

Formation of N -body polymer molecules through generalized stimulated Raman adiabatic passageFu-quan Dou,^{1,2} Li-bin Fu,^{3,4} and Jie Liu^{3,4,*}¹*College of Physics and Electronic Engineering, Northwest Normal University, Lanzhou 730070, China*²*School of Physics, Beijing Institute of Technology, Beijing 100081, 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 26 February 2013; published 25 April 2013)

We investigate the creation of stable homonuclear and heteronuclear N -body polymer molecules from ultracold atoms via a generalized stimulated Raman adiabatic passage scheme. The atom-molecule dark-state solutions for the system are obtained and are found to satisfy universal algebraic equations. We discuss the linear instability and the adiabatic fidelity of the dark state. We also explore the effects of the interparticle interactions, the atomic number of the polymer, and external field parameters on the conversion process.

DOI: [10.1103/PhysRevA.87.043631](https://doi.org/10.1103/PhysRevA.87.043631)

PACS number(s): 03.75.-b, 05.30.Jp, 67.85.-d

I. INTRODUCTION

The study of ultracold molecules is one of the most active areas of atomic and molecular physics and attracts considerable attention [1–4]. However, most of the experimental and theoretical studies in this field to date have focused on diatomic molecules [5]. A major thrust of current work in this field aims to extend the techniques and study of ultracold diatomic molecules to polyatomic molecules [6]. Cooling and trapping of polyatomic molecules would greatly enrich the scope of molecular physics and allow the study of complex chemical reactions (ultracold chemistry and superchemistry) [7], molecular decoherence [8], precision measurements [9], and molecular optics [10].

It is, in principle, possible to form ultracold molecules larger than a dimer, either by directly cooling preexisting molecules from room temperature or by indirect association from atoms or smaller molecules. Very recently, a direct approach of cooling polyatomic molecules, called Sisyphus cooling, has been reported experimentally [11]. The scheme can cool fluoromethane CH_3F molecules down to 29 mK and will allow cooling to sub-mK temperatures and beyond. It represents a beautiful demonstration of new cooling and trapping methods and their surprising application to a gas of polyatomic molecules [12]. A potentially viable technique for indirectly forming ultracold polyatomic molecules could be based on cryogenic buffer-gas cooling [13] or Stark deceleration [14] followed by magnetic trapping and subsequent sympathetic cooling with ultracold atoms [15–17]. However, methods such as Stark deceleration have not yet reached the regime of ultracold temperatures. Another promising powerful method to produce ultracold polyatomic molecules is related to magnetically tunable Feshbach resonances (FR) [18]. With the aid of FR and photoassociation (PA) [19], a generalized stimulated Raman adiabatic passage (STIRAP) scheme [20], which was first proposed to efficiently generate large amounts of deeply bound ultracold diatomic molecules by using a chirped coupling field to compensate the effects of nonlinear

interactions, is employed for producing the homonuclear and heteronuclear triatomic molecules [21]. Experimentally, the STIRAP scheme has been used to create a ground-state $^{87}\text{Rb}_2$ molecule [22], a $^{133}\text{Cs}_2$ molecule [23], and $^{40}\text{K}^{87}\text{Rb}$ [24] and $^6\text{Li}^{40}\text{K}$ [25] heteronuclear molecules. In all these experiments, one makes use of a STIRAP process to coherently transfer the extremely weakly bound Feshbach molecules to the vibrotational ground state of the ground electronic state. Very recently, the ultracold alkaline-earth-metal $^{84}\text{Sr}_2$ molecule [26] was formed from atom pairs on sites of an optical lattice using the STIRAP scheme. Near the center of the FR, a three-body Efimov resonance (ER) was predicted in the early 1970s [27] and was first observed for ultracold gases in 2006 [28]. The observation of an ER confirms the existence of weakly bound trimer states and opens up new ways [29] of experimentally and theoretically exploring the intriguing physics of few-body quantum systems. Over the last several years, the field of Efimov physics has generated a great deal of excitement in ultracold atomic gases system [30–32]. The association of three-body Efimov molecules [33] was proposed in Bose gases loaded into optical lattices. The tetramer states [34,35] were experimentally realized in ultracold gas of cesium atoms [36] and were further studied for four identical bosons [37–39]. Recent theoretical investigations have predicted a further extension of the Efimov scenario, where five-, six-, seven-, or higher-body cluster states (N -body Borromean) can be bound even if no bound subsystem exists [40–42]. Clusters with up to $N = 40$ atoms have been investigated, and the scattering-length values at which successive N -boson systems cross the corresponding atomic threshold have been calculated [41]. With the help of three- and four-body ER and PA, the generalized STIRAP scheme has been applied to forming the homonuclear and heteronuclear tetramer [43] and pentamer [44] molecules. One could easily be tempted to extend this technique to creating more complex ultracold N -body polyatomic molecules.

