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Instability, adiabaticity, and controlling effects of external fields for the dark state in a homonuclear atom–tetramer conversion system*

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In the present paper, we investigate the instability, adiabaticity, and controlling effects of external fields for a dark state in a homonuclear atom–tetramer conversion that is implemented by a generalized stimulated Raman adiabatic passage. We analytically obtain the regions for the appearance of dynamical instability and study the adiabatic evolution by a newly defined adiabatic fidelity. Moreover, the effects of the external field parameters and the spontaneous emissions on the conversion efficiency are also investigated.

Keywords: dynamical instability, adiabatic fidelity, dark state, atom–molecule conversion

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1. Introduction

Molecules offer a whole new dimension in the study of ultracold atomic physics since it opens possibilities for studying unique physical phenomena such as molecular matter waves,[1,2] high-precision molecular spectroscopy,[3] strongly interacting superfluids,[4] and so on. To investigate various novel physical features of ultracold molecules, one needs to convert cold atoms into molecules. Photoassociation (PA)[5,6] and magnetic Feshbach resonance (FR)[7,8] are two ways in which gases of ultracold atoms are connected to the molecular bound states of their underlying two-body interactions. Since such a process generally produces molecules from bosonic atoms in a vibrationally and/or energetically excited quasi-bound level and hence is not energetically stable, the stimulated Raman adiabatic passage (STIRAP) in PA[9–11] or aided by FR[12–16] has been regarded as an effective approach to create ground-state molecules. The success of the STIRAP technique requires the existence of the coherent population trapping state or dark state,[17,18] which is the superposition of the free atomic and ground molecular states and should be followed adiabatically. However, since the interparticle nonlinear collisions bring dynamical instability,[19] which could make the system not follow the *CPT* state adiabatically and lead to the low atom–molecule conversion efficiency, and make the adiabatic theorem invalid for the atom–molecule conversion system, the stability and adiabaticity analysis of the dark state in these nonlinear atom–molecule conversion systems have been widely studied.[11,12,20–26] For instance, the adiabatic theorem of the atom–dimer conversion system is investigated by linking the nonadiabaticity with the population growth in the collective excitations of the dark state,[20] and the dynamical instability of this system is studied by the emergence of complex frequencies in the collective modes.[12] An improved adiabatic condition for the above system is put forward by Itin *et al.*[21] via applying methods of classical Hamiltonian dynamics. While in our recent works,[22] the adiabaticity of the atom–trimer dark state was investigated by the newly defined adiabatic fidelity, and the dynamical instability was discussed by the linear stability theorem with classical Hamiltonian dynamics.

Recently, the STIRAP aided by Efimov resonance (ER) has been firstly generalized to create more complex molecules–tetramers.[27] In the STIRAP aided by ER in this atom–tetramer conversion system, the weakly bound trimer molecules are firstly created by ER and then photoassociated with another atom to become tetramer molecules. This is different from the traditional STIRAP in PA or aided by FR in the atom–dimer conversion system, where the excited dimers are coupled with the ground atoms with FR[9–11] or PA[12–15] and coupled with the ground dimers with PA. Motivated by this paper and in order to obtain high atom–tetramer conversion efficiency, in the present paper, we investigate the dynamical instability, adiabaticity, and controlling effects of external field parameters for the dark state in the homonuclear atom–

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tetramer conversion system. The paper is organized as follows. In Section 2, we model the systems and derive the *CPT* state solution. In Section 3, we investigate the dynamical instability of the atom–tetramer dark state by the linear stability theorem. In Section 4, we study the adiabaticity and the controlling effects of the external field parameters on the *CPT* state quantitatively based on the newly defined adiabatic fidelity. In Section 5, our conclusion is presented.

2. Mode and *CPT* state

Consider the atom–tetramer conversion system, where the intermediated trimer A_3 is formed by the three-body Efimov resonance, then these trimers, along with another atom, are photoassociated to form tetramers A_4. By denoting the atom–trimer coupling strength as λ' with detuning δ, the Rabi frequency of the trimer–tetramer coupling optical field with Ω' and detuning Δ, including s-wave scattering processes, the second quantized Hamiltonian under the rotating frame reads:

