spin-0 atoms, $3A_0 \rightarrow A_0 A_\downarrow + A_\uparrow$, a process that can be intuitively thought of as an effective, spin-dependent three-body recombination. As such, this process is quite different from both the scalar-like PA of the previous subsection [16] and the purely atomic laser-catalyzed spin mixing [17].

We found numerically that in this case the stable atom-molecule pair formation is possible, provided that the dark-state condition

$$\Omega(t) = -\chi_2 \sqrt{\frac{\rho_0 \rho_+}{\rho_g}}, \tag{8}$$

for the spin-down atomic state is satisfied [22, 23]. This result is easily confirmed from Eqs. (5), which show that when condition (8) is satisfied the spin-down atomic state remains essentially unoccupied. That situation is illustrated in Fig. 3, which shows the efficient stable creation of atom-molecule pairs in this case.

D. Role of magnetization

The initial magnetization

$$\mathcal{M} = \rho_+ - \rho_- - \rho_g \tag{9}$$

of a spin gas prepared in the state $f = [0, 1, 0]$ is $\mathcal{M} = 0$. In this subsection we consider the role of that initial magnetization in the creation of atom-molecule pairs. We find that in contrast to the case of scalar-like molecule formation, the initial magnetization now plays a significant role, as illustrated in Figs. 4 and 5.

Figure 4 shows the evolution of the population of ground-state molecules for several values of the initial

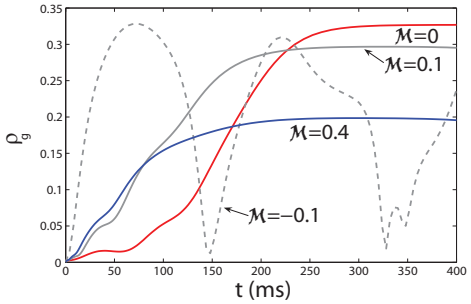

FIG. 4: (Color online) Spinor molecules population for several values of the initial magnetization $\mathcal{M}$ under the dark-state condition, with $\chi_2 = -0.01$ and $\delta = 100|\chi_2|$. The stable formation of spinor molecules is possible only for $\mathcal{M} \geq 0$.

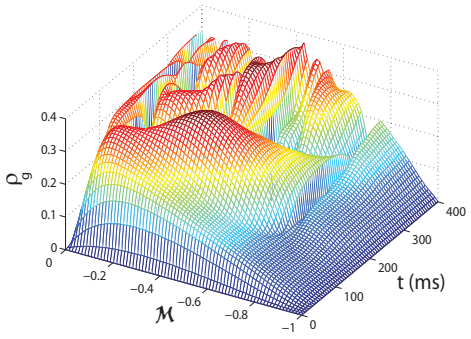

FIG. 5: (Color online) Large-amplitude oscillations of the spinor molecules population for negative values of the magnetization $\mathcal{M} < 0$. All other parameters are as in Fig. 4.

magnetization, under the generalized dark-state condition (8). For $\mathcal{M} \geq 0$ the ground-state molecules are produced efficiently and reach a steady-state population $\rho_g = (1 - \mathcal{M})/3$; for $\mathcal{M} < 0$, in contrast, this population exhibits large oscillations – see also Fig. 5, which shows more details of the oscillations of ρ_g for negative magnetizations – and do not appear to reach a steady state. This is due to the simple fact that for $\mathcal{M} < 0$, the populations of spin-down atomic state are not zero and thus that state is a "bright" state that does not remain uncoupled to the other atomic states during association.