In the present paper, we theoretically investigate the conversion problem from atom to N -body polyatomic molecule in an ultracold bosonic system by implementing the generalized STIRAP scheme, where atoms are first associated with excited polymers A_{N-1} , and the polymers are then coupled with

*liu_jie@iapcm.ac.cn

another atom by the optical field to form the bound polyatomic molecules. We first discuss the model for homonuclear and heteronuclear systems and then derive the coherent population trapping (CPT), i.e., dark-state solutions, which satisfy universal algebraic equations. We further focus on the linear instability and the adiabatic fidelity of the atom-polymer dark state in the STIRAP and find that the interparticle interactions will lead to the instability and cause the oscillation of adiabatic fidelity, forming an oscillation window, and the atomic number N of polymers and external field parameters will affect the duration and the magnitude of the oscillation.

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 process. The conclusions are given in Sec. IV.

II. MODEL AND CPT STATE

The system that we consider is illustrated in Fig. 1. It consists of ultracold Bose atoms coupled to molecular polymers A_{N-1} via a two-body FR ($N = 2$) or many-body ER ($N \geq 3$); these polymers A_{N-1} are in turn photoassociated to another atom, A or B , to form the polymer homonuclear molecule A_N or heteronuclear molecule $A_{N-1}B$.

A. Homonuclear polymer

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

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

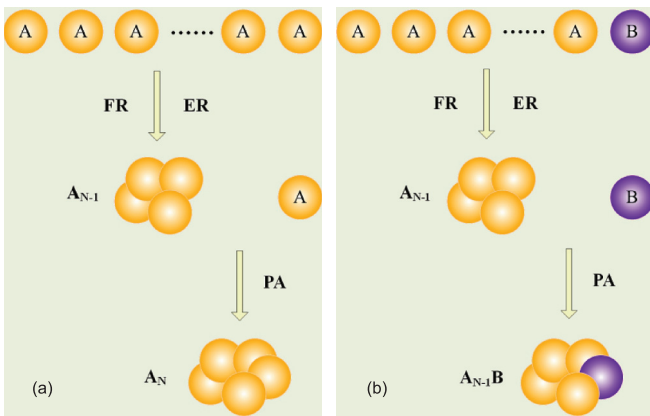


FIG. 1. (Color online) The scheme for atom-polymer conversion in an ultracold atomic system: (a) homonuclear polymer and (b) heteronuclear polymer.

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 $i, j = a, m, p$ stand for the atom, intermediate polymer A_{N-1} , and polymer A_N states, respectively.

For the systems with a large particle number and low temperature, it is usually quite well described by the mean-field approximation [45,46]. In the approximation, the number of particles tends to infinity while the density is held fixed, and the quantum and thermal fluctuations are negligible [47]. It has been successfully applied to explain the experiments of Feshbach molecules [4,22,48]. By replacing the field operators $\hat{\psi}_i$ and $\hat{\psi}_i^\dagger$ with 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 system is described by the Heisenberg equations of motion for annihilation operators $\hat{\psi}_i$:

$$\begin{aligned} \frac{d\psi_a}{dt} &= 2i \sum_j \chi_{aj} |\psi_j|^2 \psi_a + (N-1)i\lambda \psi_m \psi_a^{*N-2} - i\Omega \psi_p \psi_m^*, \\ \frac{d\psi_m}{dt} &= 2i \sum_j \chi_{mj} |\psi_j|^2 \psi_m + (i\delta - \gamma)\psi_m + i\lambda \psi_a^{N-1} \\ &\quad - i\Omega \psi_p \psi_a^*, \\ \frac{d\psi_p}{dt} &= 2i \sum_j \chi_{pj} |\psi_j|^2 \psi_p + i(\Delta + \delta)\psi_p - i\Omega \psi_m \psi_a, \end{aligned} \quad (2)$$

in which $\chi_{ij} = n\chi'_{ij}$, $\lambda = n^{\frac{N}{2}-1}\lambda'$, 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 polymer state.

To seek a steady CPT state with $|\psi_m^0| = 0$, we first neglect the particle loss by taking $\gamma = 0$ and start by considering the steady-state solutions of Eqs. (2) through the following steady-state ansatz (μ_a is the atomic chemical potential):

$$\begin{aligned} \psi_a &= |\psi_a^0| \exp[i(\theta_a - \mu_a t)], \\ \psi_m &= |\psi_m^0| \exp[(N-1)i(\theta_a - \mu_a t)], \\ \psi_p &= |\psi_p^0| \exp[Ni(\theta_a - \mu_a t)]. \end{aligned} \quad (3)$$

