Creating pentamer molecules by generalized stimulated Raman adiabatic passage

Fu-Quan Dou,[1,2,*] Sheng-Chang Li,[3] Hui Cao,[1] and Li-Bin Fu[3,4]

[1]*School of Physics, Beijing Institute of Technology, Beijing 100081, China*

[2]*Key Laboratory of Atomic and Molecular Physics and Functional Materials of Gansu Province, College of Physics and Electronic Engineering, Northwest Normal University, Lanzhou 730070, China*

[3]*National Laboratory of Science and Technology on Computation Physics, Institute of Applied Physics and Computational Mathematics, Beijing 100088, China*

[4]*Key Laboratory of High Energy Density Physics Simulation, Center for Applied Physics and Technology, Peking University, Beijing 100084, China*

(Received 28 July 2011; published 23 February 2012)

We study the formation of stable homonuclear and heteronuclear pentamers from ultracold atoms via a generalized stimulated Raman adiabatic passage scheme. The atom-molecule dark-state solutions for the system are obtained, and the linear instability and the adiabatic fidelity of the dark state are investigated. We also discuss the effects of external field parameters on the conversion efficiency.

DOI: 10.1103/PhysRevA.85.023629 PACS number(s): 03.75.−b, 05.30.Jp, 67.85.−d

I. INTRODUCTION

The production of ultracold molecular gases has attracted much attention [1–3] since it opens possibilities for studying unique physical phenomena and their applications. For example, ultracold molecules may be used in testing fundamental symmetries [4], in precision spectroscopy [5], in quantum information processing [6], and in ultracold chemistry [7]. Photoassociation (PA) [8] and magnetic Feshbach resonances (FRs) [9] are two ways in which gases of ultracold atoms may be connected to the molecular bound states of their underlying two-body interaction potentials. However, the diatomic molecules formed by a PA or FR process are usually loosely bound energetically unstable. They have to be adiabatically transferred into a tightly bound ground state via a stimulated Raman adiabatic passage (STIRAP) [10,11]. First proposed for associating nondegenerate atoms into stable molecules, FR-aided STIRAP [12–14] is considered a more efficient way of converting atomic condensates into molecular ones than the bare STIRAP technique. The success of STIRAP relies on the existence of the coherent population trapping (CPT) state [15], i.e., dark state, which should be followed adiabatically. Such a condition can be fulfilled for linear systems by appropriately choosing laser frequencies. However, for systems with interparticle interactions, the two-photon resonance condition dynamically changes when the population is transferred from atomic states to molecular states. This renders the CPT state more difficult to be followed adiabatically and leads to a low atom-molecule conversion efficiency [13]. To cope with this problem, the generalized STIRAP scheme [16] adopts a chirped coupling field to compensate for the effects of nonlinear interactions and thus to efficiently generate large amounts of deeply bound ultracold molecules.

Another interesting issue in this field is to obtain an ultracold complex molecule. From the perspective of few-body physics, a three-body Efimov resonance (ER) molecule was predicted in the early 1970's [17] and was first observed for ultracold gases in 2006 [18]. It not only confirmed the existence of weakly bound trimer states, but also opened up different ways of exploring the intriguing physics of few-body quantum systems. ER was soon generalized to four-body systems [19,20]. For four identical bosons [21–23], it was found that the Efimov trimer and tetramer states always appear as sets of states with two tetramers associated with each of the trimer levels. Experimentally, tetramer states were recently realized in an ultracold gas of cesium atoms [24]. With the help of mean-field theory, the generalized STIRAP technique has been used to obtain the homonuclear [25] and heteronuclear [26] tetramer molecules. To take another step forward, one may naturally wonder if we can assemble even more complex ultracold polyatomic molecules.

The goal of this paper is to demonstrate theoretically that the generalized STIRAP technique can in principle be employed in the generation of homonuclear and heteronuclear molecular pentamers. We first create tetramer A_4 from ultracold atoms, and then couple it with another atom to a bound pentamer A_5 or A_4B via PA. A coherent atom-molecule dark state is exploited to prevent the tetramer population from becoming significant throughout the conversion process. After deriving the atom-pentamer dark-state solution for the conversion process, we focus on the linear instability induced by the interparticle interactions and the adiabatic fidelity of the atom-pentamer dark state in the STIRAP. To choose suitable parameter values to implement efficiently the conversion from atoms to pentamers, we also discuss the effects of the single-photon detuning, and the strength and width of the Rabi pulse on the atom-pentamer conversion efficiency.

The rest of this paper is organized as follows. In Sec. II, we introduce our model and obtain the CPT state solution. In Sec. III, we study the linear instability and adiabatic fidelity of the CPT state and discuss the effect of external field parameters on the conversion. The conclusions and discussions are given in Sec. IV.

