Filtering single atoms from Rydberg blockaded mesoscopic ensembles

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We propose an efficient method to filter out single atoms from trapped ensembles with unknown number of atoms. The method employs stimulated adiabatic passage to reversibly transfer a single atom to the Rydberg state which blocks subsequent Rydberg excitation of all the other atoms within the ensemble. This triggers the excitation of Rydberg blockaded atoms to short lived intermediate states and their subsequent decay to untrapped states. Using an auxiliary microwave field to carefully engineer the dissipation, we obtain a nearly deterministic single-atom source. Our method is applicable to small atomic ensembles in individual microtraps and in lattice arrays.

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Introduction.— Deterministic preparation of single trapped atoms, or arrays thereof, is an important task required for many potentially interesting applications, including quantum computation and simulation [1–7]. Optical lattice experiments can realize Mott insulators with single atom per lattice site [8] with low particle or hole defect probability [9, 10]. Achieving single-atom occupation of spatially separated microtraps with high fidelity is more challenging. Several techniques have been explored to reduce the trap occupation number fluctuations [11–17].

Here we propose a method to filter out single atoms from trapped ensembles with unknown number of atoms. Our scheme relies on the strong, long-range interaction between atoms in high-lying Rydberg states [18, 19] which blocks resonant optical excitation of more than one Rydberg atom within a certain interatomic distance [2, 20–23]. We use stimulated adiabatic passage technique (STIRAP) [24] to transfer any one atom of the ensemble from the ground state to the Rydberg state by a two-photon transition via an intermediate excited state [22–24]. This atom then strongly suppresses Rydberg excitation of the other atoms due to the interaction-induced level shifts. The Rydberg-blockaded atoms now turn into strongly-driven two-level atoms which rapidly decay from the (intermediate) excited state back to the initial trapped ground state [25, 26] as well as to other untrapped states. After several excitation and decay cycles, all of the Rydberg-blockaded atoms are removed from the trap. The reverse adiabatic transfer of the remaining Rydberg atom back to the initial ground state completes the preparation of a single trapped atom. We achieve high efficiency and reliability of the single-atom source by carefully engineering the atomic excitation and dissipation. The former is controlled by the time-dependent amplitude and detuning of the laser field, while the latter can be accomplished by employing an auxiliary microwave field to open appropriate decay channels to the untrapped states.

Stimulated adiabatic passage with a single-atom.— For what follows, it will be helpful to recall the essence of adiabatic transfer (STIRAP) of population in an isolated three-level atom using a pair of coherent laser pulses [24]. A laser field with Rabi frequency Ωge couples the stable ground state |g⟩ to an unstable (decaying) excited state |e⟩, which in turn is coupled to another stable state |r⟩ by the second laser field of Rabi frequency Ωer [Fig. [1]a]. In the frame rotating with the frequencies of the laser fields, the atom-field interaction Hamiltonian reads

\[ \mathcal{V}_{\text{at}} = \hbar \Delta_\epsilon |e\rangle \langle e| + \hbar (\Omega_{ge} |e\rangle \langle g| + \Omega_{er} |r\rangle \langle e| + \text{H.c.}), \]

where \( \Delta_\epsilon \) is the detuning of the intermediate state |e⟩ and we assume two-photon resonance between |g⟩ and |r⟩. The eigenvalues of \( \mathcal{V}_{\text{at}}/\hbar \) are \( \lambda_0 = 0 \) and \( \lambda_\pm = \Delta_\epsilon/2 \pm \sqrt{\Omega_{ge}^2 + \Omega_{er}^2 + (\Delta_\epsilon/2)^2} \) and the corresponding eigenstates are given by

\[ |\psi_0\rangle = (\Omega_{er} |g\rangle - \Omega_{ge} |r\rangle)/\omega_0, \]
\[ |\psi_\pm\rangle = (\Omega_{ge} |g\rangle + \lambda_\pm |e\rangle + \Omega_{er} |r\rangle)/\omega_\pm, \]

