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ADIABATICITY OF THE DARK STATE IN A NONLINEAR ATOM-TRIMER CONVERSION SYSTEM

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We investigate the adiabaticity of the dark state in a nonlinear atom-trimer conversion system in a stimulated Raman adiabatic passage (STIRAP). We find that, in the absence of the nonlinear collisions, the adiabatic condition for this nonlinear system only depends on the Rabi-frequency of the dimer-trimer coupling optical field, which is different from traditional STIRAP processes. In the presence of the nonlinear collisions, the adiabatic condition also relies on the atom-dimer coupling Rabi frequency. However, its influence is really small. Moreover, we propose a more feasible two-photon STIRAP scheme that has better adiabaticity and hence could yield higher atom-trimer conversion efficiency.

Keywords: Atom-trimer conversion system; stimulated Raman adiabatic passage; dark state; adiabaticity.

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1. Introduction

Adiabatic theorem[1–4] plays a major role in various areas of physics. According to the adiabatic theorem, if the parameters of the system vary with time much more slowly than the intrinsic motion of the system, the system will undergo adiabatic evolution. For a quantum system, adiabatic evolution means that an initial nondegenerate eigenstate remains to be an instantaneous eigenstate when the Hamiltonian changes slowly compared to the level spacings. Due to the fundamental importance of adiabatic evolution in quantum theory and applications of quantum state control, the adiabatic theory has been very actively studied under various practical conditions in the past years.[5,6]

Recently, the adiabatic evolution has been extensively used to create an ultracold molecular gas or even a molecular Bose–Einstein condensate (BEC) from its atomic counterpart in the stimulated Raman adiabatic passage (STIRAP),[7–13] where the existence of the coherent population trapping (CPT) state or dark state facilitates the adiabatic coherent population transfer between atoms and molecules

with high conversion efficiency. However, different from the traditional STIRAP in an Λ-atomic system, the atom-molecule STIRAP contains nonlinearities that stem from the mean-field treatment of the interparticle interactions and the conversion process of atoms to molecules. The existence of these nonlinearities make it not justified to apply the adiabatic condition of quantum mechanics to nonlinear BEC system because of the absence of the superposition principle. Therefore, it is very difficult to analyze the adiabaticity of the atom-molecule conversion systems.

The adiabatic theory for nonlinear quantum systems, i.e. the systems governed by the nonlinear Schrödinger equation, was first discussed in Ref. 5, where adiabatic conditions and adiabatic invariants were obtained through casting the nonlinear Schrödinger equation into an effective classical Hamiltonian. Recently, Ling *et al.*[10,11] extended the above adiabatic theory to the atom-dimer conversion system by linking the nonadiabaticity with the population growth in the collective excitations of the dark state. Following the above work, an improved adiabatic condition was put forward by Itin *et al.*[12] via applying methods of classical Hamiltonian dynamics under the collisionless limit. Very recently, the atom-molecule dark state technique in STIRAP process is theoretically generalized to create more complex homonuclear or heteronuclear molecules-trimer or tetramer.[14-16] Moreover, following the approach of Ref. 10, the adiabatic condition for the atom-heteronuclear-trimer conversion process[17] is developed in the collisionless limit.

In this paper, we include the collisional mean-field interactions and use methods of classical Hamiltonian dynamics to study the adiabaticity of the coherent atom-heteronuclear-trimer conversion system,[14] where the heteronuclear trimers A_2B are formed via two different paths with an intermediate homonuclear or heteronuclear dimer state A_2 or AB. In Sec. 2, we model the system, focus on the nonlinear atom-trimer dark state solution and propose two schemes to implement the two-photon STIRAP. In Sec. 3, we cast the quantum Hamiltonian under the mean-field approximation to an effective classical Hamiltonian and take two mathematical methods to obtain the frequencies of the fixed point which corresponds to the CPT state, and further obtain the adiabatic condition from classical adiabatic dynamics. Moreover, we also discuss and compare the adiabaticity of the CPT state under two kinds of two-photon STIRAP processes. In Sec. 4, we give our conclusions.

