Coherent population transfer and quantum entanglement generation involving a Rydberg state by stimulated Raman adiabatic passage

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We study a dilute sample of cold atoms to achieve efficient population transfer from a ground state to a Rydberg state. This sample is approximately divided into many independent microspheres containing only two atoms. Each pair of atoms in a microsphere may become quantum correlated via the dipole-dipole interaction characterized by a van der Waals potential. Our numerical results show that, by modulating detunings of a pump pulse and a Stokes pulse applied in the counterintuitive order, we can drive the dilute sample either into the blockade regime or into the antiblockade regime. In the blockade regime, only one atom is allowed to be coherently transferred into the Rydberg state in a microsphere can be efficiently excited into the Rydberg state, which is not accompanied by quantum entanglement. A second maximal entangled state may also be generated if we work between the blockade regime and the antiblockade regime. Note that the existence of a quasidark state is essential for exciting both atoms in a microsphere into the Rydberg state when the van der Waals potential is nonzero.

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I. INTRODUCTION

It was recently found that an ensemble of cold atoms can be partially transferred into a desired Rydberg state with a large principal quantum number via the resonant excitation of one or two coherent lasers [1-4]. This opens an avenue to study many interesting phenomena originating from the dipole-dipole interaction [5] of neighboring cold atoms, among which the dipole blockade effect has attracted great attention due to its potential applications in quantum information processing [6–9]. The dipole blockade effect allows only one atom to be resonantly excited into a certain Rydberg state within a small volume of the cold atomic sample, which is usually defined as a blockade sphere [7,8,10-14]. This is because the dipole-dipole interaction in the presence of a single Rydberg atom is so strong that the relevant Rydberg state of all other atoms within the blockade microsphere is shifted in energy to be far detuned from the driving fields. Note that the dipole-dipole interaction depends critically on the interatomic distance and thus can be safely neglected between the single Rydberg atom inside a blockade sphere and all atoms outside this blockade sphere. So far the dipole blockade effect has been extensively investigated to attain various entangled states [15], to engineer important quantum gates [16], to perform many-particle quantum simulation [17], to generate reliable single photons [6], etc.

To transfer more cold atoms into a desired Rydberg state, however, we have to avoid or overcome the dipole blockade effect by adopting an alternative strategy for the resonant laser excitation. This may be achieved by utilizing the transient Autler-Townes splitting of a single-atom population in a two-step excitation scheme to realize the dipole antiblockade effect [18,19]. The antiblockade effect usually manifests itself in the form of a greatly enhanced Rydberg excitation for a proper principal quantum number [18] or a suitable two-photon detuning [19]. On the other hand, we note that the technique of stimulated Raman adiabatic passage (STIRAP) has been well explored to efficiently transfer cold atoms from one ground state to another ground state in a controlled fashion [20–22]. In a typical STIRAP process, two laser pulses (a pump and a Stokes) are applied in the counterintuitive order to resonantly interact with a three-level Λ system so that it adiabatically evolves in a time-dependent dark state consisting of the two ground states [23,24]. To the best of our knowledge, the STIRAP technique has not been combined with the antiblockade effect to attain efficient population transfer from a ground state to a Rydberg state in the presence of a strong dipoledipole interaction. Moreover, quantum entanglement generation via the dipole-dipole interaction is seldom discussed together with the adiabatic population transfer into a Rydberg state.

Here we study a dilute sample of cold atoms driven by a pump field and a Stokes field into the ladder configuration involving a ground state, a normal excited state, and a Rydberg state. The sample is so dilute that there is only two cold atoms in a blockade sphere on average, i.e., a cold atom only interacts with its nearest partner via the dipole-dipole interaction described by a van der Waals (vdW) potential, which then allows us to adopt a two-body model as in Refs. [19,25]. Our numerical calculations show that, with the typical STIRAP technique, we can drive the cold atomic sample either into the blockade regime or into the antiblockade regime just by modulating detunings of the pump and Stokes fields. To be more specific, only one atom in a blockade sphere can be transferred into the Rydberg state when the pump and Stokes pulses are on exact two-photon resonance; both atoms in a blockade sphere may be simultaneously excited into the Rydberg state when the vdW potential is compensated by a suitable two-photon detuning. The latter