II. MODEL AND CPT STATE

Our model consists of ultracold Bose atoms coupled to molecular tetramers from ultracold atoms, and then these tetramers, along with another atom, are photoassociated to

*doufq@nwnu.edu.cn

form pentamers. This coherent transfer process can be better understood by analyzing an abstract three-level model. The free atomic, the high excited tetramer, and the stable pentamer states form the abstract three-level system to which STIRAP can be applied.

A. Homonuclear pentamer

We consider first the creation of homonuclear pentamers. By denoting the atom-tetramer coupling strength as λ' with detuning δ, and the Rabi frequency of the photoassociation laser as Ω' with detuning Δ, in the interaction picture, the Hamiltonian describing the system reads

$$\begin{aligned}\hat{H} = -\hbar \int d\mathbf{r} \Bigg\{ & \sum_{i,j} \chi'_{i,j} \hat{\psi}_i^\dagger(\mathbf{r}) \hat{\psi}_j^\dagger(\mathbf{r}) \hat{\psi}_j(\mathbf{r}) \hat{\psi}_i(\mathbf{r}) + \delta \hat{\psi}_t^\dagger(\mathbf{r}) \hat{\psi}_t(\mathbf{r}) \\ & + \lambda' [\hat{\psi}_t^\dagger(\mathbf{r}) \hat{\psi}_a(\mathbf{r}) \hat{\psi}_a(\mathbf{r}) \hat{\psi}_a(\mathbf{r}) \hat{\psi}_a(\mathbf{r}) + \text{H.c.}] \\ & + (\Delta + \delta) \hat{\psi}_p^\dagger(\mathbf{r}) \hat{\psi}_p(\mathbf{r}) - \Omega' [\hat{\psi}_p^\dagger(\mathbf{r}) \hat{\psi}_t(\mathbf{r}) \hat{\psi}_a(\mathbf{r}) + \text{H.c.}] \Bigg\}, \end{aligned} \tag{1}$$

where $\hat{\psi}_i$ and $\hat{\psi}_i^\dagger$ are the annihilation and creation operators, $\chi'_{i,j}$ represents the two-body interaction, and the indices $i, j = a, t, p$ stand for the atom, tetramer, and pentamer states, respectively.

The particle number is very large in experiments, and we can apply the mean-field theory. In the approximation, the number of particles tends to infinity while the density is held and fixed, and the quantum and thermal fluctuations around the mean value are negligible [27]. The field operators $\hat{\psi}_i$ and $\hat{\psi}_i^\dagger$ are replaced by c-numbered order-parameter fields $\sqrt{n}\psi_i$ and $\sqrt{n}\psi_i^*$, where n is the density of the total particle number. The Heisenberg equations of motion for annihilation operators $\hat{\psi}_i$ can be written as

$$\begin{aligned} \frac{d\psi_a}{dt} &= 2i \sum_j \chi_{aj} |\psi_j|^2 \psi_a + 4i\lambda \psi_t \psi_a^{*3} - i\Omega \psi_p \psi_t^*, \\ \frac{d\psi_t}{dt} &= 2i \sum_j \chi_{tj} |\psi_j|^2 \psi_t + (i\delta - \gamma)\psi_t + i\lambda \psi_a^4 - i\Omega \psi_p \psi_a^*, \\ \frac{d\psi_p}{dt} &= 2i \sum_j \chi_{pj} |\psi_j|^2 \psi_p + i(\Delta + \delta)\psi_p - i\Omega \psi_t \psi_a, \end{aligned} \tag{2}$$

in which $\chi_{ij} = n\chi'_{ij}$, $\lambda = n\sqrt{n}\lambda'$, and $\Omega = \sqrt{n}\Omega'$ are the renormalized quantities and the term proportional to γ is introduced phenomenologically to simulate the decay of the intermediate tetramer state.

Now we demonstrate that Eqs. (2) really support a steady CPT state with $|\psi_t^0| = 0$. We start by considering the steady-state solutions of Eqs. (2) through the following steady-state ansatz:

$$\begin{aligned} \psi_a &= |\psi_a^0| \exp[i(\theta_a - \mu_a t)], \\ \psi_t &= |\psi_t^0| \exp[4i(\theta_a - \mu_a t)], \\ \psi_p &= |\psi_p^0| \exp[5i(\theta_a - \mu_a t)], \end{aligned} \tag{3}$$

where μ_a is the atomic chemical potential. Putting Eqs. (3) into Eqs. (2) and keeping the intermediate state unpopulated, one can obtain the following CPT solutions:

