Quantum Gates and Multiparticle Entanglement by Rydberg Excitation Blockade and Adiabatic Passage

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We propose to apply stimulated adiabatic passage to transfer atoms from their ground state into Rydberg excited states. Atoms a few micrometers apart experience a dipole-dipole interaction among Rydberg states that is strong enough to shift the atomic resonance and inhibit excitation of more than a single atom. We show that the adiabatic passage in the presence of this interaction between two atoms leads to robust creation of maximally entangled states and to two-bit quantum gates. For many atoms, the excitation blockade leads to an effective implementation of collective-spin and Jaynes-Cummings–like Hamiltonians, and we show that the adiabatic passage can be used to generate collective $J_x = 0$ eigenstates and Greenberger-Horne-Zeilinger states of tens of atoms.

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Entanglement is a central property of quantum mechanical systems and an important resource for applications in quantum information and quantum metrology. Entanglement is a subject of broad theoretical interest, and experimental implementations in various systems have been demonstrated. One scheme for creation of entanglement and quantum gates between neutral atoms utilizes the large dipole moments of highly excited Rydberg states resulting in a dipole-dipole interaction which is strong enough to shift the atomic energy levels and prevent more than one atom from being excited to the Rydberg state by a resonant laser field [1,2]. In alkali atoms Rydberg states with principal quantum number $n > 70$ have lifetimes $\gtrsim 100$ μs and experience a dipole-dipole interaction strength, $E/\hbar$, above $100 \times 2\pi$ MHz when atoms are separated less than 5 μm [3]. The accompanying suppression of excitation for n up to 80 [4,5] and the influence of the excitation blockade on coherent collective dynamics [6] were observed in cold gases.

We propose to combine the Rydberg blockade mechanism with the rapid adiabatic laser pulse sequence known from stimulated Raman adiabatic passage (STIRAP) [7–10]. We consider atoms with two lower levels and a Rydberg level in a ladder structure coupled by two resonant laser fields with Rabi frequencies Ω_1 and Ω_r as shown in Fig. 1(a). The STIRAP process applies the "counterintuitive" pulse sequence of Fig. 1(b) to transfer population from the ground state $|1\rangle$ to a highly excited Rydberg state $|r\rangle$ by adiabatically following [11] a dark state which never populates the intermediate state $|2\rangle$. If we assume a relative phase, $\phi_r(t)$, between the Rabi frequencies Ω_1 and Ω_r, a single atom exposed to the fields follows the dark state superposition, $|D_1\rangle = \cos\theta|1\rangle - \sin\theta e^{i\phi_r}|r\rangle$, where $\tan\theta = \Omega_1/\Omega_r$ expresses the relative strength of the two laser fields. Since the dark state has zero energy, there is no dynamic phase accumulated during the process, but if ϕ_r varies, $|D_1\rangle$ acquires the geometric phase, $\gamma_1 = -\int \sin^2\theta d\phi_r$ [12,13]. A controlled geometric phase shift on state $|1\rangle$ can be implemented if the phase difference between the fields evolves between the STIRAP pulses and a second inverted set of pulses, shown in Fig. 1(c). For atoms with a further logical state $|0\rangle$ such a pulse sequence enables robust geometric phase gates and other one-bit gates [13].

With more atoms the Rydberg blockade comes into play, but we may still identify dark states. For two atoms, initially in the product state $|11\rangle$ and subject to the same interaction with the two resonant laser fields Ω_1 and Ω_r, the evolution preserves the symmetry under interchange of atoms, and hence it is sufficient to consider the Hamiltonian in the symmetric two-atomic basis $\{|11\rangle, \frac{1}{\sqrt{2}} \times (|1r\rangle + |r1\rangle), \frac{1}{\sqrt{2}}(|12\rangle + |21\rangle), |rr\rangle, \frac{1}{\sqrt{2}}(|2r\rangle + |r2\rangle), |22\rangle\}$,

$$H(t) = \frac{\hbar}{2}\begin{bmatrix} 0 & 0 & \sqrt{2}\Omega_1^* & 0 & 0 & 0 \\ 0 & 0 & \Omega_r^* & 0 & \Omega_1^* & 0 \\ \sqrt{2}\Omega_1 & \Omega_r & 0 & 0 & 0 & \sqrt{2}\Omega_1^* \\ 0 & 0 & 0 & 2E/\hbar & \sqrt{2}\Omega_r^* & 0 \\ 0 & \Omega_1 & 0 & \sqrt{2}\Omega_r & 0 & \sqrt{2}\Omega_r^* \\ 0 & 0 & \sqrt{2}\Omega_1 & 0 & \sqrt{2}\Omega_r & 0 \end{bmatrix}, \tag{1}$$

where E denotes the energy shift of the state $|rr\rangle$ due to the dipole-dipole interaction. This Hamiltonian has one dark state for the two-atom system

$$|D_2\rangle = \frac{1}{\sqrt{\cos^4\theta + 2\sin^4\theta}}[(\cos^2\theta - \sin^2\theta)|11\rangle - \cos\theta\sin\theta e^{i\phi_r}(|1r\rangle + |r1\rangle) + \sin^2\theta|22\rangle]. \tag{2}$$