2. Models, Equations and CPT State

Following,[14,17] we consider that two species of Bosonic atoms are converted into stable heteronuclear trimers A_2B via the STIRAP, where the intermediate dimers (A_2 or AB) are formed by two different reaction paths, i.e. AA-path and AB-path, as is shown in Fig. 1. Including s-wave scattering processes, the second quantized Hamiltonian under the rotating frame reads,
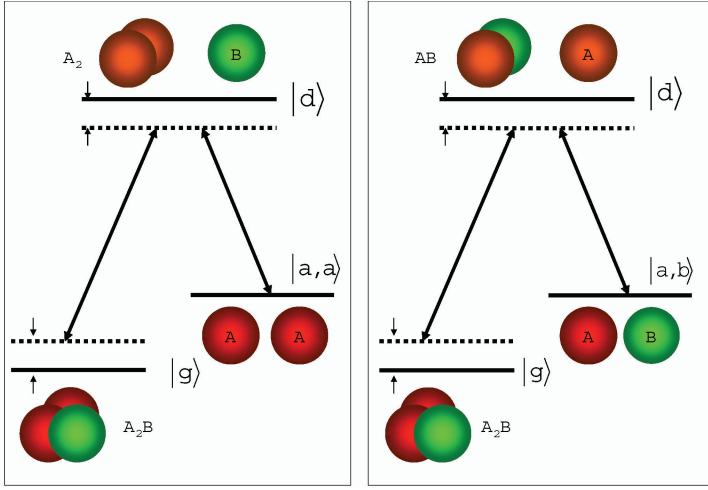
Fig. 1.   Atom-trimer conversion system coupled by two laser fields for single path AA and AB.
Ω, λ are the Rabi frequencies, δ and Δ are the one- and two-photon detunings, respectively.

$$\hat{H} = -\hbar \sum_{i,j} \chi'_{ij} \hat{\psi}_i^\dagger \hat{\psi}_j^\dagger \hat{\psi}_j \hat{\psi}_i - \hbar[\delta \hat{\psi}_d^\dagger \hat{\psi}_d + (\Delta + \delta)\hat{\psi}_g^\dagger \hat{\psi}_g$$

$$+ \lambda'(\hat{\psi}_d^\dagger \hat{\psi}_a \hat{\psi}_m + H.C.) - \Omega'(\hat{\psi}_g^\dagger \hat{\psi}_d \hat{\psi}_n + H.C.)] \,, \tag{1}$$

Here $m = a$, $n = b$ ($m = b, n = a$) for the AA-path (AB-path), $\hat{\psi}_i(\hat{\psi}_i^\dagger)$ are the annihilation (creation) operator, where the indices i, $j = a, b, d, g$ stand for atoms, dimers and trimers, and the terms proportional to χ_{ij} represent two-body collisions.

As in papers,[8,18] to guarantee the conservation of the total particle numbers, we introduce two multipliers $\hbar\mu_a$ and $\hbar\mu_b$ into the Hamiltonian in Eq. (1) and obtain the grand canonical Hamiltonian "Kamiltonian",

$$\hat{K} = \hat{H} - \hbar\mu_a \hat{N}_A - \hbar\mu_b \hat{N}_B \,, \tag{2}$$

where $\hbar\mu_A$, $\hbar\mu_B$ are identified as the chemical potentials of the corresponding atoms, and $\hat{N}_A$, $\hat{N}_B$ are the operators for the total particle number of the corresponding species.

From the Kamiltonian we can easily derive the equations of motion of the unit-scaled operators. As usual, in the mean-field treatment, $\hat{\psi}_i$ and $\hat{\psi}_i^\dagger$ are replaced by c-number $\sqrt{n}\psi_i$ and $\sqrt{n}\psi_i^*$, where n is the density of the total particle number. For the AA-path, the set of the mean-field Gross-Pitaevskii (G-P) equations (with $\hbar = 1$) is,

$$\begin{aligned}
i\dot{\psi}_a &= (\omega_a - \mu_a)\psi_a - 2\lambda\psi_a^*\psi_d \,, \\
i\dot{\psi}_b &= (\omega_b - \mu_b)\psi_b + \Omega\psi_d^*\psi_g \,, \\
i\dot{\psi}_d &= (\omega_d - 2\mu_a - i\gamma - \delta)\psi_d - \lambda\psi_a^2 + \Omega\psi_b^*\psi_g \,, \\
i\dot{\psi}_g &= (\omega_g - 2\mu_a - \mu_b)\psi_g - (\Delta + \delta)\psi_g + \Omega\psi_d\psi_b \,.
\end{aligned} \tag{3}$$

For the AB-path, it becomes,

$$
\begin{aligned}
i\dot{\psi}_a &= (\omega_a - \mu_a)\psi_a - \lambda\psi_b^*\psi_d + \Omega\psi_d^*\psi_g\,,\\
i\dot{\psi}_b &= (\omega_b - \mu_b)\psi_b - \lambda\psi_a^*\psi_d\,,\\
i\dot{\psi}_d &= (\omega_d - \mu_a - \mu_b - i\gamma - \delta)\psi_d - \lambda\psi_a\psi_b + \Omega\psi_a^*\psi_g\,,\\
i\dot{\psi}_g &= (\omega_g - 2\mu_a - \mu_b)\psi_g - (\Delta + \delta)\psi_g + \Omega\psi_d\psi_a\,,
\end{aligned}
\tag{4}
$$

