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Dynamical instability and adiabatic evolution of the atom–homonuclear–trimer dark state in a condensate system*

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This paper investigates the dynamical instability and adiabatic evolution of the atom–homonuclear–trimer dark state of a condensate system in a stimulated Raman adiabatic passage aided by Feshbach resonance. It obtains analytically the regions for the appearance of dynamical instability caused by the interparticle interactions. Moreover, the adiabatic property of the dark state is also studied in terms of a newly defined adiabatic fidelity. It shows that the nonlinear collisions have a negative effect on the adiabaticity of the dark state and hence reduce the conversion efficiency.

Keywords: stimulated Raman adiabatic passage, dark state, dynamical instability, adiabatic evolution

PACC: 3280P, 3380P, 0530J

1. Introduction

Since the experimental realization of the trapped atomic Bose–Einstein condensate (BEC),[1] the formation of an ultracold molecular gas or even a molecular BEC from its atomic counterparts has been regarded as another milestone in the field of ultracold atomic physics in recent years.[2,3] To create stable ground state molecules with high conversion efficiency, the stimulated Raman adiabatic passage (SRAP)[4−10] in photoassociation (PA)[11] or aided by Feshbach resonance (FR)[12−14] has been suggested as a more effective method than the bare PA or FR technique. The success of the SRAP relies on the existence of the coherent population trapping (CPT) state, or dark state[15,16], which can be followed adiabatically. However, the atom–molecule SRAP contains nonlinearities which arise from the mean-field treatment of the interparticle interactions and converting atoms to molecules. These nonlinearities do not justify the application of the adiabatic condition of quantum mechanics to study adiabaticity of atom–molecule coupling systems. Moreover, the nonlinear collisions could also bring forth a dynamical instability[17,18] that makes the real solution deviate rapidly from the CPT state in the adiabatic evolution, results in low atom-to-molecule conversion efficiency, and should be avoided for the success of the SRAP. Therefore, it is important to investigate the adiabatic theory and dynamical instability for such kinds of nonlinear systems.

The analysis of stability and adiabaticity for the atom–dimer conversion system in the SRAP has been presented in recent papers.[19−21] Adiabaticity means that there is no population growth in the collective excitations of the dark state, whereas the occurrence of the dynamical instability is accompanied with the emergence of complex frequencies in the collective modes. Following the above works, the adiabatic theory for the same system is further well studied by means of the classical adiabatic dynamics[22] and the adiabatic fidelity[23−26] which describes the difference between the actual evolution state and the adiabatic state (dark state). Meanwhile the nonlinear instability is also investigated by the resonance normal form

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theory.[22,27] Very recently, the SRAP technique has been generalized to create more complex molecules, trimers or tetramers.[28−30] Hence it is worthwhile to study the instability and adiabatic property of the dark state in such complex systems.

In the present paper, we investigate the dynamical instability and adiabatic evolution of the atom–homonuclear–trimer conversion system[30] in the SRAP aided by FR. Under the mean-field approximation, making use of the classical Hamiltonian dynamics and analysing the eigenvalues of the Hamiltonian–Jacobi matrix obtained via linearizing the equations of motion around the fixed point which corresponds to the CPT state, we obtained analytically the regions for the occurrence of dynamical instability of the dark state characterized by the emergence of real or complex eigenvalues of the Hamiltonian–Jacobi matrix. It shows that, as in the atom–dimer conversion system,[20] such instability is caused by the interparticle interactions. Taking the ^{87}Rb condensate system as an example, we numerically give the unstable regions. Moreover, by properly defining the fidelity for this system, the adiabatic evolution of the dark state is quantitatively studied in terms of the adiabatic fidelity. We find that the nonlinear collisions bring a negative effect to the adiabatic evolution and make the adiabaticity of the dark state worse.

