

Chirped multiphoton adiabatic passage for a four-level ladder-type Rydberg excitationJing Qian,^{*} Jingjing Zhai, Lu Zhang, and Weiping Zhang*Quantum Institute for Light and Atoms, Department of Physics, East China Normal University, Shanghai 200062, People's Republic of China*

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We develop a multiphoton adiabatic passage to realize a highly efficient Rydberg excitation in a four-level ladder-type atomic system. The adiabatic passage is based on the existence of a novel quasidark state in the cascade excitation system where the frequencies of the lasers are appropriately chirped with time. We also investigate the influence of the interatomic Rydberg interaction on the passage and extend its application to the preparation of antiblockade Rydberg atom pairs in the Rydberg blockade regime.

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I. INTRODUCTION

Recently, it has been recognized that quantum optical systems involving highly excited atomic Rydberg levels can provide a promising perspective for applications in quantum information processing, quantum metrology, and quantum simulation [1–4]. In particular, the huge polarizability of Rydberg states gives rise to giant Kerr coefficients, which enable nonlinear optical effects, such as electromagnetically induced transparency, in the single-photon level [5]. It also induces the strong dipole-dipole or van der Waals interactions between neighboring Rydberg atoms, which can be used to implement atom-photon and photon-photon phase gates [6–8], effective photon-photon interactions [9,10], and deterministic single-photon sources [11–13].

In experimental realizations of these and other applications, it will be essential to have methods available to efficiently transfer ground-state atoms into and out of the highly excited Rydberg states [14]. In the direct excitation case, except for the requirement of deep ultraviolet wavelengths of the lasers, the poor selectivity of the excitation due to the optical mixing of the nearly degenerate Rydberg states limits its use [15]. These shortcomings can be made up by a multistep excitation via some intermediate states [16–20]. In particular, stimulated Raman adiabatic passage (STIRAP) is a powerful technique that is used intensively in preparing and manipulating atomic and molecular states [21]. It is sufficiently robust to overcome the effects of the Doppler broadening, spatial laser-pulse inhomogeneities, and the spontaneous decay of the intermediate excited state. However, the formation of the dark state restricts the application of the STIRAP to the three-level excitation scheme. The multiphoton adiabatic passage and the fractional STIRAP with optimal pulse sequences are thus extensively studied [22–27] to realize highly efficient excitation in four- or more level schemes.

Compared to the general atomic excitation, strong interatomic interaction induced by the huge dipole moment of Rydberg states may result in poor effects of all above excitation strategies in the Rydberg excitation. The interaction shifts the original atomic energy levels and blocks the Rydberg excitation of the atoms whose neighboring atom has been excited to the Rydberg state, which is so-called Rydberg blockade effect [28–30]. To overcome this obstacle, one

direct solution resorts to increasing the mean interatomic distance (typically larger than $10\ \mu\text{m}$) by lowering atomic gas density. Then the probability of obtaining Rydberg-excited atoms can have clear growth [31]. An alternative way is to utilize an appropriate optical detuning with respect to the Rydberg state to compensate for the energy shift induced by the Rydberg-Rydberg interaction, which can generate antiblockade Rydberg atom pairs even in the deep Rydberg blockade regime [32–34]. A modified STIRAP scheme based on this detuning-compensated approach has also been proposed lately [35].

In the present work, we design a multiphoton adiabatic passage to realize a highly efficient Rydberg excitation in a four-level ladder-type atomic system with two intermediate excited states located between the ground state and the Rydberg state. Differing from the previous works, the present adiabatic passage is based on a quasidark state which involves only one of the intermediate excited states. This excitation scheme depends on the application of three coherent light pulses with carefully chosen Rabi frequencies, time widths, delay times, and most importantly chirped optical frequencies. The influence of the Rydberg-Rydberg interaction on this excitation scheme is investigated and discussed. We further find the detuning with respect to the other intermediate state could be an effective control knob to reduce the destructive effect of the Rydberg-Rydberg interaction to the excitation. Then the high excitation efficiency of the Rydberg atom pairs can be reached even in the deep Rydberg blockade regime, where the adiabatic passage no longer exists.

This paper is organized in the following way: In Sec. II, we establish a detailed model for the quasidark state and multiphoton adiabatic passage in a four-level ladder-type atomic system without Rydberg-Rydberg interaction. The parameters of the optical pulses are optimized to make a trade-off between the fidelity of the adiabatic passage based on the quasidark state and the spontaneous decay of the intermediate state involved. The influence of the Rydberg-Rydberg interaction on the excitation scheme is displayed and discussed with a two-atom model in Sec. III, while the crucial role for the optical detuning with respect to the other intermediate state in the strong interaction case is shown in Sec. IV.