where \( \omega_0^2 = \Omega_{ge}^2 + \Omega_{er}^2 \) and \( \omega_\pm^2 = \omega_0^2 + \lambda_\pm^2 \). The zero-energy “dark” state \( |\psi_0\rangle \) does not contain the rapidly decaying state |e⟩, which, however, contributes to the energy-shifted “bright” states \( |\psi_\pm\rangle \) making them unstable against spontaneous decay. By adiabatically changing the dark state superposition, the STIRAP process can completely transfer the population between the two stable states |g⟩ and |r⟩ without populating the unstable state |e⟩. To this end, with the atom initially in state |g⟩, one first applies the \( \Omega_{er} \) field (\( \Omega_{ge} \gg \Omega_{er} \)), resulting in \( |g\rangle |\psi_0\rangle = 1 \). This is then followed by switching on the \( \Omega_{ge} \) field, leading to \( \langle r|\psi_0\rangle = -\Omega_{ge}/\omega_0 \), which results in \( \langle r|\psi_0\rangle \approx 1 \) when \( \Omega_{ge} \gg \Omega_{er} \). For sufficiently smooth switching, \( \partial_\lambda \omega_0 \ll \omega_0 |\lambda_\pm - \lambda_0| \), the atom adiabatically follows the dark state \( |\psi_0\rangle \), and the bright states \( |\psi_\pm\rangle \), and thereby |e⟩, are never populated. The reverse process, at the end of which \( \Omega_{er} \gg \Omega_{ge} \), can transfer the atom in state |r⟩ back to state |g⟩.
The multiatom system. — Consider now an ensemble of $N$ atoms initially in the ground state $|g\rangle$ confined within a small trap. All the atoms uniformly interact with two laser fields on the transitions $|g\rangle \rightarrow |e\rangle$ and $|e\rangle \rightarrow |r\rangle$ with Rabi frequencies $\Omega_{ge}$ and $\Omega_{er}$, as shown in Fig. 1(a). For each atom $j$, the Hamiltonian reads

$$
\mathcal{H}_{ad}^{(j)} = \hbar \Delta_j \hat{\sigma}_{ee}^{(j)} + \hbar (\Omega_{ge} \sigma_{eg}^{(j)} + \Omega_{er} \sigma_{er}^{(j)} + \text{H.c.}),
$$

where $\sigma_{\mu \nu}^{(j)} \equiv \{ \mu \}_{ij} \{ \nu \}_{uj}$ are the atomic operators. The intermediate excited state $|e\rangle$, possibly detuned by $\Delta_e$, decays with the rate $\Gamma_{eg}$ to the trapped ground state $|g\rangle$, and with the rate $\Gamma_{es}$ to another untrapped lower state $|s\rangle$ which is decoupled from the driving lasers. For completeness, we also take into account the typically much smaller decay rate $\Gamma_{es}$ of the highly excited Rydberg state $|r\rangle$ and its possible dephasing rate $\gamma_r$ (with respect to the other states) due to nonradiative collisions, inhomogeneous trapping potential, external field noise, laser phase fluctuations or Doppler shifts. The corresponding Lindblad generators for the decay and dephasing processes are

$$
\hat{L}_{ge} = \sqrt{\Gamma_{es} \sigma_{ge}^{(j)}}, \quad \hat{L}_{se} = \sqrt{\Gamma_{es} \sigma_{se}^{(j)}}, \quad \hat{L}_{er} = \sqrt{\Gamma_{es} \sigma_{er}^{(j)}}, \quad \hat{L}_{r} = \sqrt{\gamma_r (2\sigma_{rr}^{(j)} - 1)},
$$

where $\sigma_{\mu \nu}^{(j)} \equiv \{ \mu \}_{ij} \{ \nu \}_{uj}$.

The long-range atom-atom interactions are described by the Hamiltonian

$$
\mathcal{V}_{aa}^{ij} = \hbar \Delta (d_{ij}) \sigma_{rr}^{(i)} \otimes \sigma_{rr}^{(j)},
$$

where $\Delta (d_{ij}) = C_6 / d_6^{12}$ is the van der Waals potential between pairs of atoms $i, j$ in the Rydberg state $|r\rangle$ separated by distance $d_{ij}$. When the interaction-induced level shifts $\Delta (d_{ij})$ of states $|r_i, r_j\rangle$ exceed the two-photon excitation linewidth of the Rydberg state $w \sim \Omega_{ge}^2 + \Omega_{er}^2$, (\Gamma = \Gamma_{eg} + \Gamma_{es}$), double (multiple) Rydberg excitations are blocked. We can then define the blockade distance via $\Delta (d_b) \gtrsim w_{\max}$ leading to $d_b = \sqrt{\Gamma / w_{\max}}$. To achieve nearly complete blockade of more than one Rydberg excitation, we assume that all the atoms are initially trapped within a volume of linear dimension $L \lesssim \frac{2}{\Gamma} \Delta (d_b)$ corresponding to $\Delta (d_{ij}) \gtrsim 10w_{\max}$ for $i, j \in [1, N]$.