III. NON-RESONANT REGIME

The dynamics of atom-molecule pair formation in the case of negative magnetization indicates that the presence or absence of an atomic dark state plays a key role in that process. In this section we further investigate the "non-resonant" situation where no dark state exists. We consider specifically two examples: The first one is an 'adiabatic' case characterized by an approximate dark-state condition. In this case the system dynamics can be understood in terms of an effective nonrigid pendulum model that permits to discuss an antiferromagnetic instability of the atom-molecule pendulum. In a second

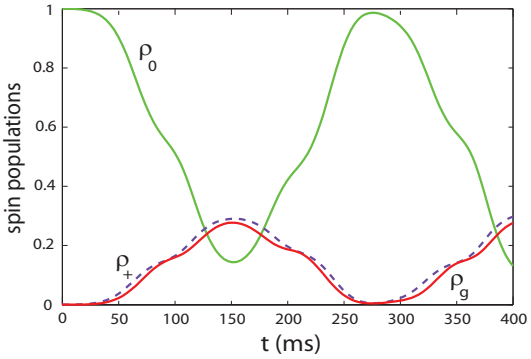

FIG. 6: (Color online) Coherent atom-molecule oscillations as a function of time for ^{87}Rb atoms. The dashed line is the population of the spin-up atoms. The initial atomic state is $f = [0, 1, 0]$, $\Omega = 0.75|\chi_2|$, and the other parameters are as in Fig. 2.

example, we briefly discuss a situation where the dark-state condition is strongly violated.

A. Adiabatic case

Figure 6 shows an example of atom-molecule-pair oscillations for a non-resonant situation and starting from spin-0 ^{87}Rb atoms. (Note that pair formation implies that $\rho_+ \simeq \rho_g$.) As would be intuitively expected, the numerical integration of Eqs. (5) confirms that the creation of atom-molecule pairs is only possible for PA field strengths that allow for the simultaneous occurrence of spin-exchange collisions and atom-molecule conversion. For the initial atomic state $f = [0, 1, 0]$, we find that the Rabi frequencies of the PA fields should be such that

$$\Omega = -\chi_2\sqrt{\rho_0} \leq |\chi_2|$$

or equivalently

$$\Omega_1\Omega_2 \leq |N\delta c_2|,$$

which gives $\Omega_1 \leq 0.3\pi$MHz, and $\Omega_2 \leq 0.6\pi$MHz for the case $\Omega_2/\Omega_1 = 2$ considered here.

We remark that for an atomic condensate initially prepared in the spin-0 state, and assuming that the dark-state condition (8) is approximately satisfied, the first derivatives of the slowly-varying amplitudes for spin-down atoms can be neglected, $i\hat{a}_- \approx 0$ [24, 25]. It is then possible to describe the system by the approximate effective three-state Hamiltonian

$$\begin{aligned}\hat{\mathcal{H}}_{\rm eff} &= \chi_3(\hat{a}_0^{\dagger 3}\hat{a}_+\hat{g} + \hat{a}_0^3\hat{a}_+^\dagger\hat{g}^\dagger \\ &+ \frac{1}{\Gamma}(\Omega^2\hat{\rho}_0\hat{\rho}_g + \chi_2^2\hat{\rho}_0^2\hat{\rho}_+) + \chi_2\hat{\rho}_0\hat{\rho}_+, \qquad (10)\end{aligned}$$

where $\chi_3 = \Omega\chi_2/\Gamma$. The first term in this Hamiltonian describes the creation of atom-molecule pairs from three spin-0 atoms through a laser-induced effective three-body recombination [26].

For short enough times, one could neglect the depletion of the spin-0 population and to treat $\hat{a}_0$ as a c-number, $\hat{a}_0 \to N^{1/2}$. Linearizing the Hamiltonian (10), the second line reduces then to a self-interacting contribution, with $\Gamma/\rho_0 \simeq \omega - \chi_2$, and the Heisenberg equations of motion for the remaining operators $\hat{a}_+$ and $\hat{g}$ have the solution

$$\begin{aligned} \hat{a}_+(t) &= \hat{a}_+(0)\cosh\chi_3' t - i\hat{g}^\dagger(0)\sinh\chi_3' t, \\ \hat{g}(t) &= \hat{g}(0)\cosh\chi_3' t - i\hat{a}_+^\dagger(0)\sinh\chi_3' t, \end{aligned} \tag{11}$$

with $\chi_3' = N^{3/2}\chi_3$. These solutions are well-known to be indicative of quantum entanglement of the created atom-molecule pairs. As such this system is formally a matter-wave analog of optical parametric down conversion in quantum optics [1, 24].