We first assume $e^{i\phi_r} = 1$ and apply the counterintuitive pulse sequence of Fig. 1(b). Initially $\cos\theta = 1$ and the system is in $|D_2\rangle = |11\rangle$. Adiabaticity ensures that we remain in $|D_2\rangle$ and after the pulses $\sin\theta = 1$ and the system is in

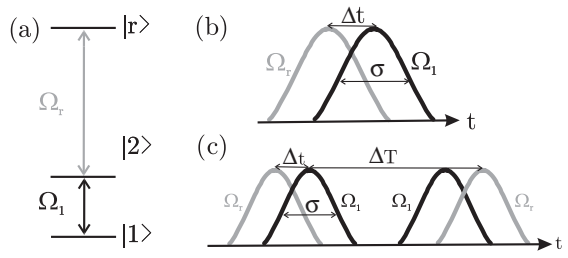

FIG. 1. (a) Three-level ladder system and two laser fields with Rabi frequencies Ω_1 and Ω_r. (b) STIRAP pulse sequence. The FWHM of each pulse is σ and the delay between pulses within one process is Δt. (c) Pulse sequence consisting of two STIRAP processes separated by ΔT in time.

$$|D_2\rangle = \frac{1}{\sqrt{2}}(-|11\rangle + |22\rangle). \tag{3}$$

Equation (2) shows that while the STIRAP process in the single atom case ensures that $|2\rangle$ is never populated, due to the Rydberg blockade the pair of atoms is adiabatically steered into a state populating $|22\rangle$. Moreover, (3) is a maximally entangled state of the two atoms, generated robustly irrespective of the precise pulse shapes, field strengths, and the precise value of the Rydberg interaction energy. Figure 2(a) shows the evolution of populations for realistic experimental parameters, obtained from a numerical propagation of the Schrödinger equation (see caption). The decay due to the finite lifetime of the $|r\rangle$ states, populated during the process, is incorporated as a decay out of the system, whereas the $|1\rangle$ and $|2\rangle$ states are treated as long-lived hyperfine sublevels. This implies that if the levels are coupled by single photon transitions, the field coupling $|1\rangle$ and $|2\rangle$ will have a frequency in the radio

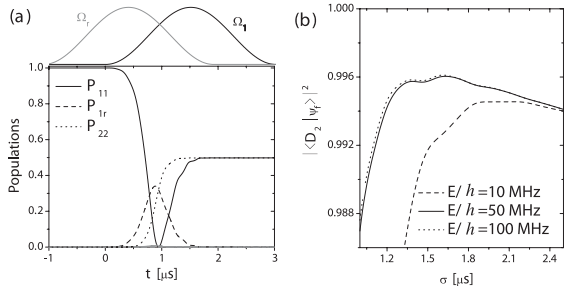

FIG. 2. (a) Time evolution of the population in $|11\rangle$, $\frac{1}{\sqrt{2}}(|1r\rangle + |r1\rangle)$ and $|22\rangle$. (b) Fidelity of the entanglement creation as a function of pulse width for different values of dipole-dipole interaction E. $|\psi_f\rangle$ was found by propagating the Schrödinger equation (1) with initial state $|11\rangle$. We show the norm square of the overlap with the target state $|D_2\rangle$. The laser pulses are modeled by $\sin^2$ pulses, $\Omega_j(t) = \Omega_{\max,j}\sin^2(\frac{\pi(t-t_{sj})}{2\sigma})$ for $t_{sj} < t < t_{sj} + 2\sigma$, where t_{sj} is the instant of time when the pulse starts and σ the FWHM. The simulations are made with parameters $\Omega_{\max,1}/2\pi = \Omega_{\max,2}/2\pi = 10$ MHz, $\Delta t = 1.1$ μs, lifetime of the $|r\rangle$ state, $\tau_r = 100$ μs, and in (a) $\sigma = 1.5$ μs and $E/\hbar = 100 \times 2\pi$ MHz.

frequency range, while the laser field coupling $|2\rangle$ and $|r\rangle$ has a wavelength ≈ 300 nm for Cs and Rb. Alternatively the couplings can be obtained with two-photon transitions involving four optical fields. By appropriately choosing the laser detunings to compensate for the time-dependent ac Stark shifts during the pulses, the corresponding five level system of equations with two intermediate optically excited states can be effectively reduced to the present three-level ladder system.