$$\begin{aligned} |\psi_a^0|^2 &= \frac{-2 \times 15^{\frac{1}{3}} + 2^{\frac{1}{3}} \left[45\frac{\lambda}{\Omega} + \sqrt{60 + \left(45\frac{\lambda}{\Omega}\right)^2}\right]^{\frac{2}{3}}}{30^{\frac{2}{3}} \frac{\lambda}{\Omega} \left[45\frac{\lambda}{\Omega} + \sqrt{60 + \left(45\frac{\lambda}{\Omega}\right)^2}\right]^{\frac{1}{3}}}, \\ |\psi_t^0|^2 &= 0, \\ |\psi_p^0|^2 &= \frac{1}{5}(1 - |\psi_a^0|^2), \end{aligned} \tag{4}$$

and the conserved total particle number, $|\psi_a|^2 + 4|\psi_t|^2 + 5|\psi_p|^2 = 1$. The chemical potential and the generalized two-photon resonance conditions [16] are

$$\mu_a = -2\chi_{aa}|\psi_a^0|^2 - 2\chi_{ap}|\psi_p^0|^2 \tag{5}$$

and

$$\Delta = -\delta + (10\chi_{aa} - 2\chi_{pa})|\psi_a^0|^2 + (10\chi_{ap} - 2\chi_{pp})|\psi_p^0|^2. \tag{6}$$

A population distribution such as Eqs. (4) has a remarkable property of allowing all the atoms to be converted into pentamers as λ/Ω changes from 0 to ∞ as long as the two-photon resonance condition (6) can be maintained dynamically.

B. Heteronuclear pentamer

We now turn to the situation of heteronuclear pentamer formation. The dynamics of the system can be described by the Hamiltonian

$$\begin{aligned}\hat{H} = -\hbar \int d\mathbf{r} \Bigg\{ & \sum_{i,j} \chi'_{i,j} \hat{\psi}_i^\dagger(\mathbf{r}) \hat{\psi}_j^\dagger(\mathbf{r}) \hat{\psi}_j(\mathbf{r}) \hat{\psi}_i(\mathbf{r}) + \delta \hat{\psi}_t^\dagger(\mathbf{r}) \hat{\psi}_t(\mathbf{r}) \\ & + \lambda' [\hat{\psi}_t^\dagger(\mathbf{r}) \hat{\psi}_a(\mathbf{r}) \hat{\psi}_a(\mathbf{r}) \hat{\psi}_a(\mathbf{r}) \hat{\psi}_a(\mathbf{r}) + \text{H.c.}] \\ & + (\Delta + \delta) \hat{\psi}_p^\dagger(\mathbf{r}) \hat{\psi}_p(\mathbf{r}) - \Omega' [\hat{\psi}_p^\dagger(\mathbf{r}) \hat{\psi}_t(\mathbf{r}) \hat{\psi}_b(\mathbf{r}) + \text{H.c.}] \Bigg\}, \end{aligned} \tag{7}$$

where the subscript b represents atom B. Under the mean-field approximation, the system is described by the mean-field equations of motion

$$\begin{aligned} \frac{d\psi_a}{dt} &= 2i \sum_j \chi_{aj} |\psi_j|^2 \psi_a + 4i\lambda \psi_t \psi_a^{*3}, \\ \frac{d\psi_b}{dt} &= 2i \sum_j \chi_{bj} |\psi_j|^2 \psi_b - i\Omega \psi_p \psi_t^*, \\ \frac{d\psi_t}{dt} &= 2i \sum_j \chi_{tj} |\psi_j|^2 \psi_t + (i\delta - \gamma)\psi_t + i\lambda \psi_a^4 - i\Omega \psi_p \psi_b^*, \\ \frac{d\psi_p}{dt} &= 2i \sum_j \chi_{pj} |\psi_j|^2 \psi_p + i(\Delta + \delta)\psi_p - i\Omega \psi_t \psi_b, \end{aligned} \tag{8}$$

where $\chi_{ij} = n\chi'_{ij}$, $\lambda = n\sqrt{n}\lambda'$, and $\Omega = \sqrt{n}\Omega'$ are the renormalized quantities and the decay rate γ accounts for the loss of untrapped tetramers.

By using the steady-state ansatz

$$\begin{aligned}\psi_a &= |\psi_a^0| \exp[i(\theta_a - \mu_a t)],\\ \psi_b &= |\psi_b^0| \exp[i(\theta_b - \mu_b t)],\\ \psi_t &= |\psi_t^0| \exp[4i(\theta_a - \mu_a t)],\\ \psi_p &= |\psi_p^0| \exp\{i\,[4\theta_a + \theta_b - (4\mu_a + \mu_b)t]\},\end{aligned} \tag{9}$$

where μ_b is the chemical potential of atom B, one finds that the following CPT solutions exist:

$$\begin{aligned}|\psi_b^0|^2 &= \frac{-5\times 3^{\frac{1}{3}} + 5^{\frac{1}{3}}\left[72\frac{\lambda}{\Omega} + \sqrt{75 + \left(72\frac{\lambda}{\Omega}\right)^2}\right]^{\frac{2}{3}}}{16\times 15^{\frac{2}{3}}\frac{\lambda}{\Omega}\left[72\frac{\lambda}{\Omega} + \sqrt{75 + \left(72\frac{\lambda}{\Omega}\right)^2}\right]^{\frac{1}{3}}},\\ |\psi_a^0|^2 &= 4|\psi_b^0|^2,\\ |\psi_t^0|^2 &= 0,\\ |\psi_p^0|^2 &= \frac{1}{5} - |\psi_b^0|^2,\end{aligned} \tag{10}$$

where we have used the condition of conserved particle number: $|\psi_a|^2 + |\psi_b|^2 + 4|\psi_t|^2 + 5|\psi_p|^2 = 1$. The atomic chemical potentials and generalized two-photon resonance condition are

$$\begin{aligned}\mu_a &= -2\chi_{aa}|\psi_a^0|^2 - 2\chi_{ab}|\psi_b^0|^2 - 2\chi_{ap}|\psi_p^0|^2,\\ \mu_b &= -2\chi_{ba}|\psi_a^0|^2 - 2\chi_{bb}|\psi_b^0|^2 - 2\chi_{bp}|\psi_p^0|^2,\end{aligned} \tag{11}$$

and

$$\begin{aligned}\Delta = -\delta &+ (8\chi_{aa} + 2\chi_{ba} - 2\chi_{pa})|\psi_a^0|^2\\ &+ (8\chi_{ab} + 2\chi_{bb} - 2\chi_{pb})|\psi_b^0|^2\\ &+ (8\chi_{ap} + 2\chi_{bp} - 2\chi_{pp})|\psi_p^0|^2.\end{aligned} \tag{12}$$

Similarly, from Eqs. (10) and (12), we can find that, by dynamically maintaining the resonance condition, the population can be concentrated in atomic and pentamer bound states under the respective limits $\lambda/\Omega \to 0$ and $\lambda/\Omega \to \infty$.

III. LINEAR INSTABILITY AND ADIABATIC FIDELITY OF CPT STATE

The existence of the CPT state, however, does not guarantee that this state can be followed adiabatically. In the section, we investigate the stability properties and the adiabatic fidelity for the atom-pentamer CPT state.

We adopt the linear stability analysis, i.e., we add a small fluctuation to steady-state CPT solutions and follow the dynamical evolution of the system to see whether the fluctuation remains insignificant. For this purpose, we linearize the equations of motion including the chemical potential and obtain the Jacobi matrix around the fixed point (CPT state) for the atom-pentamer conversion system. The excitation frequencies (corresponding to the eigenvalues of the Jacobi matrix) of the linearized equation other than the zero-frequency mode (corresponding to the Goldstone mode [28]) can be found analytically as

$$\omega = \pm\sqrt{\frac{(B \pm \sqrt{B^2 - 4C})}{2}}, \tag{13}$$

where

$$B = \begin{cases} 32\lambda^2|\psi_a^0|^6 + 2\left(|\psi_a^0|^2 - |\psi_p^0|^2\right)\Omega^2 + A^2, & \text{homonuclear},\\ 32\lambda^2|\psi_a^0|^6 + 2\left(|\psi_b^0|^2 - |\psi_p^0|^2\right)\Omega^2 + A^2, & \text{heteronuclear},\end{cases} \tag{14}$$

$$C = \begin{cases} \left(16\lambda^2|\psi_a^0|^6 + \left(|\psi_a^0|^2 - |\psi_p^0|^2\right)\Omega^2\right)^2\\ \quad - 4A\left[8\lambda\Omega(\chi_{aa} - \chi_{ap})|\psi_a^0|^5|\psi_p^0|\right.\\ \quad \left. + \Omega^2(\chi_{aa} - 2\chi_{ap} + \chi_{pp})|\psi_a^0|^2|\psi_p^0|^2 + 16\lambda^2\chi_{aa}|\psi_a^0|^8\right],\\ \quad \text{homonuclear},\\ \left(16\lambda^2|\psi_a^0|^6 + (|\psi_b^0|^2 - |\psi_p^0|^2)\Omega^2\right)^2\\ \quad - 4A\left[8\lambda\Omega(\chi_{ab} - \chi_{ap})|\psi_a^0|^4|\psi_b^0||\psi_p^0|\right.\\ \quad \left. + \Omega^2(\chi_{bb} - \chi_{bp} + \chi_{pp})|\psi_b^0|^2|\psi_p^0|^2 + 16\lambda^2\chi_{aa}|\psi_a^0|^8\right],\\ \quad \text{heteronuclear},\end{cases} \tag{15}$$

