GENERAL

Dynamical instability of the dark state in the conversion of Bose—Fermi mixtures into stable molecules

Dynamical instability of the dark state in the conversion of Bose–Fermi mixtures into stable molecules*

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In this paper, we investigate the dynamical instability of the dark state in the conversion of Bose–Fermi mixtures into stable molecules through a stimulated Raman adiabatic passage aided by Feshbach resonance. We analytically obtain the regions where the dynamical instability appears and find that such instability in the Bose–Fermi mixture system is caused not only by bosonic interparticle interactions but also by Pauli blocking terms, which is different from the scenario of a pure bosonic system where instability is induced by nonlinear interparticle collisions. Taking a ^{40}K–^{87}Rb mixture as an example, we give the unstable regions numerically.

Keywords: Bose–Fermi mixture, dynamical instability, dark state

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1. Introduction

Experiments on the Bose–Einstein condensates (BECs) of neutral atomic gases, first realized in 1995 and different from a pure bosonic system where the instability is induced by nonlinear interparticle collisions,[1−3] have led to a profound revolution in modern physics, from low-temperature physics to atom optics. In particular, there has much recent interest in the creation of a molecular BEC via the external field of its atomic counterpart.[4−8] The use of a stimulated Raman adiabatic passage (STIRAP) by photo-association[9−14] or aided by Feshbach resonance[15−17] to form ultracold molecules is an attractive idea, due mainly to the promise of a high yield. A key element of this scheme is to exploit a coherent population trapping (CPT) state or dark state,[18,19] i.e., a superposition of the free atomic and ground molecular states that, once formed, can effectively suppress the population loss of the excited molecular state and transfer free atoms to ground molecules with a high conversion efficiency during the adiabatic evolution.

However, the existence of the dark state in atom–molecule STIRAP does not guarantee that it can always be followed adiabatically since interparticle nonlinear collisions may bring dynamical instability[20−23] to the system, which is driven by the emergence of the complex intrinsic frequencies of the system in some parameter space of the interaction strength. This fact means that there is an eigenmode growing exponentially and that the condensate becomes unstable against infinitesimal perturbations, and hence makes the real solution deviate away rapidly from the dark state even in the adiabatic limit. Therefore, it is crucial for the success of STIRAP to avoid these unstable regimes when the route of adiabatic passage is designed. So far, the dynamical instability of the atom–molecule dark state has only been studied in pure bosonic systems.[13,15] In recent years much attention has been paid to converting Bose–Fermi[24] or Fermi–Fermi[25,26] mixture atoms to their compounded molecules. To obtain a high atom-to-

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molecule transfer efficiency, it is essential to study the dynamical instability of atom–molecule conversion systems consisting of fermions.

In this paper, we investigate the dynamical instability of a dark state in the conversion of Bose–Fermi mixtures into stable molecules through stimulated Raman adiabatic passage aided by Feshbach resonance.[16] Based on our theoretical analysis, the region where the dynamical instability appears is obtained analytically. We show that the dynamical instability in this system is caused not only by bosonic interparticle interactions but also by Pauli blocking terms, which is different from the scenario of a pure bosonic system where instability is induced by nonlinear collisions.[13,15] Moreover, taking a ^{40}K–^{87}Rb mixture as an example, we give the unstable region numerically and analyse the effects of the Pauli blocking terms on the unstable regime.

2. Model: CPT state

Our model is schematically outlined in Fig. 1. A large number of two-species atoms, A and B, are initially prepared in a trapped state $|a,b\rangle$. Here we use $|a\rangle$ and $|b\rangle$ to denote the ground states of the fermionic and bosonic atoms in the open channel, and $|m\rangle$ and $|g\rangle$ to represent the quasibound and ground molecular states in the closed channel, respectively. The quasibound molecular state $|m\rangle$ is coupled with $|a\rangle$ and $|b\rangle$ through a magnetic field with coupling strength λ' and detuning δ, while the quasibound state $|m\rangle$ and the target state $|g\rangle$ are coupled by a laser field with Rabi frequency Ω' and detuning Δ. For this Bose–Fermi mixture system, as given in Ref. [16], the energy density corresponding to the Hamiltonian describing the above system in the interaction picture under the Hartree approximation is written as