II. CHIRPED MULTIPHOTON ADIABATIC PASSAGE

As shown in Fig. 1(a), we consider a four-level ladder-type atomic system with a ground state $|g\rangle$ and a Rydberg state

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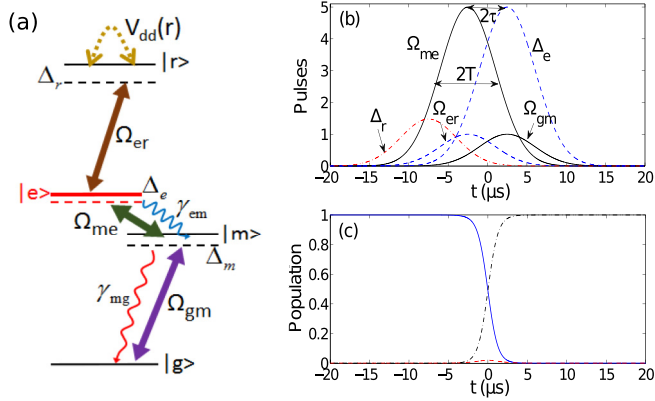


FIG. 1. (Color online) (a) Scheme of the four-level ladder-type system interacting with the three laser fields with Rabi frequencies Ω_{gm} , Ω_{me} , and Ω_{er} , and detunings Δ_m , Δ_e , and Δ_r from the intermediate state $|m\rangle$, the intermediate state $|e\rangle$, and the Rydberg state $|r\rangle$, respectively. The two intermediate states are unstable and suffer from decay of rates γ_{em} and γ_{mg} . V_{dd} represents the interaction between two neighboring Rydberg atoms. (b) Two pairs of pulse sequences with counterintuitive character and their time-dependent Rabi frequencies denoted by $\Omega_{gm}(t)$ and $\Omega_{me}(t)$ (black solid curves), $\Delta_e(t)$ and $\Omega_{er}(t)$ (blue [gray] dashed curves), driving atoms from ground state $|g\rangle$ to Rydberg state $|r\rangle$ via a chirped multiphoton adiabatic passage. (c) The time evolution of the quasidark state $|D_4\rangle$ with the population probabilities of the components $|g\rangle$, $|r\rangle$, and $|e\rangle$ labeled by the blue (gray) solid curve, the black dash-dotted curve, and the red (gray) dashed curve, respectively. The population probability of $|e\rangle$ has been largely suppressed by optimized pulses.

$|r\rangle$, which can be excited via the two intermediate states $|m\rangle$ and $|e\rangle$. The transition Rabi frequencies between states $|i\rangle$ and $|j\rangle$ are denoted by Ω_{ij} , with $ij = gm, me, er$, and optical detunings $\Delta_{m,e,r}$ may be applied with respect to the corresponding levels. All the Rabi frequencies are assumed to be real and positive in this work. The Rydberg and the two intermediate excited states decay by spontaneous emission of radiation, and we assume that the lifetime of the Rydberg state is much longer than the other two so that we can neglect its decay. In the present section, we also neglect the dipole-dipole interaction V_{dd} between the Rydberg atoms, which may induce the Rydberg blockade effect, and focus on the single-atom excitation mechanism. Then in the rotating-wave frame, the Hamiltonian describing Rydberg excitation of such a single four-level atom is given by (where $\hbar = 1$ throughout the work)

$$\mathcal{H}_0 = \Delta_m |m\rangle\langle m| + \Delta_e |e\rangle\langle e| + \Delta_r |r\rangle\langle r| + (\Omega_{gm} |m\rangle\langle g| + \Omega_{me} |e\rangle\langle m| + \Omega_{er} |r\rangle\langle e| + \text{H.c.}), \quad (1)$$

and the transition dynamics and the dissipations can be described by the master equation of single atom density matrix $\hat{\rho}$,

$$\partial_t \hat{\rho}(t) = -i[\mathcal{H}_0(t), \hat{\rho}(t)] + \mathcal{L}[\hat{\rho}(t)], \quad (2)$$

where the Lindblad operator $\mathcal{L}[\hat{\rho}(t)]$, referring to the spontaneous decays from the two intermediate unstable states $|m\rangle$

