

## Adiabatic fidelity for atom-molecule conversion in a nonlinear three-level $\Lambda$ system

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By generalizing the definition of fidelity for the nonlinear system, we investigate the dynamics and adiabaticity of the population transfer for atom-molecule three-level  $\Lambda$  system on a stimulated Raman adiabatic passage (STIRAP). We find that the adiabatic fidelity for the coherent population trapping state or dark state, as the function of the adiabatic parameter, approaches to unit in a power law. The power exponent, however, is much less than the prediction of the linear adiabatic theorem. We further discuss how to achieve higher adiabatic fidelity for the dark state through optimizing the external parameters of the STIRAP.

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In the field of ultracold atomic physics, conversion of an atomic pair to a molecule by means of photoassociation [1] or magnetic Feshbach resonances [2,3] is a hot topic both in experiment and in theory. Photoassociation creates molecules in the excited electronic level, while magnetoassociation creates molecules in a high vibrational quantum state. For fermionic atoms with a feshbach resonance, the resulting molecules can be long lived, while for bosonic atoms, the molecules are energetically unstable and suffer from large inelastic loss rate.

One possible scheme to overcome this difficulty is to employ the stimulated Raman adiabatic passage (STIRAP) [4–6], whose success relies on the existence of the coherent population trapping (CPT) state or dark state [4–6]. For the dark state, the population on the excited electronic level is zero, hence the adiabatic evolution will reduce the spontaneous emission losses in the excited electronic level and create ultracold stable molecules with high conversion efficiency. Therefore the STIRAP scheme is a possible way to achieve the ground molecular Bose-Einstein condensates from its atomic counterpart [7] or extremely weakly bound molecular gases [8] in experiments.

Many recent theoretical works [6,9–11] have been devoted to studying the dynamics, proving the adiabatic condition and improving the conversion efficiency of the atom-molecule coupling model. Indeed, the adiabatic evolution of a system can be thoroughly studied quantitatively by employing adiabatic fidelity which describes the distance between the actual evolution state and the adiabatic state (dark state). However, different from the traditional STIRAP in an atomic system, the atom-molecule STIRAP contains nonlinearity. Creating more difficulties, the system is no longer invariant under U(1)-transformation. Hence the definition of the adiabatic fidelity, which is based on the a U(1) invariance, is no longer suitable for the atom-molecule coupling system.

In the present paper, we properly define an adiabatic fidelity in the nonlinear  $\Lambda$  system. Using this function, we study the adiabatic evolution in the atom-molecule conver-

sion system quantitatively. We find that the adiabatic fidelity for the dark state, as the function of the adiabatic parameter, approaches to unit in a power law. However, the power exponents are much less than the prediction from the linear adiabatic theorem. Then, we further discuss how to optimize the external parameters of the STIRAP process to achieve higher adiabatic fidelity for the dark state.

Our model is schematically sketched in Fig. 1(a). The initial state  $|a\rangle$  (atomic state) and the intermediate state  $|e\rangle$  (excited molecular state) are coupled by pump laser with Rabi frequency  $\Omega_2$ , while state  $|e\rangle$  and the target state  $|g\rangle$  (ground molecular state) are coupled by Stokes laser with Rabi frequency  $\Omega_1$ . The frequencies of the applied lasers are expressed in terms of the single- and two-photon detunings  $\Delta$  and  $\delta$ , respectively. Without loss of generality, we assume that the Rabi frequencies  $\Omega_{1,2}$  are real and positive. Under the two-photon resonance condition ( $\delta=0$ ), the Hamiltonian in second quantized form reads [9,11],

$$H_{am} = -\hbar\Delta\hat{\psi}_e^\dagger\hat{\psi}_e + \frac{\hbar}{2}(-\Omega_2\hat{\psi}_e^\dagger\hat{\psi}_a\hat{\psi}_a + \Omega_1\hat{\psi}_e^\dagger\hat{\psi}_g + \text{H.c.}), \quad (1)$$

where  $\hat{\psi}_i$  and  $\hat{\psi}_i^\dagger$  are the annihilation and creation operators for state  $|i\rangle$ , respectively. Under the mean-field approxima-