To investigate the criterion of adiabaticity and the role of the Rydberg interaction energy E, in Fig. 2(b), we show the fidelity of the creation of the entangled state, $F = |\langle D_2|\psi_f\rangle|^2$, where $|\psi_f\rangle$ is the final state calculated by propagation of the state vector with the time-dependent Hamiltonian (1). The simulations show that, as long as E is sufficiently large to block the population of states with more than one Rydberg excitation, the exact value of E is not important. With Rabi frequencies $\Omega_{\max,1}/2\pi = \Omega_{\max,r}/2\pi = 10$ MHz, a Rydberg energy shift of $E/\hbar = 50 \times 2\pi$ MHz is sufficient. The time window where $|r\rangle$ is populated is determined by the pulse width and it is desirable to use the smallest possible width that does not violate adiabaticity, yielding a total time of entanglement generation of 3–4 μs, which is short compared with the radiative lifetime of the highly excited Rydberg state $\gtrsim 100$ μs.

The two-atom entanglement scheme can be modified to create a two-qubit phase gate. To this end we apply a second STIRAP sequence with phase shifted Rabi frequencies and the pulses in reversed order [cf. Fig. 1(c)] that transfers the population back to the $|11\rangle$. With a nonvanishing relative phase $\phi_r(t)$ between Ω_1 and Ω_r the dark state (2) acquires the geometric phase [12]

$$\gamma_2 = -\int \frac{\cos^2\theta\sin^2\theta}{\cos^4\theta + 2\sin^4\theta} d\phi_r.$$

When we supplement the atomic level scheme with another qubit state $|0\rangle$, which is uncoupled from the STIRAP pulses, the gate performed by the two STIRAP processes amounts to multiplication of all two-bit register states by phase factors corresponding to a controlled two-qubit phase gate with phase $\Delta\varphi = \gamma_2 - 2\gamma_1$. γ_2 is only acquired when the pulses overlap, while γ_1 is acquired between the two pulse sequences, and $\Delta\varphi$ can, e.g., be controlled by adjusting ΔT.

We now show that when more than two atoms in $|1\rangle$ are subject to the STIRAP pulse sequence they also become entangled. Provided all atoms are localized within a region of a few μm, the transition towards the Rydberg states is restricted to the coupling of collective states with either zero or one Rydberg atom and we implement this by truncating the basis so it only includes states with zero or one atom in the $|r\rangle$ state. We write the symmetric basis states of the system as $\{|n_1, n_2 = N - n_1, 0\rangle, |n_1, n_2 = N - n_1 - 1, 1\rangle\}$, where N is the total number of atoms, n_1 and n_2 the number of atoms in $|1\rangle$ and $|2\rangle$, and 0 or 1 indicates if $|r\rangle$ is populated with zero or a single atom. The

basis consists of $N+1$ states with no Rydberg excitation and N with a single excitation. The interaction with the radiation fields Ω_1 and Ω_r, $\sum_{j=1}^{N} -\frac{1}{2}(\Omega_1(t)|2\rangle_j\langle 1| + \Omega_r(t)|r\rangle_j\langle 2| + \text{h.c.})$, can now be rewritten

$$H(t) = H_{J_x}(t) + H_{\text{JC}}(t), \tag{4}$$

with variable strengths, representing the coupling by the fields driving the lower and the upper transition, respectively. The dynamics of the lower levels can be rewritten in a collective spin description, and in the accompanying Schwinger oscillator description,

$$H_{J_x}(t) = -\hbar\Omega_1(t)J_x(t) = -\tfrac{1}{2}\hbar\Omega_1(t)(a_1^\dagger a_2 + a_1 a_2^\dagger), \tag{5}$$

where $a_i^{(\dagger)}$ are annihilation (creation) operators, with conventional oscillator commutator relations, for the number of atoms in $|i\rangle$. The upper transition is described by

$$H_{\text{JC}}(t) = -\tfrac{1}{2}\hbar\Omega_r(t)(a_2\sigma^+ + a_2^\dagger\sigma^-), \tag{6}$$

