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A deterministic free-space source of single photons using Rydberg atoms

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We propose an efficient free-space scheme to create single photons in a well-defined spatio-temporal mode. To that end, we first prepare a single source atom in an excited Rydberg state. The source atom interacts with a large ensemble of ground-state atoms via laser-mediated dipole-dipole exchange interaction. Using adiabatic passage with a chirped laser pulse, we produce a spatially-extended spin-wave of a single Rydberg excitation in the ensemble, accompanied by the transition of the source atom to another Rydberg state. The collective atomic excitation can then be converted to a propagating optical photon via a coherent coupling field. In contrast to previous approaches, our single-photon source does not rely on strong coupling of a single emitter to a resonant cavity, nor does it require heralding of collective excitation or complete Rydberg blockade of multiple excitations in the atomic ensemble.

Single photons can serve as flying qubits for many important applications, including all-optical quantum computation and long-distance quantum communication and cryptography [1, 2]. Various sources of single photons are being explored, most of them use single emitters coupled to resonant cavities or waveguides [3–10]. Free-space schemes typically rely on the DLCZ protocol [11] for low-efficiency heralded preparation of a collective spin excitation of an atomic ensemble followed by its stimulated Raman conversion into a photon [12–14]. Creating a deterministic source of single photons without requiring coupling to resonant optical structures remains an outstanding challenge. Here we show how, in a free-space setting, the remarkable properties of Rydberg atoms can be used to map a single atomic excitation on a single photon emitted into a well defined spatial and temporal mode.

Atomic Rydberg states with high principal quantum numbers $n \gg 1$ have long lifetimes $\tau \propto n^3$ and strong electric dipole moments $\rho \propto n^2$ [15]. The resulting long-range, resonant (exchange) and nonresonant (dispersive or van der Waals) dipole-dipole interactions between the atoms can suppress more than one Rydberg excitations within a certain blockade distance [16–19]. An ensemble of atoms in the blockaded volume can be viewed as an effective two-level Rydberg superatom [16, 19–23]. A single collective excitation of the superatom can be created by resonant laser(s) and then converted to a photon in a well-defined spatio-temporal mode [24–29].

There are several complications associated with the efficient creation of a single coherent Rydberg excitation in an atomic ensemble and its deterministic conversion into a photon. Creating only a single excitation requires a completely blockaded atomic ensemble. Efficient conversion of the excitation into a photon in a well-defined spatio-temporal mode requires large optical depth. Hence, the blockaded volume should accommodate many atoms, which presumes strong, long-range, isotropic interactions. Van der Waals interactions between Rydberg excited atoms can be nearly isotropic [30], but a blockade range much beyond 10 $\mu$m is difficult to achieve. Large optical depth of the blockaded volume requires high atomic density which, however, leads to strong decoherence of the Rydberg-state electrons [31], and may involve molecular resonances of Rydberg excited atoms [32]. Dipole-dipole interactions have longer range, which allows using larger atomic ensembles with lower densities. Compared to the van der Waals interaction, however, the dipole-dipole interactions are “softer”, leading to incomplete suppression of multiple Rydberg excitations within the blockade distance [33].

In contrast, preparing a single, isolated atom in an excited state with high fidelity is relatively easy. We propose an efficient free-space technique to convert this excitation into a single photon in a well defined mode, without resorting to strong coupling of the atom to a single cavity mode [3–6]. Instead, we use long-range dipole-dipole exchange interactions between the Rydberg states of atoms to map the Rydberg excitation of the single “source” atom onto a collective Rydberg excitation of an ensemble of “medium” atoms. The mapping efficiency is boosted by the collectively enhanced coupling of the source atom to many medium atoms, but complete Rydberg blockade or strong interactions among the medium atoms are not required. Subsequently, using a coupling laser pulse, the collective excitation of the ensemble of medium atoms having large optical depth can be converted into a single photon propagating in a phase-matched direction.

We note a closely related recent proposal to realize a chiral light-atom interface by transferring the state of a single atom to a spatially ordered array of atoms which acts as a phased-array optical antenna for photon emission into a well-defined spatial mode [34].