Putting Eqs. (3) into Eqs. (2) and keeping the intermediate state unpopulated, one can find that the following CPT solutions exist:

$$|\psi_m^0|^2 = 0, \quad (4)$$

$$|\psi_p^0|^2 = \frac{1}{N}(1 - |\psi_a^0|^2), \quad (5)$$

where $|\psi_a^0|^2$ satisfies the following algebraic equation:

$$|\psi_a^0|^2 + N \left(\frac{\lambda}{\Omega} \right)^2 (|\psi_a^0|^2)^{N-2} - 1 = 0 \quad (6)$$

and the conserved total particle number $|\psi_a|^2 + (N-1)|\psi_m|^2 + N|\psi_p|^2 = 1$. The chemical potential and the generalized two-photon resonance conditions [20] are

$$\mu_a = -2(\chi_{aa} + \chi_{ap})|\psi_a^0|^2 \quad (7)$$

and

$$\Delta = -\delta + 2(N\chi_{aa} - \chi_{pa})|\psi_a^0|^2 + 2(N\chi_{ap} - \chi_{pp})|\psi_p^0|^2. \quad (8)$$

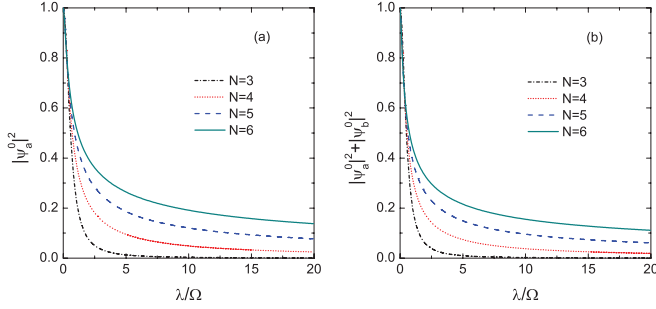


FIG. 2. (Color online) Atomic populations of CPT as functions of λ/Ω for (a) homonuclear and (b) heteronuclear systems.

Figure 2(a) shows the atomic population $|\psi_a|^2$ satisfying Eq. (6) as a function of λ/Ω for different N (with $N = 3, 4, 5, 6$ as examples). Clearly, for increasing λ/Ω we find that $|\psi_a|^2$ tends to zero monotonically, and a smaller N accelerates this decrease. This implies not only that the population distribution has the remarkable property of allowing all the atoms to be converted into polymers as λ/Ω changes from zero to infinity as long as the two-photon resonance condition (8) can be maintained dynamically but also that the conversion efficiency from ultracold atoms to more complex polymers is much lower. Since the temporal dependence of Ω uniquely determines the time evolution of the population, we can design *a priori* the temporal evolution of the laser detuning Δ in accordance with $\Omega(t)$ such that the resonance condition (8) is maintained dynamically at any time. It is noted that although, in principle, the detuning δ can be varied temporally to optimize the conversion efficiency, in actual experiments, we can fix δ at a finite value; it not only simplifies the experimental procedure, but more importantly, it also allows us to avoid strong condensate losses near the exact resonance ($\delta = 0$).

B. Heteronuclear polymer

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

$$\begin{aligned} \hat{H} = & -\hbar \int d\mathbf{r} \left\{ \sum_{i,j} \chi'_{ij} \hat{\psi}_i^\dagger(\mathbf{r}) \hat{\psi}_j^\dagger(\mathbf{r}) \hat{\psi}_j(\mathbf{r}) \hat{\psi}_i(\mathbf{r}) + \delta \hat{\psi}_m^\dagger(\mathbf{r}) \hat{\psi}_m(\mathbf{r}) \right. \\ & + \lambda \{ \hat{\psi}_m^\dagger(\mathbf{r}) [\hat{\psi}_a(\mathbf{r})]^{N-1} + \text{H.c.} \} + (\Delta + \delta) \hat{\psi}_p^\dagger(\mathbf{r}) \hat{\psi}_p(\mathbf{r}) \\ & \left. - \Omega' [\hat{\psi}_p^\dagger(\mathbf{r}) \hat{\psi}_m(\mathbf{r}) \hat{\psi}_b(\mathbf{r}) + \text{H.c.}] \right\}, \end{aligned} \quad (9)$$

where the subscript b represents atom B and the indices $i, j = a, m, p$ stand for the atom, intermediate polymer A_{N-1} , and polymer $A_{N-1}B$ states, respectively. Using the mean-field approximation, the system is described by the equations of motion

$$\begin{aligned} \frac{d\psi_a}{dt} &= 2i \sum_j \chi_{aj} |\psi_j|^2 \psi_a + i(N-1)\lambda \psi_m \psi_a^{*N-2}, \\ \frac{d\psi_b}{dt} &= 2i \sum_j \chi_{bj} |\psi_j|^2 \psi_b - i\Omega \psi_p \psi_m^*, \end{aligned}$$

$$\begin{aligned} \frac{d\psi_m}{dt} &= 2i \sum_j \chi_{mj} |\psi_j|^2 \psi_m + (i\delta - \gamma) \psi_m + i\lambda \psi_a^N \\ &\quad - i\Omega \psi_p \psi_b^*, \\ \frac{d\psi_p}{dt} &= 2i \sum_j \chi_{pj} |\psi_j|^2 \psi_p + i(\Delta + \delta) \psi_p - i\Omega \psi_m \psi_b, \end{aligned} \quad (10)$$