with $A = \delta + 2(\chi_{at} - 4\chi_{aa})|\psi_a^0|^2 + 2(\chi_{tp} - 4\chi_{ap})|\psi_p^0|^2$ for the homonuclear atom-pentamer system and $A = \delta + 2(\chi_{at} - 4\chi_{aa})|\psi_a^0|^2 + 2(\chi_{bt} - 4\chi_{ab})|\psi_b^0|^2 + 2(\chi_{tp} - 4\chi_{ap})|\psi_p^0|^2$ for the heteronuclear atom-pentamer system. When ω becomes complex, the corresponding CPT state is dynamically unstable. Hence, the unstable regime is given by either $C < 0$ or $C > B^2/4$. We see that the stability properties of the CPT state strongly depend on the nonlinear interactions. The typical results from the stability analysis based on the parameters of our interest are summarized in Fig. 1, where the (Ω,δ) space is divided into the stable (white) and the unstable (blue/dark gray) regions. There are two unstable regions: Region I is thin along the δ dimension and corresponds to the unstable region obtained by setting $C > B^2/4$; region II is the unstable region obtained by setting $C < 0$. In order to convert atoms into stable pentamers, it is crucial to avoid these unstable regions when designing the route of adiabatic passage.

In our calculations, we have taken the parameters for ^{133}Cs and ^{87} Rb atoms and have created the molecular pentamers Cs_5 and Cs_4Rb. The s-wave scattering lengths for cesium and

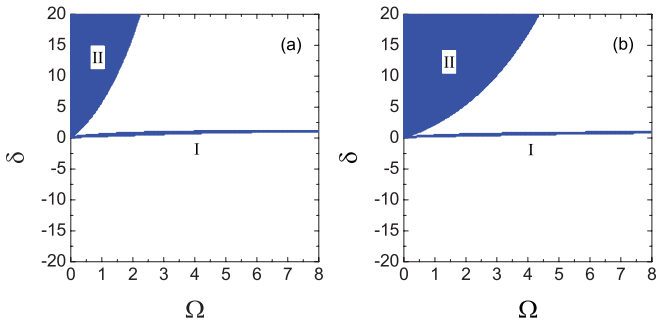

FIG. 1. (Color online) Instability diagrams in (Ω,δ) space for (a) homonuclear and (b) heteronuclear atom-pentamer systems. The blue (dark gray) areas correspond to the unstable regions.

rubidium atoms are $a = -374a_0$ [24] and $a = 100a_0$ [29] (a_0 is Bohr's radius). We take the atom density $n = 6 \times 10^{19}$ m^{-3} and $\lambda = 1.961 \times 10^4$ s^{-1}. This gives rise to the parameters $\chi_{aa} = 0.182\lambda$, $\chi_{bb} = 0.074\lambda$, and other interaction parameters are taken as 0.055λ [30].

Samples of our results on the atom-pentamer conversion are shown in Figs. 2(a) and 2(b) for the homonuclear and heteronuclear cases, where we have numerically solved Eqs. (2) and (8) including the loss term with a time-varying Rabi frequency given by

$$\Omega(t) = \Omega_0 \operatorname{sech}\frac{t}{\tau}, \tag{16}$$

where Ω_0 and τ are the strength and the width of the Rabi pulse. Here δ is chosen so that the system remains in the stable regime. We note that the time is in units of $1/\lambda$, and other quantities are in units of λ. Also plotted in the figure are the analytical CPT solutions of Eqs. (4) and (10) for homonuclear and heteronuclear pentamers. We find that the stable formation of pentamers is always possible by optimizing the parameters of the system.

In the stable regions, the existence of the CPT state facilitates the adiabatic coherent population transfer between

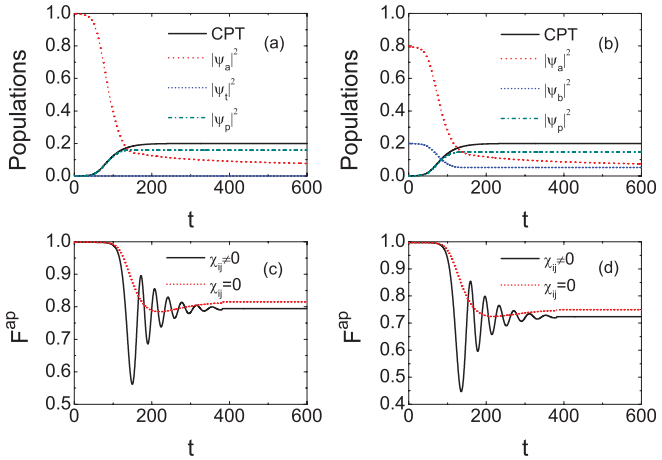

FIG. 2. (Color online) Population (upper) and adiabatic fidelity (lower) as functions of time with $\delta = -2.0$ for homonuclear (left) and heteronuclear (right) systems. The adiabatic fidelities without considering two-body interactions are also shown in (c) and (d), respectively. The other parameters are $\Omega_0 = 50$, $\tau = 20$, and $\gamma = 1.0$.