Numerical simulations. — We simulate the dissipative dynamics of the system of $N$ atoms using the quantum stochastic (Monte Carlo) wavefunctions [39], which allows us to treat nontrivial numbers $N \leq 10$ of strongly-interacting atoms exactly, without truncating the correspondingly large Hilbert space. In such simulation, the state of the system $|\Psi\rangle$ evolves according to the Schrödinger equation $\partial_t |\Psi\rangle = -i \mathcal{H} |\Psi\rangle$ with an effective Hamiltonian

$$
\hat{\mathcal{H}} = \mathcal{H} - \frac{i}{2} \hat{L}^2,
$$

where

$$
\mathcal{H} = \sum_j \mathcal{V}_{ad}^{(j)} + \sum_{i<j} \mathcal{V}_{aa}^{ij},
$$

is the usual (Hermitian) Hamiltonian, while

$$
\hat{L}^2 = \sum_j \left[ \hat{L}_{ge}^{(j)} \sigma_{ge} + \hat{L}_{se}^{(j)} \sigma_{se} + \hat{L}_{er}^{(j)} \sigma_{er} + \hat{L}_{r}^{(j)} \right] = \sum_j \left[ (\Gamma_{eg} + \Gamma_{es}) \sigma_{ee} + \Gamma_{re} \sigma_{rr} + \gamma_r \right]
$$

is the non-Hermitian part which does not preserve the norm of $|\Psi\rangle$. The evolution is interrupted by random quantum jumps $|\Psi\rangle \rightarrow \hat{L}_{\mu \nu} |\Psi\rangle$ of individual atoms with probabilities determined by the corresponding weights $\langle \Psi | \hat{L}_{\mu \nu} | \Psi \rangle$. In a single quantum trajectory, the normalized wavefunction of the system at any time $t$ is given by $|\Psi(t)\rangle = |\Psi(t)\rangle / \sqrt{\langle \Psi(t) | \Psi(t) \rangle}$. The expectation value of any observable $\hat{O}$ of the system is then obtained by averaging over many, $M \gg 1$, independently simulated trajectories, $\langle \hat{O} \rangle = \text{Tr}[\rho \hat{O}] = \frac{1}{M} \sum_M \langle \Psi_m | \hat{O} | \Psi_m \rangle$, while the density operator is given by $\rho(t) = \frac{1}{M} \sum_M | \Psi_m(t) \rangle \langle \Psi_m(t) |$.

Starting from all the atoms in the trapped ground state, $|\Psi(0)\rangle = |g_1, g_2, \ldots, g_N\rangle$, we assume that during the evolution the atoms that decay to state $|s\rangle$ decouple from the laser field and are eventually lost from the

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{Level scheme of atoms driven by a pair of laser fields with Rabi frequencies $\Omega_{ge}$ and $\Omega_{er}$ on the transitions $|g\rangle \rightarrow |e\rangle$ and $|e\rangle \rightarrow |r\rangle$. The atoms in the ground state $|g\rangle$ are confined within a volume of linear dimension $L \approx 2 \mu m$ smaller than the blockade distance $d_b$ determined by the interaction $\mathcal{V}_{aa}$ between their Rydberg states $|r\rangle \equiv nS_{1/2}$ with large principal quantum number $n$ ($\approx 50$) and small decay $\Gamma_{re}$ ($\approx 3$ kHz) and dephasing $\gamma_r$ ($\lesssim 0.1$ MHz) rates. In (a) the ground state of $^{87}$Rb is $|g\rangle \equiv 5S_{1/2} (F = 1, m_F = 1)$ while the intermediate state $|e\rangle \equiv 5P_{1/2} (F = 2, m_F = 2)$ decays back to $|g\rangle$ with the rate $\Gamma_{eg} = \frac{1}{4} \Gamma_e$ ($\Gamma_e = 36$ MHz) and to other untrapped states $|s\rangle \equiv 3S_{1/2} (F = 2, m_F = 1, 2)$ with the rate $\Gamma_{es} = \frac{1}{4} \Gamma_e$ (branching ratio $\Gamma_{es}/\Gamma_{eg} = 1$). In (b) the transition between the ground state $|g\rangle \equiv 5S_{1/2} (F = 2, m_F = 2)$ and intermediate $|e\rangle \equiv 5P_{3/2} (F = 3, m_F = 3)$ states is closed ($|e\rangle$ decays only to $|g\rangle$ with rate $\Gamma_e = 38$ MHz), but a microwave field coupling $|e\rangle$ to, e.g., $|e'\rangle \equiv 3P_{3/2} (F = 2, m_F = 2)$ with Rabi frequency $\Omega_{er}$ can open a weak decay channel $\Gamma_{es} \approx \frac{\gamma_0}{\Delta e} \sim 2.4$ MHz ($\ll \Gamma_{eg} = \Gamma_e + \frac{\gamma_0}{\Delta e} \approx 40$ MHz) to the untrapped states $|s\rangle \equiv 3S_{1/2} (F = 1, 2, m_F = 1)$. The hyperfine and/or Zeeman shifts of states $|s\rangle$ suppress their coupling to the $\Omega_{ge}$ field, while facilitating selective interaction with a pushing laser of appropriate frequency and polarization.}
\end{figure}
The probability that a trapped atom is uncertain, with the Poisson distribution