$$\begin{aligned} E = \hbar\Bigg[&\frac{1}{2}\sum_{i\neq j}\chi'_{ij}\left|\psi_i\right|^2\left|\psi_j\right|^2 + \delta\psi_m^*\psi_m + \Delta\psi_g^*\psi_g \\ &+ \frac{\lambda'}{2}\left(\psi_m^*\psi_a\psi_b + \text{H.c.}\right) - \frac{\Omega'}{2}\left(\psi_g^*\psi_m + \text{H.c.}\right) \\ &+ \frac{1}{2}\chi'_{bb}\left|\psi_b\right|^4 + \frac{3}{5}\sum_{i=\{a,m,g\}}A'_i\left|\psi_i\right|^{10/3}\Bigg], \end{aligned} \tag{1}$$

where ψ_i is the complex probability amplitude of the i-th component, the coefficients $\chi'_{ii} = 4\pi\hbar a_i/m_i$ and $\chi'_{ij} = \chi'_{ji} = 2\pi\hbar a_{ij}/m_{ij}$ (a_i and a_{ij} are the s-wave scattering lengths, m_i is the mass of species i, and m_{ij} is the reduced mass between states i and j) characterize the bosonic intrastate and interstate interaction strengths, respectively, and the term proportional to A'_i represents the effective self-interaction related to fermions and is called the Pauli blocking term with $A'_i = \hbar^2(6\pi^2)^{2/3}/2m_i$.

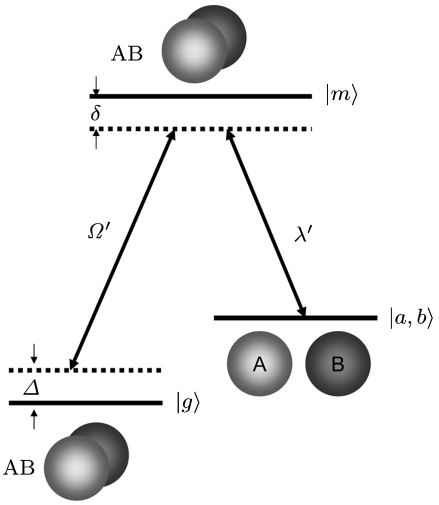

Fig. 1. The energy diagram of three-level atom–molecule system involving free-quasibound–bound transitions. Conversion of atoms in $|a,b\rangle$ is accomplished by Feshbach resonance, while the coupling between the quasibound molecular state $|m\rangle$ and the ground molecular state $|g\rangle$ is provided by laser light. δ and Δ are the one-photon and two-photon detunings, respectively.

As in Refs. [9], [16], and [17], in order to investigate the dynamics conveniently and to guarantee the conservation of the total particle numbers for different species, we introduce the mean-field Lagrange density with two Lagrange multipliers $\hbar\mu_a$ and $\hbar\mu_b$,

$$\begin{aligned} L = \hbar\sum_i\left[\frac{\mathrm{i}\hbar}{2}\left(\psi_i^*\frac{\partial\psi_i}{\partial t} - \psi_i\frac{\partial\psi_i^*}{\partial t}\right)\right] \\ - E - \hbar\mu_a N_a - \hbar\mu_b N_b, \end{aligned} \tag{2}$$

where $N_a = |\psi_a|^2 + |\psi_m|^2 + |\psi_g|^2$ and $N_b = |\psi_b|^2 + |\psi_m|^2 + |\psi_g|^2$ are the total particle numbers of the corresponding species, and $\hbar\mu_a$ and $\hbar\mu_b$ are identified as the chemical potentials of the corresponding atoms. Substituting the above mean-field Lagrangian density into the Euler–Lagrange equation

$$\frac{\partial L}{\partial\psi_i^*} - \partial_\nu\left(\frac{\partial L}{\partial\left(\partial_\nu\psi_i^*\right)}\right) = 0,$$