where we use Pauli matrices, σ^+ and σ^-, to describe the transfer of population between states with zero and one Rydberg excitation in the atomic ensemble. This Hamiltonian is the quantum optical Jaynes-Cummings (JC) Hamiltonian, introduced originally to describe the resonant interaction between a two-level atom and quantized light [14]. JC dynamics has been implemented in strong coupling cavity QED experiments, where nonclassical states of light, such as Fock states and quantum superposition states, are produced efficiently [15]. H_{JC} also describes the motion of laser driven trapped ions where it has been used to generate various nonclassical states [16,17]. Following these proposals, JC dynamics is sufficient to produce a variety of interesting states of an atomic ensemble by the Rydberg blockade.

The Hamiltonian in (4) has a dark state with zero valued energy eigenvalue throughout the STIRAP process. This follows because the "parity" operator $\Theta = (-1)^{\hat{n}_2}$, which inverts the sign of the operators a_2 and $a_2^\dagger$, anticommutes with H, $H\Theta = -\Theta H$. Any eigenstate $|\psi\rangle$ of H with energy eigenvalue $\mathcal{E}$ then has a partner $\Theta|\psi\rangle$ with eigenvalue $-\mathcal{E}$ and the energy eigenvalue spectrum is symmetric around zero. The number of eigenstates is odd because N atoms induce $N+1$ different states with no atoms in $|r\rangle$ and N states with one atom in $|r\rangle$, and hence there must always be a state with eigenvalue zero. The full curves in Fig. 3 show the 13 energy eigenvalues for 6 atoms found from a numerical diagonalization of Eq. (4), and they clearly confirm the existence of the dark state throughout the pulse sequence.

The STIRAP process starts with $\Omega_1 = 0$ and hence initially $H(t) = H_{\text{JC}}(t) = -\frac{1}{2}\hbar\Omega_r(t)(a_2\sigma^+ + a_2^\dagger\sigma^-)$ with the eigenvalues $\{0, \pm\frac{1}{2}\hbar\Omega_r(t)\sqrt{n_2+1}\}$, as shown with the squares in Fig. 3. The dark state is $|n_1 = N, n_2 = 0, n_r = 0\rangle$ with all atoms in state $|1\rangle$. Adiabaticity ensures that we remain in the dark state of the full Hamiltonian, and when Ω_1 is turned on and Ω_r reduced, the system ends up in the

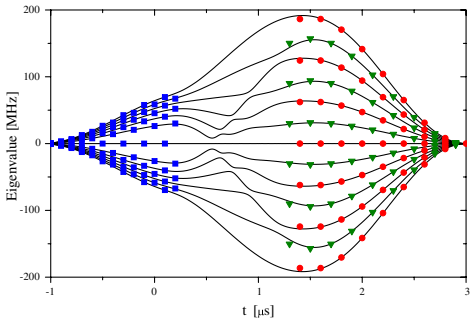

FIG. 3 (color online). Eigenvalues of $H(t)/\hbar$ of (4) for $N = 6$. Black solid curves show the results of a numerical diagonalization of the full Hamiltonian. The squares on the left show the analytical eigenvalues of $H_{\text{JC}}(t)/\hbar$ and the bullets and triangles on the right show the analytical eigenvalues of $H_{J_x}(t)/\hbar$ for zero and one Rydberg excitation, respectively. Parameters used are $\Omega_{\max,1}/2\pi = 10$ MHz, $\Omega_{\max,r}/2\pi = 10$ MHz, $\sigma = 1.5$ μs, $\Delta t = 1.1$ μs.

dark state of $H(t) = H_{J_x}(t) = -\hbar\Omega_1(t)J_x$. In this state, the system can have either no or a single Rydberg excitation leaving $K = N$ or $K = N-1$ atoms in the $|1\rangle$ and $|2\rangle$ states. The eigenvalues of $-\Omega_1(t)J_x$ are $-\Omega_1(t)\times\{-K/2, -K/2+1, \ldots, K/2-1, K/2\}$ and $\mathcal{E} = 0$ occurs for K even. Figure 3 shows the eigenvalues of $-\Omega_1(t)J_x$ when $|r\rangle$ is not populated (bullets) and when one atom is excited to $|r\rangle$ (triangles) for $N = 6$. For N even, the dark state does not populate $|r\rangle$, while for N odd, the final dark state is the state with one Rydberg excitation and $N-1$ atoms in the $J_x = 0$ eigenstate. In general we write the final dark state

$$|D_N\rangle = \begin{cases} |J_x = 0\rangle & \text{if } N \text{ is even} \\ (|J_x = 0\rangle \otimes |r\rangle)_{\text{sym}} & \text{if } N \text{ is odd,} \end{cases} \tag{7}$$

where $(\cdot)_{\text{sym}}$ indicates that the state is symmetrized with respect to $|r\rangle$, such that any atom is Rydberg excited with equal weight.