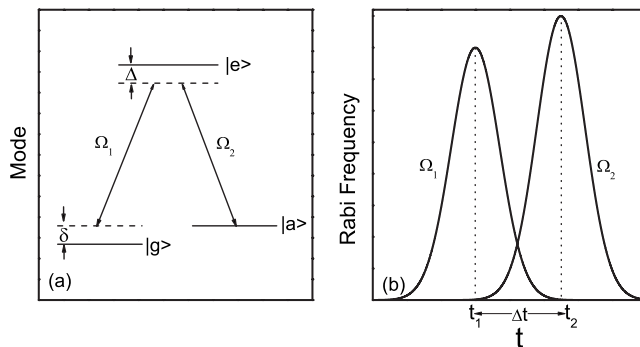


FIG. 1. (a) Three-level system coupled by two lasers.  $\Omega_1, \Omega_2$  are the Rabi frequencies for the pump and Stokes laser,  $\Delta$  and  $\delta$  are one and two-photon detunings, respectively. (b) Time dependence of  $\Omega_1, \Omega_2$ .  $t_1, t_2$  are the centers of the two pulses, respectively, and  $\Delta t$  is the time delay between the two pulses.

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tion, i.e.,  $\hat{\psi}_i$  and  $\hat{\psi}_i^\dagger$  are replaced by  $c$  number  $\psi_i$  and  $\psi_i^*$ , the Schrödinger equations (with  $\hbar=1$ ) are

$$i\dot{\psi}_a = -\Omega_2 \psi_a^* \psi_e, \quad (2a)$$

$$i\dot{\psi}_e = -\Delta \psi_e - \frac{\Omega_2}{2} \psi_a^2 + \frac{\Omega_1}{2} \psi_g, \quad (2b)$$

$$i\dot{\psi}_g = \frac{\Omega_1}{2} \psi_e. \quad (2c)$$

In the above model, the nonlinear collisions between particles are neglected, so the only nonlinearity comes from the fact that it takes two atoms to form a molecule. Mathematically, the Hamiltonian in the above Schrödinger equations is a functional of the instantaneous wave function as well as its conjugate.

The adiabatic theory for nonlinear quantum systems has been set up recently in Ref. [12], where new adiabatic conditions and adiabatic invariants are given. However, these discussions are restricted to systems that have U(1) invariance. For the atom-molecule  $\Lambda$  system, because the Hamiltonian is a functional of both the wave function and its conjugate, the U(1) invariance is broken. Instead, the system is invariant under the following transformation:

$$U(\phi) = e^{i\Theta(\phi)}, \quad \Theta(\phi) = \begin{pmatrix} \phi & 0 & 0 \\ 0 & 2\phi & 0 \\ 0 & 0 & 2\phi \end{pmatrix}. \quad (3)$$

Under this transformation,  $|\psi\rangle = (\psi_a, \psi_e, \psi_g)^T \rightarrow |\psi'\rangle = U(\phi)|\psi\rangle = (\psi_a e^{i\phi}, \psi_e e^{i2\phi}, \psi_g e^{i2\phi})^T$ . In fact, when the diagonal terms in the above matrix are identical, the transformation  $U(\phi)$  degenerates to the U(1) transformation.

This new kind of symmetry allows the following stationary states with chemical potential  $\mu$ :

$$\psi_a = |\psi_a| e^{i\theta_a} e^{-i\mu t}, \quad \psi_{e,g} = |\psi_{e,g}| e^{i\theta_{e,g}} e^{-i2\mu t}, \quad (4)$$

where

$$2\theta_a - \theta_e = \text{const}, \quad \theta_g - \theta_e = \text{const}. \quad (5)$$

Under the normalized condition ( $|\psi_a|^2 + 2|\psi_e|^2 + 2|\psi_g|^2 = 1$ ), we can show, as in the atomic counterpart, the atom-molecule  $\Lambda$  system supports a CPT eigenstate [9,10] with zero eigenvalue;

$$|\text{CPT}\rangle = \left( \frac{(\Omega_1 \Omega_{eff}^{nl} - \Omega_1^2)^{1/2}}{2\Omega_2}, 0, \frac{\Omega_{eff}^{nl} - \Omega_1}{4\Omega_2} \right)^T, \quad (6)$$

where  $\Omega_{eff}^{nl} = \sqrt{\Omega_1^2 + 8\Omega_2^2}$ .