atoms and pentamers. The adiabatic evolution of our system can be thoroughly studied quantitatively by employing the adiabatic fidelity [31,32] which describes the distance between the adiabatic solution and the actual one. Here we define the adiabatic fidelity of the CPT state for atom-pentamer conversion system as

$$F^{ap}(t) = |\langle\overline{\psi(t)}|\overline{\mathrm{CPT}}\rangle|^2, \tag{17}$$

where $|\psi(t)\rangle$ is the exact solution of the Schrödinger equation. $|\overline{\psi(t)}\rangle$ and $|\overline{\mathrm{CPT}}\rangle$ are the rescaled wave functions of the $|\psi(t)\rangle$ and CPT state, respectively. The actual state of the system is

$$|\overline{\psi(t)}\rangle = \begin{cases} \left(\frac{\psi_a^5}{|\psi_a|^4}, 2\frac{\psi_a\psi_t}{|\psi_a|}, \sqrt{5}\psi_p\right)^T, & \text{homonuclear,} \\ \left(\frac{\psi_a^4\psi_b}{|\psi_a|^3|\psi_b|}, \frac{\psi_a^4\psi_b}{|\psi_a|^4}, \frac{2\psi_b\psi_t}{|\psi_b|}, \sqrt{5}\psi_p\right)^T, & \text{heteronuclear.} \end{cases} \tag{18}$$

If the system can adiabatically evolve along the CPT state, then the value of the adiabatic fidelity should be close to 1. The variation of the adiabatic fidelity with time for homonuclear and heteronuclear pentamers via the tetramer-intermediated dark-state scheme is shown in Figs. 2(c) and 2(d). One can see that the system adiabatically evolves along the CPT state at the initial time. The fidelity diminishes to minimal values of 0.56 and 0.45 at times 150 and 135 for the homonuclear and heteronuclear pentamer systems, respectively, which implies that the system deviates from the CPT state distinctly at that time. Although the fidelity begins to fluctuate later on, its final value is still no more than 0.795 and 0.723, respectively. The corresponding results without considering the interparticle interactions are also shown. We find that the interactions suppress the conversion.

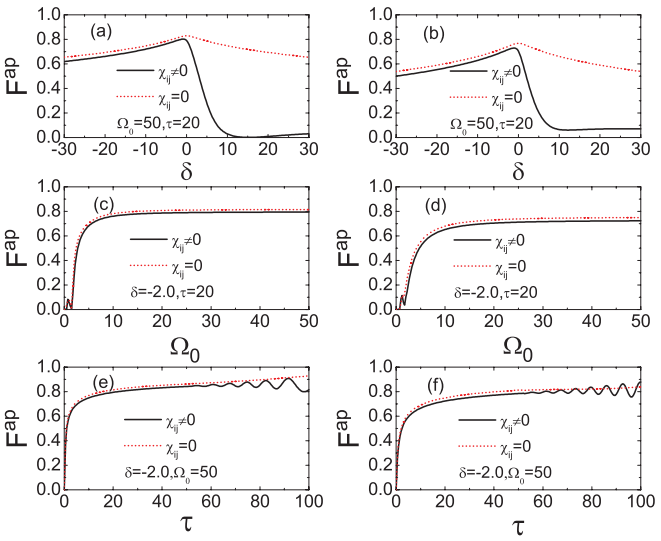

FIG. 3. (Color online) Effects of the external field parameters on the conversion efficiency for homonuclear (left) and heteronuclear (right) systems. (a), (b) The adiabatic fidelity vs the detuning δ. (c), (d) The adiabatic fidelity vs the Rabi pulse strength Ω_0. (e), (f) The adiabatic fidelity vs the pulse width τ. The corresponding results without considering two-body interactions are also shown as short-dashed red (light gray) lines. The parameter $\gamma = 1.0$.

Furthermore, we find that the final adiabatic fidelity $F^{ap}(\infty)$ is used to indicate the conversion efficiency because $F^{ap}(\infty)$ is the final value of adiabatic fidelity $F^{ap}(t)$ in the process of evolution, and the larger is $F^{ap}(\infty)$, the higher is the conversion efficiency.