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

Taking the time derivative in Eqs. (3) and (4) to be zero and making use of the two population conversion conditions of species a and b for different paths, we can show that, Eqs. (3) and (4) admit the following stationary-state solutions with no dimer population, i.e. the generalized CPT state,

$$
\begin{aligned}
|\psi_g|^2 &= \frac{k(\lambda/\Omega)^2}{3(1 + k(\lambda/\Omega)^2)}\,,\\
|\psi_a|^2 &= 2|\psi_b|^2 = \frac{2}{3}(1 - 3|\psi_g|^2)\,,
\end{aligned}
\tag{5}
$$

where $k = 4$ ($k = 1$) for the AA-path (AB-path). In the above equations, $N_A = 2N_B = 2/3$. The two-photon resonance conditions and the chemical potentials corresponding to the CPT solution for the two paths are the same, i.e.

$$
\begin{aligned}
\mu_a &= -2(\chi_{aa}|\psi_a|^2 + \chi_{ab}|\psi_b|^2 + \chi_{ag}|\psi_g|^2)\,,\\
\mu_b &= -2(\chi_{ab}|\psi_a|^2 + \chi_{bb}|\psi_b|^2 + \chi_{bg}|\psi_g|^2)\,,\\
\Delta_{AA} = \Delta_{AB} &= -\delta + 2(2\chi_{ag} + \chi_{bg} - \chi_{gg})|\psi_g|^2\\
&\quad + (4\chi_{aa} - 2\chi_{ag} + 4\chi_{ab} - \chi_{bg})|\psi_a|^2\,.
\end{aligned}
\tag{6}
$$

This suggests that, by dynamically maintaining the two-photon resonance condition, population can be concentrated in atomic states and trimer bound states under the respective limit $\lambda/\Omega \to 0$ and $\lambda/\Omega \to \infty$.

In the following discussions, we will consider two schemes to implement the two-photon STIRAP, where a pair of atoms is first associated to the molecular dimer via PA, the dimer molecule is then photoassociated with another atom to the bound trimer molecule. In scheme (i), the atom-dimer coupling Rabi frequency λ is constant, and the dimer-trimer coupling Rabi frequency is modulated as $\Omega(t) = \Omega_0 \,\mathrm{sech}\, t/\tau$; In scheme (ii), the atom-dimer coupling Rabi frequency is controlled as $\lambda = \lambda_0 \cosh t/\tau$, and the dimer-trimer coupling Rabi frequency Ω is fixed. In both cases, $\lambda/\Omega \sim \cosh t/\tau$ satisfies $\lambda/\Omega \to 0$ as $t \to 0$ and $\lambda/\Omega \to \infty$ as $t \to \infty$, which facilitates adiabatic coherent population transfer between atoms and trimers.

3. Classical Hamiltonian Dynamics and Adiabatic Condition

The existence of the CPT state facilitates adiabatic coherent population transfer between atoms and trimers. However, owing to the invalidation of the superposition principle in the nonlinear atom-trimer conversion system, it is not justified to apply the adiabatic condition of quantum mechanics to study the adiabatic evolution of the CPT state. To overcome this difficulty, in this section, we attempt to obtain the adiabatic criterion for this nonlinear system with classical adiabatic dynamics[5,12] through casting the mean-field Hamiltonian to an effective classical one. Therefore, the adiabatic parameter of this nonlinear system can be defined as the ratio between the change rate of the external parameters and the fundamental frequencies of periodic orbits around the fixed point which corresponds to the CPT state.[5] These frequencies can be evaluated by linearizing equations of motion of the classical Hamiltonian around the fixed point. Here we take two mathematical methods to obtain these frequencies. On the one hand, in the collisionless limit, they are obtained by the linearization process with reducing the degrees of freedom of the classical system. On the other hand, in the collision case, they are obtained through solving the eigenvalues of the Hamiltonian–Jacobi matrix.

3.1. *Collisionless model*

3.1.1. *Intrinsic frequencies of the classical Hamiltonian system for the AA-path*

In the absence of nonlinear collisions, i.e. $\chi_{ij} = 0$, the two-photon resonance condition and the chemical potentials in Eq. (6) are simplified to $\Delta + \delta = 0$ and $\mu_a = \mu_b = 0$. Then, $K = H$. Taking use of the canonical transformation: $\psi_a = x_1 + iy_1$, $\psi_b = x_2 + iy_2$, $\psi_d = x_3 + iy_3$, $\psi_g = x_4 + iy_4$, the mean-field Hamiltonian for the AA-path can be cast into the following classical Hamiltonian,

$$H = 2\Omega[x_3(x_2x_4 + y_2y_4) - y_3(x_4y_2 - x_2y_4)]$$
$$- 2\lambda[x_3(x_1^2 - y_1^2) + 2x_1y_1y_3] + \delta(x_3^3 + y_3^3). \tag{7}$$