The STIRAP protocol produces a $|J_x = 0\rangle$ multiparticle entangled state, and precisely this state reaches the Heisenberg limit of phase sensitivity in entanglement enabled precision metrology [18,19]. If the two lower states are, e.g., the hyperfine states of the Cs clock transition, the presented entanglement scheme thus constitutes an ideal preparation of the system for an atomic clock. Note that $|J_x = 0\rangle$ is produced even when no knowledge of the exact atom number is available implying, e.g., that a mixed initial state is transferred to a mixed final state with $J_x = 0$ exactly fulfilled. To avoid decay into the states $|1\rangle$ and $|2\rangle$, which would slightly perturb the $J_x = 0$ property, one may field ionize the Rydberg excited component after the STIRAP process.

The state (7) has none or a single Rydberg excited atom, depending on the number of atoms initially in the $|1\rangle$ state, n_1, being even or odd. Following [20], this can be used to prepare a Greenberger-Horne-Zeilinger (GHZ) state [21] of the system. We first prepare all our atoms in a spin

coherent state, i.e., a product of superpositions of two ground states $|0\rangle$ and $|1\rangle$,

$$\left(\frac{|0\rangle + |1\rangle}{\sqrt{2}}\right)^{\otimes N} = \sum_{n_1=0}^{N} \sqrt{\binom{N}{n_1}} \left(\frac{1}{\sqrt{2}}\right)^{N} |n_0, n_1\rangle, \quad (8)$$

where n_1 is the number of atoms in the state $|1\rangle$ and $n_0 = N - n_1$. Applying the pair of STIRAP processes in Fig. 1(c) to the entire system transfers each component $|n_0, n_1\rangle$ to a state with none or a single Rydberg excitation and back. An energy shift of the Rydberg state or a phase shift of the laser coupling the Rydberg state can now be used to provide states populating the Rydberg state with a phase, $i = e^{i\pi/2}$. We can write this phase, $e^{i\eta(n_1)} = [e^{i\pi/4} + (-1)^{n_1} e^{-i\pi/4}]/\sqrt{2}$ leading after the second STIRAP process to the final GHZ state,

$$|\psi_f\rangle = \sum_{n_1=0}^{N} \sqrt{\binom{N}{n_1}} \left(\frac{1}{\sqrt{2}}\right)^{N} e^{i\eta(n_1)} |n_0, n_1\rangle$$
$$= \frac{e^{i\pi/4}}{\sqrt{2}} \left(\frac{|0\rangle + |1\rangle}{\sqrt{2}}\right)^{\otimes N} + \frac{e^{-i\pi/4}}{\sqrt{2}} \left(\frac{|0\rangle - |1\rangle}{\sqrt{2}}\right)^{\otimes N}. \quad (9)$$

We have investigated the preparation of the $|J_x = 0\rangle$ (7) and GHZ (9) states using the $\sin^2$ pulses and taking into account the coupling to states with two Rydberg excited atoms and the accompanying energy shift. For $N = 10$ we find a population of the $|J_x = 0\rangle$ or GHZ state above 0.995 for peak Rabi frequencies of $10 \times 2\pi$ MHz, a Rydberg interaction, $E/2\pi = 400$ MHz, and pulse widths of 50 μs. These numbers require the use of Rydberg levels with $n \geq 100$ where a lifetime above 1 ms can be achieved for atoms in a cryogenic environment [3]. Increasing N further will require a stronger Rydberg interaction to prevent multiple Rydberg excitations as well as longer pulses and higher Rabi frequencies to ensure adiabaticity. As indicated by the energy spectra in Fig. 3, there is a critical crossing region at $t \simeq 0.5$ μs, where special care should be taken, and we anticipate that control theory may be used to find optimal pulse shapes.

In conclusion, we have demonstrated that the Rydberg excitation blockade mechanism in conjunction with rapid adiabatic passage processes provides rich opportunities to prepare two-atom and multiatom entangled states with confined samples of atoms, e.g., in optical dipole traps or small lattice arrays. Our calculations indicate the possibility of high fidelity generation of entangled states and quantum superpositions states with tens of atoms, and we propose to apply control theory methods to optimize pulse shapes and reach even larger systems.

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