and $|e\rangle$, is readily written as

$$\begin{aligned} \mathcal{L}[\hat{\rho}] = & \frac{\gamma_{mg}}{2} (2\hat{\sigma}_{gm}\hat{\rho}\hat{\sigma}_{gm}^\dagger - \{\hat{\sigma}_{mm}, \hat{\rho}\}) \\ & + \frac{\gamma_{em}}{2} (2\hat{\sigma}_{me}\hat{\rho}\hat{\sigma}_{me}^\dagger - \{\hat{\sigma}_{ee}, \hat{\rho}\}) \end{aligned} \quad (3)$$

with $\hat{\sigma}_{ij} = |i\rangle\langle j|$ denoting the atomic transition operator and $\gamma_{em(mg)}$ the spontaneous decay rates with respect to states $|e\rangle$ ($|m\rangle$). An efficient population transfer from the ground state $|g\rangle$ to the Rydberg state $|r\rangle$ can be achieved relatively easily in experiments via a multiple photon cascade excitation, with dissipation from the intermediate states being a major impediment [36].

Recently the technique of STIRAP is employed widely in the Rydberg excitation of three-level ladder atomic systems due to its advantage of immunity to the decay of the intermediate state [37,38]. This feature arises from the existence of a “dark” eigenstate $|D_3\rangle$ in a coherent coupled three-level system, with eigenenergy $\lambda_3 = 0$ and eigenvector

$$|D_3\rangle = \cos\theta |g\rangle - \sin\theta |r\rangle, \quad (4)$$

where the mixing angle θ is given by $\tan\theta = \Omega_g/\Omega_r$ with $\Omega_{g(r)}$ being the Rabi frequency between the intermediate state and the state $|g\rangle$ ($|r\rangle$). It does not have any contribution from the unstable intermediate state. Then a complete transfer from $|g\rangle$ to $|r\rangle$ can be achieved by adiabatically changing the dark state superposition, that is, performing a pair of optical pulses in a counterintuitive order [21,39].

For a four-level atomic system considered in the present work, by diagonalizing Hamiltonian (1) we find due to the different symmetry from the three-level system, there is no dark state which is immune to the two intermediate states $|m\rangle$ and $|e\rangle$. However, as long as the condition

$$\Omega_{er}^2 = \Delta_e \Delta_r \quad (5)$$

is fulfilled, there exists a novel eigenstate $|D_4\rangle$ with its eigenenergy $\lambda_4 = 0$ and eigenvector

$$|D_4\rangle = \cos\varphi |g\rangle + \sin\varphi \cos\phi |e\rangle - \sin\varphi \sin\phi |r\rangle, \quad (6)$$

which involves one of the intermediate states, $|e\rangle$, and is called the quasidark state by us. In comparison to state $|D_3\rangle$, $|D_4\rangle$ possesses two mixing angles, which are given by

$$\tan\varphi = -\frac{\Omega_{gm}}{\Omega_{me}} \sqrt{1 + \frac{\Delta_e^2}{\Omega_{er}^2}}, \quad (7)$$

$$\tan\phi = \frac{\Delta_e}{\Omega_{er}}. \quad (8)$$

Similar to the three-level case, one can build a multiphoton adiabatic passage via state $|D_4\rangle$. The atom initially prepared in the ground state $|g\rangle$ ($\langle g|D_4\rangle = 1$ for $\cos\varphi = 1$ and $\cos\phi = 0$) will finally settle in the Rydberg state $|r\rangle$ ($\langle r|D_4\rangle = 1$ for $\sin\varphi = 1$ and $\sin\phi = 1$) by slowly adjusting the two mixing angles. It is a bit special that except for the three Rabi frequencies Ω_{gm} , Ω_{me} , and Ω_{er} , the detunings Δ_e and Δ_r should also change over time to meet the quasidark condition (5). This could be achieved through coupling state $|m\rangle$ and state $|e\rangle$ and coupling state $|e\rangle$ and state $|r\rangle$ using two frequency-chirped optical pulses, the technology of which has been widely investigated in the theory and the experiment of

atomic and molecular state control [40–45]. The time sequence of pulses and the time dependence of detunings for our chirped multiphoton adiabatic passage are displayed in Fig. 1(b).