where $\chi_{ij} = n\chi'_{ij}$, $\lambda = n^{\frac{N}{2}-1}\lambda'$, and $\Omega = \sqrt{n}\Omega'$ are the renormalized quantities and the decay rate γ accounts for the loss of untrapped polymers. By using the steady-state ansatz

$$\begin{aligned} \psi_a &= |\psi_a^0| \exp[i(\theta_a - \mu_a t)], \\ \psi_b &= |\psi_b^0| \exp[i(\theta_b - \mu_b t)], \\ \psi_m &= |\psi_m^0| \exp[i(N-1)(\theta_a - \mu_a t)], \\ \psi_p &= |\psi_p^0| \exp(i\{[(N-1)\theta_a + \theta_b] - [(N-1)\mu_a + \mu_b]t\}), \end{aligned} \quad (11)$$

where μ_b is the chemical potential of atom B , one can obtain the following CPT solutions:

$$|\psi_b^0|^2 = \frac{|\psi_a^0|^2}{(N-1)}, \quad (12)$$

$$|\psi_m^0|^2 = 0, \quad (13)$$

$$|\psi_p^0|^2 = \frac{1}{N} - \frac{|\psi_a^0|^2}{N-1}, \quad (14)$$

and $|\psi_a|^2$ satisfies the following algebraic equation:

$$\frac{N}{N-1} |\psi_a^0|^2 + N(N-1) \left(\frac{\lambda}{\Omega} \right)^2 (|\psi_a^0|^2)^{N-2} - 1 = 0, \quad (15)$$

where we have used the condition of conserved particles number, $|\psi_a|^2 + |\psi_b|^2 + (N-1)|\psi_m|^2 + N|\psi_p|^2 = 1$. The atomic chemical potentials and generalized two-photon resonance condition are

$$\begin{aligned} \mu_a &= -2(\chi_{aa} |\psi_a^0|^2 + \chi_{ab} |\psi_b^0|^2 + \chi_{ap} |\psi_p^0|^2), \\ \mu_b &= -2(\chi_{ba} |\psi_a^0|^2 + \chi_{bb} |\psi_b^0|^2 + \chi_{bp} |\psi_p^0|^2), \end{aligned} \quad (16)$$

and

$$\begin{aligned} \Delta &= -\delta + 2[(N-1)\chi_{aa} + \chi_{ba} - \chi_{pa}] |\psi_a^0|^2 \\ &\quad + 2[(N-1)\chi_{ab} + \chi_{bb} - \chi_{pb}] |\psi_b^0|^2 \\ &\quad + 2[(N-1)\chi_{ap} + \chi_{bp} - \chi_{pp}] |\psi_p^0|^2. \end{aligned} \quad (17)$$

The atomic population $|\psi_a|^2 + |\psi_b|^2$ satisfying Eqs. (12) and (15) as a function of λ/Ω for different N is illustrated in Fig. 2(b). Similarly, we can find that, by dynamically maintaining the resonance condition, the population can be concentrated in atomic and polymer bound states under the respective limits $\lambda/\Omega \rightarrow 0$ and $\lambda/\Omega \rightarrow \infty$.

III. LINEAR INSTABILITY AND ADIABATIC FIDELITY OF THE CPT STATE

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

It is important to avoid the occurrence of instability in the generalized STIRAP. To this end, we linearize the equations of motion including the chemical potential and obtain the Jacobi matrix around the fixed point (CPT state) for the atom-polymer 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 [49]) can be obtained analytically as

$$\omega = \pm \sqrt{\frac{(B \pm \sqrt{B^2 - 4C})}{2}}, \quad (18)$$

where

$$B = \begin{cases} 2N(N-2)\lambda^2|\psi_a^0|^{2(N-2)} + 2|\psi_a^0|^2\Omega^2 + A^2, & \text{homonuclear,} \\ 2(N-1)(N-2)\lambda^2|\psi_a^0|^{2(N-2)} + \frac{2}{N-1}\Omega^2|\psi_a^0|^2 + A^2, & \text{heteronuclear,} \end{cases} \quad (19)$$

$$C = \begin{cases} [N(N-2)\lambda^2|\psi_a^0|^{2(N-2)} + |\psi_a^0|^2\Omega^2]^2 + 4A\lambda^2(N^2\chi_{aa} - 2N\chi_{ap} + \chi_{pp})|\psi_a^0|^{2(N-1)}, & \text{homonuclear,} \\ [(N-1)(N-2)\lambda^2|\psi_a^0|^{2(N-2)} + \frac{1}{N-1}|\psi_a^0|^2\Omega^2]^2 + 4A\lambda^2|\psi_a^0|^{2(N-1)}[(N-1)\chi_{aa} + 2(N-1)\chi_{ab} - 2(N-1)\chi_{ap} + \chi_{bb} - 2\chi_{bp} + \chi_{pp}], & \text{heteronuclear,} \end{cases} \quad (20)$$