2. Model, CPT state

Our model system consists of a Bose condensate of atoms coupled to molecular dimers (A_2) via a FR; these dimers are in turn photoassociated to the atoms to form bound trimers (A_3).[30] Denoting the strength of the atom–dimer coupling by λ' with detuning δ, the Rabi frequency of the dimer–trimer coupling optical field by Ω' with detuning Δ, including s-wave scattering processes, we get the second quantized Hamiltonian under the rotating frame,

$$\begin{aligned}\hat{H} = -\hbar\Big\{ & \sum_{i,j}\chi_{ij}\hat{\psi}_i^{\dagger}\hat{\psi}_j^{\dagger}\hat{\psi}_j\hat{\psi}_i + \delta\hat{\psi}_{\rm d}^{\dagger}\hat{\psi}_{\rm d} \\ & +(\Delta+\delta)\hat{\psi}_{\rm g}^{\dagger}\hat{\psi}_{\rm g} + \lambda'[\hat{\psi}_{\rm d}^{\dagger}\hat{\psi}_{\rm a}^2 + {\rm H.c.}] \\ & -\Omega'[\hat{\psi}_{\rm g}^{\dagger}\hat{\psi}_{\rm d}\hat{\psi}_{\rm a} + {\rm H.c.}]\Big\}, \end{aligned} \tag{1}$$

where $\hat{\psi}_i$ ($\hat{\psi}_i{}^{\dagger}$) are the bosonic annihilation (creation) operators, the collision terms are proportional to χ_{ij} and describe s-wave collisions, the indices $i, j = {\rm a}, {\rm d}, {\rm g}$ stand for the atoms, dimers and trimers, respectively.

As in Refs.[7, 24], to guarantee the conservation of the total particle numbers, we introduce a multiplier $\hbar\mu$ into the Hamiltonian in Eq.(1) and obtain the grand canonical Hamiltonian, 'Kamiltonian',

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

where $\hbar\mu$ is identified as the chemical potentials of atoms, $\hat{N} = \hat{\psi}_{\rm a}{}^{\dagger}\hat{\psi}_{\rm a} + 2\hat{\psi}_{\rm d}{}^{\dagger}\hat{\psi}_{\rm d} + 3\hat{\psi}_{\rm g}{}^{\dagger}\hat{\psi}_{\rm g}$ is the operator for the total particle number.

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-numbers ψ_i and ψ_i^* respectively, the Schrödinger equations are,

$$\begin{aligned} {\rm i}\dot{\psi_{\rm a}} &= (\omega_{\rm a}-\mu)\,\psi_{\rm a} - \lambda\psi_{\rm d}\psi_{\rm a}^* + \Omega\psi_{\rm d}^*\psi_{\rm g}, \\ {\rm i}\dot{\psi_{\rm d}} &= (\omega_{\rm d}-\delta-{\rm i}\gamma-2\mu)\,\psi_{\rm d} - \lambda\psi_{\rm a}^2 + \Omega\psi_{\rm g}\psi_{\rm a}^*, \\ {\rm i}\dot{\psi_{\rm g}} &= [\omega_{\rm g}-(\Delta+\delta)-3\mu]\,\psi_{\rm g} + \Omega\psi_{\rm d}\psi_{\rm a}, \end{aligned} \tag{3}$$

where $\omega_i = -2n\sum_j \chi_{ij}\,|\psi_j|^2$, $\Omega = \Omega'\sqrt{n}$, $\lambda = \lambda'\sqrt{n}$ are the renormalized quantities, and the phenomenological parameter γ is introduced to characterize the decay of the quasibound molecules (dimer).

To seek the stationary solution, we take $\dot{x} \approx 0, x = \psi_{\rm a}, \psi_{\rm d}, \psi_{\rm g}$. Under the normalized condition $|\psi_{\rm a}|^2 + 2|\psi_{\rm d}|^2 + 2|\psi_{\rm g}|^2 = 1$, one can easily derive the following CPT solution ($\psi_{\rm d} = 0$):

$$|\psi_{\rm g}|^2 = \frac{(\lambda/\Omega)^2}{1+3\,(\lambda/\Omega)^2} = \frac{1}{3}\left(1-|\psi_{\rm a}|^2\right), \tag{4}$$

with the following 'two-photon' resonance condition and chemical potentials,

$$\begin{aligned} \Delta &= -\delta + (6\chi_{\rm ag}-2\chi_{\rm gg})\,|\psi_{\rm g}|^2 \\ &\quad + (6\chi_{\rm aa}-2\chi_{\rm ag})\,|\psi_{\rm a}|^2, \\ \mu_{\rm a} &= -2\left(\chi_{\rm aa}|\psi_{\rm a}|^2 + \chi_{\rm ag}|\psi_{\rm g}|^2\right). \end{aligned} \tag{5}$$

From Eqs.(4) and (5), we can conclude that, as long as the two-photon resonance condition can be maintained dynamically, population can be concentrated in the atomic state and trimer state under the respective limit $\lambda/\Omega \to 0$ and $\lambda/\Omega \to \infty$, which facilitates the adiabatic coherent population transfer between atoms and trimers.