Since the quasidark state $|D_4\rangle$ involves intermediate state $|e\rangle$ of nonignorable decay, which may break the adiabatic transfer, we first calculate the accumulated population probability on the component state $|e\rangle$ of state $|D_4\rangle$ over the whole transfer process, whose value can briefly estimate that destructive effect. For generality, we assume all the pulses and chirped detunings are Gaussian, with the same width T and the same duration τ ,

$$\begin{aligned}\Omega_{gm}(t) &= \Omega_{gm}^{\max} e^{-(t-\tau)^2/T^2}, \\ \Omega_{me}(t) &= \Omega_{me}^{\max} e^{-(t+\tau)^2/T^2}, \\ \Delta_e(t) &= \Delta_e^{\max} e^{-(t-\tau)^2/T^2}, \\ \Omega_{er}(t) &= \Omega_{er}^{\max} e^{-(t+\tau)^2/T^2}, \\ \Delta_r(t) &= \frac{(\Omega_{er}^{\max})^2}{\Delta_e^{\max}} e^{8\tau^2/T^2} e^{-(t+3\tau)^2/T^2},\end{aligned}\quad (9)$$

where the quantities with superscript ‘‘max’’ represent the Gaussian peak values. $\tau > 0$ is required to ensure the counter-intuitive feature of STIRAP pulses. Then according to Eq. (6), the time evolution of the population probabilities on the three components of quasidark state $|D_4\rangle$ is plotted in Fig. 1(c), and the accumulated population probability on the component state $|e\rangle$ is given by

$$\int_{-\infty}^{+\infty} |\sin \varphi \cos \phi|^2 dt = \frac{T^2}{8\tau} f(\chi_1, \chi_2), \quad (10)$$

where we have defined

$$f(\chi_1, \chi_2) = \int_{-\infty}^{+\infty} d\xi \frac{\chi_1^2}{\chi_1^2 \chi_2^2 e^{\xi} + e^{-\xi} + \chi_1^2}, \quad (11)$$

as a function of the two peak value ratios $\chi_1 = \Omega_{gm}^{\max}/\Omega_{me}^{\max}$ and $\chi_2 = \Delta_e^{\max}/\Omega_{er}^{\max}$. We plot $f(\chi_1, \chi_2)$ in Fig. 2, which clearly shows that the condition $\chi_2 > \chi_1$ is preferred for a small accumulated population probability and thus a weak dissipation effect. This is due to the condition that gives $\Delta_e^{\max} > \Omega_{gm}^{\max} \Omega_{er}^{\max}/\Omega_{me}^{\max}$, which means a far-off resonant excitation to the intermediate state $|e\rangle$. According to Eq. (10), it seems that a short pulse width and a long pulse duration can

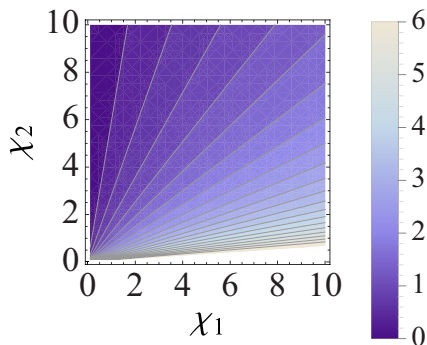


FIG. 2. (Color online) The accumulated population probability on the excited state $|e\rangle$ is estimated by the characteristic function $f(\chi_1, \chi_2)$. Areas marked by darker color mean smaller values of f .

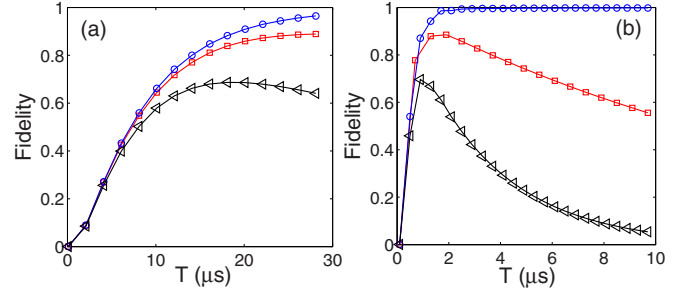


FIG. 3. (Color online) Fidelity $F^{(\infty)}$ as a function of pulse width T for different spontaneous decay rates of state $|e\rangle$: $\gamma_{em} = 0$ (blue [gray] curve with circles), 0.1 MHz (red [gray] curve with squares), and 0.5 MHz (black curve with triangles). (a) $\Omega_{gm}^{\max} = \Omega_{er}^{\max} = 1.0$ MHz, $\Omega_{me}^{\max} = \Delta_e^{\max} = 5.0$ MHz and (b) $\Omega_{gm}^{\max} = \Omega_{er}^{\max} = \Omega_{me}^{\max} = \Delta_e^{\max} = 5.0$ MHz. In simulations other parameters are $\gamma_{mg} = 0.1$ MHz, $\tau = 0.5T$, $\Delta_m = 0$, $\Delta_r = \Omega_{er}^2/\Delta_e$.

also reduce the accumulated population probability. However, it means a rapidly changed pulse sequence, which goes against the adiabatic passage theory [46].