B. Antiferromagnetic instability

Within the mean-field approach, the spatial part of the atomic and molecular wave functions can be written as $\sqrt{N}e^{-i\mu t/\hbar}\zeta$, where $\zeta\sqrt{\rho_i}e^{i\theta_i}$ or $\sqrt{\rho_g}e^{i\theta_g}$ and θ_i represents the phase of the i-th Zeeman state [3]. Within this description the dynamics of the system can be expressed in terms of the coupled equations

$$\begin{aligned} \dot{\rho}_0 &= 3\chi_3\rho_0^{3/2}\sqrt{(1-\rho_0)^2-\mathcal{M}^2}\sin\theta, \\ \dot{\theta} &= -\Theta + \chi_2(1+\mathcal{M}-2\rho_0) + \frac{1}{\Gamma}[\chi_2^2\rho_0(3+3\mathcal{M}-4\rho_0) \\ &+ \Omega^2(\frac{3}{2}-\frac{3m}{2}-\frac{5\rho_0}{2}) + \Omega^2 + (\Delta+\delta-\delta')\Gamma] \\ &+ \frac{\Omega\chi_2}{2\Gamma}\frac{\sqrt{\rho_0}[(1-\rho_0)(9-13\rho_0)-9\mathcal{M}^2]}{\sqrt{(1-\rho_0)^2-\mathcal{M}^2}}\cos\theta, \end{aligned} \tag{12}$$

where

$$\theta = 3\theta_0 - (\theta_+ + \theta_g) \tag{13}$$

and

$$\Theta = E_g + E_+ - 3E_0. \tag{14}$$

These nonlinear equations support the two phase-independent fixed-point solutions $\rho_0 = 0$ and $\rho_0 = 1 - |\mathcal{M}|$, as well as phase-dependent solutions for $\theta = 0$ or π.

Equations (12) describe a nonrigid pendulum with energy functional

$$\mathcal{E} = \lambda_1\cos\theta + \lambda_2, \tag{15}$$

where

$$\begin{aligned} \lambda_1 &= 3\chi_3\rho_0^{3/2}\sqrt{(1-\rho_0)^2-\mathcal{M}^2}, \\ \lambda_2 &= \frac{\rho_0}{\Gamma}\left[\chi_2^2\rho_0\left(\frac{3}{2}+\frac{3}{2}\mathcal{M}-\frac{4}{3}\rho_0\right)\right. \\ &+ \left.\frac{\Omega^2}{2}\left(3-3\mathcal{M}-\frac{5}{2}\rho_0\right)\right] \\ &- \rho_0\left(\Theta+\frac{\Omega^2}{\Gamma}+\Delta+\delta-\delta'\right) \\ &+ \chi_2\rho_0(1+\mathcal{M}-\rho_0). \end{aligned} \tag{16}$$

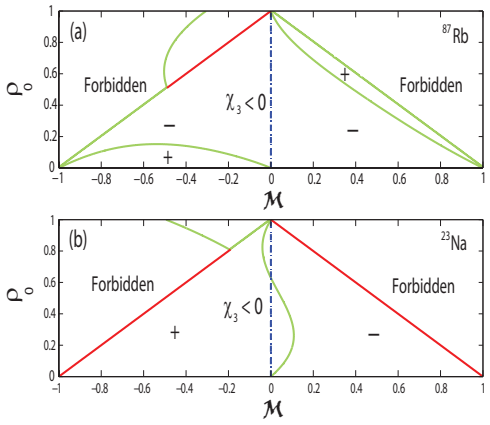

FIG. 7: (Color online) Surfaces of $d\mathcal{E}/d\mathcal{M} = 0$ (green solid lines) for (a) ferromagnetic ^{87}Rb atoms ($\theta = 0$, $\chi_2 = -0.01$); and (b) anti-ferromagnetic ^{23}Na atoms ($\theta = \pi$, $\chi_2 = 0.01$). The red forbidden line is determined by the condition of conserved total atomic number or $\rho_0 + |\mathcal{M}| \leq 1$ (see Ref. [27]).