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FIG. 1. (Color online) (a) A dilute sample of cold atoms (blue circles) illuminated by two laser fields, denoted by the red zone. In this sample, only two atoms are contained in a blockade sphere of radius r_d on average. (b) Relevant states of atoms A and B in a blockade sphere. The two atoms are driven by a pump field Ω_p and a Stokes field Ω_s and may interact via a vdW potential V_d . (c) As in a typical STIRAP process, the pump and Stokes fields are modulated into two Gaussian pulses applied in the counterintuitive order.

referring to the antiblockade effect is attained, in fact, when the two-body system evolves in a quasidark state to which the normal excited state contributes little. In addition, we have identified two different entangled states with the largest negativity [26] after the STIRAP process, which benefits from either a perfect blockade effect or an imperfect antiblockade effect.

II. MODEL AND EQUATIONS

We consider here a cold atomic sample where only two atoms are contained in a blockade sphere of radius r_b on average, as shown in Fig. 1(a). This can be attained in experiment by greatly reducing the volume density, e.g., of cold ⁸⁷Rb atoms, to the order of $10^9 - 10^{10}$ cm⁻³ as in Ref. [27]. Figure 1(b) shows that each atom under consideration has a ground state $|1\rangle$ of lifetime longer than 1.0 ms, a normal excited state $|2\rangle$ of lifetime shorter than 1.0 μ s, and a Rydberg state $|r\rangle$ of lifetime about 100 μ s. The three states are driven into the ladder configuration by two laser fields (the pump and the Stokes) with Rabi frequencies Ω_p and Ω_s , respectively. The pump (Stokes) field may also be detuned in frequency from the dipole-allowed transition $|1\rangle \leftrightarrow |2\rangle$ $(|2\rangle \leftrightarrow |r\rangle)$ by $\Delta_p = \omega_p - \omega_{21}$ ($\Delta_s = \omega_s - \omega_{r2}$). The two atoms labeled A and B in a blockade sphere, if excited into the Rydberg state $|r\rangle$, will experience a large vdW potential V_d and therefore become quantum correlated due to the dipole-dipole interaction [19,27,28]. In this case, we can write down the two-body Hamiltonian

$$H_{AB} = H_A + H_B + H_{\text{int}},\tag{1}$$

with

$$H_{A(B)} = \hbar \Delta_p |2_{A(B)}\rangle \langle 2_{A(B)}| + \hbar \Delta |r_{A(B)}\rangle \langle r_{A(B)}| + \hbar [\Omega_p |2_{A(B)}\rangle \langle 1_{A(B)}| + \Omega_s |r_{A(B)}\rangle \langle 2_{A(B)}| + \text{H.c.}]$$
(2)

being the Hamiltonian of atom A (B) and

$$H_{\rm int} = \hbar V_d |r_A\rangle |r_B\rangle \langle r_B |\langle r_A| \tag{3}$$

being the Hamiltonian shared by both of them. In the above, we have defined $\Delta = \Delta_p + \Delta_s$ as the two-photon detuning between state $|1\rangle$ and state $|r\rangle$.

The optical response of both atoms in a blockade sphere can be examined by solving the master equation of two-body density operator ρ_{AB} ,

$$i\hbar\frac{\partial\rho_{AB}}{\partial t} = [H_{AB}, \rho_{AB}], \qquad (4)$$

where the population decay rates and the coherence dephasing rates should be phenomenologically added. Equation (4), when extended into the atomic system shown in Fig. 1(b), turns into a set of dynamical equations for 9×9 density matrix elements $\rho_{ij,ij}$, with the first *i* and *j* denoting states of atom A and the second *i* and *j* denoting states of atom B. To examine the optical response per atom, we can compute the partial traces of $\rho_{ij,ij}$ with respect to both atoms A and B to attain $\rho_{ij} = (\rho_{ij}^A + \rho_{ij}^B)/2 = [\text{Tr}^{(B)}(\rho_{ij,ij}) + \text{Tr}^{(A)}(\rho_{ij,ij})]/2.$

In the following, we only consider the case where the pump field and the Stokes field are modulated into two Gaussian pulses separated by a time delay τ so that we have

$$\Omega_p(t) = \Omega_p^{\max} e^{-(t-\tau/2)^2/T_p^2},$$

$$\Omega_s(t) = \Omega_s^{\max} e^{-(t+\tau/2)^2/T_s^2},$$
(5)

with $2T_{p,s}$ being the time widths and $\Omega_{p,s}^{\max}$ being the peak Rabi frequencies. In a typical STIRAP process, the two Gaussian light pulses are required to be applied in the counterintuitive order characterized by $\tau \approx T_p + T_s > 0$, which is necessary to guarantee the adiabaticity of population transfer between two ground states and will also be adopted here.