To investigate the influence of these relevant parameters on the chirped multiphoton adiabatic passage (CMPAP) more precisely, we define the fidelity of the quasidark state $F^{(t)} = \langle D_4(t) | \hat{\rho}(t) | D_4(t) \rangle$ and simulate the master equation (2) to obtain its value at $t \rightarrow \infty$ for various parameters. Notice that $F^{(\infty)}$ is just the population of the Rydberg state $|r\rangle$. As shown in Fig. 3, when there is no dissipation of the intermediate state $|e\rangle$ ($\gamma_{em} = 0$, which means $|D_4(t)\rangle$ is a perfect dark state), the curves labeled by circles show that the value of $F^{(\infty)}$ can reach unity as long as the applied pulse for optical excitation is sufficiently long so that the dark state can be adiabatically followed. In the presence of nonzero γ_{em} , we compare the case in which $\Omega_{gm}^{\max} = \Omega_{er}^{\max} = 1.0$ MHz and $\Omega_{me}^{\max} = \Delta_e^{\max} = 5.0$ MHz [Fig. 3(a)], which meets the decay-suppressed condition $\chi_2 > \chi_1$, with the case in which $\Omega_{gm}^{\max} = \Omega_{er}^{\max} = \Omega_{me}^{\max} = \Delta_e^{\max} = 5.0$ MHz [Fig. 3(b)], which does not meet the condition. In both cases, the fidelity decreases as the decay rate γ_{em} increases. As the pulse width T grows, an obvious single-peak structure of the fidelity appears in the second case, which is different from the typical STIRAP case. This is because our CMPAP is based on a quasidark state. For short pulses the adiabatic following cannot be guaranteed, while for long pulses the influence of the decay of state $|e\rangle$ becomes significant, which will also breaks the adiabaticity. In a real experimental realization, the rubidium atom system as used in Rydberg excitation experiments [18] and [47] is a suitable candidate to adopt our approach. The selected four levels of rubidium atoms in those experiments can build a ladder-type atomic system required by the CMPAP, e.g., $5S_{1/2}(|g\rangle)$, $5P_{3/2}(|m\rangle)$, $6S_{1/2}$ or $5D_{5/2}(|e\rangle)$, and $6P_{3/2}(|r\rangle)$. The natural linewidths of the two intermediate states are in the order of several MHz, which is consistent with the values applied in our numerical simulations.

We conclude that without the Rydberg-Rydberg interaction it is possible to achieve an easy and high-fidelity CMPAP to coherent transfer ground-state atoms into the high-lying Rydberg state in a four-level ladder system.

III. INTERACTING RYDBERG ATOMS

With two or more atoms the Rydberg blockade effect comes into play, which will give rise to an energy shift on the single-atom Rydberg state. To investigate its influence to the performance of the CMPAP, we consider a two-atom model whose Hamiltonian is written as

$$\mathcal{H}_I = \mathcal{H}_0 \otimes \mathcal{I} + \mathcal{I} \otimes \mathcal{H}_0 + V_{dd}|rr\rangle\langle rr|, \quad (12)$$

where \mathcal{H}_0 is the four-level single-atom Hamiltonian given by Eq. (1), \mathcal{I} is the identity matrix, and V_{dd} represents the dipole-dipole interaction strength between two atoms both on the Rydberg state. The corresponding master equation becomes

$$\partial_t \hat{\rho}_I(t) = -i[\mathcal{H}_I(t), \hat{\rho}_I(t)] + \sum_{j=1,2} \mathcal{L}_j[\hat{\rho}_I(t)], \quad (13)$$

where the form of the Lindblad operator \mathcal{L}_j is same as Eq. (3) but with the atomic transition operators for atom j only. The subscript “ I ” indicates the case for two interacting atoms.

The eigenstates of \mathcal{H}_I may be quite different from the single-atom case due to the Rydberg interaction term so as to threaten the existence of the quasidark state $|D_4\rangle$. In the weak interaction case where $|V_{dd}| \ll \Omega$ with single-atom characteristic frequency $\Omega \in \{\Omega_{gm}^{\max}, \Omega_{me}^{\max}, \Omega_{mr}^{\max}, \Delta_e^{\max}\}$, the CMPAP for Rydberg excitation of both atoms are sustained with the two-atom eigenstate, which can be approximately represented as $|D_4\rangle_I = |D_4\rangle \otimes |D_4\rangle$. Then there still exists an adiabatic passage between the ground state $|gg\rangle$ and the double Rydberg excited state $|rr\rangle$. However, this is not the case for $|V_{dd}| \sim \Omega$ or even $|V_{dd}| > \Omega$, where the target state for the CMPAP could be no longer the state $|rr\rangle$ due to the Rydberg blockade effect [38].