The dependence of the conversion efficiency on the external field parameters is investigated and shown in Fig. 3. We see that that stable creation of pentamers is always possible for red detuning ($\delta < 0$), whereas for blue detuning ($\delta > 0$) the final conversion efficiency is very small. However, no matter what the detuning is, there always exists higher efficiency without considering the two-body interactions. The conversion efficiency increases quickly with increasing Rabi pulse strength and width for smaller Ω_0 ($\Omega_0 < 10$) and τ ($\tau < 10$) while it increases very slowly for larger Ω_0 and τ. Compared with the results disregarding two-body interactions, the atom-pentamer conversion efficiency with two-body interactions becomes lower, i.e., the two-body interactions suppress the conversion of pentamers. It is clear that the conversion efficiency from ultracold atoms to pentamers can be controlled effectively by the external field parameters δ, Ω_0, and τ.

IV. CONCLUSIONS AND DISCUSSIONS

In conclusion, we have investigated the homonuclear and heteronuclear molecular pentamer conversion problem via the generalized STIRAP scheme in an ultracold bosonic system that includes interparticle interactions and have derived the CPT state solution. The linear instability and the adiabatic fidelity of the atom-pentamer dark state in the STIRAP have been studied via an analysis of linear stability and the proper definition of adiabatic fidelity, respectively. We have also discussed the effects of the single-photon detuning, and the strength and the width of the Rabi pulse on the atom-pentamer conversion efficiency, and found that optimized parameter regions for stable conversion always exist. Our results not only provide a possible route for creating homonuclear and heteronuclear pentamers experimentally in the future, but also can be extended to the assembly of more complex ultracold polyatomic molecules.

It should be pointed out that hitherto we have not discussed how to form the intermediate tetramer state in our scheme. Recently, the experimental realization of tetramer states with universal properties tied to Efimov physics in an ultracold gas of cesium atoms [24] was reported, which may provide a possible way to form the intermediate tetramer state. However, the fully realization of coherent atom-pentamer conversion is experimentally challenging, and some conditions for the validity of our analysis have been hard to confirm. Nevertheless, in view of the rapid progress in the production and manipulation of ultracold molecules [33–35], such as the creation of long-lifetime, high-efficiency molecules in optical lattices [36,37], and the formation of stable dimers, trimers, and tetramers in low-dimensional geometries [38], we hope it may become possible in the future.

ACKNOWLEDGMENTS

We thank J. Liu for helpful discussions. The work is supported by the National Fundamental Research Program of China (Contract No. 2011CB921503), the National Natural Science Foundation of China (Contracts No. 91021021 and No. 11075020), and the Scientific Research Foundation of NWNU (No. NWNU-LKQN-10-24).

[1] L. D. Carr, D. DeMille, Roman V. Krems, and J. Ye, New J. Phys. **11**, 055049 (2009); E. R. Hudson, H. J. Lewandowski, B. C. Sawyer, and J. Ye, Phys. Rev. Lett. **96**, 143004 (2006).

[2] S. Ospelkaus, K.-K. Ni, D. Wang, M. H. G. de Miranda, B. Neyenhuis, G. Quéméner, P. S. Julienne, J. L. Bohn, D. S. Jin, and J. Ye, Science **327**, 853 (2010); D. DeMille, Phys. Rev. Lett. **88**, 067901 (2002).

[3] L. B. Fu and J. Liu, Ann. Phys. **325**, 2425 (2010).

[4] J. J. Hudson, B. E. Sauer, M. R. Tarbutt, and E. A. Hinds, Phys. Rev. Lett. **89**, 023003 (2002).

[5] V. V. Flambaum and M. G. Kozlov, Phys. Rev. Lett. **99**, 150801 (2007).

[6] A. Micheli, G. K. Brennen, and P. Zoller, Nat. Phys. **2**, 341 (2006); P. Rabl, D. DeMille, J. M. Doyle, M. D. Lukin, R. J. Schoelkopf, and P. Zoller, Phys. Rev. Lett. **97**, 033003 (2006).

[7] C. Chin, T. Kraemer, M. Mark, J. Herbig, P. Waldburger, H. C. Nägerl, and R. Grimm, Phys. Rev. Lett. **94**, 123201 (2005); T. V. Tscherbul and R. V. Krems, *ibid.* **97**, 083201 (2006).

[8] H. R. Thorsheim, J. Weiner, and P. S. Julienne, Phys. Rev. Lett. **58**, 2420 (1987); P. D. Lett, K. Helmerson, W. D. Phillips, L. P. Ratliff, S. L. Rolston, and M. E. Wagshul, *ibid.* **71**, 2200 (1993).

[9] S. Inouye, M. R. Andrews, J. Stenger, H. J. Miesner, D. M. Stamper-Kurn, and W. Ketterle, Nature (London) **392**, 151 (1998); J. Stenger, S. Inouye, M. R. Andrews, H. J. Miesner, D. M. Stamper-Kurn, and W. Ketterle, Phys. Rev. Lett. **82**, 2422 (1999).

[10] A. Vardi, D. Abrashkevich, E. Frishman, and M. Shapiro, J. Chem. Phys. **107**, 6166 (1997).