Here x_k are canonical momenta, while y_k are the coordinates. In fact, the system has only 2 degrees of freedom (d.o.f.), because there exists two integrals of motion, i.e. $x_1^2 + y_1^2 + 2(x_3^2 + y_3^2 + x_4^2 + y_4^2) = N_A$, $x_2^2 + y_2^2 + x_4^2 + y_4^2 = N_B$. For simplicity, we consider the case $\delta = 0$. Let $H = 0$. On this manifold, dynamics at constant parameters λ, Ω is completely integrable. Indeed, $I_3 \equiv x_3/y_3$ is the additional integral of motion. As a result, $I_1 \equiv I_3(y_1^2 - x_1^2) - 2x_1y_1$ and $I_2 \equiv I_3(x_2x_4 + y_2y_4) - (x_4y_2 - x_2y_4)$ are also integrals of motion. In case the external parameters λ, Ω changes with time, the system may leave the $H = 0$ manifold. However, if $I_{1,2}$ are initially equal to zero, then even with time-dependent parameters dynamics will be confined to the initial $H = 0$ manifold. The case where all population are initially atoms a and b is of this type, i.e. initially $x_{3,4} = y_{1,2,3,4} = 0$, hence $H = 0$.

With $H = 0$, we can reduce the system to 1 d.o.f. classical Hamiltonian system. Without loss of generality, let $I_3 = 0$, then we find $\dot{x}_3 = 0$, $\dot{y}_1 = 0$, and ψ_b, ψ_g have the same phase, i.e. $y_2/x_2 = y_4/x_4 = \tan\theta$. Therefore, we can make the following transformation: $x_2 = z_2\cos\theta$, $y_2 = z_2\sin\theta$, $x_4 = z_4\cos\theta$, $y_4 = z_4\sin\theta$, where $z_2 = |\psi_b|$ and $z_4 = |\psi_g|$. It is readily to demonstrate that $\dot{\theta} = 0$, $\dot{z}_2 = -\Omega y_3 z_4$, $\dot{z}_4 = \Omega y_3 z_2$. Additionally, owing to the dependence of z_2 and z_4 with the population conversation condition of atom b: $z_2^2 + z_4^2 = N_B$, we can further lessen $1/2$ d.o.f. of the system, then the equations of motion reduce to $\dot{x}_1 = -2\lambda x_1 y_3$, $\dot{y}_3 = \lambda x_1^2 - \Omega z_2\sqrt{N_B - z_2^2}$, $\dot{z}_2 = -\Omega y_3\sqrt{N_B - z_2^2}$. From the particle-conversation condition of atom a: $x_1^2 + 2y_3^2 = 2z_2^2$, we can make transformations: $x_1 = \sqrt{2}z_2\cos\phi$, $\sqrt{2}y_3 = \sqrt{2}z_2\sin\phi$. Then d.o.f. of the system can be lessen to 1 with the following equations of motion,

$$
\begin{aligned}
\dot{z}_2 &= -\Omega z_2\sqrt{\frac{1}{3} - z_2^2}\,\sin\phi\,, \\
\dot{\phi} &= \left(2\lambda z_2 - \Omega\sqrt{\frac{1}{3} - z_2^2}\right)\cos\phi\,.
\end{aligned}
\tag{8}
$$

Let the right-hand sides of the above equations be zero, then the fixed point is easily obtained,

$$
z_2 = \frac{\Omega}{\sqrt{12\lambda^2 + 3\Omega^2}}\,, \qquad \phi = 0\,, \qquad \pi\,.
\tag{9}
$$

This fixed point corresponds to the CPT state in Eq. (5). For small-amplitude oscillations, linearizing motions of equation in Eq. (8) around the fixed point in Eq. (9), the linearized matrix can be obtained,

$$
M = \begin{pmatrix}
\dfrac{(6z_2^2 - 1)\Omega\sin(\phi)}{\sqrt{3 - 9z_2^2}} & -z_2\sqrt{\dfrac{1}{3} - z_2^2}\,\Omega\cos(\phi) \\
2\lambda + \dfrac{z_2\Omega\cos(\phi)}{\sqrt{1/3 - z_2^2}} & \sqrt{\dfrac{1}{3} - z_2^2}\,\Omega\sin(\phi)
\end{pmatrix}\,.
\tag{10}
$$