one can obtain a set of equations for the complex probability amplitudes (with $\hbar = 1$),

$$\begin{aligned} \mathrm{i}\dot{\phi_a} &= \left(\sum_{i\neq a}\chi_{ai}\left|\phi_i\right|^2 + A_a\left|\phi_a\right|^{4/3} - \mu_a\right)\phi_a \\ &\quad + \frac{\lambda}{2}\phi_b^*\phi_m, \\ \mathrm{i}\dot{\phi_b} &= \left(\sum_i\chi_{bi}\left|\phi_i\right|^2 - \mu_b\right)\phi_b + \frac{\lambda}{2}\phi_a^*\phi_m, \end{aligned}$$

$$
\begin{aligned}
\mathrm{i}\dot{\phi_m} &= \Big(\sum_{i\neq m}\chi_{mi}\left|\phi_i\right|^2 + A_m\left|\phi_m\right|^{4/3} \\
&\quad + \delta - \mu_a - \mu_b\Big)\phi_m + \frac{\lambda}{2}\phi_a\phi_b - \frac{\Omega}{2}\phi_g, \\
\mathrm{i}\dot{\phi_g} &= \Big(\sum_{i\neq g}\chi_{gi}\left|\phi_g\right|^2 + A_g\left|\phi_g\right|^{4/3} \\
&\quad + \Delta - \mu_a - \mu_b\Big)\phi_g - \frac{\Omega}{2}\phi_m, \qquad (3)
\end{aligned}
$$

where $\phi_i = \psi_i/\sqrt{N}$, $\chi_{ij} = n\chi'_{ij}$, $\lambda_i = \lambda'_i\sqrt{n}$, $\Omega_i = \Omega'_i$, $A_i = A'_i n^{2/3}$ are the renormalized quantities with the density of the total particle number $N = N_a + N_b$.

We now consider the stationary states. We know that the existence of the stationary solutions of Eq. (3) requires that $\dot{x} = 0, x = \phi_a, \phi_b, \phi_m, \phi_g$. However, it is difficult to find the exact solutions of the above dynamical equations when $\dot{x} = 0$. By analogy to the closely related models,[16,17] we assume that the system supports a CPT state with $\phi_m = 0$. Then, one can easily obtain the following CPT solutions,

$$
\begin{aligned}
&|\phi_a^0|^2 = |{\phi_b}^0|^2 = -(\Omega^2 + \sqrt{\Omega^4 + 2\lambda^2\Omega^2})/(2\lambda^2), \\
&|\phi_g^0|^2 = 1/2 - |\phi_a^0|^2, \qquad (4)
\end{aligned}
$$

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

$$
\begin{aligned}
\mu_a &= \chi_{ab}|\phi_b^0|^2 + \chi_{ag}|\phi_g^0|^2 + A_a|\phi_a^0|^{4/3}, \\
\mu_b &= \chi_{ab}|\phi_a^0|^2 + \chi_{bb}|\phi_b^0|^2 + \chi_{bg}|\phi_g^0|^2), \\
\Delta &= (\chi_{ab} - \chi_{ag})\left|\phi_a^0\right|^2 + (\chi_{bb} + \chi_{ab} - \chi_{bg})\left|\phi_b^0\right|^2 \\
&\quad + (\chi_{ag} + \chi_{bg})\left|\phi_g^0\right|^2 + A_a|\phi_a^0|^{4/3} - A_g|\phi_g^0|^{4/3}. \qquad (5)
\end{aligned}
$$

In the above calculations, we take the total particle numbers of fermionic atoms A and bosonic atoms B to be equal. From Eq. (4), we can see that the population distribution of the CPT state allows all the atoms to be converted into ground molecules as λ/Ω changes from 0 to ∞ as long as the two-photon resonance condition can be maintained dynamically. Then when the Rabi laser is ramped up adiabatically, i.e., Ω slowly varies in time, the state that is initially prepared as the CPT state is expected to remain close to the instantaneous CPT state throughout the entire process.