A similar example of this is in the three-level Rydberg excitation system. When the detuning with respect to the intermediate state is zero, the single-atom dark state $|D_3\rangle$ is replaced by a new two-atom quasidark state [48],

$$|D_3\rangle_I = \frac{1}{\sqrt{\cos^4 \theta + 2 \sin^4 \theta}} [(\cos^2 \theta - \sin^2 \theta)|gg\rangle - \cos \theta \sin \theta(|gr\rangle + |rg\rangle) + \sin^2 \theta |mm\rangle], \quad (14)$$

which means while the STIRAP process in the single-atom case ensures that the Rydberg state $|r\rangle$ is finally populated, due to the Rydberg-Rydberg interaction the pair of atoms will be adiabatically steered into a superposition state of the ground state $|gg\rangle$ and the intermediate state $|mm\rangle$. The situation can be modified by applying a finite and appropriate detuning from the intermediate state, which might generate a new eigenstate that involves both $|gg\rangle$ and $|rr\rangle$, leading to a near-unity final population of $|rr\rangle$ [49]. However, since in the case of our four-level Rydberg atoms excited by the CMPAP, the detunings Δ_m and Δ_e are both time dependent, the same approach cannot be adopted directly. We then first perform a numerical simulation on the excitation dynamics by the two-atom master equation (13) to investigate the influence of the interaction strength V_{dd} on the two-atom eigenstates and on the CMPAP.

We focus on the several two-atom eigenstates $|E_j\rangle$, which can serve as the double Rydberg excitation passages, that is, $|E_j(t = -\infty)\rangle = |gg\rangle$ and $|E_j(t = +\infty)\rangle = |rr\rangle$, when the

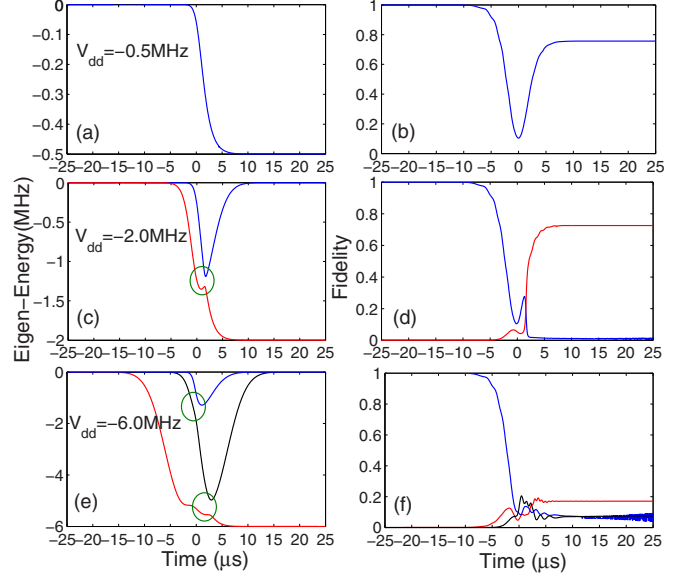


FIG. 4. (Color online) Left column: Time evolution of the energies of the eigenstates involved in the transition from $|gg\rangle$ to $|rr\rangle$ for the cases, from top to bottom, $V_{dd} = -0.5$, -2.0 , and -6.0 MHz. Right column: Corresponding fidelity (population) in these eigenstates. Other parameters are same as in Fig. 3(b); note that $\Delta_m = 0$.