3. Dynamical instability of the CPT state

The existence of the CPT state cannot guarantee that it can always be followed adiabatically. This is because the interparticle nonlinear collisions may

bring forth dynamical instability that makes the real solution deviate rapidly from the CPT state even in the adiabatic limit and hence results in low conversion efficiency. Therefore, it is important to avoid the occurrence of dynamical instability of the CPT state in the SRAP. For this purpose, we now investigate the dynamical instability of the atom–homonuclear–trimer dark state by using the classical Hamiltonian dynamics.[31−33]

Under the mean-field approximation, substituting $\psi_{\rm a} = x_{\rm a} + {\rm i}y_{\rm a}$, $\psi_{\rm d} = x_{\rm d} + {\rm i}y_{\rm d}$, $\psi_{\rm g} = x_{\rm g} + {\rm i}y_{\rm g}$ into Eq.(2), we cast the grand canonical Hamiltonian in the following form of a classical Hamiltonian:

$$\begin{aligned} K = & -\{\chi_{\rm aa}\big(x_{\rm a}^4 + 2x_{\rm a}^2y_{\rm a}^2 + y_{\rm a}^4\big) \\ & +\chi_{\rm dd}(x_{\rm d}^4 + 2x_{\rm d}^2y_{\rm d}^2 + y_{\rm d}^4) \\ & +\chi_{\rm gg}(x_{\rm g}^4 + 2x_{\rm g}^2y_{\rm g}^2 + y_{\rm g}^4) \\ & +2\chi_{\rm ad}(x_{\rm a}^2 + y_{\rm a}^2)(x_{\rm d}^2 + y_{\rm d}^2) \\ & +2\chi_{\rm ag}(x_{\rm a}^2 + y_{\rm a}^2)(x_{\rm g}^2 + y_{\rm g}^2) \\ & +2\chi_{\rm dg}(x_{\rm d}^2 + y_{\rm d}^2)(x_{\rm g}^2 + x_{\rm g}^2) \\ & +\delta(x_{\rm d}^2 + y_{\rm d}^2) + (\Delta + \delta)(x_{\rm g}^2 + y_{\rm g}^2) \\ & +2\lambda\big[x_{\rm d}(x_{\rm a}^2 + y_{\rm a}^2) + 2x_{\rm a}y_{\rm a}y_{\rm d}\big] \\ & -2\Omega\big[x_{\rm g}(x_{\rm a}x_{\rm d} - y_{\rm a}y_{\rm d}) \\ & +x_{\rm g}(x_{\rm a}y_{\rm d} + y_{\rm a}x_{\rm d})\big]\} \\ & -\mu\big[x_{\rm a}^2 + y_{\rm a}^2 + 2\big(x_{\rm d}^2 + y_{\rm d}^2\big) + 3\big(x_{\rm g}^2 + y_{\rm g}^2\big)\big]. \end{aligned} \quad (6)$$

Here x_i are the canonical momenta, while y_i are the coordinates. They are governed by the differential equations: $\dot{x_i} = \dfrac{\partial K}{\partial y_i}, \dot{y_i} = -\dfrac{\partial K}{\partial x_i}$. By setting $\dot{x_i} = \dot{y_i} = 0$, we can obtain the fixed point which corresponds to the CPT state: $x_{\rm a} = |\psi_{\rm a}|$, $y_{\rm a} = 0$, $x_{\rm d} = 0$, $y_{\rm d} = 0$, $x_{\rm g} = |\psi_{\rm g}|, y_{\rm g} = 0$ with the same chemical potential and the two-photon resonance condition in Eq.(5).