Substituting Eq. (9) into the above matrix and solving the eigenvalues of M, we can obtain the intrinsic frequencies of the system,

$$
\omega_\pm^{AA} = \pm\frac{\Omega}{\sqrt{3}}\,.
\tag{11}
$$

As we know, these frequencies can be real, complex or pure imaginary. Only the real frequencies correspond to the stable fixed points. Others imply the unstable ones. Since $\omega_\pm^{AA}$ are all real, the fixed point in Eq. (9) is stable. For a classical system with one degree of freedom, the stable fixed points are the elliptic points. From Fig. 2 where the phase portrait of Eq. (8) is plotted, we see that, as the parameter λ/Ω changes from 0 to ∞, the stable fixed point moves from $z_2 = 1/\sqrt{3}$ to 0.
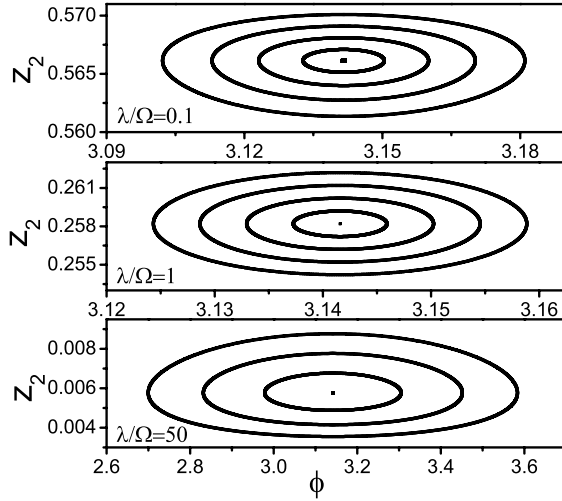
Fig. 2. Single path AA: Phase portrait of (8). As parameter λ/Ω increases, the stable fixed point (corresponding to the dark state) moves from $z_2 = 1/\sqrt{3}$ to 0.

3.1.2. *Intrinsic frequencies of the classical Hamiltonian system for the AB-path*

In the absence of interparticle interactions, we can similarly cast the mean-field Hamiltonian for the AB-path into the form of a classical system under the two-photon resonance condition (i.e. $\Delta + \delta = 0$),

$$H = 2\Omega[x_3(x_2x_4 + y_2y_4) - y_3(x_4y_2 - x_2y_4)]$$
$$- 2\lambda[x_3(x_1^2 - y_1^2) + 2x_1y_1y_3] + \delta(x_3^3 + y_3^3). \tag{12}$$

The conditions of conserved particle number are $x_1^2 + y_1^2 + x_3^2 + y_3^2 + 2(x_4^2 + y_4^2) = N_A$, $x_2^2 + y_2^2 + x_3^2 + y_3^2 + x_4^2 + y_4^2 = N_B$. Let $H = 0$. On the manifold of $H = 0$, dynamics at constant parameters λ, Ω is completely integrable. Here $I_3 \equiv x_3/y_3$, $I_1 \equiv I_3(x_1x_2 - y_1y_2) + (x_1y_2 + x_2y_1)$ and $I_2 \equiv I_3(x_1x_4 + y_1y_4) + (x_1y_4 - y_1x_4)$ are the three integrals of motion. In case initial particles are all atoms a and b, i.e. initially $x_{3,4} = y_{1,2,3,4} = 0$, $H = 0$.

With $H = 0$, we further reduce the system to 1 d.o.f. classical Hamiltonian. Let $I_3 = 0$, we find that $\dot{x}_1 = 0$, ψ_a, ψ_g have the same phase θ, and ψ_b has the inverse phase $-\theta$ (i.e. $y_1/x_1 = -y_2/x_2 = y_4/x_4 = \tan\theta$). Making the transformation: $x_1 = z_1\cos\theta$, $y_1 = z_1\sin\theta$, $x_2 = z_2\cos(-\theta)$, $y_2 = z_2\sin(-\theta)$, $x_4 = z_4\cos\theta$, $y_4 = z_4\sin\theta$, where $z_1 = |\psi_a|$, $z_2 = |\psi_b|$, $z_4 = |\psi_g|$, we can prove that $\dot{\theta} = 0$ and equations of motion reduce to $\dot{z}_1 = -y_3(\Omega z_4 + \lambda z_2)$, $\dot{z}_2 = -\lambda z_1 y_3$, $\dot{z}_4 = \Omega z_1 y_3$, $\dot{y}_3 = z_1(\lambda z_2 - \Omega z_4)$. From the two population conversation conditions, we find $y_3^2 + 2z_2^2 = z_1^2$ and $z_4 = \sqrt{1/3 - (z_1^2 - z_2^2)}$. Therefore, we can make the transformations: $y_3 = z_1\cos\phi$, $\sqrt{2}z_2 = z_1\sin\phi$. Then we reduce the system to 1 d.o.f. with equations of motion,

$$\dot{z}_1 = \left(-\frac{z_1^2}{\sqrt{2}} \lambda \sin\phi - \Omega z_1 \sqrt{\frac{1}{3} + \frac{z_1^2}{2} \sin^2\phi - z_1^2} \right) \cos\phi\,,$$

$$\dot{\phi} = -\frac{z_1\lambda}{\sqrt{2}} - \frac{z_1\lambda}{\sqrt{2}} \cos^2\phi + \Omega \sqrt{\frac{1}{3} + \frac{z_1^2}{2} \sin^2\phi - z_1^2} \sin\phi\,. \tag{13}$$