same pulse sequences and the chirped detunings as discussed in the last section are applied. The time evolutions of their eigenenergies and fidelities $F_j^{(t)} = \langle E_j(t) | \hat{\rho}_I(t) | E_j(t) \rangle$ under the different Rydberg interaction strengths are displayed in Fig. 4. In the weak interaction case ($|V_{dd}| \ll \Omega$) we find there exists a single two-atom eigenstate which can connect $|gg\rangle$ and $|rr\rangle$. However, its eigenenergy changes over time so that it is not a dark or even quasidark state. The fidelity of this state reveals a deep at $t = 0$ at which the effective optical coupling approaches its maximal value, and after that the fidelity quickly revives and becomes as high as 0.76 in the region where the eigenstate turns into $|rr\rangle$, which means a large population on the double Rydberg excited state. In the intermediate case ($|V_{dd}| \sim \Omega$) we still find a large final population probability on $|rr\rangle$ but it is not due to an adiabatic passage. In this case there is not one single two-atom eigenstate which connects $|gg\rangle$ and $|rr\rangle$ as time going, but two different states $|E_g(t)\rangle$ and $|E_r(t)\rangle$ which are initially $|gg\rangle$ and finally $|rr\rangle$, respectively. Moreover, as marked by a circle in the figure, at the region near $t = 0$ there is an avoided crossing with a small energy gap between these two states, which may result in an efficient population transfer into the final state $|rr\rangle$. In the strong interaction case ($|V_{dd}| \gg \Omega$), $|E_g(t)\rangle$ and $|E_r(t)\rangle$ can no longer be connected directly by an avoided crossing. It instead requires another eigenstate to play the role as the bridge (see the black curve) between them. As the interaction strength $|V_{dd}|$ increases, more eigenstates are involved in the population transfer dynamics, which scatters the population and finally results in a poor final population on $|rr\rangle$.

Our simulation shows that the CMPAP is very effective in the Rydberg excitation with a weak Rydberg-Rydberg interaction. When the interaction is comparable with the

characteristic energy of the single atom, the adiabatic passage will be taken apart, but due to the small energy gap between the parts the Rydberg excitation efficiency of the CMPAP is still pretty robust. Finally the CMPAP will fail in an atomic system with dominant Rydberg-Rydberg interaction.

IV. INTERMEDIATE-STATE DETUNINGS

Recent researches illustrate that the optical detuning is an effective control knob in the Rydberg excitation mechanism. Typical examples include the antiblockade excitation with the detuning to the Rydberg state compensating for the frequency shift caused by the Rydberg-Rydberg interaction [32,33]

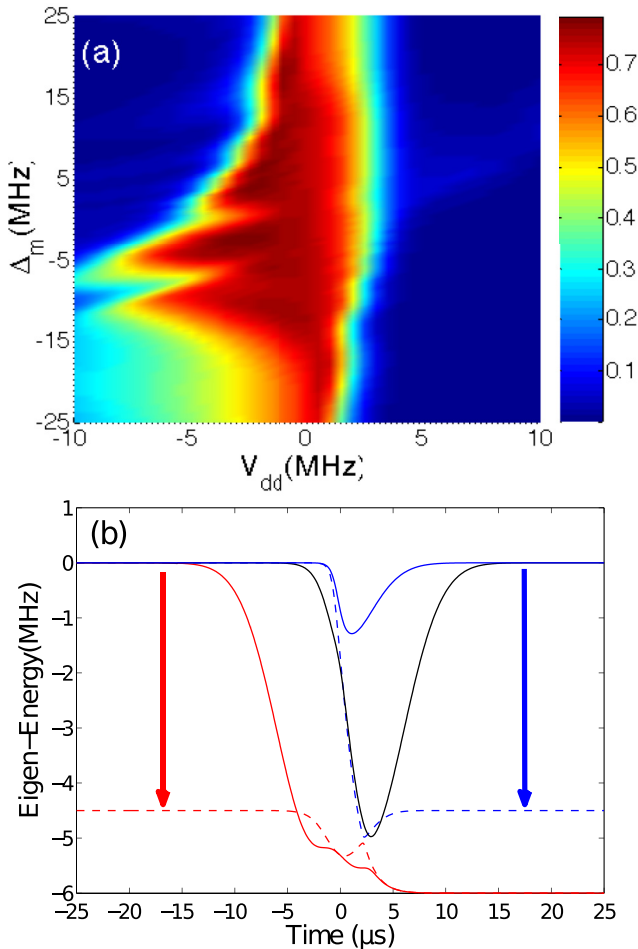


FIG. 5. (Color online) (a) Fidelity F_{rr}^{∞} standing for the probability of antiblockade atom pair production in the parameter space of V_{dd} and Δ_m with other parameters $\Omega_{me}^{\max} = \Omega_{gm}^{\max} = \Omega_{er}^{\max} = \Delta_e^{\max} = 5.0$ MHz, $T = 5 \mu\text{s}$, and $\gamma_{em} = \gamma_{mg} = 0.1$ MHz. (b) The time evolution of the energy of the eigenstates involved in the excitation. The solid curves are for the case $\Delta_m = 0$ and the dashed curves are for $\Delta_m = -4.5$ MHz. The Rydberg-Rydberg coupling strength $|V_{dd}|$ ($= 6$ MHz) is larger than the characteristic frequency Ω ($= 5$ MHz). Eigenstates $|E_g\rangle, |E_m\rangle$ (assistant eigenstate), and $|E_r\rangle$ are marked with blue (top solid and dashed) curves, black (middle solid) curve and red (bottom solid and dashed) curves, respectively. At time $t = -25 \mu\text{s}$, $|E_r\rangle \approx (|me\rangle - |em\rangle)/\sqrt{2}$; at $t = 25 \mu\text{s}$, $|E_g\rangle \approx (|gm\rangle - |mg\rangle)/\sqrt{2}$.