When the Rabi laser pulses are ramped up adiabatically, i.e.,  $\Omega_{1,2}$  vary in time slowly, the state that is initially prepared as the CPT state is expected to remain close to the instantaneous CPT state throughout the entire process. The problem is how close is the above adiabatic approximation? To clarify the above question and formulate it quantitatively, we introduce two physical quantities, namely the adiabatic fidelity and the adiabatic parameter.

For the linear system, adiabatic fidelity is introduced as

$F_{ad} = |\langle \psi(t) | \psi_{ad} \rangle|^2$ , where  $|\psi_{ad}\rangle$  is the adiabatic approximation solution, i.e., the instantaneous eigenstate of the Hamiltonian and  $|\psi(t)\rangle$  is the real solution which has evolved under the Hamiltonian from  $|\psi(0)\rangle$ . The adiabatic fidelity approaches to unit in a power law of the adiabatic parameter [13], i.e.,  $1 - F_{ad} \sim \epsilon^2$ . Here, the adiabatic parameter  $\epsilon$  is the ratio between the rate of the energy change and the level spacing. For linear quantum systems, evaluating the adiabatic fidelity gives a good estimation on how close the adiabatic solution is to the real solution [14].

For the nonlinear atom-molecule conversion system, the traditional definition of fidelity is no longer suitable because the system is not invariant under the U(1)-transformation. We need to define new fidelity for such systems. For convenience, we denote the fidelity of two states  $|\psi_1\rangle$  and  $|\psi_2\rangle$  as  $F^{am}(|\psi_1\rangle, |\psi_2\rangle)$ . This definition should not only satisfy  $F^{am}(|\psi\rangle, |\psi\rangle) = 1$  for any  $|\psi\rangle$  but also fulfill  $F^{am}(|\psi\rangle, U(\phi)|\psi\rangle) = 1$  for any  $\phi$  [15]. With this consideration, we define the adiabatic fidelity as  $F^{am}(|\psi_1\rangle, |\psi_2\rangle) = |\langle \bar{\psi}_1 | \bar{\psi}_2 \rangle|^2$ , where  $|\bar{\xi}\rangle = (\xi_a^2 / |\xi_a|, \sqrt{2}\xi_e, \sqrt{2}\xi_g)^T$  is the rescaled wave function of  $|\xi\rangle = (\xi_a, \xi_e, \xi_g)^T$ . For any wave function  $|\xi\rangle$ , it is readily proved that  $F^{am}(|\xi\rangle, |\xi\rangle) = |\xi_a|^2 + 2|\xi_e|^2 + 2|\xi_g|^2 = 1$  and  $F^{am}(|\xi\rangle, U(\phi)|\xi\rangle) = \|\xi\rangle, U(\phi)|\xi\rangle\|^2 = |(\xi_a^{*2} / |\xi_a^*|, \sqrt{2}\xi_e^*, \sqrt{2}\xi_g^*)^T (\xi_a^{-2} e^{2i\phi} / |\xi_a|, \sqrt{2}\xi_e e^{2i\phi}, \sqrt{2}\xi_g e^{2i\phi})|^2 = |\xi_a|^2 + 2|\xi_e|^2 + 2|\xi_g|^2 = 1$ . Hence the above two conditions are satisfied. On the other hand, this definition also fulfills  $F^{am}(|\psi_1\rangle, U(\phi)|\psi_2\rangle) = F^{am}(U(\phi)|\psi_1\rangle, |\psi_2\rangle)$ , which means the phase transformation given in Eq. (3) does not vary the magnitude of fidelity. Because we are only concerned with the adiabatic evolution of the CPT state throughout, we denote the adiabatic fidelity as  $F^{am} = |\langle \psi(t) | \text{CPT} \rangle|^2$  where  $|\psi(t)\rangle = (\psi_a, \psi_e, \psi_g)^T$  is the exact solution of the Schrödinger equation.  $|\bar{\psi}\rangle$  and  $|\text{CPT}\rangle$  are the rescaled wave functions of the  $|\psi\rangle$  and CPT state respectively.