with $A = -\delta + 2[(N-1)\chi_{aa} - \chi_{am}]\psi_a^0|^2 + 2[(N-1)\chi_{ap} - \chi_{mp}]\psi_p^0|^2$ for the homonuclear atom-polymer system and $A = -\delta + 2[(N-1)\chi_{aa} - \chi_{am}]\psi_a^0|^2 + 2[(N-1)\chi_{ab} - \chi_{bm}]\psi_b^0|^2 + 2[(N-1)\chi_{ap} - \chi_{mp}]\psi_p^0|^2$ for the heteronuclear atom-polymer system. When ω becomes complex, the corresponding CPT state is dynamically unstable. Hence, the unstable regime is given by either $C < 0$ or $C > B^2/4$. The typical results from the stability analysis based on the parameters of interest here are summarized in Fig. 3, where the (Ω, δ) space is divided into the stable (white) and the unstable (colored) regions. There are two unstable regions: Region I is thin along the Ω dimension and corresponds to the unstable region obtained by setting $C > B^2/4$, which expands

and shifts as N increases; region II occurs at small Ω and is the unstable region obtained by setting $C < 0$, whose width becomes thin with increasing N for the homonuclear system, while it is almost unchanged for the heteronuclear system. In order to convert atoms into stable polymer molecules, 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 polymers Cs_N and Cs_{N-1}Rb . The s -wave scattering lengths for cesium and rubidium atoms are $a = -374a_0$ [36] and $a = 100a_0$ [50] (a_0 is Bohr's radius). We choose $\lambda = 1.961 \times 10^4 \text{ s}^{-1}$ and the atom density $n = 6 \times 10^{19} \text{ m}^{-3}$. 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 [44]. We note that the time is in units of $1/\lambda$ and all other coefficients are in units of λ .

In the stable regions, the existence of the CPT state facilitates the adiabatic coherent population transfer between atoms and polymers. The time evolution of the population can be obtained by solving Eqs. (2) and (10). We plot the numerical results of the population $|\psi_p|^2$ in Fig. 4. Also plotted in Fig. 4 are the corresponding analytical CPT solutions of Eqs. (5) and (14). As can be seen, the exact population dynamics follows closely the prediction of the CPT solutions at the initial time. However, the discrepancies occur at later times, and the deviations will increase when N increases. In our calculations, the Rabi frequency is modulated as

$$\Omega(t) = \Omega_0 \text{sech} \frac{t}{\tau}, \quad (21)$$

where Ω_0 is the pulse strength and τ is the width of the pulse. The value of δ is chosen so that the system remains in the stable regions.

The deviation between the actual evolution state and the CPT state implies that the system cannot maintain adiabaticity completely. The reason can be attributed to the loss of adiabaticity and the decay of the intermediate polymer state. We neglect the particle loss by taking $\gamma = 0$ for calculating the CPT solution but include it by taking the decay parameter $\gamma = 1.0$ for the numerical result. In fact, the adiabatic evolution of our system can be thoroughly studied quantitatively by employing the adiabatic fidelity [44,51,52], which describes the distance between the adiabatic solution and the actual one.

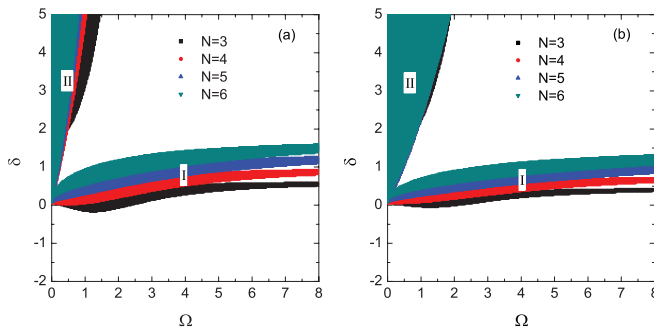


FIG. 3. (Color online) Instability diagrams in (Ω, δ) space for (a) homonuclear and (b) heteronuclear atom-polymer systems. The colored areas correspond to the unstable regions. All parameters are in units of λ .

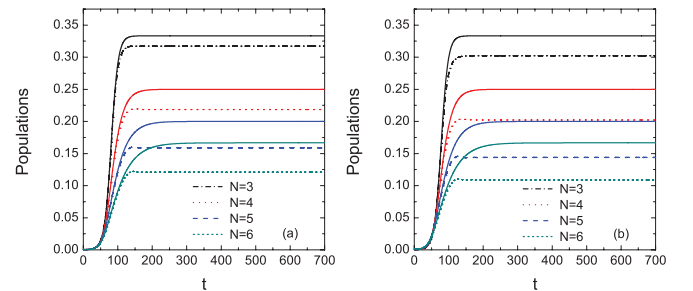


FIG. 4. (Color online) Populations as functions of time with $\delta = -2.0$ for (a) homonuclear and (b) heteronuclear systems. The solid lines correspond to the CPT, and other lines indicate the populations $|\psi_p|^2$. The other parameters are $\Omega_0 = 50$, $\tau = 20$, and $\gamma = 1.0$. The time is in units of $1/\lambda$, and all the other parameters are in units of λ .