Consider the system shown schematically in Fig. 1. We assume that a single source atom, having a strong dipole-allowed microwave transition with frequency $\omega_{ud}$ between the Rydberg states $|u\rangle$ and $|d\rangle$, is trapped in a...
the Hamiltonian for the system is
\[
\hat{H}_1/\hbar = -\sum_{j=1}^{N} \left[ \Delta |u_j\rangle \langle i| + \delta |s_j\rangle \langle s| \otimes |d_s\rangle \langle d| \right] \\
+ (\Omega e^{i\kappa_0 \cdot r_j} |i_j\rangle \langle g| + \text{H.c.}) \\
- [D(R_j) |s_j\rangle \langle i| \otimes |d_s\rangle \langle u| + \text{H.c.}], \tag{1}
\]
where index \( j \) enumerates the medium atoms at positions \( r_j, \kappa_0 \parallel e_z \) is the wave vector of the laser field, and \( \delta \equiv \Delta + \Delta_{sa} = \omega + \omega_{ud} - \omega_{sg} \) is the detuning of the product state \(|s\rangle \langle d| \). Since the intermediate Rydberg level \(|i\rangle \) is strongly detuned, \( \Delta \simeq -\Delta_{sa} \gg |\Omega|, D, |\delta| \), we can eliminate it adiabatically. We then obtain an effective Hamiltonian
\[
\hat{H}_1/\hbar = -\sum_{j} \left[ \delta_j |s_j\rangle \langle s| \otimes |d_s\rangle \langle d| \right] \\
+ (\hat{D_j} e^{i\kappa_0 \cdot r_j} |i_j\rangle \langle g| \otimes |d_s\rangle \langle u| + \text{H.c.}), \tag{2}
\]
where \( \delta_j \equiv \delta(R_j) = \delta + |(j-1)\Delta D(R_j)|^2/\Delta \) is the shifted detuning of \(|s_j\rangle \langle d| \) and \( \hat{D_j} \equiv \hat{D}(R_j) = -D(R_j)|\delta_j|/\Delta \) is the second-order coupling between \(|g_j\rangle \langle u| \) and \(|s_j\rangle \langle d| \). \( \delta(R) \) has a weak position dependence stemming from the level shift \( |D(R)|^2/\Delta \) of \(|g\rangle \) due to the non-resonant dipole-dipole coupling, while the level shift \( |\Omega|^2/\Delta \) of \(|g\rangle \) is uniform.

Let us for the moment neglect the (weak) spatial dependence of \( \delta \), i.e., assume that all the medium atoms have the same \(|g\rangle \rightarrow |s\rangle \) transition frequency. Since by flipping the source atom \(|u\rangle \rightarrow |d| \) we can create at most one Rydberg excitation in the medium, we introduce the ensemble ground state \(|G\rangle \equiv |g_1, g_2, \ldots, g_N\rangle \) and a single collective Rydberg excitation state \(|S,d\rangle \equiv \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |D_j|^2 \rangle \rangle \langle G_1, g_2, \ldots, s_j, \ldots, g_N| \) with \( \hat{D} \equiv (\sum_j |D_j|^2)^{1/2} \). The Hamiltonian (2) then reduces to that for a two-level system,
\[
\hat{H}_1 = -\hbar \begin{pmatrix} 0 & \hat{D} \\ \hat{D}^\dagger & \delta \end{pmatrix}, \tag{3}
\]
in the basis of states \{ |G,u\rangle, |S,d\rangle \}. The eigenstates and corresponding eigenvalues of this Hamiltonian are \(|\pm\rangle \equiv [ \mp \lambda_\pm \langle G,u \rangle \pm \hat{D} (|S,d\rangle) / \sqrt{\lambda_+^2 + \hat{D}^2} \rangle \rangle \), \( \lambda_\pm = \delta \pm \sqrt{\delta^2 + 4\hat{D}^2}/2 \). In the limit of large negative detuning \(-\delta \gg \hat{D} \), \(|+\rangle \simeq |G,u\rangle \) with \( \lambda_+ \simeq 0 \) coincides with the ensemble ground state. In the opposite limit of large positive detuning \(\delta \gg \hat{D} \), \(|+\rangle \simeq |S, d\rangle \) with \( \lambda_+ \simeq 0 \) corresponds to the collective Rydberg excited state of the ensemble. We can thus use adiabatic passage [36] to convert the system from the initial ground state \(|G,u\rangle \) to the Rydberg excited state \(|S,d\rangle \), by applying a laser pulse with a chirped frequency \( \omega = \omega_0 + \alpha(t - t_0) \), such that \( \delta = \alpha(t - t_0) \) is large and negative at early times \( t < t_0 \) and is large and positive at later times \( t > t_0 \).
Provided the chirp rate satisfies $\alpha < \tilde{D}^2$, the probability $P_{\uparrow \downarrow} \rightarrow |\downarrow \rangle = e^{-2\pi \tilde{D}^2/\alpha}$ of non-adiabatic Landau-Zener transition to the eigenstate $|\downarrow \rangle$ will be small [36]. Since the coupling $\tilde{D}$ is collectively enhanced by the number $N \gg 1$ of the medium atoms interacting with the source atom, we can use high chirp rates $\alpha$ to prepare the system with nearly unit probability in state $|S,d\rangle$. Moreover, we can verify the successful preparation of the medium in the collective state $|S\rangle$ by detecting the source atom in state $|d\rangle$ [44]. We note that adiabatic preparation of spatially-ordered Rydberg excitations of atoms in a lattice using chirped laser pulses was studied in [37–41].