3. Dynamical instability of the CPT state

The existence of the CPT state in the atom–molecule STIRAP cannot ensure that the system can always follow it adiabatically since bosonic interparticle mean-field interactions could cause the dynamical instability that makes the quantum evolution deviate from the dark state rapidly, even in the adiabatic evolution.[15] Therefore, it is important to avoid the occurrence of the dynamical instability of the CPT state for the success of the STIRAP. For this purpose, we now investigate the dynamical instability of the dark state by casting the model into an effective classical one and analysing the eigenvalues of the Hamiltonian–Jacobi matrix obtained by linearizing the equations of motion around the fixed point corresponding to the CPT state.[27–30]

Equations (3) are equivalent to the Hamiltonian equations of motion of the effective canonical classical Hamiltonian

$$
\begin{aligned}
K &= \chi_{ab}(x_a^2 + y_a^2)(x_b^2 + y_b^2) \\
&\quad + \chi_{am}(x_a^2 + y_a^2)(x_m^2 + y_m^2) \\
&\quad + \chi_{ag}(x_a^2 + y_a^2)(x_g^2 + y_g^2) \\
&\quad + \chi_{bm}(x_b^2 + y_b^2)(x_m^2 + y_m^2) \\
&\quad + \chi_{bg}(x_b^2 + y_b^2)(x_g^2 + y_g^2) \\
&\quad + \chi_{mg}(x_m^2 + y_m^2)(x_g^2 + y_g^2) \\
&\quad + \frac{1}{2}\chi_{bb}(x_b^4 + 2x_b^2y_b^2 + y_b^4) - \Omega\left(x_m x_g + y_m y_g\right) \\
&\quad + \lambda\left[x_m(x_a x_b - y_a y_b) + y_m(x_a y_b + x_b y_a)\right] \\
&\quad + \delta(x_m^2 + y_m^2) + \Delta(x_g^2 + y_g^2) + \frac{3}{5}A_a\left(x_a^2 + y_a^2\right)^{5/3} \\
&\quad + \frac{3}{5}\left[A_m\left(x_m^2 + y_m^2\right)^{5/3} + A_g\left(x_g^2 + y_g^2\right)^{5/3}\right] \\
&\quad - \mu_a\left(x_a^2 + y_a^2 + x_m^2 + y_m^2 + x_g^2 + y_g^2\right) \\
&\quad - \mu_b\left(x_b^2 + y_b^2 + x_m^2 + y_m^2 + x_g^2 + y_g^2\right). \qquad (6)
\end{aligned}
$$

Here, x_i are canonical momenta, while y_i are the coordinates, being related with the old 'variables'-ϕ_i (complex numbers) as $\phi_a = x_a + \mathrm{i}y_a$, $\phi_b = x_b + \mathrm{i}y_b$, $\phi_d = x_m + \mathrm{i}y_m$, $\phi_g = x_g + \mathrm{i}y_g$. They are governed by the differential equations: $\dot{x_i} = \partial K/\partial y_i$, $\dot{y_i} = -\partial K/\partial x_i$. By setting $\dot{x_i} = \dot{y_i} = 0$, we can obtain the fixed point, which corresponds to the CPT state: $x_a = |\phi_a^0|$, $y_a = 0$, $x_b = |\phi_b^0|$, $y_b = 0$, $x_m = 0$, $y_m = 0$, $x_g = |\phi_g^0|$, $y_g = 0$ with the same chemical potentials and two-photon resonance condition in Eq. (5).