To study the stability of the fixed point, we have to analyse the eigenvalues of the Hamiltonian–Jacobi matrix[31] obtained by linearizing equations of motion of the classical Hamiltonian around the fixed point. These eigenvalues can be real, complex and pure imaginary. However, only pure imaginary ones correspond to the stable fixed points. Others indicate the unstable ones. Let $x_{\rm a} = z_1$, $y_{\rm a} = z_2$, $x_{\rm d} = z_3$, $y_{\rm d} = z_4$, $x_{\rm g} = z_5$, $y_{\rm g} = z_6$, then elements of the Hamiltonian–Jacobi matrix can be written elegantly as $J_{ij} = \dfrac{(-1)^i}{2}\dfrac{\partial^2 K}{\partial z_i \partial z_{j\pm1}}|_{\rm CPT}$. Here i, j are respectively indexes of rows and columns, and the plus (minus) sign is for odd (even) j. Substituting the CPT state into the matrix elements, we find the Hamiltonian–Jacobi matrix J around the fixed point (CPT state) for the atom–homonuclear–trimer conversion system. Then we solve the eigenvalues of J and obtain analytically the eigenvalues ϖ_i other than the zero-mode frequency of J.

$$\begin{aligned} & \varpi_{1,2\pm} = {\rm i}\omega_{1,2\pm} = \pm\frac{\rm i}{\sqrt{2}}\sqrt{b \pm \sqrt{b^2 - c}}, \\ & b = \xi^2 + 2\Omega^2, \\ & c = \frac{4\Omega^2\left(9\lambda^4 + 2(3 - 20\chi_{\rm aa} + 12\chi_{\rm ag})\Omega^2\lambda^2 + \Omega^4\right)}{\left(3\lambda^2 + \Omega^2\right)^2}, \end{aligned} \quad (7)$$

where $\xi = \delta + 2(\chi_{\rm ad} - 2\chi_{\rm aa})|\psi_{\rm a}|^2 + 2(\chi_{\rm dg} - 2\chi_{\rm ag})|\psi_{\rm g}|^2$, $\omega_{1,2\pm}$ are the intrinsic frequencies of the system and are identical to the Bogoliubov excitation frequencies of the system.[33] When ϖ becomes real or complex, the corresponding CPT state is dynamically unstable. Here $b = \xi^2 + 2\Omega^2 > 0$, hence the unstable region is given by either $c < 0$ or $c > b^2$. When the nonlinear collisions are absent, i.e., $\chi_{ij} = 0$, $c = 4\Omega^2$ satisfying $c < b^2$, $\varpi_\pm$ are all pure imaginary. Therefore, the property of the dynamics of the CPT state is always stable in this case. When the nonlinear collisions are present, i.e., $\chi_{ij} \neq 0$, there are some parameters which make $c < 0$ or $c > b^2$, as will be shown below. Hence it can be concluded that the dynamical instability of the atom–trimer dark state is caused by interparticle interactions.

In the following discussions, we consider the special ^{87}Rb condensate system, with the s-wave scattering length 5.77 nm. The parameters are chosen as: $\lambda = 4.718 \times 10^4\ {\rm s}^{-1}$, $\Omega = \Omega_0{\rm sech}(t/\tau)$ with $\Omega_0/\lambda = 20, \lambda\tau = 20$. As in Ref.[30], the collisional parameter $\chi_{\rm aa} = 0.5303$, and other collisions are taken as 0.0938 (all in units of λ/n).

Figures 1(a) and 1(b) respectively show the instability diagrams and a sample of the occurrence of instability with the parameter in the unstable regions. In Fig.1(a), the dark areas are the unstable regions corresponding to the two cases $c < 0$ or $c > b^2$. Region I corresponds to the unstable region obtained by setting $c > b^2$, whose width shrinks as Ω increases. Region II is the unstable region obtained by setting $c < 0$, whose width grows with increasing δ. When $\delta = 3$, the dynamics of the system is unstable once $0 < \Omega < 2$. That is the reason why the population dynamics deviate from the CPT solution as Ω decreases to 2 at the latter stage of evolution in Ref.[30] (Fig.1 with $\delta = 3$). For the success of SRAP, it is crucial to avoid these unstable regions when we design the route of adiabatic passage.