Purposely setting $V_d = 0$, we find that Eq. (1) has an eigenstate of zero eigenvalue, i.e., a dark state,

$$|D_{1}(t)\rangle = [\cos\theta(t)|1_{A}\rangle + \sin\theta(t)|r_{A}\rangle]$$

$$\otimes [\cos\theta(t)|1_{B}\rangle + \sin\theta(t)|r_{B}\rangle], \qquad (6)$$

with $\tan \theta(t) = \Omega_p(t) / \Omega_s(t)$ in the case of $\Delta = 0$. Thus it is possible to achieve the complete population transfer from state $|1\rangle$ to state $|r\rangle$ for both atoms in a blockade sphere by rotating the dark state $|D_1(t)\rangle$. In the presence of dipole-dipole interaction ($V_d \neq 0$), however, the dark state corresponding to $\Delta = 0$ turns into

$$|D_2(t)\rangle = \frac{\cos 2\theta(t)|11\rangle - \sin 2\theta(t)[|1r\rangle + |r1\rangle]/2 + \sin^2 \theta(t)|22\rangle}{\sqrt{\cos^4 \theta(t) + 2\sin^4 \theta(t)}},\tag{7}$$

as in Ref. [15]. Thus it seems impossible to simultaneously transfer atom A and atom B into state $|r\rangle$ from state $|1\rangle$ via a STIRAP process because state $|2\rangle$ is involved in the dark state $|D_2(t)\rangle$.

To achieve the complete population transfer from state $|1\rangle$ to state $|r\rangle$ for both atoms in a blockade sphere, one feasible way is to find the following eigenstate:

$$|D_{3}(t)\rangle = c_{1}(t)|11\rangle + c_{2}(t)|1r\rangle + c_{3}(t)|r1\rangle + c_{4}(t)|rr\rangle + c_{5}(t)|22\rangle + c_{6}(t)|12\rangle + c_{7}(t)|21\rangle + c_{8}(t)|2r\rangle + c_{9}(t)|r2\rangle,$$
(8)

constrained by $|c_1(t)|^2 + |c_2(t)|^2 + \dots + |c_9(t)|^2 = 1$ and $[|c_1(t)|^2 + |c_2(t)|^2 + |c_3(t)|^2 + |c_4(t)|^2] \gg [|c_5(t)|^2 + |c_6(t)|^2 + |c_7(t)|^2 + |c_8(t)|^2 + |c_9(t)|^2]$. The existence of $|D_3(t)\rangle$ can be verified by numerically solving the secular equation of the two-body Hamiltonian H_{AB} to attain the smallest absolute eigenvalue $|\lambda_{\min}(t)|$, with its mean value

$$\overline{\lambda} = \frac{\int_{-2T_s - \tau/2}^{+2T_p + \tau/2} |\lambda_{\min}(t)| dt}{2T_s + 2T_p + \tau}$$

being very close to zero. This eigenstate may be regarded as a quasidark state because it approximately excludes the intermediate state $|2\rangle$ and could be attained when $\Delta \approx -V_d/2$, i.e., the two-photon detuning compensates half of the vdW potential.

When atoms A and B in a blockade sphere are in the quasidark state $|D_3(t)\rangle$, they should be quantum correlated to a certain extent as a result of dipole-dipole interaction. To evaluate the relevant entanglement, we can resort to the negativity [26], defined as

$$\mathcal{N}(\rho_{AB}) = \sum_{i} |\mu_{i}|, \tag{9}$$

with ρ_{AB} being the density matrix corresponding to $|D_3(t)\rangle$ and μ_i being a negative eigenvalue of the partial transposition of ρ_{AB} . Note that the possible value of negativity \mathcal{N} is between 0.0 and 0.5, with $\mathcal{N} = 0$ denoting the complete separability [29] and $\mathcal{N} = 0.5$ denoting the maximal entanglement.

