Efficient excitation of a symmetric collective atomic state with a single-photon through dipole blockade

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In the famous quantum communication scheme developed by Duan et al.[L.M. Duan, M.D. Lukin, J.I. Cirac, and P. Zoller, Nature (London) 414 413 (2001)], the probability of successful generating a symmetric collective atomic state with a single-photon emitted have to be far smaller than 1 to obtain an acceptable entangled state. Because of strong dipole-dipole interaction between two Rydberg atoms, more than one simultaneous excitations in an atomic ensembles are greatly suppressed, which makes it possible to excite a mesoscopic cold atomic ensemble into a singly-excited symmetric collective state accompanied by a signal photon with near unity success probability, at the same higher-order excitations can be significantly inhibited.

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I. INTRODUCTION

Quantum information science holds promise for many fascinating potential applications such as the factorization of large numbers[1], secure communication[2], and spectroscopic techniques with enhanced sensitivity [3]. As a promising candidate for quantum state engineering and for quantum information processing, recently atomic ensembles with a large number of identical atoms have received much attention. Such a system can be used, e.g., for generation of significant spin squeezing [4–6] and entanglement [7–11], for storage of quantum light [12–15], and for long-distance quantum communication [16–18]. As against quantum information schemes employing single particles, those based on atomic ensembles have two assets: first, to manipulate atomic ensemble with laser is easier than to control a single particle; second, the coupling between atoms and some light mode in atomic ensembles can be greatly enhanced by a factor of $\sqrt{N_a}$ for certain atomic level structure because of many-atom interference effects [16, 19], where $N_a$ denotes the atom number in the ensemble. Due to this coupling enhancement, the atom coupling to light does not require a high-finesse microcavity, which, in spite of recent experimental advances [20, 21], remains a demanding technology.

In the Duan-Lukin-Cirac-Zoller (DLCZ) protocol for long-distance quantum communication [16], entanglement in the elementary links is created by recording a single photon produced indistinguishably by one of two atomic ensembles. The probability $p$ of successful generating one excitation in two ensembles is related to the fidelity of the entanglement, leading to the condition $p \ll 1$ to guaranty an acceptable entanglement quality. But when $p \to 0$, some experimental imperfections such as stray light scattering and detector dark counts will contaminate the entangled state increasingly [22], and subsequent processes including quantum swap and quantum communication become more challenging for finite coherent time of quantum memory [10]. To solve this problem, protocols based on single photon sources [10, 23] were suggested.

For the generation of a single collective excitation in the atomic ensemble with high success probability $p$ at the same time with high fidelity, we describe a method based on the strong dipole-dipole interaction between the atoms in the Rydberg state. When an atom is in the Rydberg state, the transition of another atom to this Rydberg level is inhibited due to the level shift arising from the strong dipole-dipole interaction between Rydberg atoms, which is the so-called dipole blockade [24–26]. This blockade can be used to generate entanglement between two atoms and therefor to accomplish two-bit quantum gates in individual-atom systems [27]. Recently Brion et al. propose to encode quantum information on the collective state of multilevel atomic ensembles and implement one and two-bit gates through collective internal state transitions in the presence of the dipole blockade [28]. This blockade can also be employed for a controlled generation of collective atomic state, for nonclassical photonic states, and for a scalable quantum logic gates [24]. In this paper, we will show that a single-excitation symmetric collective state of a mesoscopic cold atomic ensemble accompanied by a signal photon can be created with near unity success probability, while higher-order excitations in the ensemble can be significantly suppressed in the presence of the strong dipole blockade.

There have been many models describing the interaction between atomic ensembles and optical beams, such as the cavity-QED models [4–6, 12–16], one-dimensional light propagation models [7, 13], and three-dimensional free-space perturbation theory [19] which can describe most of the current experiments on the atomic ensembles [6, 8, 14, 15]. Here, we describe the interaction of light and mesoscopic cold atomic ensembles with strong dipole-dipole interaction also in the three-dimensional free-space configuration but without applying perturbation expansion.
II. THE LIGHT-ATOMIC ENSEMBLE INTERACTION