For the atom-molecule conversion system, owing to the invalidation of the concept of an orthogonal set of energy eigenstates and the linear superposition principle involving these states, the adiabatic parameter is not related to the energy level spacing [12]. Moreover, since the eigenstates correspond to extremum points or fixed points of the system energy, the fundamental frequencies of periodic orbits around the fixed points serves as the adiabatic parameter. These frequencies can be evaluated by linearizing Eq. (2) about the fixed points and are identical to the Bogoliubov excitation spectrum of the corresponding eigenstate, as is demonstrated in [9,12], the adiabatic parameter is

$$\epsilon^{am} = \left| \frac{\dot{\Omega}_1 \Omega_2 - \Omega_1 \dot{\Omega}_2}{\Omega_1 + \Omega_{eff}^{nl}} \right| \frac{1}{\Omega_{eff}^{nl} \Omega_1 / 2}. \quad (7)$$

For the atom-molecule  $\Lambda$  system, STIRAP can be implemented as usual by using two laser pulses applied in counterintuitive order. We choose Gaussian pulses of the form

$$\Omega_{1,2} = \Omega'_{1,2} e^{-(t-t_{1,2})^2/\tau^2}, \quad (8)$$

where the pulse width  $\tau$  has been taken as units for time in the following discussions.

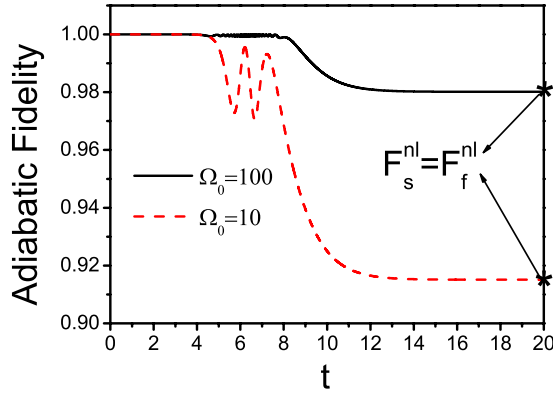


FIG. 2. (Color online) Time evolution of the adiabatic fidelity for the atom-molecule three-level system with  $\Delta=0$ ,  $t_1=5.0$ ,  $t_2=6.0$ ,  $\Delta t=1.0$ ,  $\Omega_0=10, 100$ .

First, for convenience, we consider the case when the two pulses have equal amplitudes, i.e.,  $\Omega'_1=\Omega'_2=\Omega_0$ . Substituting Eq. (8) into Eq. (7), we obtain the adiabatic parameter of the atom-molecule conversion system:

$$\epsilon^{am} \sim \frac{1}{\Omega_0}. \quad (9)$$

In the STIRAP, it is required that  $t_1 < t_2$  and only level  $|a\rangle$  is populated initially, then the system evolves under  $H_{am}$ . In our calculation, we solve the nonlinear Schrödinger equation using the fourth-fifth order Runge-Kutta adaptive-step algorithm.

To demonstrate the dynamics of the population transfer, in Fig. 2, we show a typical change of the adiabatic fidelity with time for the three-level atom-molecule conversion system. We focus on two quantities to describe dynamics of the atom-molecule conversion system: the smallest adiabatic fidelity  $F_s^{am}$  and the final adiabatic fidelity  $F_f^{am}$ . The former ( $F_s^{am}$ ) can be used to depict the adiabaticity of motion, since  $F_f^{am}$  denotes the maximal deviation of  $|\psi(t)\rangle$  from the CPT state during the evolution, and the closer  $F_f^{am}$  to 1 is, the better the adiabaticity is. The latter is used to indicate the conversion efficiency because  $F_f^{am}$  is the final value of adiabatic fidelity  $F(t)$  in the process of evolution, and the larger  $F_f^{am}$  is, the higher the conversion efficiency is. These two points are labeled \* in Fig. 2. For this case,  $F_s^{am} = F_f^{am}$  which can be found in this figure. In addition, this figure also demonstrates that  $F_s^{am}(\Omega_0=100) > F_s^{am}(\Omega_0=10)$ . Then the relationship between the smallest adiabatic fidelity  $F_s^{am}$  and the amplitude  $\Omega_0$  becomes interesting because it denotes the relation between the adiabaticity and the amplitude. In this way, we can further find whether there is an analogous relation between the adiabatic fidelity and the adiabatic parameter as in the linear system.