and a new two-atom adiabatic passage in the three-level atomic system with an appropriate detuning to the intermediate state as proposed by Rao and Mølmer [49]. In our four-level excitation scheme, although the detunings to the Rydberg state $|r\rangle$ and to the intermediate state $|e\rangle$ are both limited by the quasidark-state condition (5), the detuning to the other intermediate state $|m\rangle$ is a good candidate to optimize the effect of our CMPAP in the strong interaction case.

In Fig. 5(a) we show the fidelity $F_{rr}^{\infty} = \langle rr | \hat{\rho}_I | rr \rangle$, i.e., the final probability in state $|rr\rangle$ as a function of the detuning Δ_m and the interaction V_{dd} under the same parameters as used in Fig. 4. When $V_{dd} = 0$ it shows that the value of the fidelity is high and has less dependence on Δ_m . This can be attributed to the adiabatic passage based on the two-atom quasidark state $|D_4\rangle_I$ which does not involve the intermediate state $|m\rangle$. As the interaction V_{dd} becomes remarkable, things have certainly changed. High fidelity disappears in most of the area but still survives in lower left corner of the figure. For example, when $V_{dd} = -6.0$ MHz and $\Delta_m = -4.5$ MHz, $F_{rr}^{\infty} \approx 0.71$. Notice that when $\Delta_m = 0$ under the same value of the interaction V_{dd} the fidelity is very poor ($F_{rr}^{\infty} \approx 0.17$), the cause of which has been analyzed in Figs. 4(e) and 4(f); the original adiabatic passage is split and other eigenstates are involved in the excitation. Since in the strong interaction case the adiabatic passage is no longer based on the quasidark state, we attribute the improvement of the fidelity to the change of the two-atom eigenstates and their eigenenergies with a nonzero Δ_m . To display the change, in Fig. 5(b) we plot the time evolution of the eigenenergy spectrum of the involved eigenstates under the different values of Δ_m . The solid curves are for the case $\Delta_m = 0$, which is same as Fig. 4(e), while the dashed curves are for the case $\Delta_m = -4.5$ MHz. We find the eigenstate $|E_g(t)\rangle$ is initially $|gg\rangle$ and in the end almost equals $(|gm\rangle - |mg\rangle)/\sqrt{2}$ while the eigenstate $|E_r(t)\rangle$ changes from $(|em\rangle - |me\rangle)/\sqrt{2}$ to $|rr\rangle$ as time goes on. For the zero detuning case in the optical excitation region (about $t = 0$), there is a large energy gap between these two states and another eigenstate is involved to fill the passage to the double Rydberg excitation, which results in the final low fidelity. Since the evolution of both state $|E_g(t)\rangle$ and state $|E_r(t)\rangle$ involves the intermediate state $|m\rangle$, an appropriate value of detuning Δ_m may reduce the energy gap between them as identified by the dashed curves, and then the CMPAP recovers a high Rydberg excitation rate.

V. CONCLUSION

In summary, we present a highly efficient three-photon cascade excitation scheme for Rydberg atoms. In contrast to the typical two-photon excitation scheme with the STIRAP, our scheme is based on a novel quasidark eigenstate, which appears when the frequencies of the optical pulses are appropriately chirped. It enables a CMPAP between the ground state and the Rydberg state in the four-level atomic system. When the interatomic Rydberg interaction is considered, we find the two-atom quasidark state approach is only feasible when the interaction strength is relatively weak compared to the peak value of the optical Rabi frequencies. When the two become comparable or even when the Rydberg interaction is dominant,

the adiabatic excitation passage to the Rydberg atom pairs will be broken. Moreover, we find an appropriate optical detuning with respect to the intermediate state can reduce the energy gap between the fragment states of the broken adiabatic excitation passage. With its help the CMPAP gives rise to an enhancement of the production of antiblockade Rydberg atom pairs even in the strong Rydberg blockade regime.

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