$$\begin{aligned}\hat{H} = -\hbar\Bigg[&\sum_{i,j}\chi'_{ij}\hat{\psi}_i^{\dagger}\hat{\psi}_j^{\dagger}\hat{\psi}_j\hat{\psi}_i+\delta\hat{\psi}_{\rm m}^{\dagger}\hat{\psi}_{\rm m}+(\Delta+\delta)\hat{\psi}_{\rm g}^{\dagger}\hat{\psi}_{\rm g}\\&+\lambda'(\hat{\psi}_{\rm m}^{\dagger}\hat{\psi}_{\rm a}\hat{\psi}_{\rm a}\hat{\psi}_{\rm a}+{\rm H.c.})\\&-\Omega'(\hat{\psi}_{\rm g}^{\dagger}\hat{\psi}_{\rm m}\hat{\psi}_{\rm a}+{\rm H.c.})\Bigg],\end{aligned}\tag{1}$$

where $\hat{\psi}_i$ and $\hat{\psi}_i^{\dagger}$ are the annihilation and creation operators, χ_{ij} represents the two-body collisions, and the indices i, j = a, m, g stand for the atom, trimer, and tetramer, respectively.

As in Refs. [9], [13], [14], and [22], considering the conservation of the total particle numbers, we put Eq. (1) into the grand canonical Hamiltonian "Kamiltonian" by adding the conserved particle number into the Hamiltonian,

$$\hat{K}=\hat{H}-\hbar\mu\hat{N},\tag{2}$$

where $\hbar\mu$ is the atomic chemical potential, and N is the operators for the total particle number with $\hat{N}=\hat{\psi}_{\rm a}^{\dagger}\hat{\psi}_{\rm a}+3\hat{\psi}_{\rm m}^{\dagger}\hat{\psi}_{\rm m}+4\hat{\psi}_{\rm g}^{\dagger}\hat{\psi}_{\rm g}$.

From the Kamiltonian we can easily derive the equations of motion of the unit-scaled operators. Under the mean-field approximation, i.e., $\hat{\psi}_i$ and $\hat{\psi}_i^{\dagger}$ are replaced by c number $\sqrt{n}\psi_i$ and $\sqrt{n}\psi_i^*$, where n is the density of the total particle number. We can obtain the set of mean-field equations ($\hbar=1$),

$$\begin{aligned}{\rm i}\dot{\psi}_{\rm a}&=(\omega_{\rm a}-\mu_{\rm a})\,\psi_{\rm a}-3\lambda\,\psi_{\rm m}\psi_{\rm a}^{*2}+\Omega\,\psi_{\rm g}\psi_{\rm m}^*,\\{\rm i}\dot{\psi}_{\rm m}&=(\omega_{\rm m}-3\mu_{\rm a})\,\psi_{\rm m}-({\rm i}\gamma+\delta)\,\psi_{\rm m}-\lambda\,\psi_{\rm a}^3+\Omega\,\psi_{\rm g}\psi_{\rm a}^*,\\{\rm i}\dot{\psi}_{\rm g}&=\left(\omega_{\rm g}-4\mu_{\rm a}\right)\psi_{\rm g}-(\Delta+\delta)\psi_{\rm g}+\Omega\,\psi_{\rm m}\psi_{\rm a}.\end{aligned}\tag{3}$$

In the above sets of equations (3), $\omega_i=-2\sum_j\chi_{ij}|\psi_j|^2$, $\chi_{ij}=\chi'_{ij}n$, $\lambda=\lambda'\sqrt{n}$, $\Omega=\Omega'\sqrt{n}$ are the renormalized quantities, and the term proportional to γ is introduced phenomenologically to simulate the loss of intermediate trimers.

To seek the *CPT* solution, we take $\dot{x}\approx 0$, $x=\psi_{\rm a}$, $\psi_{\rm m}$, $\psi_{\rm g}$, and $\psi_{\rm m}=0$, then one can easily derive the following *CPT* solutions:

$$\begin{aligned}|\psi_{\rm a}^0|^2&=2/(1+\sqrt{1+16(\lambda/\Omega)^2}),\\|\psi_{\rm g}^0|^2&=(1-|\psi_{\rm a}^0|^2)/4.\end{aligned}\tag{4}$$

with the following chemical potentials and two-photon resonance condition:

$$\begin{aligned}\mu&=-2\left(\chi_{\rm aa}|\psi_{\rm a}^0|^2+\chi_{\rm ag}|\psi_{\rm g}^0|^2\right),\\\Delta&=-\delta+\left(8\chi_{\rm aa}-2\chi_{\rm ag}\right)|\psi_{\rm a}^0|^2+\left(8\chi_{\rm ag}-2\chi_{\rm gg}\right)|\psi_{\rm g}^0|^2.\end{aligned}\tag{5}$$

From Eqs. (5), we can conclude that, by dynamically maintaining the two-photon resonance condition, population can be concentrated in atomic states and bound molecular states under the respective limit $\lambda/\Omega\to 0$ and $\lambda/\Omega\to\infty$, which facilitates the adiabatic coherent population transfer between atoms and tetramer molecules.