We consider a cold atomic ensemble of \( N_a \) identical atoms contained in a volume \( V \) with level structure shown in Fig. 1, ground state \(|g\rangle\), metastable state \(|s\rangle\), and Rydberg state \(|r\rangle\). Hereafter we will assume that the ground state and the Rydberg state are generated in a Doppler free configuration \([29]\). All atoms are initialized in the ground state \(|g\rangle\). The \(|g\rangle \rightarrow |r\rangle\) transition is coupled by a classical Raman pumping laser \( \epsilon_p \), of central frequency \( \omega_0 \) and Rabi frequency \( \Omega(r, t) \equiv eE(r, t) \cdot r_{eg}/\hbar \), where \( e \) is the electron charge. \( E(r, t) \) is the slowly varying envelope of the pumping field amplitude, and \( r_{eg} \) is the electric dipole moment of the corresponding atomic transition. The \(|r\rangle \rightarrow |s\rangle\) transition is coupled to the spontaneous emission field \( \epsilon_s \), which can be expanded into plane wave modes. In the interaction picture the Hamiltonian that models our system is thus

\[
H(t) = \left\{ \sum_{i=1}^{N_a} \sigma_{r, g}^i g(k) a_k^i e^{-i(k \cdot r_{i} - \omega_0 t)} + \sum_{i=1}^{N_a} \Omega(r_i, t) \sigma_{r, g}^i e^{i(k \cdot r_{i})} + H.c. \right\} + \Delta \sum_{i=1}^{N_a} \sigma_{r, s}^i 
\]

where the pumping field \( \epsilon_p \) is assumed to propagate along the \( z \) direction, \( \Delta = \omega_r - \omega_0 \) with \( \omega_0 = 0 \), the pumping laser frequency detuning, \( \sigma_{r, g}^i = |\alpha_i\rangle \langle \beta |, \beta = s, g, r \), the transition operators of the \( i \)th atom, and the coupling factor \( g(k) \) is dependent on the dipole moments of the corresponding transitions and on the direction of the wave vector \( k \) of the spontaneous emission field \( \epsilon_s \), which has a carrier frequency \( \omega_0 - \omega_s \) and frequency width determined approximately by the natural width \( \Gamma \) of the Rydberg state \(|r\rangle\). Thus the modes \( \omega_k \) with \( \omega_k - (\omega_0 - \omega_s) \gg \Gamma \) have scarcely any effect on the system evolution. For the purpose specified in the literatures \([16, 18, 19]\), the spontaneous emission back to the ground state \(|g\rangle\) is negligible.

On the condition that the detuning \( \Delta \) substantially larger than the natural width \( \Gamma \) of the level \(|r\rangle\) and the frequency spreading of the classical field \( \epsilon_p \), we may use standard methods \([30]\) to adiabatically eliminate the level \(|r\rangle\) from the system dynamics. The resulting Hamiltonian takes the form

\[
H(t) = \left\{ \sum_{i=1}^{N_a} \sigma_{r, g}^i \Delta \frac{\Omega(r_i, t)g(k)}{\Delta} a_k^i e^{-i(k \cdot r_{i} - \omega_0 t)} \right\} + H.c. \]  

\[
- \frac{1}{\Delta} \sum_{i=1}^{N_a} \sigma_{r, s}^i \Delta \frac{\Omega(r_i, t)g(k)}{\Delta} a_k^i + H.c. \]

where \( \Delta \omega_k = \omega_k - (\omega_0 - \omega_s) \) and \( \Delta k = k - k_0 z_0 \) with the \( z \) direction unit vector \( z_0 \). Because the spontaneous emission field is greatly weaker than the pumping field, the Stark shift of the level \(|s\rangle\) is far smaller than the other terms and has been omitted in the Eq. (2). The last term in Eq. (2) corresponding to the Stark shift of the level \(|g\rangle\) can be removed via a phase rotation of the basis \(|g\rangle, |s\rangle\) which will map \( \sigma_{r, g}^i \) into \( \sigma_{r, s}^i \). Thus the Hamiltonian can be written in the form

\[
H(t) = -\frac{1}{\Delta} \sum_{i=1}^{N_a} \sigma_{r, s}^i \Delta \frac{\Omega(r_i, t)g(k)}{\Delta} a_k^i + H.c. \]  