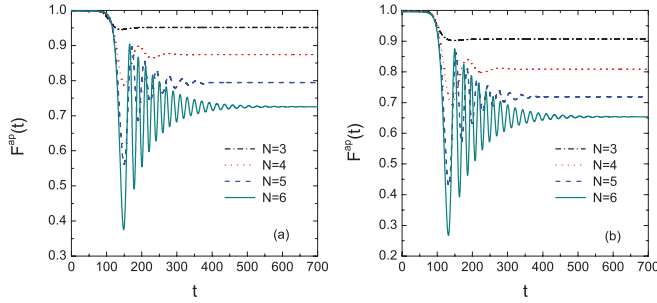


FIG. 5. (Color online) Adiabatic fidelity as a function of time with $\delta = -2.0$ for (a) homonuclear and (b) heteronuclear systems. The other parameters are the same as in Fig. 4. The time is in units of $1/\lambda$, and all the other parameters are in units of λ .

Here we define the adiabatic fidelity of the CPT state for atom-polymer conversion systems as

$$F^{ap}(t) = |\langle \overline{\psi}(t) | \overline{\psi}_{\text{CPT}}(t) \rangle|^2, \quad (22)$$

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

$$|\overline{\psi}(t)\rangle = \begin{cases} \left(\frac{\psi_a^N}{|\psi_a|^{N-1}}, \sqrt{N-1} \frac{\psi_a \psi_m}{|\psi_a|}, \sqrt{N} \psi_p \right)^T, \\ \text{homonuclear,} \\ \left(\frac{\psi_a^{N-1} \psi_b}{|\psi_a|^{N-2} |\psi_b|}, \frac{\psi_a^{N-1} \psi_b}{|\psi_a|^{N-1}}, \sqrt{N-1} \frac{\psi_b \psi_m}{|\psi_b|}, \sqrt{N} \psi_p \right)^T, \\ \text{heteronuclear.} \end{cases} \quad (23)$$

The variation of the adiabatic fidelity with time for homonuclear and heteronuclear polymers via the polymer-intermediated dark-state scheme is shown in Figs. 5(a) and 5(b). The evolution process is divided into three windows along the time dimension. The first window perches at the initial time, and the magnitude of the adiabatic fidelity is about 1. This implies that the system can adiabatically evolve along the CPT state in the region. Subsequently, it arrives at the oscillation window; the magnitude of the adiabatic fidelity first decreases sharply to minimal values and then oscillates with gradually damped amplitude with time. Finally, the adiabatic fidelity approaches a steady value which is smaller than 1, corresponding to the third window. The evolution process is strongly dependent on the atomic number N , interparticle interactions, and external field parameters. As N increases, whether for homonuclear or heteronuclear systems, the oscillation window will broaden, and the magnitude of the oscillation will increase, while the final value will decrease. This indicates the conversion efficiency from ultracold atoms to more complex polymers is much lower under the same conditions. Further studies show that (1) the oscillation of

adiabatic fidelity is due to interparticle interactions and (2) as the width τ of the pulse increases, both the first and second windows will expand, the magnitude of the oscillation will decrease, and the final value of adiabatic fidelity will increase. This means the stable formation of polymers is always possible by optimizing the parameters of the system.

Furthermore, we find that the dependence of the conversion efficiency on the other external field parameters is similar to the case of the pentamers [44]. Stable creation of polymers 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 a higher efficiency without considering the two-body interactions. It is clear that the conversion efficiency from ultracold atoms to N -body polymers can be controlled effectively by the external field parameters δ, Ω_0 , and τ .

IV. CONCLUSIONS

In conclusion, we have investigated the homonuclear and heteronuclear molecular N -body polymer conversion problem via the generalized STIRAP scheme in an ultracold bosonic system that includes interparticle interactions. We have derived a universal algebraic equation that satisfies the CPT state solution. The linear instability and the adiabatic fidelity of the atom-polymer dark state in the STIRAP have been studied via analysis of linear stability and proper definition of adiabatic fidelity, respectively. We have also discussed the effects of the interparticle interactions, the atomic number of the polymer, and external field parameters on the conversion process. We have found that interparticle interactions will lead to the instability of the CPT state, induce the oscillation of adiabatic fidelity, and suppress the conversion efficiency of polymers; the atomic number will affect the regions of both instability and oscillation, and the width of pulse τ will influence both the region and the magnitude of the oscillation of adiabatic fidelity. Our scheme not only includes previous methods for creation of ultracold trimer, tetramer, and pentamer molecules but also provides a possible route for the formation of ultracold N -body polyatomic molecules experimentally in the future.