3. Instability of *CPT* state

The existence of the *CPT* state cannot guarantee that it can always be followed adiabatically. In this section, we investigate the stability properties for the homonuclear atom–tetramer *CPT* state.

As in Refs. [22], [23], and [26], we make use of the linear stability analysis through casting a nonlinear Schrödinger equation into an effective classical Hamiltonian and analyzing the eigenvalues of the Hamiltonian–Jacobi matrix obtained by linearizing the equations of motion around the fixed point, which corresponds to the *CPT* state.[21,28,29] A straightforward calculation of the eigenvalues (other than the zero-mode frequency) of the Hamiltonian–Jacobi matrix with an analytic expression can be obtained,

$$\begin{aligned}\omega_{1,2\pm}&=\pm\frac{1}{\sqrt{2}}\sqrt{b\pm\sqrt{b^2-c}},\\b&=-\xi^2+2\Omega^2(|\psi_{\rm g}^0|^2-|\psi_{\rm a}^0|^2)-18\lambda^2|\psi_{\rm a}^0|^4,\\c&=(\xi-1)(\xi^3-4\xi\Omega^2|\psi_{\rm g}^0|^2)+72\lambda^2\left(2\chi_{\rm aa}-\Omega^2\right)|\psi_{\rm a}^0|^6\\&\quad+324\lambda^4|\psi_{\rm a}^0|^8+4\Omega^4|\psi_{\rm g}^0|^4+4|\psi_{\rm a}^0|^4\Big[9\lambda^2\xi(\xi-1)\\&\quad+\Omega^4+24\lambda\Omega|\psi_{\rm g}^0|(\chi_{\rm aa}+\chi_{\rm ag})-18\lambda^2\Omega^2|\psi_{\rm g}^0|^2\Big]\\&\quad+4\Omega|\psi_{\rm a}^0|^2\Big[2(2\chi_{\rm aa}+4\chi_{\rm ag}+2\chi_{\rm gg}+\Omega^2)|\psi_{\rm g}^0|^2\\&\quad-\xi(\xi-1)\Big],\end{aligned}\tag{6}$$

where $\xi=(6\chi_{\rm aa}-2\chi_{\rm am})\,|\psi_{\rm a}^0|^2+\left(6\chi_{\rm ag}-2\chi_{\rm mg}\right)|\psi_{\rm g}^0|^2$. When $\omega_{\pm}$ becomes complex, the corresponding *CPT* state is dynamically unstable. We can see from Eq. (6) that the unstable regime is given by either $c<0$ or $c>b^2$, and the instability here strongly depends on the nonlinear collisions.

In order to support the above stability analysis, we numerically solve Eq. (6) and the results are shown in Fig. 1, where the parameters are taken for ^{87}Rb atoms with the s-wave scattering length 5.77 nm. Following the similar routine of Refs. [15], [27], and [30], the atom density n is about 5×10^{20} m^{-3}, and the Rabi frequency is given by $\Omega=\Omega_0\,\mathrm{sech}\,t/\tau$, where Ω_0 and τ are the strength and the width of the Rabi pulse, and $\lambda=4.718\times10^4$ s^{-1}. Here time is in units of $1/\lambda$, and other parameters are in units of λ. Moreover, as in Refs. [15], [27], and [30], the collisional parameter χ_{aa} is taken as 0.5303, and other collisional parameters are 0.0938. In Fig. 1, the black (white) areas correspond to the unstable (stable) regions. From this figure, we see that there are two unstable regions corresponding to the two cases $c>b^2$ or $c<0$, respectively. Region I corresponds to the unstable region obtained by setting $c<0$, whose width shrinks as Ω increases; region II is the unstable region obtained by setting $c>b^2$, whose width becomes fat with increasing δ. In order to obtain high conversion efficiency, it is crucial for adiabatic evolution to avoid these unstable regimes when designing the route of adiabatic passage.

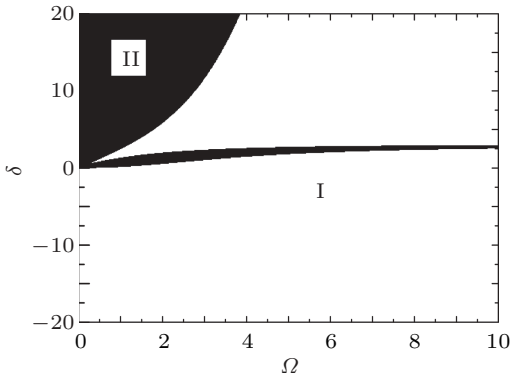

Fig. 1. Instability diagrams for ^{87}Rb atom–tetramer conversion systems, where the black areas correspond to the unstable regions.