The instability of the fixed points relies on the eigenvalues of the Hamiltonian–Jacobi matrix, which can be real, complex or purely imaginary. However, only purely imaginary eigenvalues correspond to the stable fixed points; others indicate unstable ones. Let $x_a = z_1$, $y_a = z_2$, $x_b = z_3$, $y_b = z_4$, $x_m = z_5$, $y_m = z_6$, $x_g = z_7$, $y_g = z_8$, then the elements of the

Hamiltonian–Jacobi matrix can be written elegantly as

$$J_{ij} = \frac{(-1)^i}{2} \frac{\partial^2 K}{\partial z_i \partial z_{j\pm1}}\bigg|_{\rm CPT},$$

where i, j are, respectively, indexes of rows and columns, and the plus (subtraction) sign is for odd (even) j. Substituting the CPT state into the matrix elements, we find the Hamiltonian–Jacobi matrix around this fixed point (CPT state) for the atom–molecule conversion system,

$$J = \begin{pmatrix} 0 & \rho & 0 & \alpha & 0 & -\mu & 0 & \beta \\ 0 & 0 & 0 & 0 & \mu & 0 & 0 & 0 \\ 0 & \alpha & 0 & \varsigma & 0 & -\nu & 0 & \gamma \\ 0 & 0 & 0 & 0 & \nu & 0 & 0 & 0 \\ 0 & -\mu & 0 & -\nu & 0 & -\xi & 0 & \zeta \\ \mu & 0 & \nu & 0 & \xi & 0 & -\zeta & 0 \\ 0 & \beta & 0 & \gamma & 0 & \zeta & 0 & \varrho \\ 0 & 0 & 0 & 0 & -\zeta & 0 & 0 & 0 \end{pmatrix}, \quad (7)$$

where

$$\begin{aligned} &\alpha = -2\chi_{ab}|\phi_a^0||\phi_b^0|, && \beta = -2\chi_{ag}|\phi_a^0||\phi_g^0|, \\ &\gamma = -2\chi_{bg}|\phi_b^0||\phi_g^0|, && \mu = (\lambda/2)|\phi_b^0|, \\ &\nu = (\lambda/2)|\phi_a^0|, && \zeta = \Omega/2, \\ &\rho = -\frac{4}{3}A_a|\phi_a^0|^{4/3}, && \varrho = -\frac{4}{3}A_g|\phi_g^0|^{4/3}, \\ &\varsigma = -2\chi_{bb}|\phi_b^0|^2, \end{aligned}$$

and

$$\begin{aligned} \xi = {} & (\chi_{am} - \chi_{ab})\,|\phi_a^0|^2 + (\chi_{bm} - \chi_{ab} - \chi_{bb})\,|\phi_b^0|^2 \\ & + (\chi_{mg} - \chi_{ag} - \chi_{bg})\,|\phi_g^0|^2 + \delta - A_a|\phi_g^0|^{4/3}. \end{aligned}$$

We see that the Hamiltonian–Jacobi matrix J of Eq. (7) is in a simple form with many zero matrix elements. After calculation, we can analytically obtain the eigenvalues other than the zero-mode frequency of the above matrix J,

$$\begin{aligned} \omega_{1,2\pm} &= \pm\frac{\mathrm{i}}{\sqrt{2}}\sqrt{b \pm \sqrt{b^2 - c}}, \\ b &= 2\xi^2 + \Omega^2 + 2\lambda^2|\phi_a^0|^2, \\ c &= \Omega^4 + 4\left(\lambda^4 - 12\chi_{ab}\xi\lambda^2\right)|\phi_a^0|^4 \\ &\quad + 4\lambda|\phi_a^0|^2\left[\lambda\left(\Omega^2 - 4\xi^2\right) - 6\chi_{bb}\lambda\xi|\phi_b^0|^2\right. \\ &\quad \left. + 12(\chi_{ag} + \chi_{bg})\xi\Omega|\phi_g^0|\right] - 8\xi^4 - 8\Omega^2\xi^2 \\ &\quad - 16A_a\xi\lambda^2|\phi_a^0|^{10/3} - 16A_g\xi\Omega^2|\phi_g^0|^{4/3}. \end{aligned} \quad (8)$$