[11] M. Mackie, R. Kowalski, and J. Javanainen, Phys. Rev. Lett. **84**, 3803 (2000).

[12] E. A. Donley, N. R. Claussen, S. T. Thompson, and C. E. Wieman, Nature (London) **417**, 529 (2002); J. Herbig, T. Kraemer, M. Mark, T. Weber, C. Chin, H. C. Nägerl, and R. Grimm, Science **301**, 1510 (2003).

[13] S. J. J. M. F. Kokkelmans, H. M. J. Vissers, and B. J. Verhaar, Phys. Rev. A **63**, 031601 (2001).

[14] M. Mackie, Phys. Rev. A **66**, 043613 (2002).

[15] G. Alzetta, A. Gozzini, L. Moi, and G. Oriolis, Nuovo Cimento B **36**, 5 (1976); L. Moi and G. Orriols, *ibid.* **52**, 209 (1979).

[16] H. Y. Ling, H. Pu, and B. Seaman, Phys. Rev. Lett. **93**, 250403 (2004).

[17] V. Efimov, Phys. Lett. B **33**, 563 (1970).

[18] T. Kraemer, M. Mark, P. Waldburger, J. G. Danzl, C. Chin, B. Engeser, A. D. Lange, K. Pilch, A. Jaakkola, H.-C. Näerl, and R. Grimm, Nature (London) **440**, 315 (2006).

[19] H. W. Hammer and L. Platter, Eur. Phys. J. A **32**, 113 (2007).

[20] J. von Stecher, J. P. D'Incao, and C. H. Greene, Nat. Phys. **5**, 417 (2009).

[21] S. E. Pollack, D. Dries, and R. G. Hulet, Science **326**, 1683 (2009).
[22] R. Schmidt and S. Moroz, Phys. Rev. A **81**, 052709 (2010).
[23] M. T. Yamashita and D. V. Fedorov, and A. S. Jensen, Phys. Rev. A **81**, 063607 (2010).
[24] F. Ferlaino, S. Knoop, M. Berninger, W. Harm, J. P. D'Incao, H.-C. Nägerl, and R. Grimm, Phys. Rev. Lett. **102**, 140401 (2009).
[25] H. Jing and Y. Jiang, Phys. Rev. A **77**, 065601 (2008).
[26] G. Q. Li and P. Peng, Phys. Rev. A **83**, 043605 (2011).
[27] A. S. Parkins and D. F. Walls, Phys. Rep. **303**, 1 (1998).
[28] H. Y. Ling, P. Maenner, W. P. Zhang, and H. Pu, Phys. Rev. A **75**, 033615 (2007).
[29] A. Widera, O. Mandel, M. Greiner, S. Kreim, T. W. Hä nsch, and I. Bloch, Phys. Rev. Lett. **92**, 160406 (2004).
[30] To the best of our knowledge, there are so far no good estimates on the polyatomic molecular scattering length. So we take the interaction coefficients involving the polyatomic molecular to have the same magnitudes as the atomic-molecular interaction. In our calculations, all quantities are renormalized to be dimensionless by λ.
[31] L. H. Lu and Y. Q. Li, Phys. Rev. A **77**, 053611 (2008).
[32] S. Y. Meng, L. B. Fu, and J. Liu, Phys. Rev. A **78**, 053410 (2008); S. Y. Meng, L. B. Fu, J. Chen, and J. Liu, *ibid.* **79**, 063415 (2009).
[33] J. von Stecher, Phys. Rev. Lett. **107**, 200402 (2011).
[34] M. R. Hadizadeh, M. T. Yamashita, Lauro Tomio, A. Delfino, and T. Frederico, Phys. Rev. Lett. **107**, 135304 (2011).
[35] O. Dulieu and C. Gabbanini, Rep. Prog. Phys. **72**, 086401 (2009); E. Kuznetsova, P. Pellegrini, R. Côté, M. D. Lukin, and S. F. Yelin, Phys. Rev. A **78**, 021402 (2008).
[36] G. Thalhammer, K. Winkler, F. Lang, S. Schmid, R. Grimm, and J. H. Denschlag, Phys. Rev. Lett. **96**, 050402 (2006).
[37] T. Volz, N. Syassen, D. M. Bauer, E. Hansis, S. Dürr, and G. Rempe, Nat. Phys. **2**, 692 (2006).
[38] B. Wunsch, N. T. Zinner, I. B. Mekhov, S.-J. Huang, D.-W. Wang, and E. Demler, Phys. Rev. Lett. **107**, 073201 (2011); N. T. Zinner, B. Wunsch, I. B. Mekhov, S.-J. Huang, D.-W. Wang, and E. Demler, Phys. Rev. A **84**, 063606 (2011).