Figure 3 shows the smallest adiabatic fidelity  $F_s^{am}$  as a function of  $\Omega_0$  for the three-level atom-molecule conversion system. This figure indicates that the smallest adiabatic fidelity grows with the increasing amplitude accompanied by small oscillation, and we can obtain the asymptotic behaviors of the lower bounds of  $F_s^{am}$  which can be expressed as

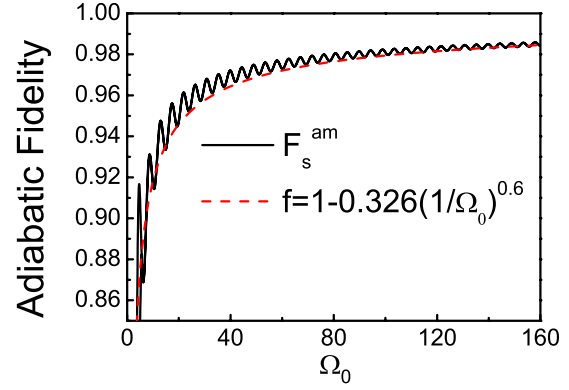


FIG. 3. (Color online) Smallest adiabatic fidelity for the atom-molecule three-level system as a function of the amplitude  $\Omega_0$  with  $\Delta=0$ ,  $t_1=5.0$ ,  $t_2=6.0$ ,  $\Delta t=1.0$ .

$$1 - F_s^{am} = 0.326 \left( \frac{1}{\Omega_0} \right)^{0.6} \sim (\epsilon^{am})^{0.6}. \quad (10)$$

As in the linear system where the adiabatic fidelity as a function of the adiabatic parameter approaches to a power law, i.e.,  $1 - F \sim \epsilon^2$ , there also exists a power law relationship between the adiabatic fidelity and the adiabatic parameter in the atom-molecule conversion system, but the exponent is 0.6 rather than 2. Here the power exponent is not universal, but depends on the time delay  $\Delta t$ . For example, when  $\Delta t = 0.8$ , it is 0.5, and when  $\Delta t = 1.2$ , it is 1.3.

We have discussed the case when the two pulses have equal amplitudes  $\Omega_0$  with  $\Delta t = 1.0$ , and find that the larger the amplitude  $\Omega_0$  is, the better the adiabaticity is, and hence the higher the conversion efficiency is. Now we consider how the adiabatic fidelity changes with the time delay. In Fig. 4(a), the smallest adiabatic fidelity  $F_s^{am}$  is plotted as a function of the pulse delay  $\Delta t$  for several different values of  $\Omega_0$  in the atom-molecule three-level system. This figure shows that, as the delay  $\Delta t$  increases, the smallest adiabatic fidelity first increases, then reaches a steady stage depending on the amplitude  $\Omega_0$ , and finally decreases beyond certain values of

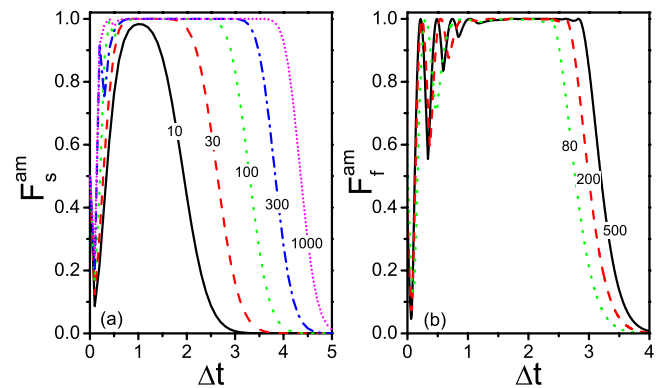


FIG. 4. (Color online) (a) Smallest adiabatic fidelity of atom-molecule three-level system plotted against the pulse delay  $\Delta t$  for different  $\Omega_0$ . (b) Final adiabatic fidelity of atom-molecule three-level system plotted against the pulse delay  $\Delta t$  for different  $\Omega_0^2$  at  $\Omega_1^2=10$ .