Once the nonzero-frequency eigenvalues ω_i become real or complex, the fixed point corresponding to the CPT state is dynamically unstable. From Eq. (8), we can see that $b > 0$. Hence the unstable regime is given by either $c < 0$ or $c > b^2$. In the absence of both the nonlinear collisions and the Pauli blocking terms, i.e., $\chi_{ij} = 0$ and $A_i = 0$, we find $b = 2\delta^2 + \Omega^2 + 2\lambda^2|\phi_a^0|^2$ and $c = \Omega^4 + 4\Omega^2\lambda^2|\phi_a^0|^2 + 4\lambda^4|\phi_a^0|^4$, which satisfy $c < b^2$. Therefore, the eigenvalue ω_i is purely imaginary and implies that the CPT state of the system is always stable. However, once either of them are included, i.e., $\chi_{ij} \neq 0$ or $A_i \neq 0$, ω_i may become real or complex, and hence brings instability to the population dynamics of the system. In the case $\chi_{ij} = 0, A_i \neq 0$, the dynamical instability is induced by the Pauli blocking terms. When $A_i = 0, \chi_{ij} \neq 0$, as in the pure bosonic system, this instability is caused by nonlinear collisions.[13] Therefore, we can conclude that unlike the pure bosonic system where the instability is only induced by interparticle interactions,[13,15] the instability here is not only caused by bosonic nonlinear collisions but also by the fermionic Pauli blocking terms.

4. Numerical results

Now we consider the concrete Bose–Fermi mixture system and concentrate on the dynamically unstable regions and the effects of the Pauli blocking terms on the instable regimes. The fermionic atoms A and the bosonic atoms B are taken as ^{40}K and ^{87}Rb, respectively. For this Bose–Fermi mixture system, we adopt the time-dependent Rabi frequency

$$\Omega(t) = \Omega_0\left[1 - \tanh\left(\frac{t - t_0}{\tau}\right)\right]. \quad (9)$$

The s-wave scattering length for a ^{40}K–^{87}Rb mixture determined through Feshbach spectroscopy is about $-185a_0$ with a_0 being the Bohr radius.[32] The resonance width is about $-3G$.[32] Then we can obtain the atom-to-molecule coupling strength $\lambda' = 9.07 \times 10^{-39}$ J. The s-wave scattering for ^{87}Rb is about $100a_0$ and the condensate density n is taken as 10^{20} m^{-3}. It is then easy to obtain $\chi_{ab} = 0.23\lambda$, $\chi_{bb} = 0.0056\lambda$, $A_a = 0.3\lambda$, $A_m = A_g = 0.09\lambda$.

Figures 2(a) and 2(b) respectively show the instability diagrams and a sample of the occurrence of instability in the population dynamics. In Fig. 2(a), in order to see the effects of Pauli blocking terms on the unstable regime, we have considered two different situations with and without the Pauli blocking terms, i.e., $A_i \neq 0$ and $A_i = 0$, where the dark gray and light

gray areas respectively correspond to unstable regions for the two cases $A_i \neq 0$ and $A_i = 0$. From this figure, we see that regardless of whether the Pauli blocking terms are considered or not, there are two unstable regions I (I′) and II (II′) corresponding to the two cases $c < 0$ or $c > b^2$, respectively. Here I and II are the unstable regions when $A_i \neq 0$, while I′ and II′ are the unstable regions when $A_i = 0$. Moreover, we find that region I (I′) corresponds to the unstable region with $c > b^2$, whose width shrinks as Ω increases, and region II (II′) is the unstable region with $c < 0$, whose width becomes fat with increasing δ. In the absence of Pauli blocking terms, i.e., $A_i = 0$, the unstable regions (I′ and II′) are only caused by bosonic nonlinear collisions, as in the pure bosonic system.[13] In the presence of Pauli blocking terms, i.e., $A_i \neq 0$, we find that the scopes of the two unstable areas (I and II) are larger than that of the case $A_i = 0$. For example, region II, being composed of II′ and H, is much larger than region II′. Hence, region H is the unstable space induced by the Pauli blocking terms. Therefore, we can conclude that the Pauli blocking terms enlarge the unstable areas. For the ^{40}K–^{87}Rb mixture condensate system, once the parameters enter into the unstable regions I and II, the instability could occur and make the quantum evolution deviate from the dark state rapidly even in the adiabatic evolution, as is shown in Fig. 2(b), where initially the population of the ground molecules follows the CPT solution, but significant deviation starts to occur at about $t = 1193$ (with $\Omega = 1.05$) when the system goes into the unstable regime. Therefore, for the success of STIRAP, it is crucial to avoid these unstable regimes when designing the route of adiabatic passage.