We have performed exact numerical simulations of the dynamics of the system using realistic atomic parameters [44]. We place $N \gg 1$ ground state $|g\rangle_j$ atoms in an elongated volume at random positions $\mathbf{r}_j$ normally distributed around the origin, $x, y, z = 0$, with standard deviations $\sigma_x = \sigma_y > \sigma_z$. The source atom initially in state $|u\rangle$ is placed at a position $\mathbf{r}_s$ close to the center of the longitudinal extent of the volume and well outside its transverse width. We apply to the atoms a pulsed and chirped laser field with the Rabi frequency $\Omega$ and two-photon detuning $\delta$ shown in Fig. 2(a). We simulate the evolution of the state vector of the system $|\Psi_t\rangle = c_0 |G\rangle \otimes |u\rangle + \sum_{j=1}^{N} c_j e^{i k_0 \cdot \mathbf{r}_j} |s_j\rangle \otimes |d\rangle$ taking into account the decay and dephasing of the Rydberg states [44]. The resulting dynamics of populations $P_G = |c_0|^2$ and $P_S \equiv \sum_j |c_j|^2$ are shown in Fig. 2(b). We obtain a single collective Rydberg excitation $|S\rangle$ of the atomic ensemble with high probability $P_S \gg 0.977$, which is slightly smaller than unity mainly due to the decay and dephasing. In Fig. 2(c) we show the final spatial distribution of the Rydberg excitation density $p_s (x) \propto |D(\mathbf{r} - \mathbf{r}_s)|^2$ which follows the dipole-dipole interaction strength. Our simulations verify that we can reliably prepare a spin wave of single collective Rydberg excitation $|S\rangle$ with the spatial wavefunction

$$S(\mathbf{r}) \simeq -\frac{\sqrt{\rho(\mathbf{r})} D(\mathbf{r} - \mathbf{r}_s)}{\sqrt{\int dr^3 \rho(\mathbf{r}) D^2(\mathbf{r} - \mathbf{r}_s)}} e^{i k_0 \cdot \mathbf{r}}.$$ 

Consider now the conversion of the collective Rydberg excitation of the atomic medium into a photon. To that end, we use a control laser field with wave vector $\mathbf{k}_c$ and frequency $\omega_c = c k_c$ acting resonantly on the transition $|s\rangle \rightarrow |e\rangle$ with the Rabi frequency $\Omega_s$. The atomic transition $|e\rangle \rightarrow |g\rangle$ is coupled with strengths $g_{k,\sigma}$ to the quantized radiation field modes $\hat{a}_{k,\sigma}$ characterized by the wave vectors $\mathbf{k}$, polarization $\sigma$ and frequencies $\omega_{k,\sigma} = c \mathbf{k}$. We take $\mathbf{k}_c$ parallel to $\mathbf{k}_0$ so as to achieve resonant emission of the photon in the phase-matched direction, $\mathbf{k} = k_0 - k_c$. The frequency and wave number of the Rydberg microwave transition can be neglected in comparison with those of the optical transitions. In the frame rotating with frequencies $\omega_{r,g}$ and $\omega_c = \omega_{r,e}$ (interaction picture), the Hamiltonian reads

$$H_2/\hbar = -\sum_{j=1}^{N} \sum_{k,\sigma} g_{k,\sigma} \hat{a}_{k,\sigma} e^{ik \cdot \mathbf{r}_j} e^{-i(\omega_{k,\sigma} - \omega_c)t} |e\rangle_j\langle g| + \Omega_s e^{ik_0 \cdot \mathbf{r}_s} |s\rangle_j\langle e| + H.c.,$$