It is easy to obtain the fixed point of the above equations which corresponds to the CPT state, i.e.

$$z_1 = \frac{\sqrt{2}\Omega}{\sqrt{3(\lambda^2 + \Omega^2)}}\,, \qquad \phi = \frac{\pi}{2}\,, \tag{14}$$

and the intrinsic frequencies of the system by linearizing Eq. (13) around this fixed point,

$$\omega_\pm^{AB} = \pm\sqrt{\frac{2}{3}}\Omega\,. \tag{15}$$

Since $\omega_\pm^{AB}$ are all real, we can conclude that the fixed point is stable. As times increase, λ/Ω grows, and the fixed point goes from $|z_1| = \sqrt{2/3}$ to 0, which is not shown.

3.2. *Collisional model*

In the system with interparticle interactions, the situation is more complicated and we cannot utilize the trick with the energy manifold because $H \neq 0$. For this case, we can obtain the frequencies of the atom-trimer conversion system with solving the eigenvalues of the Hamiltonian–Jacobi matrix[19] obtained by linearizing the equations of motion around the fixed point corresponding to the CPT state.

By making use of the same canonical transformation in the above discussions, i.e. $\psi_a = x_1 + iy_1$, $\psi_b = x_2 + iy_2$, $\psi_d = x_3 + iy_3$, $\psi_g = x_4 + iy_4$, we cast the mean-field grand canonical Hamiltonian $K = H - \hbar\mu_a N_A - \hbar\mu_b N_B$ into a classical grand Hamiltonian. Here x_i and y_i 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: $x_a = |\psi_a|$, $y_a = 0$, $x_b = |\psi_b|$, $y_b = 0$, $x_d = 0$, $y_d = 0$, $x_g = |\psi_g|$, $y_g = 0$ with the same chemical potentials and two-photon resonance conditions in Eq. (6).

The frequencies of the fixed points can be obtained by solving the eigenvalues of the Hamiltonian–Jacobi matrix. Let $x_1 = z_1$, $y_1 = z_2$, $x_2 = z_3$, $y_2 = z_4$, $x_3 = z_5$, $y_3 = z_6$, $x_4 = z_7$, $y_4 = z_8$, then elements of the Hamiltonian–Jacobi matrix can be written elegantly as $J_{ij} = [(-1)^i/2]\partial^2 K/\partial z_i \partial z_{j\pm1}|_{\text{CPT}}$, 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-trimer conversion system J is

$$
\begin{pmatrix}
0 & 4\chi_{aa}|\psi_a|^2 & 0 & 4\chi_{ab}|\psi_a||\psi_b| & 0 & \alpha & 0 & 4\chi_{ag}|\psi_a||\psi_g| \\
0 & 0 & 0 & 0 & \beta & 0 & 0 & 0 \\
0 & 4\chi_{ab}|\psi_a||\psi_b| & 0 & 4\chi_{bb}|\psi_b|^2 & 0 & \gamma & 0 & 4\chi_{bg}|\psi_b||\psi_g| \\
0 & 0 & 0 & 0 & \eta & 0 & 0 & 0 \\
0 & \alpha & 0 & \gamma & 0 & -\zeta & 0 & -\nu \\
\beta & 0 & \eta & 0 & \zeta & 0 & \nu & 0 \\
0 & 4\chi_{ag}|\psi_a||\psi_g| & 0 & 4\chi_{bg}|\psi_b||\psi_g| & 0 & -\nu & 0 & 4\chi_{gg}|\psi_g|^2 \\
0 & 0 & 0 & 0 & \nu & 0 & 0 & 0
\end{pmatrix}
\tag{16}
$$

where $\alpha = -\beta = 2\lambda|\psi_a|$, $\gamma = \eta = -\Omega|\psi_g|$, $\nu = \Omega|\psi_b|$, $\zeta = (4\chi_{aa} - 2\chi_{ad})|\psi_a|^2 + (4\chi_{ab} - 2\chi_{bd})|\psi_b|^2 + (4\chi_{ag} - 4\chi_{dg})|\psi_g|^2 - \delta$ for the AA-path, while $\alpha = \lambda|\psi_b| - \Omega|\psi_g|$, $\beta = -\lambda|\psi_b| - \Omega|\psi_g|$, $\gamma = -\eta = -\lambda|\psi_a|$, $\nu = \lambda|\psi_a|$, $\zeta = (\chi_{aa} + \chi_{ab} - 2\chi_{ad})|\psi_a|^2 + (\chi_{ab} + \chi_{bb} - 2\chi_{bd})|\psi_b|^2 + (\chi_{ag} + \chi_{bg} - 2\chi_{dg})|\psi_g|^2 - \delta$ for the AB-path.