ACKNOWLEDGMENTS

We thank Dr. H. Cao and Dr. S. C. Li for helpful discussions. The work is supported by the NFRP (Grants No. 2011CB921503 and No. 2013CB834100), the NNSF of China (Grants No. 91021021, No. 11075020, No. 11274051, and No. 11005055), the Fundamental Research Funds for the Higher Education Institutions of Gansu Province of China, and the Scientific Research Foundation of NWN (Grant No. NWN-LKQN-10-24).

[1] T. Köhler, K. Góral, and P. S. Julienne, *Rev. Mod. Phys.* **78**, 1311 (2006); L. D. Carr, D. DeMille, R. V. Krems, and J. Ye, *New J. Phys.* **11**, 055049 (2009); B. Friedrich and J. M. Doyle, *Chem. Phys. Chem.* **10**, 604 (2009).

[2] S. Ospelkaus, K.-K. Ni, D. Wang, M. H. G. de Miranda, B. Neyenhuis, G. Quémener, P. S. Julienne, J. L. Bohn, D. S. Jin, and J. Ye, *Science* **327**, 853 (2010).

[3] D. DeMille, *Phys. Rev. Lett.* **88**, 067901 (2002).

- [4] J. Liu, B. Liu, and L. B. Fu, *Phys. Rev. A* **78**, 013618 (2008); J. Liu, L. B. Fu, B. Liu, and B. Wu, *New J. Phys.* **10**, 123018 (2008); L. B. Fu and J. Liu, *Ann. Phys. (NY)* **325**, 2425 (2010).
- [5] E. S. Shuman, J. F. Barry, and D. DeMille, *Nature (London)* **467**, 820 (2010); I. Manai, R. Horchani, H. Lignier, P. Pillet, D. Comparat, A. Fioretti, and M. Allegrini, *Phys. Rev. Lett.* **109**, 183001 (2012); B. K. Stuh, M. T. Hummon, M. Yeo, G. Quéméner, J. L. Bohn, and J. Ye, *Nature (London)* **492**, 396 (2012).
- [6] J. Küpper, F. Filsinger, and G. Meijer, *Faraday Discuss.* **142**, 155 (2009); D. Patterson, E. Tsikata, and J. Doyle, *Phys. Chem. Chem. Phys.* **12**, 9736 (2010).
- [7] H. Sabbath, L. Biennier, I. R. Sims, Y. Georgievskii, S. J. Klippenstein, and I. W. M. Smith, *Science* **317**, 102 (2007); J. J. Larsen, I. Wendt-Larsen, and H. Stapelfeldt, *Phys. Rev. Lett.* **83**, 1123 (1999).
- [8] K. Hornberger, S. Uttenthaler, B. Brezger, L. Hackermüller, M. Arndt, and A. Zeilinger, *Phys. Rev. Lett.* **90**, 160401 (2003).
- [9] V. V. Flambaum and M. G. Kozlov, *Phys. Rev. Lett.* **99**, 150801 (2007).
- [10] R. A. Bartels, T. C. Weinacht, N. Wagner, M. Baertschy, C. H. Greene, M. M. Murnane, and H. C. Kapteyn, *Phys. Rev. Lett.* **88**, 013903 (2001); T. Seideman, *Phys. Rev. A* **64**, 042504 (2001); C. A. R. Sá de Melo, *Phys. Today* **61**(10), 45 (2008).
- [11] M. Zeppenfeld, B. G. U. Englert, R. Glöckner, A. Prehn, M. Mielenz, C. Sommer, L. D. van Buuren, M. Motsch, and G. Rempe, *Nature (London)* **491**, 570 (2012).
- [12] J. F. Barry and D. Demille, *Nature (London)* **491**, 539 (2012).
- [13] J. D. Weinstein, R. deCarvalho, T. Guillet, B. Friedrich, and J. M. Doyle, *Nature (London)*, **395**, 148 (1998).
- [14] R. Fulton, A. I. Bishop, and P. F. Barker, *Phys. Rev. Lett.* **93**, 243004 (2004).
- [15] M. T. Hummon, T. V. Tscherbul, J. Klos, H.-I. Lu, E. Tsikata, W. C. Campbell, A. Dalgarno, and J. M. Doyle, *Phys. Rev. Lett.* **106**, 053201 (2011).
- [16] A. O. G. Wallis and J. M. Hutson, *Phys. Rev. Lett.* **103**, 183201 (2009).
- [17] T. V. Tscherbul, H.-G. Yu, and A. Dalgarno, *Phys. Rev. Lett.* **106**, 073201 (2011).
- [18] 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).
- [19] 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).
- [20] H. Y. Ling, H. Pu, and B. Seaman, *Phys. Rev. Lett.* **93**, 250403 (2004).
- [21] H. Jing, J. Cheng, and P. Meystre, *Phys. Rev. Lett.* **99**, 133002 (2007); *Phys. Rev. A* **77**, 043614 (2008).