III. RESULTS AND DISCUSSION

We first plot in Fig. 2 the single Rydberg excitation ρ_{rr} and the double Rydberg excitation $\rho_{rr,rr}$ at time $t = 4 \ \mu s$ (just after the STIRAP process) as a function of the single-photon detuning Δ_p and the two-photon detuning Δ . To be more specific, $\rho_{rr,rr} = \langle rr | \rho_{AB} | rr \rangle$ is the total probability of both atoms and $\rho_{rr} = (\rho_{rr}^A + \rho_{rr}^B)/2$ is the average probability of one atom being populated into the Rydberg state $|r\rangle$ in a blockade sphere of radius r_b . In the absence of a vdW potential, we always have $\rho_{rr,rr} = \rho_{rr}^2$ because $\rho_{rr}^A \equiv \rho_{rr}^B$, so that it is enough to examine only ρ_{rr} as given in Fig. 2(a). As expected, the atomic population can be efficiently transferred from the ground state $|1\rangle$ into the Rydberg state $|r\rangle$ near the two-photon resonance $\Delta = 0$ via the STIRAP technique. With the increase of single-photon detuning Δ_p , the region of two-photon detuning Δ for achieving efficient single Rydberg excitation $(\rho_{rr} \approx 1)$ becomes narrower and narrower, producing a *fanlike* pattern. When the vdW potential is nonzero, however, we may have $\rho_{rr,rr} \neq \rho_{rr}^2$ because ρ_{rr}^A could be very different from ρ_{rr}^B



FIG. 2. (Color online) (a) Single Rydberg excitation ρ_{rr} as a function of detunings Δ_p and Δ in the absence of a vdW potential V_d . (b) Single Rydberg excitation ρ_{rr} as a function of detunings Δ_p and Δ in the presence of a vdW potential V_d . (c) Double Rydberg excitation $\rho_{rr,rr}$ as a function of detunings Δ_p and Δ in the presence of a vdW potential V_d . (c) Double Rydberg excitation $\rho_{rr,rr}$ as a function of detunings Δ_p and Δ in the presence of a vdW potential V_d . The relevant parameters are $\Gamma_{21} = 1.5$ MHz, $\Gamma_{r2} = 0.05$ MHz, $V_d = 100$ MHz, $\Omega_p^{max} = \Omega_s^{max} = 100$ MHz, $T_p = T_s = 1.0 \ \mu$ s, and $\tau = 2.0 \ \mu$ s. In numerical calculations, the Stokes pulse is turned on at time $t = -2T_s - \tau/2$, while the pump pulse is turned off at time $t = +2T_p + \tau/2$.

as a result of the dipole blockade effect. It is why we have plotted ρ_{rr} and $\rho_{rr,rr}$ in Figs. 2(b) and 2(c), respectively. From Fig. 2(b), we can see that the fanlike region indicating efficient single Rydberg excitation ($\rho_{rr} \approx 1$) moves left, with its center located at $\Delta = -V_d/2$. This implies, in fact, an antiblockade effect, as discussed in Ref. [19], where two continuous-wave fields are applied instead. Note also that the single Rydberg excitation ρ_{rr} reduces to less than 0.5 near the two-photon resonance $\Delta = 0$ as a signature of the dipole blockade effect. The existence of both blockade and antiblockade effects is further verified by Fig. 2(c), where we find $\rho_{rr,rr} \approx 1$ in a fanlike region centered at $\Delta = -V_d/2$ but $\rho_{rr,rr} \approx 0$ near the two-photon resonance $\Delta = 0$.

To attain efficient double Rydberg excitation $\rho_{rr,rr} \approx 1$, we should try our best to avoid populating the intermediate state $|2\rangle$ with a large decay rate Γ_{21} at any time. That is, the atomic system under consideration should evolve in a quasidark state $|D_3(t)\rangle$ approximately excluding the contribution of state $|2\rangle$ during the STIRAP process. In Fig. 3, we plot the mean value $\overline{\lambda}$ as a function of the single-photon detuning Δ_p and the two-photon detuning Δ . It is clear that $\overline{\lambda}$ is very small when $\Delta \approx -V_d/2 = -50$ MHz, so we can claim that the efficient double Rydberg excitation in Fig. 2(c) is attained when the two-body system evolves in a quasidark state $|D_3(t)\rangle$. To gain deeper insight, we plot populations $\rho_{11,11}$, $\rho_{rr,rr}$, and $\rho_{22,22}$ in Fig. 4(a) and populations ρ_{11} , ρ_{rr} , and ρ_{22} in Fig. 4(b) as a function of time t with $\Delta_p = 200$ MHz and $\Delta = -50$ MHz. As we can see, the population transfer from state $|1\rangle$ to state $|r\rangle$