Now we solve the eigenvalues η_i of J. We can obtain analytically the eigenvalues (η_i) of Eq. (16) other than the zero-mode frequency of the matrix J.

$$
\begin{aligned}
\eta_{1,2\pm} &= i\omega_{1,2\pm} = \pm\frac{i}{\sqrt{2}}\sqrt{b \pm \sqrt{b^2 - c}}\,, \\
b &= \zeta^2 + 2\nu^2 - 2\alpha\beta - 2\gamma\eta\,, \\
c &= 4(\nu^4 - 2\alpha\beta\nu^2 - 2\gamma\eta\nu^2 + \alpha^2\beta^2 + \gamma^2\eta^2 + 2\alpha\beta\gamma\eta \\
&\quad + 4\chi_{aa}\beta^2\zeta|\psi_a|^2 + 4\chi_{bb}\zeta\eta^2|\psi_b|^2 + 4\chi_{gg}\zeta\nu^2|\psi_g|^2 \\
&\quad + 8\chi_{ab}\beta\zeta\eta|\psi_a||\psi_b| + 8\chi_{ag}\beta\zeta\nu|\psi_a||\psi_g| \\
&\quad + 8\chi_{bg}\nu\zeta\eta|\psi_b||\psi_g|)\,.
\end{aligned}
\tag{17}
$$

Here $\omega_{1,2\pm}$ are the intrinsic frequencies of the system and are identical to the Bogoliubov excitation frequencies of the system.[5] When $\omega_{1,2\pm}$ become pure imaginary or complex, the corresponding CPT state is dynamically unstable. For the two paths, we find $b = \delta^2 + 2l\Omega^2/3 > 0$. Here $l = 1$ ($l = 2$) for the AA- (AB-)path. Hence the unstable regime is given by either $c < 0$ or $c > b^2$. When $\chi_{ij} = 0$ and $\delta = 0$, eigenvalues η_i reduce to $\omega_{1,2\pm} = \pm\sqrt{l/3}\Omega$. This is consistent with the results in Eqs. (11) and (15).

3.3. *Adiabatic condition*

For a classical system, the adiabatic evolution[3] requires that the external parameter R changes slowly compared to the intrinsic frequency of the system, i.e. the adiabatic parameter satisfy:

$$
\varepsilon = \frac{2\pi}{\omega} \cdot \left|\frac{\dot{R}}{R}\right| \ll 1\,,
\tag{18}
$$

where ω is the frequency of the system, and $\varepsilon \to 0$ corresponds to the adiabatic limit.

For the atom-trimer conversion system, if the STIRAP process is implemented by scheme (i), then $R = \Omega$; if the STIRAP process is carried out by scheme (ii), then $R = \lambda$. In both schemes, $|\dot{R}/R| = |\tanh(t/\tau)| \in (0, 1)$. Hence $2\pi/\omega$ can be used to scale the adiabatic parameter completely, i.e. $2\pi/\omega \to 0$ corresponds to the adiabatic limit. For this atom-trimer conversion system which has multiple freedom degrees, ω is the smallest value of the magnitude of the frequencies of the system in Eq. (17), i.e. $\omega = \text{Min}\{|\omega_{1+}|, |\omega_{1-}|, |\omega_{2+}|, |\omega_{2-}|\}$. When $\chi_{ij} = 0$, $\omega = (1/\sqrt{6})\sqrt{3\delta^2 + 2l\Omega^2 - \sqrt{9\delta^4 + 12l\Omega^2\delta^2}}$. Here we can see that ω only relies on the dimer-trimer coupling Rabi frequency Ω and has no relation with the atom-dimer coupling Rabi frequency λ. Owing to the relation: $\varepsilon \sim 2\pi/\omega$, this frequency makes the adiabatic condition of the atom-trimer dark state only depend upon Ω. This adiabatic condition is different from other STIRAP processes,[10,11] where the adiabatic condition of dark state is not only related with the Rabi frequency of the Stokes laser but also the Rabi frequency of the pump laser. In the presence of the nonlinear collisions, i.e. $\chi_{ij} \neq 0$, the adiabatic parameter also relies on the atom-dimer coupling Rabi frequency λ. However, its influence is really small, as demonstrated below.