Thus the Hamiltonian can be written in the form

\[
H(t) = -\left\{ \frac{1}{\Delta} \sum_{i=1}^{N_a} \sigma_{r, s}^i \Omega(r_i, t)e^{i(k \cdot r_{i})} + H.c. \right\} - \left\{ \Delta \sum_{i=1}^{N_a} \sigma_{r, s}^i \Omega(r_i, t)e^{i(k \cdot r_{i})} + H.c. \right\} 
\]

Typically, the energy level \( \omega_s \) is either around GHz or zero, which relies on whether the hyperfine level or the Zeeman sublevel is chosen for the level \(|s\rangle\). \([19]\). The magnitude of the wavevector \( k \) of the spontaneous emission field in the summation \( \sum_k \) is about \( kc = \omega_k \in [\omega_0 - \omega_s - v/2, \omega_0 - \omega_s + v/2] \), where \( v \), the bandwidth of the field, is in the order of the natural width \( \Gamma \) of the excited level \(|r\rangle\) and can be smaller than 1GHz, \( c \) is the speed of light in the vacuum. Thus we may assume \( \omega_k = \omega_0 - \omega_s \). The length of the atomic ensemble \( L \) is assumed to be smaller than \( L \leq 10 \mu m \), then we have \( (k - k_0)L \leq 3.5 \times 10^3 \). The amplitude \( E_p \) of the pumping laser field can be adjusted to be independent of the coordinate \( r_i \) of the atoms in the ensemble, i.e., \( \Omega(r_i, t) = \Omega(t) \). Thus, the Hamiltonian of the system can be rewritten in the form

\[
H(t) = -\left\{ \frac{1}{\sqrt{N_a}} \Omega(t) S_s a_s^\dagger + H.c. \right\} 
\]

\[
-\left\{ \Delta \sum_{i=1}^{N_a} \sigma_{r, s}^i \Delta \frac{\Omega(r_i, t)g(k)}{\Delta} a_k^i e^{i(k \cdot r_{i})} + H.c. \right\} 
\]

\[
\cdot \sum_k g(k) a_k^i e^{-i(k \cdot r_{i} - \omega_0 t)} + H.c. \right\} 
\]

where

\[
S_s = \frac{1}{\sqrt{N_a}} \sum_{i=1}^{N_a} |g\rangle \langle s| 
\]
\[
\Omega'(t) = \frac{1}{\Delta} \Omega(t) e^{i \frac{1}{\hbar} \int \left[ \mathbf{a}^\dagger \mathbf{r} \right]^2 d\mathbf{r} g(kz_0),}
\]

\(a_s\) is the annihilation operator of the signal field mode which is the spontaneous emission field mode propagating in the \(z\) direction, and \(\sum_{\mathbf{k}}\) denotes that the summation of the wavevector \(\mathbf{k}\) does not include the signal field mode.

We see that the Hamiltonian can be divided into two parts: the first part describes the coherent collective interaction between the atomic ensemble and the signal field with an enhancing factor \(\sqrt{N_a}\), the second part describes the incoherent interaction between individual atoms and the spontaneous emission field (not including the signal field). To contain about \(N_a \sim 10^4\) cold alkali atoms within \(V < (10\mu m)^3\) through magnetic or optical traps is within the reach of current technologies [24,28]. Note that such an atomic ensemble is still dilute enough to fulfill the condition \(\kappa / \sqrt{\beta} \gtrsim 1\), which ensures that there is no superradiance, where \(\kappa\) is the atomic number density. According to the the literature [16], the signal-to-noise ratio \(\rho_{sn}\) between the coherent interaction rate and the incoherent rate can be estimated as \(\rho_{sn} \sim 3\rho^2 / \kappa^2\). To estimate the magnitude of the incoherent interaction, we assume \(3p = 10^2 \mu m^{-3}\), \(L = 8\mu m\), and \(\lambda = 0.5\mu m\). Then we have \(\rho_{sn} \sim 1.5 \times 10^2\). Note that the incoherent action only affects the success probability and doesn’t influence the fidelity of the obtained signal state [16]. Thus the incoherent individual atom-light interaction can be negligible compared with the enhanced coherent collective interaction, leading to a simpler Hamiltonian

\[
H(t) = - \left( \sqrt{N_a} \Omega(t) S^1_{r} a^\dagger_s + H.c. \right). \tag{7}
\]