FIG. 3. (Color online) Mean value $\overline{\lambda}$ of the smallest absolute eigenvalue $|\lambda_{\min}(t)|$ as a function of detunings Δ_p and Δ in the presence of a vdW potential $V_d = 100$ MHz. Other parameters are the same as in Fig. 2.

is approximately adiabatic, i.e., without essentially involving state $|2\rangle$ (the only source of energy dissipation). This once again verifies that the efficient double Rydberg excitation $\rho_{rr,rr} \approx 1$ is attained when the two-body system evolves in a quasidark state $|D_3(t)\rangle$.

Based on the above discussions, we can say that the vdW potential V_d may be well compensated by introducing an appropriate two-photon detuning $\Delta \approx -V_d/2$ to convert the blockade effect into the antiblockade effect. Thus simply modulating detunings of the pump and Stokes pulses, we can let the two-body system work either in the blockade region or in the antiblockade region. Note, in particular, that the antiblockade region extends a little into the blockade region (see Fig. 2) because Ω_p^{max} and Ω_s^{max} are as large as V_d . If we choose $\Omega_p^{\text{max}} \approx \Omega_s^{\text{max}} \ll V_d$, it is viable to well separate the blockade region and the antiblockade region.

As mentioned in the last section, the population transfer from state $|1\rangle$ to state $|r\rangle$ may be accompanied by the



FIG. 4. (Color online) Population transfer dynamics in the antiblockade regime denoted by $\Delta_p = 200$ MHz, $\Delta = -50$ MHz, and $V_d = 100$ MHz. The black solid, red dashed, and blue dotted curves refer to $\rho_{11,11}$, $\rho_{rr,rr}$, and $\rho_{22,22}$ in (a) and ρ_{11} , ρ_{rr} , and ρ_{22} in (b), respectively. Other parameters are the same as in Fig. 2.



FIG. 5. (Color online) Single Rydberg excitation ρ_{rr} (red dashed line), double Rydberg excitation $\rho_{rr,rr}$ (black solid line), and twobody negativity \mathcal{N} (blue dotted line) as a function (a) of two-photon detuning Δ with $\Delta_p = 200$ MHz, (b) of single-photon detuning Δ_p with $\Delta = -50$ MHz, and (c) of single-photon detuning Δ_p with $\Delta = 0.0$ MHz. Other parameters are the same as in Fig. 2.

generation of quantum entanglement in the case of $V_d \neq 0$. In Fig. 5(a) we plot the single Rydberg excitation ρ_{rr} , the double Rydberg excitation $\rho_{rr,rr}$, and the two-body negativity \mathcal{N} as a function of the two-photon detuning Δ with a fixed single-photon detuning $\Delta_p = 200$ MHz. It is clear that we have $\rho_{rr} \approx \rho_{rr,rr} \approx 1.0$ and $\mathcal{N} \approx 0.0$ at $\Delta = -V_d/2 = -50$ MHz, which means that both atoms can be transferred from state $|1\rangle$ into state $|r\rangle$ when the vdW potential is compensated by a suitable two-photon detuning. In this case, atoms A and B should be in the separable state $|rr\rangle$ after the STIRAP process. In addition, we have $\rho_{rr} \approx 0.5$, $\rho_{rr,rr} \approx 0.0$ and $\mathcal{N} \approx 0.5$ at $\Delta = 0$, which means that only one atom can be transferred from state $|1\rangle$ into state $|r\rangle$ when the pump and Stokes pulses are kept on two-photon resonance. In this case, atoms A and B are expected to be in the maximal entangled state $[|1r\rangle + |r1\rangle]/\sqrt{2}$ after the STIRAP process. We also find that $ho_{rr} pprox
ho_{rr,rr} pprox 0.5$ and $\mathcal{N} pprox 0.5$ at $\Delta pprox -88$ MHz, which can be attained only if the two atoms are in the maximal entangled state $[|11\rangle + |rr\rangle]/\sqrt{2}$. Thus it is possible to prepare two different maximal entangled states involving a Rydberg state just by modulating detunings of the pump and Stokes pulses applied in the counterintuitive order.