The state vector of the system can be expanded as $|\Psi_2\rangle = \sum_{j=1}^{N} \sum_{k,\sigma} \hat{a}_{k,\sigma} e^{ik_0 \cdot \mathbf{r}_j} |s\rangle_j + b_j |e\rangle_j \otimes |0\rangle + \sum_{k,\sigma} a_{k,\sigma} |G\rangle \otimes |1_{k,\sigma}\rangle$, where $|e\rangle_j \equiv |g_1, g_2, \ldots, e_j, \ldots, g_N\rangle$ and $|1_{k,\sigma}\rangle \equiv e^{ik_0 \cdot \mathbf{r}_s} |0\rangle$ denotes the state of the radiation field with one photon in mode $k, \sigma$. Using the standard procedure [44],

![Fig. 2. Preparation of a single collective Rydberg excitation of an atomic ensemble. (a) Time dependence of Rabi frequency $\Omega$ (left vertical axis) and two-photon detuning $\delta$ (right vertical axis) of the driving laser. (b) Dynamics of populations $P_G$ and $P_S$ of the collective ground and single Rydberg excitation states of the atoms. (c) Final spatial distribution of the Rydberg excitation in an elongated volume containing the atoms: Main panel shows the density of excitation $p_s(x)$ along the longer $x$ axis of the volume (integrated over the transverse $x, y$ directions), while the left and right insets show the densities $p_s(x)$ and $p_s(y)$ along $x$ and $y$. We average over 2000 independent realizations of the ensemble of $N = 1000$ atoms with the peak density $\rho_{max} \simeq 10^{-3}$ and Gaussian distribution with $\sigma_{x,y} = 1 \mu$m and $\sigma_z = 6 \mu$m. The source atom is placed at $x_s = 7 \mu$m, $y_s = 0$, and $z_s = 0$. In the simulations, we take the peak Rabi frequency $\Omega_{max} = 2 \pi \times 10$ MHz, the Rydberg state decay $\Gamma_s = 10$ kHz and dephasing $\gamma_{eq} = 10$ kHz, and we use the atomic parameters corresponding to a Cs source atom with $|u\rangle \equiv 70 \rho_{1/2}$ and $|d\rangle \equiv 70 S_{1/2}$ and $\rho_{max}$ of $10$ kHz, and $\gamma_{eq}$ of $10$ kHz. We take the medium atoms with $|i\rangle \equiv 58 \rho_{3/2}$ and $|\chi\rangle \equiv 57 S_{3/2}$ with the intermediate state detuning $\Delta = 9.4 \Omega_{max}$ and the dipole-dipole interaction coefficient $C_3 = 11.7$ GHz $\mu$m$^3$, the medium atoms near the cloud center experience the strongest interaction $\tilde{D} \simeq 2 \pi \times 5.4$ MHz. The corresponding second-order transition rate is $\tilde{D}_{max} \simeq 2 \pi \times 0.6$ MHz, while the collective coupling rate is $\tilde{D} \simeq 2 \pi \times 13$ MHz.](image-url)
we obtain that the atomic state $|e\rangle$ spontaneously decays with rate $\Gamma_e$. Assuming $\Gamma_e \gg |\Omega|$, and eliminating $b_j$ leads to the solution for the amplitudes of photon emission into states $|1_k\rangle$,

$$a_k(t) = -\tilde{g}_k(t) \sum_j c_j(0) e^{i(k_0 - k_c - k) \cdot r_j},$$  \hspace{1cm} (5)$$

where $\tilde{g}_k(t) \equiv g_k \int_0^t dt' \frac{\Omega^2(t')}{\varepsilon} e^{i(\omega_k - \omega_{eg})t'} e^{- \int_0^t dt'' \frac{\delta(k_{e2}c)}{\varepsilon}}$ and for simplicity we drop the polarization index $\sigma$ assuming scalar and isotropic emission by individual atoms. In the case of a time-independent control field $\Omega_c$, the dimensionless coupling reduces to $\tilde{g}_k \sim \frac{g_k}{\Gamma_s(\omega_k - \omega_{eg})/2 + |\Omega_c|^2}$ for $t \gg w^{-1}$. The probability distribution of emitted photon $P_k = |a_k|^2$ is thus strongly peaked at frequency $\omega_k = \omega_{eg}$ with a narrow linewidth $w = \frac{|\Omega_c|^2}{\Gamma_s}$, and has a narrow angular distribution around $k = k_0 - k_c \parallel e_z$ as shown in Fig. 3. Our simulations reveal that the state of the emitted photon $|\psi_{ph}\rangle = \sum_k a_k |1_k\rangle$ is largely insensitive to the microscopic details of various realizations of the atomic ensemble. Note that efficient conversion of the atomic excitation into a photon requires a collinear geometry $k_c \parallel k_0$ for resonant emission at frequency $\omega_p = c|k_0 - k_c| \approx \omega_{eg}$, while even a small inclination $k_c \parallel k_0 \neq 0$ disturbs the phase-matching in a spatially-extended atomic medium and reduces the probability of photon emission into the well-defined spatial direction. In the presence of the control field $\Omega_c$, the resonant photon propagating in the optically dense medium in the $z$ direction is subject to electromagnetically induced transparency (EIT) [12] which suppresses photon reabsorption and scattering.