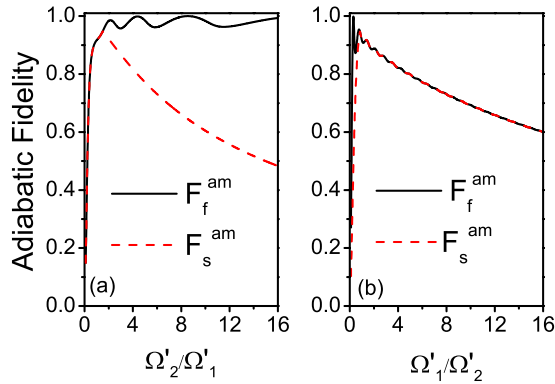


FIG. 5. (Color online) Final and smallest adiabatic fidelity for atom-molecule three-level system (a) as a function of  $\Omega'_2/\Omega'_1$  with  $\Omega'_1=10$  and (b) as a function of  $\Omega'_1/\Omega'_2$  with  $\Omega'_2=10$ .  $\Delta=0$ ,  $t_1=5.0$ ,  $t_2=6.0$ ,  $\Delta t=1.0$ .

$\Delta t$ . So, for the atom-molecule  $\Lambda$ -coupling system, there is an ideal length for time delay  $\Delta t$ , which increases as the amplitude  $\Omega_0$  grows, optimizing the adiabaticity and hence the conversion efficiency.

Second, we consider a more complicated case, i.e., the two pulses having different amplitudes, and study the adiabatic property and the conversion efficiency of the system. Now, the smallest adiabatic fidelity  $F_s^{am}$  is no longer equal to the final adiabatic fidelity  $F_f^{am}$ . In the following discussions, we are concerned with these two quantities because the former can be used to describe the adiabaticity of the system and the latter can be used to investigate the conversion efficiency.

Figure 5 shows  $F_s^{am}$  and  $F_f^{am}$  as a function of  $\Omega'_2/\Omega'_1$  and  $\Omega'_1/\Omega'_2$  with  $\Delta t=1.0$ . In Fig. 5(a), as  $\Omega'_2$  increases, both  $F_s^{am}$

and  $F_f^{am}$  increase until reaching a critical point,  $F_s^{am}$  begins to decrease while  $F_f^{am}$  stays close to 1. This means the adiabaticity of the system becomes worse, but the process still realizes complete population transfer. The physics behind this is that the effective coupling between  $|a\rangle$  and  $|e\rangle$  scaled as  $\Omega_2\psi_a$  is weakened by the decreasing atom population on  $|a\rangle$ , hence a higher pump Rabi frequency can remedy this kind of weakening and enhance the two-photon process. However, this case is different in Fig. 5(b) where both  $F_s^{am}$  and  $F_f^{am}$  decrease after the critical points as  $\Omega'_1$  increases. The adiabaticity and conversion efficiency are both poor. This case is not considered below.

The high conversion efficiency with increasing  $\Omega'_2/\Omega'_1$  drives us to study whether there is a segment or points optimizing the conversion efficiency for different  $\Omega'_2/\Omega'_1$ . In Fig. 4(b), the final adiabatic fidelity  $F_f^{am}$  is plotted as a function of the pulse delay  $\Delta t$  for several different values of  $\Omega'_2$  with  $\Omega'_1=10$ . We find that, for the case of equal amplitudes, there is also a segment of time delay  $\Delta t$  making the conversion efficiency of the atom-molecule system optimal. Moreover, the segment increases as the amplitude of the pump laser  $\Omega'_2$  grows.

In conclusion, we investigated the adiabatic fidelity for the CPT state in a nonlinear system on the STIRAP quantitatively, and discuss how to achieve higher adiabatic fidelity for the dark state through optimizing the external parameters of STIRAP.

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