This Hamiltonian describing a two-level system dynamics with the corresponding level \(|0_s, \rho\rangle\) and \(|S^1_{r} a^\dagger_s|0_s, \rho\rangle\) (Fig. 1b), where \(|0_s\rangle\) denotes the ground state of the atomic ensemble \(\otimes |g\rangle_l\) and \(|0_s\rangle\), the vacuum state of the signal light mode. The single-quantum collective excitation \(S^1_{r} a^\dagger_s|0_s, \rho\rangle\) can now be generated by a \(\pi\) pulse, \(\int^{T} \Omega(t) dT = \pi\).

### III. THE PROBABILITY OF HIGHER-ORDER EXCITATIONS

Now we discuss the higher-order excitations in the atomic ensemble during the process of generating single-quantum collective excitation. Because of the strong dipole-dipole interaction, the energy level of Rydberg excited atoms with separations of several \(\mu m\) \(|r\rangle\) is strongly shifted. Thus the presence of one Rydberg atom is enough to inhibit the excitation of all other atoms in the atomic ensemble. Doubly Rydberg excited collective states is defined as [24]

\[
|S^2_{r}\rangle \equiv \sqrt{\frac{N_a}{2(N_a - 1)}} S^1_{r} |0_s\rangle,
\]

where

\[
S_{r} = \frac{1}{\sqrt{N_a}} \sum_{i=1}^{N_a} |g_i\rangle_i \langle r|.
\]

The probability \(p_d\) of populating the doubly excited state \(|S^2_{r}\rangle\) is smaller than 1% for the situation where about \(N_a \sim 10^4\) cold alkali atoms is contained in \(V < (10\mu m)^3\) and \(T < 100\) ns [24,31]. According to a recent experimental report [25], the double-excitation probability for two Rydberg atoms with \(n = 90\) and separated by 10.2 \(\mu m\) is 0.02. Considering the probability \(p_{1r}\) of exciting a Rydberg state of an individual atom in the ensemble is far smaller than that of generating a single excited collective state \(|S^1_{r}\rangle\), the probability \(p_{2r}\) of exciting two atoms into Rydberg states (not collective state) is about \(p_{2r} \sim p^2_{1r}\) [16], and thus can be absolutely negligible.

Further, considering that the Raman pumping laser is far detuned from the transition \(|g\rangle \leftrightarrow |r\rangle\), the probability \(p_{1}\) of an atom populates the level \(|r\rangle\) is very low, and the probability of two atoms are excited in the Rydberg state are far more smaller than \(p_{1}\). Thus the doubly-excited collective state is absolutely negligible, if the duration \(T\) of the pumping laser field is no longer than the life-span of the Rydberg state \(|r\rangle\), which rules out the possibility with which two atoms in the ensemble are excited into the level \(|r\rangle\) in sequence. Thus we can generate a single-excited collective state \(S^1_{r} a^\dagger_s|0_s, \rho\rangle\) with near unity success probability at the same time with near unity fidelity.

Because the Rydberg states, in the ideal limit, never doubly populated, the scheme does not involve mechanical interaction between atoms and leaves the atomic internal state decoupled from atomic motion [24]. Thus the temperature only have to meet the condition that the atomic distribution should not vary substantially on the duration of the Raman pumping laser, which is the case for the temperature to be as high as a few mK [24].

### IV. CONCLUSION

We have present a method to describe the interaction between cold atomic ensembles and optical beams in the present of strong dipole blockade. The single-excited collective atomic state with a forward-scattered Stokes photon propagating along the same direction as the pumping laser does can be excited with a near unity success probability, at the same time, multi-excitation states can be significantly suppressed in the presence of the strong dipole blockade. With this advantage, this system can be used as an efficient on-demand single-photon source [31,32] which would give access to arbitrary small absorptions [33], improve the quality and the generation rate of random numbers [34], and play a fundamental role in quantum information processing, from Bell’s inequality test [35], quantum teleportation [36], linear-optics quantum computing [37,38], and quantum communication [10], to quantum cryptography [39]. Particularly, this scheme may open up a new possibility for scalable long-distance quantum communication.
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