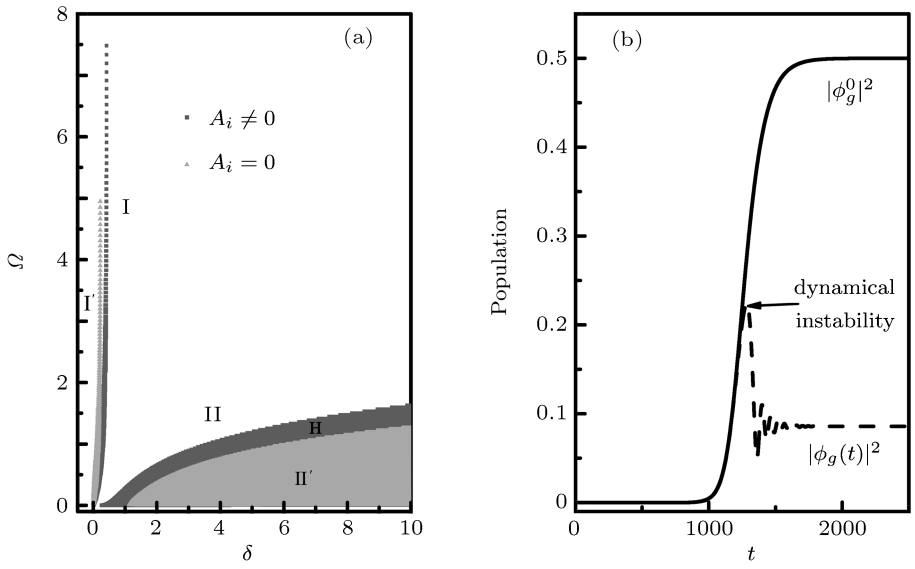

Fig. 2. (a) Unstable regions for ^{40}K–^{87}Rb mixture system for two cases ($A_i = 0$ and $A_i \neq 0$), where the dark gray areas I and II (light gray areas I′ and II′) correspond to the unstable regions of the case $A_i \neq 0$ ($A_i = 0$), and region II is made of II′ and H; (b) A sample of instability in the population dynamics with $\delta = 4$, $\Omega_0 = 200$, $\tau = 200$, $t_0 = 600$, here the solid line shows the CPT value of populations of the ground molecules $|\phi_g^0|^2$, and the dashed line shows the population evolution of ground-state molecules $|\phi_g(t)|^2$. The evolution of atoms A, atoms B, and quasibound molecules AB are not shown. The parameters are $A_a = 0.3$, $A_m = A_g = 0.09$, $\chi_{ab} = 0.23$, $\chi_{bb} = 0.0056$, $\chi_{am} = \chi_{ag} = \chi_{bm} = \chi_{bg} = \chi_{mg} = 0$. Time is in units of λ^{-1} and all other parameters are in units of λ.

5. Conclusion

In conclusion, we investigated the dynamical instability of a dark state in the conversion of Bose–Fermi mixtures into stable molecules in STIRAP aided by Feshbach resonance. We analytically obtain the regions for the occurrence of dynamical instability. We find that both bosonic interparticle interactions and fermionic Pauli blocking terms could induce dynamical instability in some parameter regions. This is different from pure bosonic atom–molecule conversion systems in which the dynamical instability is only caused by nonlinear interparticle interactions. Taking a ^{40}K–^{87}Rb mixture condensate system as an example, we map out the unstable regions. Our discussions are helpful to avoid the unstable regions and hence

successfully implement the STIRAP process and obtain high atom–molecule conversion efficiency in experiment.

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