\[ P_{\text{init}}(N) = \frac{\bar{N}^N e^{-\bar{N}}}{N!} \]

around some mean \( \bar{N} \). Then, the probabilities \( P(n) \) for \( n \) atoms surviving in the trap are given by \( P(n) = \sum NP_{\text{init}}(N)P_N(n) \).

We have performed numerical simulations for various initial number \( N \) of atoms in the trap, results of which are shown in Figs. 2-4. In all simulations, we first apply the field coupling states \(|e\rangle\) and \(|r\rangle\) with the Rabi frequency \( \Omega_{er} \) which stays constant throughout the process, and then send a smooth pulse driving transition \(|g\rangle \rightarrow |e\rangle\) with the Rabi frequency \( \Omega_{ge}(t) \) whose peak value is much larger than \( \Omega_{er} \). We thus expect that a single atom in the trap will be adiabatically transferred from the ground state \(|g\rangle\) to the Rydberg state \(|r\rangle\) and block Rydberg excitations of all the other atoms initiating thereby their optical pumping through the intermediate excited state \(|e\rangle\) to the decoupled and untrapped state(s) \(|s\rangle\). At the end of the process, when \( \Omega_{ge} \) is reduced to zero, the remaining Rydberg-state atom will be transferred back to the trapped ground state \(|g\rangle\).

**Results.**—Consider first the resonant excitation, \( \Delta_e = 0 \), of the atoms with the \( \Omega_{ge} \) and \( \Omega_{er} \) fields shown in the upper panel of Fig. 2. The branching ratio for the decay of the atoms from the intermediate excited state \(|e\rangle\) to the untrapped state \(|s\rangle\) and back to \(|g\rangle\) is large \( \Gamma_{es}/\Gamma_{eg} = 1 \), as per Fig. 1(a), while we neglect for the moment the dephasing of the Rydberg state, \( \gamma_r = 0 \). We find that even before the \( \Omega_{ge} \) pulse attains its maximum, most of the atoms are optically pumped into the untrapped state(s) \(|s\rangle\), while the mean number of atoms in the trap drops below unity: \( P(0) \approx 0.3 \) and \( P(1) \approx 0.7 \), while \( P(n > 1) \approx 0 \). The reason for this unexpected and unfortunate behavior is that each quantum jump of a blocked atom from state \(|e\rangle\) to either \(|g\rangle\) or \(|s\rangle\) projects a single atom to the Rydberg state \(|r\rangle\) whose overlap with the dark state \(|\psi_0\rangle\rangle = (\Omega_{ge}/\Omega_{ge}^2 + \Omega_{er}^2) |\psi_0\rangle\rangle \) is smaller than unity, unless \( \Omega_{ge} \gg \Omega_{er} \). Since for \( \Omega_{ge} \ll \Omega_{er} \) the Rydberg atom is not in the dark eigenstate, but has large overlap with the bright eigenstates as well, \(|\psi_\pm\rangle\rangle = (\Omega_{ge}^2 + 2\Omega_{er}^2) |\psi_\pm\rangle\rangle \), it is coupled to the state \(|e\rangle\) from where it can decay to \(|g\rangle\) or \(|s\rangle\). In a closed three-level system, \( \Omega_{ge} \) to \(|g\rangle\) would simply restart the excitation process of this or any other atom by the fields \( \Omega_{ge} \) and \( \Omega_{er} \). In the present case, however, the decay of all the atoms but one to the decoupled state \(|s\rangle\) may leave the last atom in the Rydberg state \(|r\rangle\) before the \( \Omega_{ge}(t) \) pulse reaches its peak value \( \Omega_{ge} \gg \Omega_{er} \).

The probability of loosing this last remaining atom can thus be estimated as

\[ \frac{\Gamma_{es}}{\Gamma_{es} + \Gamma_{eg}} (1 - |\langle \psi_0 |r \rangle|^2) \approx \frac{\Gamma_{es} \Omega_{er}^2}{(\Gamma_{es} + \Gamma_{eg})(\Omega_{ge}^2 + \Omega_{er}^2)}, \]

which, with \( \Gamma_{es} = \Gamma_{eg} \) and \( \Omega_{ge} \ll \Omega_{er} \) is about 1/4, consistent with \( \langle n \rangle \) and \( P(0) \) in Fig. 2.