In Figs. 5(b) and 5(c) we further plot the single Rydberg excitation $\rho_{rr,rr}$, the double Rydberg excitation $\rho_{rr,rr}$, and the two-body negativity \mathcal{N} as a function of the single-photon detuning Δ_p . The two-photon detuning Δ is set to be -50 MHz

in Fig. 5(b) but 0.0 MHz in Fig. 5(c). Figure 5(b) shows that we always have $\rho_{rr} \approx \rho_{rr,rr}$, although their values depend critically on Δ_p . This means that the two atoms in a blockade sphere have similar probabilities of being transferred from state $|1\rangle$ into state $|r\rangle$, which is a typical feature of the antiblockade effect. In particular, we can achieve the maximal negativity $\mathcal{N} \approx 0.5$ at $\Delta_p \approx \pm 680$ MHz and therefore the maximal entangled state $[|11\rangle + |rr\rangle]/\sqrt{2}$ due to the imperfect antiblockade effect. Figure 5(c) shows that we have $\rho_{rr} \approx 0.5$ and $\rho_{rr,rr} \approx 0$ when $\Delta_p < -50$ MHz or $\Delta_p > 250$ MHz. This means that only one atom can be transferred from state $|1\rangle$ into state $|r\rangle$ as a result of the dipole blockade. In this case, atoms A and B are in the maximal entangled state $[|1r\rangle + |r1\rangle]/\sqrt{2}$ as proved by $\mathcal{N} = 0.5$. When $\Delta_p \approx 0$, however, all three quantities ρ_{rr} , $\rho_{rr,rr}$, and \mathcal{N} are approaching zero because state $|2\rangle$ is essentially populated. In addition, we have $\rho_{rr} \approx \rho_{rr,rr} \approx 1$ and $\mathcal{N} \approx 0.0$ at $\Delta_p \approx 200$ MHz because the antiblockade region extends into the blockade region, as shown in Fig. 2.

IV. CONCLUSIONS

In summary, we have demonstrated a feasible way to achieve efficient population transfer from the ground state $|1\rangle$ into the Rydberg state $|r\rangle$ without essentially populating the excited state $|2\rangle$ in a cold atomic sample. The sample is assumed to be so dilute that each atom excited into the Rydberg state interacts only with its nearest partner via a vdW potential, which then allows us to divide the sample into a lot of microspheres containing only two atoms. We find by numerical calculations that the population transfer from the ground state $|1\rangle$ to the Rydberg state $|r\rangle$ is very sensitive to detunings of the two laser pulses are on two-photon resonance, only a half of all cold atoms can be transferred into

the Rydberg state as a result of the blockade effect. But when the two-photon detuning compensates the vdW potential, all atoms may be efficiently transferred into the Rydberg state as a result of the antiblockade effect. This is achieved because the two-atom system has a quasidark state approximately excluding the excited state $|2\rangle$ and can adiabatically evolve from state $|11\rangle$ into state $|rr\rangle$ without generating $|12\rangle$, $|21\rangle$, $|22\rangle$, $|2r\rangle$, and $|r2\rangle$ at any time. In the blockade regime, the two-atom system evolves in fact into a maximal entangled state $[|1r\rangle + |r1\rangle]/\sqrt{2}$ as verified by $\mathcal{N} \approx 0.5$. In the antiblockade regime, the two-atom system goes into state $|rr\rangle$ with $\mathcal{N} = 0.0$ instead. We also find that, if the two-photon detuning is modulated into an imperfect antiblockade regime, the twoatom system may also go into another maximal entangled state $[|1r\rangle + |r1\rangle]/\sqrt{2}$, as verified by the largest negativity $\mathcal{N} \approx 0.5$.

We expect that an antiblockade regime benefiting the efficient population transfer between a ground state and a Rydberg state can also be attained in the case where three or more atoms are inside a blockade sphere. For such manybody systems, we may attain more complicated multipartite quantum entangled states by working between the blockade regime and the antiblockade regime.

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