Figure 3 shows the Rabi frequencies, the adiabatic parameters and the population dynamics as a function of time for the AA-path. In this example the
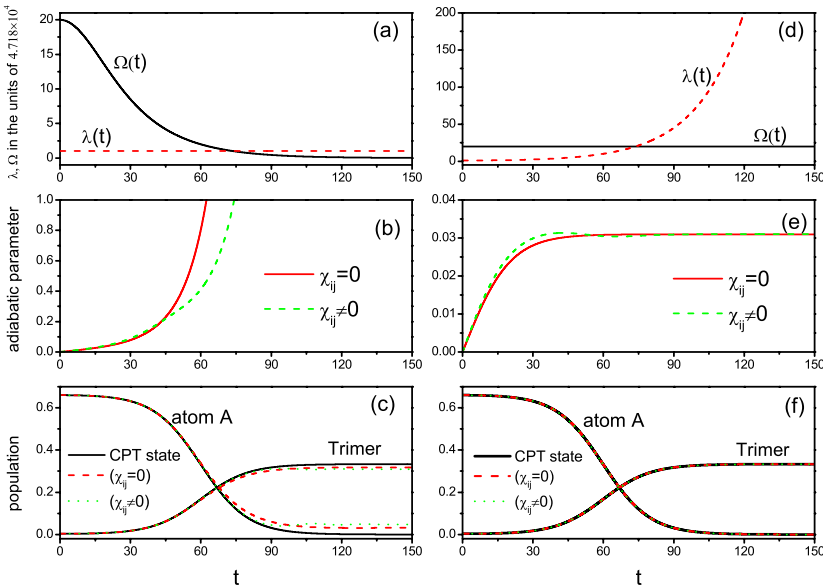

Fig. 3. Single path AA: (a) and (d) Rabi frequencies, (b) and (e) adiabatic parameters and (c) and (f) population dynamics as functions of time with and without nonlinear collisions for $\gamma = 0$, $\delta = -3$. In the left figures, the parameters are $\Omega = \Omega_0 \text{sech} \, t/\tau$, λ is constant. Time is in units of λ^{-1} (δ is in units of λ). In the right figures, the parameters are $\lambda = \lambda_0 \cosh t/\tau$, Ω is constant. Time is in units of λ_0^{-1} (δ is in units of λ_0).

various parameters are approximate for ^{41}K (A atom) and ^{87}Rb (B atom). In the left columns, the STIRAP is finished by scheme (i), where the external fields are shown in Fig. 3(a). The parameters are chosen as: $\lambda = 4.718 \times 10^4$ s^{-1}, $\Omega = \Omega_0 \operatorname{sech} t/\tau$ with $\Omega_0/\lambda = 20$, $\lambda\tau = 20$. In the right columns, the STIRAP is finished by scheme (ii), where the external fields are shown in Fig. 3(e). The parameters are chosen as: $\lambda = \lambda_0 \cosh t/\tau$ with $\lambda_0 = 4.718 \times 10^4$ s^{-1}, $\lambda_0\tau = 20$, $\Omega/\lambda_0 = 20$. For both the above cases, as in Ref. 14, the collisional parameters are taken as $\chi_{aa} = 0.3214$, $\chi_{bb} = 0.5303$, $\chi_{ab} = 0.8731$ and other collisional parameters are 0.0938, all in units of λ/n (λ_0/n) in scheme (i) (scheme (ii)).

From Fig. 3(b), one can see that, under scheme (i), no matter whether the interparticle interactions are included, the adiabatic condition is approximately satisfied at the initial time of evolution, i.e. $\varepsilon \ll 1$, which implies the system can adiabatically evolve along the CPT state. At a latter time, the adiabatic condition begins not to be fulfilled, which denotes the system deviate from the CPT state at that time, as can be seen in Fig. 3(c). From Fig. 3(e), under scheme (ii), one can see that the adiabatic conditions with and without interparticle interactions are both fulfilled during the entire evolution. For example, when $\chi_{ij} = 0$, the adiabatic parameter is smaller than 0.035 throughout the evolution process. Therefore, the system can follow the CPT state completely, as is shown in Fig. 3(f). In comparison with the results in scheme (i) and scheme (ii), we find that the adiabatic condition of the atom-trimer dark state in scheme (ii) is much easier to satisfy than the one in scheme (i). Therefore, the adiabaticity of the system in the second scheme is better than the first one.

For the AB channel, we obtain the similar conclusions as in the AA-path, and figures are not shown here.

4. Conclusion

In conclusion, using the methods of classical Hamiltonian dynamics, we investigate the adiabaticity of the dark state in the atom-trimer conversion system in the STIRAP process. We show that, in the absence of the nonlinear collisions, the adiabatic condition for the atom-trimer dark state only depends on the Rabi frequency of the dimer-trimer coupling optical field and has no relation with the atom-dimer coupling Rabi frequency. This is different from the STIRAP processes in the linear system and the atom-dimer conversion system in which the adiabatic parameters not only depend on the Rabi-frequency of the Stokes laser but also the Rabi-frequency of the pump laser. In the presence of the nonlinear collisions, the adiabatic condition also relies on the atom-dimer coupling Rabi frequency. However, its influence is very small. Moreover, we propose a more feasible two-photon STIRAP scheme that has better adiabaticity and is more effective in obtaining higher atom-trimer conversion efficiency.

Acknowledgments

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