- [22] K. Winkler, F. Lang, G. Thalhammer, P. v. d. Straten, R. Grimm, and J. H. Denschlag, *Phys. Rev. Lett.* **98**, 043201 (2007); F. Lang, K. Winkler, C. Strauss, R. Grimm, and J. Hecker Denschlag, *ibid.* **101**, 133005 (2008).
- [23] J. G. Danzl, E. Haller, M. Gustavsson, M. J. Mark, R. Hart, N. Bouloufa, O. Dulieu, H. Ritsch, and H.-C. Näerl, *Science* **321**, 1062 (2008).
- [24] K.-K. Ni, S. Ospelkaus, M. H. G. de Miranda, A. Peer, B. Neyenhuis, J. J. Zirbel, S. Kotochigova, P. S. Julienne, D. S. Jin, and J. Ye, *Science* **322**, 231 (2008); S. Ospelkaus, A. Peer, K.-K. Ni, J. J. Zirbel, B. Neyenhuis, S. Kotochigova, P. S. Julienne, J. Ye, and D. S. Jin, *Nat. Phys.* **4**, 622 (2008).
- [25] F. M. Spiegelhalder, A. Trenkwalder, D. Naik, G. Kerner, E. Wille, G. Hendl, F. Schreck, and R. Grimm, *Phys. Rev. A* **81**, 043637 (2010).
- [26] S. Stellmer, B. Pasquiou, R. Grimm, and F. Schreck, *Phys. Rev. Lett.* **109**, 115302 (2012).
- [27] V. Efimov, *Phys. Lett. B* **33**, 563 (1970).
- [28] 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).
- [29] E. Braaten, H.-W. Hammer, and M. Kusunoki, *Phys. Rev. Lett.* **90**, 170402 (2003).
- [30] S. Knoop, F. Ferlaino, M. Mark, M. Berninger, H. Schöel, H.-C. Näerl, and R. Grimm, *Nat. Phys.* **5**, 227 (2009).
- [31] C. Chin, T. Kraemer, M. Mark, J. Herbig, P. Waldburger, H. C. Nägerl, and R. Grimm, *Phys. Rev. Lett.* **94**, 123201 (2005).
- [32] O. Machtey, Z. Shotan, N. Gross, and L. Khaykovich, *Phys. Rev. Lett.* **108**, 210406 (2012).
- [33] M. Stoll and T. Köhler, *Phys. Rev. A* **72**, 022714 (2005).
- [34] H. W. Hammer and L. Platter, *Eur. Phys. J. A* **32**, 113 (2007).
- [35] J. von Stecher, J. P. D’Incao, and C. H. Greene, *Nat. Phys.* **5**, 417 (2009).
- [36] F. Ferlaino, S. Knoop, M. Berninger, W. Harm, J. P. D’Incao, H.-C. Nägerl, and R. Grimm, *Phys. Rev. Lett.* **102**, 140401 (2009).
- [37] S. E. Pollack, D. Dries, and R. G. Hulet, *Science* **326**, 1683 (2009).
- [38] R. Schmidt and S. Moroz, *Phys. Rev. A* **81**, 052709 (2010).
- [39] M. R. Hadizadeh, M. T. Yamashita, L. Tomio, A. Delfino, and T. Frederico, *Phys. Rev. Lett.* **107**, 135304 (2011).
- [40] J. von Stecher, *J. Phys. B* **43**, 101002 (2010); *Phys. Rev. Lett.* **107**, 200402 (2011); M. Thøersen, D. V. Fedorov, and A. S. Jensen, *Europhys. Lett.* **83**, 30012 (2008).
- [41] G. J. Hanna and D. Blume, *Phys. Rev. A* **74**, 063604 (2006).
- [42] M. T. Yamashita, D. V. Fedorov, and A. S. Jensen, *Phys. Rev. A* **81**, 063607 (2010).
- [43] H. Jing and Y. Jiang, *Phys. Rev. A* **77**, 065601 (2008); G. Q. Li and P. Peng, *ibid.* **83**, 043605 (2011).
- [44] F. Q. Dou, S. C. Li, H. Cao, and L. B. Fu, *Phys. Rev. A* **85**, 023629 (2012).
- [45] E. M. Graefe, H. J. Korsch, and A. E. Niederle, *Phys. Rev. Lett.* **101**, 150408 (2008).
- [46] S. C. Li, J. Liu, and L. B. Fu, *Phys. Rev. A* **83**, 042107 (2011).
- [47] A. S. Parkins and D. F. Walls, *Phys. Rep.* **303**, 1 (1998).
- [48] F. A. van Abeelen and B. J. Verhaar, *Phys. Rev. Lett.* **83**, 1550 (1999).
- [49] H. Y. Ling, P. Maenner, W. P. Zhang, and H. Pu, *Phys. Rev. A* **75**, 033615 (2007).
- [50] A. Widera, O. Mandel, M. Greiner, S. Kreim, T. W. Hänsch, and I. Bloch, *Phys. Rev. Lett.* **92**, 160406 (2004).
- [51] L. H. Lu and Y. Q. Li, *Phys. Rev. A* **77**, 053611 (2008).
- [52] 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).