This approach allows one to study simply the stability of the magnetic domain structure of the system. Specifically, we follow the approach of Ref. [27] and consider instabilities associated with a change in the sign of $d\mathcal{E}/d\mathcal{M}$. For example, $dE/d\mathcal{M} > 0$ for $\mathcal{M} > 0$ and $dE/d\mathcal{M} < 0$ for $\mathcal{M} < 0$ implies that the magnetization always oscillates around zero, and no domain forms. Following this approach we find that, in contrast to the situation for purely atomic gases [27, 28], an instability of the domain structure can occur for both ferromagnetic and anti-ferromagnetic atoms. One finds readily from Eqs. (15) and (16),

$$\begin{aligned} \frac{d\mathcal{E}}{d\mathcal{M}} &= \frac{3\chi_3}{2}\mathcal{M}\left[1-\frac{\rho_0^{3/2}cos\theta}{\sqrt{(1-\rho_0)^2-\mathcal{M}^2}}\right] + \chi_2\rho_0 \\ &+ \frac{3\rho_o}{2\Gamma}(\chi_2^2\rho_0 - \Omega^2). \end{aligned} \tag{17}$$

Figure 7 shows the resulting surfaces of $d\mathcal{E}/d\mathcal{M} = 0$ for the ferromagnetic and anti-ferromagnetic cases. The plus or minus sign denotes $d\mathcal{E}/d\mathcal{M} > 0$ or $d\mathcal{E}/d\mathcal{M} < 0$. Here the condensate size is already assumed to be much larger than the healing length $\mathcal{L}_s = 2\pi/\sqrt{2M|c_2'|n}$ at least in one direction so that instability-induced domains can appear [27].

As already mentioned, for $d\mathcal{E}/d\mathcal{M} < 0$ an increase in $\mathcal{M}$ leads to lower energy while for $d\mathcal{E}/d\mathcal{M} > 0$ it leads to a higher energy. Hence the (+, -) boundary delimitates the domain of dynamic instability (see e.g. Ref. [27] for more details). We observe that in contrast to the case of a pure sample of ^{87}Rb atoms, which is characterized by a wide instability region [27], in the case at hand this region can be significantly reduced by an appropriate tuning of the lasers. We also note that in the case of anti-ferromagnetic atoms such as ^{23}Na, where no dynamical instability exists for a pure atomic sample,

for our hybrid system, an instability can now develop for a wide range of parameters, see Fig. 7b.

One point to emphasize is that the antiferromagnetic instability can be experimentally observed without any laser fields, i.e. for $\Omega = 0$ – although these fields are of course required for the formation of molecules. We also remark that the spin mixing of spin-2 molecules is slow enough in comparison with the effective three-body recombination process that it can be safely ignored here. However, thermalization and spontaneous decay of the ground-state molecules are expected to be major challenges for the observation of coherent oscillations of atom-molecule pairs [5].

C. Violation of the dark-state condition

As a final special case we now consider the situation when $|\Omega/\chi_2| > 1$, in which case the dark-state condition (8) is completely violated. Figure 8 shows that for increasing values of Ω/χ_2, the amplitude of the oscillations in molecular population first increase, and then decreases until $|\Omega/\chi_2| = 1$. Beyond that critical value the molecular oscillations become strongly damped, and eventually population transfer to the molecular ground state essentially disappears, as illustrated in the figure for $|\Omega/\chi_2 = 1.5$. As illustrated in Fig. 8(b) the population oscillations of spin-0 atoms is also strongly suppressed in that regime of strong PA.