4. Adiabaticity and controlling effects of external fields for the *CPT* state

In the stable region, the existence of the *CPT* state facilitates the adiabatic coherent population transfer between atoms and tetramers. However, owing to the invalidation of the superposition principle in this nonlinear system, it is not justified to apply the adiabatic condition of quantum mechanics to study the adiabatic evolution of the *CPT* state. In fact, the adiabatic evolution of a system can be thoroughly studied quantitatively by employing adiabatic fidelity[11,22,23] which describes the distance between the adiabatic solution and the actual one. Here we define the fidelity for atom–tetramer conversion system as

$$f(|\psi_1\rangle,|\psi_2\rangle)=|\langle\overline{\psi_1}|\overline{\psi_2}\rangle|^2, \tag{7}$$

where $|\overline{\psi}\rangle$ is the rescaled wavefunction of the form $|\psi\rangle=(\psi_a,\psi_m,\psi_g)^T$,

$$|\overline{\psi}\rangle=\left(\frac{{\psi_a}^4}{|\psi_a|^3},\sqrt{3}\frac{\psi_a\psi_m}{|\psi_a|},2\psi_g\right). \tag{8}$$

Because we are only concerned with the adiabatic evolution of the *CPT* state throughout, we denote the adiabatic fidelity of the dark state as $F=|\langle\overline{\psi(t)}|\overline{CPT}\rangle|^2$ where $|\psi(t)\rangle$ is the exact solution of the Schrödinger equation in Eq. (3). $|\overline{\psi(t)}\rangle$ and $|\overline{CPT}\rangle$ are the rescaled wavefunctions of $|\psi(t)\rangle$ and the *CPT* state, respectively. If the system can adiabatically evolve along the *CPT* state, then the value of the adiabatic fidelity should be close to 1.

Figure 2 shows the adiabatic fidelity of the *CPT* state as functions of time with and without the nonlinear collisions. In this figure, we choose $\delta=-3$. This is because the system is dynamically stable for an arbitrary value of Ω when $\delta=-3$, as shown in Fig. 1. We assume that spontaneous emission $\gamma=1$ has the same magnitude with the coupling strength λ. If we choose $\gamma=0$, we can obtain a higher conversion efficiency. As can be seen in Fig. 2, no matter whether the interparticle interactions are considered, the magnitude of adiabatic fidelity is about 1 at the initial time, but begins to decrease at some later time, then diminishes to the minimal value, and finally approaches to a steady value which is smaller than 1. The smallest adiabatic fidelity F^s that can be used to describe the adiabaticity of the system is 0.67 (0.85) at time $t=113$ ($t=160$) with $\chi_{ij}\neq0$ ($\chi_{ij}=0$). Therefore, the adiabaticity of the system is poorer with the interparticle interactions. This induces the final adiabatic fidelity F^f which can be used to describe the conversion efficiency[11] being lower with the nonlinear collisions. Hence we can conclude that the interparticle collisions suppress the conversion. This conclusion is also true for other cases such as $\Omega_0=20$ with $\tau=40$, $\Omega_0=40$ with $\tau=20$, and so on, provided these external parameters satisfy the adiabatic evolution condition.

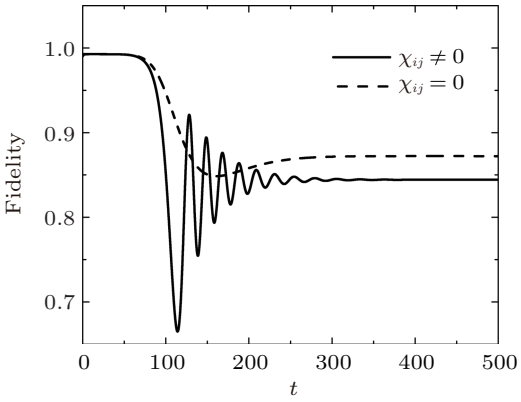

Fig. 2. Adiabatic fidelity as a function of time with and without the nonlinear collisions. For the case with nonzero nonlinear interactions, the collisional parameters are $\chi_{aa}=0.5303$, $\chi_{ab}=\chi_{ag}=\chi_{bb}=\chi_{bg}=\chi_{gg}=0.0938$. The other parameters are $\delta=-3$, $\Omega_0=20$, $\tau=20$, and $\gamma=1$. Time is in units of λ^{-1}. All other parameters are in units of λ.