Similar to cavity QED schemes [3–6] employing stimulated Raman adiabatic passage [35, 36], we can create a single photon directly, populating only virtually the Rydberg state $|s\rangle$. To that end, we assume a constant control field $|\Omega_c| < \Gamma_s/2$ and small decay and two-photon detuning of the Rydberg state $\Gamma_s, \delta_k \ll w$. We then obtain a solution for the photon amplitudes as in Eq. (5) with $c_j(0) \rightarrow -D_j/\Delta$ and $\tilde{g}_k(t) \equiv g_k \int_0^t dt' \frac{\Omega^2(t')}{\varepsilon} e^{i(\omega_k - \omega_{eg})t'} e^{- \int_0^t dt'' \frac{\delta(k_{e2}c)}{\varepsilon}}$ [44]. Thus, with the source atom in state $|u\rangle$, the medium atoms in the collective ground state $|G\rangle$, and the control field $\Omega_c \neq 0$, by turning on the excitation laser $\Omega$ we produce a single photon on the atomic transition $|e\rangle \rightarrow |g\rangle$. This single-photon wavepacket is emitted with high probability into the direction of $k \approx k_0 - k_c \parallel e_z$, while its temporal shape can be manipulated by the time-dependence of $\Omega(t)$. The emission of the optical photon is accompanied by the transition of the source atom to state $|d\rangle$, which terminates the conversion process, even if $\Omega \neq 0$. To produce another photon, we have to reset the source atom to state $|u\rangle$.

To summarize, we have presented a new scheme for efficient single-photon production, controlled by a single source atom prepared in an appropriate Rydberg state and playing the role of a switch. The dipole-dipole exchange interaction with the source atom enables single collective Rydberg excitation of the atomic ensemble without the requirement of full blockade of the entire ensemble. Detailed numerical simulations with realistic experimental parameters demonstrate that this excitation can be converted into a single photon emitted into a well-defined spatio-temporal mode with better than 70% probability. The probability that the photon is coherently emitted into the small solid angle $\Delta\Omega \approx 0.15 \, \text{sr}$ (see Fig. 3) is given approximately by $P_{\Delta\Omega} \approx \eta N \Delta\Omega/4\pi$ [25], where $\eta \approx 0.6$ is the effective fraction of the medium atoms participating in the collective Rydberg excitation $|S\rangle$ (see Fig. 2(c)). This probability can be enhanced by increasing the atom number $N$, which, however, lead to increased Rydberg state dephasing.

In our analysis, we assumed an ensemble of atoms at random positions and with moderate density and neglected the vacuum field-mediated interactions between the atoms on the optical transitions. Recently Grankin et al. [34] have shown that imprinting an appropriate spatial amplitude and phase on an array of atoms with subwavelength spacing and coupled to a single atom in the same way as in our proposal can further enhance the photon emission probability into the predefined Gaussian (paraxial) mode of the radiation field, which can be used for quantum state transfer between distant atoms in free-space quantum networks. We finally note that our method to convert a single atomic excitation to an optical photon can be used for microwave to optical conversion.
in hybrid quantum interfaces [42, 43]. For example, the source atom can be replaced by a superconducting qubit, which can strongly couple to the Rydberg states of the medium atoms by a microwave transition.

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[34] A. Grankin, P. O. Guimond, D. V. Vasilyev, B. Vermersch, F. Robicheaux, P. Zoller, ‘Free-Space’ Pho-
tonic Quantum Link and Chiral Quantum Optics, arXiv:1802.05592.


[44] See Supplementary Material, which includes Refs. [12, 15, 28, 45–53], for the details of derivations of the results and parameters and further discussions of the performance of the single-photon source.