Finally Fig. 8(b) also illustrates how different choices of the initial atomic state result in different dynamics of the spinor atom-molecule system. In particular, an atomic sample initially in the spin-0 state remains completely unperturbed by the strong PA fields (far from the dark-sate resonance condition). Note that the scalar-like atom-molecule conversion illustrated in Fig. 2 corresponds to fields that strongly violate the condition (8), with $|\Omega/\chi_2| = 1.44 \times 10^4 \gg 1$. In that case the only parameters of practical relevance are the initial atomic state and the strengths of the PA fields.

IV. SUMMARY AND CONCLUSION

In conclusion, we have studied a number of aspects of coherent photoassociation in a spinor Bose condensate, with emphasis on the creation of atom-molecule pairs from the initial spin-zero atoms. This process, which involves *two* two-body interactions, can be conveniently described by an effective three-body spin-dependent recombination mechanism – the term "three-body recombination" being used here to differentiate our proposal from the recent two-color PA experiment (that involves the scalar-like association of spinor atoms) [16]. We have shown in particular that the spin-down atoms can be kept in a dark state for appropriate conditions in both the initial states of the atoms and PA fields, leading to the formation of atom-molecule pairs. For comparison we also considered the regimes with PA fields strong enough to violate the dark-state condition.

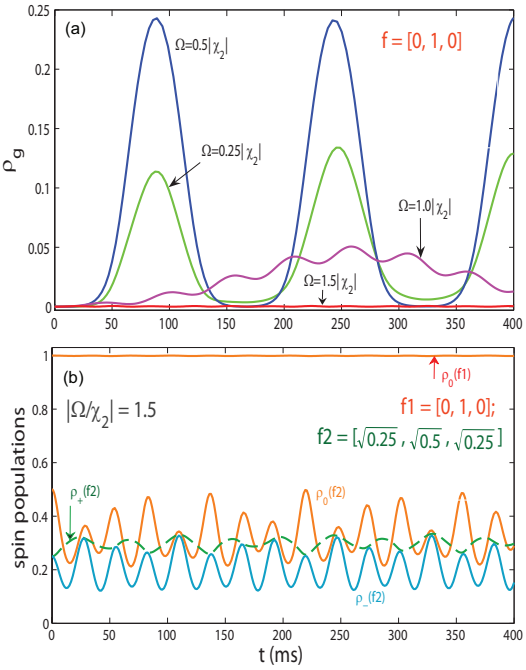

FIG. 8: (Color online) (a) Molecular oscillations for several values of $|\Omega/\chi_2|$, which label the curves, and the initial atomic state $|0, 1, 0\rangle$. (b) Atomic spin populations for the initial atomic states $|f_1\rangle = |0, 1, 0\rangle$ and $|f_2\rangle = |\sqrt{0.25}, \sqrt{0.5}, \sqrt{0.25}\rangle$, and for $|\Omega/\chi_2| = 1.5$. Other parameters are as in Fig. 2.

Although it shares the similar usage of PA fields and spin-dependent collisions, the present work is different from previous results on laser-catalyzed atomic spin oscillations [17], which did not involve the formation of molecules. In addition, the simulations of experimentally observed scalar-like features in associating spinor atoms, the study of the roles of magnetization and of the initial atomic state, and the antiferromagnetic instability of a hybrid atom-molecule system are also the new results.

In view of the rapid experimental advances in all-optical association of laser-cooled atoms [19], it can be expected that the coherent PA of quantum spin gases, in particular, the atom-molecule pair formation in a spinor sample, should become experimentally observable in the near future [16]. Laser-controlled spinor reactions can provide a new testing ground to address a number of questions in many-body physics, cold chemistry, and quantum information science. Future work will study the creation of heteronuclear spinor molecules from a two-species atomic spin gas [29], and the spinor reactions in an optical lattice [30], with and without the long-range dipole-dipole interactions [13]. We also plan to study the cavity-assisted amplification of spinor molecules [31], the bistability of a spinor atom-molecule "pendulum" [32], and the spinor trimer formation [20, 33].

This work is supported by the U.S. Office of Naval Research, by the U.S. National Science Foundation, by the

U.S. Army Research Office, and by the National Science Foundation of China under Grant Numbers 10874041 and 10974045.

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