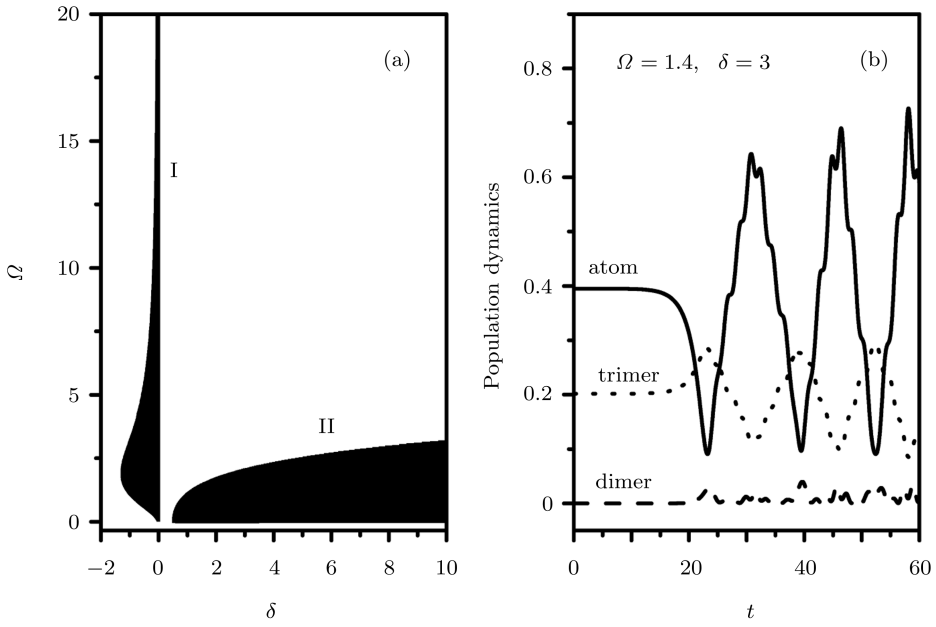

Fig.1. (a) Instability diagrams in δ, Ω space, in which the black areas are the unstable regions. (b) Sample of instability in the population dynamics with the parameters in (a). δ and Ω are in units of λ, time is in units of λ^{-1}.

4. Adiabatic evolution of the CPT state

Now we are in a position to study the adiabatic evolution of the CPT state. The atom–molecule conversion systems contain nonlinearities originating from the interparticle interactions and converting atoms to molecules, hence it is not justified to apply the adiabatic condition of quantum mechanics to study the adiabaticity of such systems because of the absence of the superposition principle. In fact, adiabatic evolution of a system can be thoroughly studied quantitatively by employing adiabatic fidelity[23–26] which describes the difference between the actual evolution state and the adiabatic solution (i.e., dark state). However, for the atom–homonuclear–trimer conversion system, because the Hamiltonian in the nonlinear Schrödinger equation (3) is a functional of both the wavefunction and its conjugate, the $U(1)$-invariance is broken. Therefore, the traditional definition of fidelity based on the $U(1)$ invariance is no longer suitable for this nonlinear system. Mathematically, we see that the Hamiltonian of the atom–homonuclear–trimer conversion system is invariant under the following transformation:

$$\boldsymbol{U}(\phi) = \begin{pmatrix} \mathrm{e}^{\mathrm{i}\phi} & 0 & 0 \\ 0 & \mathrm{e}^{\mathrm{i}2\phi} & 0 \\ 0 & 0 & \mathrm{e}^{\mathrm{i}3\phi} \end{pmatrix}. \tag{8}$$

With this transformation, $|\psi\rangle = (\psi_\mathrm{a}, \psi_\mathrm{d}, \psi_\mathrm{g})^\mathrm{T} \rightarrow |\psi'\rangle = U(\phi)|\psi\rangle$, they represent the same state. Therefore, if we denote the fidelity of two states $|\psi_1\rangle$ and $|\psi_2\rangle$ as $F(|\psi_1\rangle, |\psi_2\rangle)$, then this definition should not only satisfy $F(|\psi\rangle, |\psi\rangle) = 1$ but also fulfill $F(|\psi\rangle, U(\phi)|\psi\rangle) = 1$ for any ϕ. With this consideration, we define the fidelity for the atom–homonuclear–trimer conversion system as

$$F^\mathrm{am}(|\psi_1\rangle, |\psi_2\rangle) = \left|\langle\overline{\psi_1}|\,\overline{\psi_2}\rangle\right|^2, \tag{9}$$

where $|\overline{\psi}\rangle = \left(\psi_\mathrm{a}^2/|\psi_\mathrm{a}|, \sqrt{2}\psi_\mathrm{d}, \sqrt{3}\psi_\mathrm{g}\right)^\mathrm{T}$ is the rescaled wavefunction of $|\psi\rangle = (\psi_\mathrm{a}, \psi_\mathrm{d}, \psi_\mathrm{g})^\mathrm{T}$. As in Refs.[23] and [24], one can easily prove that this definition satisfies the above conditions and other conditions for the definition of fidelity.[34–36]