The dependence of the final adiabatic fidelity $F^{\rm f}$ on the external field parameters and the spontaneous emissions is shown in Fig. 3. As in the discussion about the parameters in Fig. 2, here we choose the parameters $\delta = -3$, $\Omega_0 = 40$, $\gamma = 1$, and $\tau = 20$ when they are fixed. From this figure, we see that the stable creation of tetramers is always possible for $\delta < 0$ no matter whether the nonlinear collisions are included. However, once the interparticle interactions are included, the conversion efficiency is near to zero for $\delta > 0$. Moreover, as the Rabi pulse amplitude Ω_0 and width τ increase, the conversion efficiency first increases quickly, then reaches a steady optimal value which is close to 1. We also see that no matter whether the nonlinear collisions are considered, the bigger the spontaneous emission is, the lower the conversion efficiency is. After comparing the results with and without the two-body interactions, we find the two-body interactions suppress the conversion of tetramers. It is clear that we can improve the conversion efficiency by choosing the optimal external field parameters δ, Ω_0, and τ.

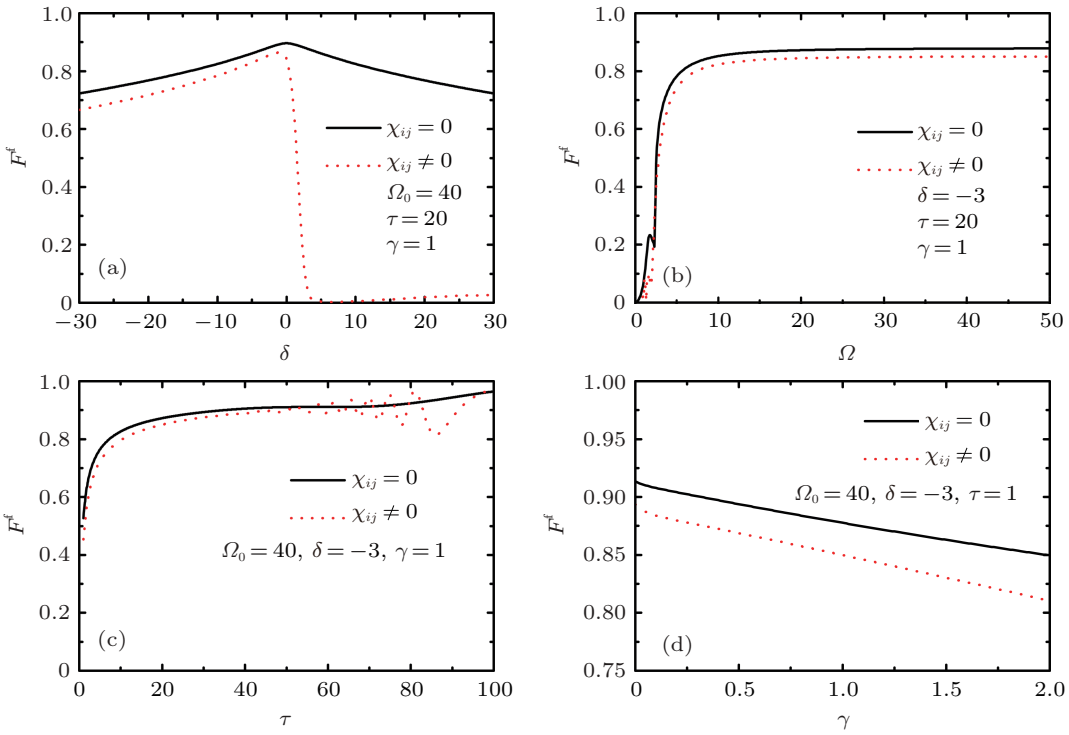

Fig. 3. (color online) Effects of the external field parameters and the spontaneous emissions on the conversion efficiency. The adiabatic fidelity versus (a) the detuning δ, (b) the Rabi pulse strength Ω_0, (c) the pulse width τ, and (d) the spontaneous emissions γ.

5. Conclusion

In conclusion, we investigated the instability, adiabaticity, and controlling effects of the external fields of the atom–tetramer dark state in the stimulated Raman adiabatic passage aided by Efimov resonance. By making use of the linear stability analysis, we analytically obtained the regions for the appearance of dynamical instability. Taking ^{87}Rb atom–molecule conversion systems as an example, we gave the unstable regions numerically. Moreover, the effects of the external field parameters and spontaneous emissions on the conversion efficiency were studied by the newly defined adiabatic fidelity. We found that one can improve the conversion efficiency by optimizing the single-photon detuning, the strength, and the width of the Rabi pulse.

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