Including now in the simulations realistic dephasing, \( \gamma_r \neq 0 \), amounts to additional coherence relaxation between the long-lived states \(|g\rangle\) and \(|r\rangle\) with the rate \( \gamma_{rg} = \frac{1}{2}\Gamma_{re} + 2\gamma_r \). As is well known [34], the STIRAP in a three-level atom is very sensitive to such dephasing, since the decoherence of the dark-state superposition leads to additional population of the bright states [34], which in the present system can decay to the untrapped state(s) \(|s\rangle\). Hence, in the inset of Fig. 2 we observe further decrease of the survival probability \( P(1) \approx 0.63 \) for a single trapped atom.

To increase the single atom survival probability \( P(1) \), we should therefore postpone the decay of atoms from state \(|e\rangle\) to \(|s\rangle\) until \( \Omega_{ge} \) is much larger than \( \Omega_{er} \). We thus take a finite detuning \( \Delta_e \) of intermediate level \(|e\rangle\) and switch-on \( \Omega_{ge} \) slightly faster. The results of simulations are shown in Fig. 3 were we indeed notice significant improvement of the single-atom filtering, \( P(1) \approx \)}
0.87 or 0.8 with additional dephasing $\gamma_r$. These results are obtained after some optimization of both $\Omega_{ge}$ and $\Delta_e$, but for the given atomic level scheme further substantial improvements do not seem possible.

To achieve nearly perfect filtering, we now consider the closed three-level system of Fig. 1(b), where state $|e\rangle$ decays spontaneously only to the trapped state $|g\rangle$, while we can engineer an additional decay channel to the untrapped states $|s\rangle$. To this end, we use a microwave field which weakly couples $|e\rangle$ to an auxiliary state $|e'\rangle$ with the Rabi frequency $\Omega_{ee'}$ that is small compared to the total decay rate $\Gamma_{ee'}$ of $|e'\rangle$. Upon adiabatic elimination of $|e'\rangle$, we obtain a small additional decay on the transition $|e\rangle \rightarrow |g\rangle$ with rate $4\Omega_{ee'}^2/3\Gamma_{ee'}$, and a decay to the untrapped states with rate $\Gamma_{es} = 8\Omega_{ee'}^2/3\Gamma_{ee'}$, which can be controlled via the microwave field intensity $\Omega_{ee'}^2$ (the numerical prefactors are related to the branching ratio of the decay of state $|e'\rangle$ to $|g\rangle$ and $|s\rangle$, $\Gamma_{es}/\Gamma_{ee'} = 1/2$). Figure 4 shows the results of our simulations demonstrating dramatically improved single atom filtering, $P(1) \approx 0.96$ [$P(0) \approx 0.02$ and $P(2) \approx 0.01$, $P(3) \approx 0 \times 10^{-4}$], at the expense of a longer duration of the process. Even sizable dephasing $\gamma_r$ of the Rydberg state with respect to the ground state does not significantly reduce the efficiency of the single-atom filtering, $P(1) \approx 0.92$ [$P(0) \approx 0.05$], which is due to the robustness of adiabatic passage in a Rydberg-blockaded ensemble composed of atoms with nearly-closed transitions [36].

We finally note that in obtaining the averaged probabilities $P(n)$ of the final number of atoms, we assumed the initial Poisson distribution $P_{\text{init}}(N)$ of the atom number in the trap with the mean $\bar{N} = 5$, for which $P_{\text{init}}(0) \approx 0.007$ and $\sum_{N>10} P_{\text{init}}(N) \approx 0.02$, and performed simulations for $N \leq 10$. The initial 2% population of the trap not included in our simulation can only increase the final single atom probability. Our method should work even better for larger initial number of atoms in the trap, which is, however, prohibitively difficult to simulate numerically.

Summary.— To conclude, we have proposed and analyzed a viable scheme to filter out single atoms from mesoscopic ensembles containing unknown number of initially trapped atoms. We have shown that the combination of two-photon adiabatic transfer of the atoms to the strongly interacting Rydberg state and carefully engineered decay of atoms from the intermediate excited state can lead to nearly-deterministic production of single trapped atoms in individual microtraps or optical lattice potentials, which is an important prerequisite for quantum computation with atoms serving as qubits or quantum simulations of many-body spin systems. Moreover, our studies represent an important and interesting new facet of the very active field of dissipative generation of states and processes in open many-body systems [40, 41].