Making use of the above new definition of fidelity, we study quantitatively the adiabatic evolution of the CPT state. The adiabatic fidelity of the dark state can be denoted as $F = \left|\langle\overline{\psi(t)}|\overline{\mathrm{CPT}}\rangle\right|^2$ where $|\psi(t)\rangle$ is the exact solution of the Schrödinger equation in Eq.(3). $|\overline{\psi(t)}\rangle$ and $|\overline{\mathrm{CPT}}\rangle$ are respectively the rescaled

wavefunctions of $|\psi(t)\rangle$ and CPT state. If the system can adiabatically evolve along the CPT state, then the value of the adiabatic fidelity should be close to unity. In our calculation, we solve the nonlinear Schrödinger equation using the fourth–fifth order Runge–Kutta adaptive-step algorithm.

The time dependence of the external fields λ, Ω and adiabatic fidelity of the CPT state for the ^{87}Rb condensate system are shown in Figs.2(a) and 2(b), respectively. Both the cases for the nonlinear collisions being absent and present are considered. From Fig.2(b), we see that no matter whether the nonlinear collisions are included, the magnitude of adiabatic fidelity is about 1 at the initial time, but begins to decrease at the time 63, then diminishes to the minimal value, and finally approaches a steady value. In the absence of the nonlinear collisions, i.e., $\chi_{ij} = 0$, the minimum of adiabatic fidelity is 0.94 at the time $t = 121$, and the final value is 0.95. In the presence of the nonlinear collisions, i.e., $\chi_{ij} \neq 0$, the minimal value of adiabatic fidelity is 0.91 at the time $t = 107$, and the final value is about 0.93. In both the above cases, the minimal (final) adiabatic fidelity which can be used to describe the adiabaticity (coversion efficiency) of the system[23] is close to 1; this fact implies that the system can adiabatically evolve along the CPT state. However, the minimal (final) value of adiabatic fidelity in the latter case is smaller than that in the former case, hence the adiabaticity (conversion efficiency) of the system in the latter case is worse (lower) than that in the former case. It can be found that this negative effect is caused by the interparticle interactions.

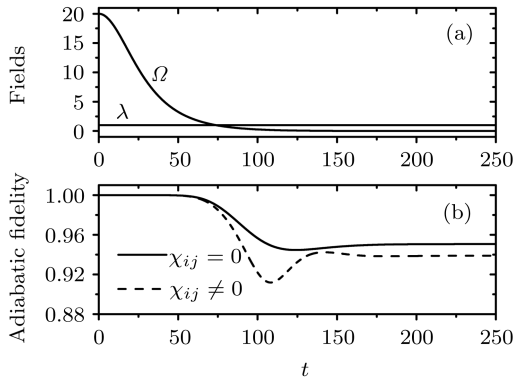

Fig.2. External fields (a) and adiabatic fidelity (b) as functions of time with and without nonlinear collisions for $\delta = -3, \gamma = 1$. Here λ is in units of 4.718×10^4 s^{-1}, and time is in units of λ^{-1} (δ and γ are in units of λ). Other parameters are defined in Section 3.

5. Conclusion

In conclusion, we investigate the dynamical instability and adiabatic evolution of an atom–homonuclear–trimer dark state in a condensate system in the SRAP aided by FR. We find that, as in the atom–dimer coupling system, the interparticle interactions may cause dynamical instability in some parameter regions. In addition, we obtain analytically the regions for the occurrence of dynamical instability. Taking the ^{87}Rb condensate system as an example, we numerically give the unstable regions. Moreover, we properly define the fidelity for the system. In calculating the adiabatic fidelity of the dark state, we study quantitatively the adiabatic evolution of the system. We show that the interparticle interactions have negative influence on the adiabaticity and conversion efficiency of the system.

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