Dephasing of multiparticle Rydberg excitations for fast entanglement generation

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An approach to fast entanglement generation based on Rydberg dephasing of collective excitations (spin waves) in large, optically thick atomic ensembles is proposed. Long range $1/r^3$ atomic interactions are induced by microwave mixing of opposite-parity Rydberg states. Required long coherence times are achieved via four-photon excitation and read-out of long wavelength spin waves. The dephasing mechanism is shown to have favorable, approximately exponential, scaling for entanglement generation.

Alkali atoms excited to Rydberg levels are attracting increasing attention as candidates for quantum computation on the MHz scale [1]. Various protocols for quantum computation and multiparticle entanglement using Rydberg level interactions have been proposed in recent years [2, 3]. These proposals rely on variations of the Rydberg blockade mechanism, where the presence of an excited Rydberg atom prevents (blocks) another atom, or atoms, from being excited [4, 5]. This approach has already been used to generate entanglement of pairs of Rb atoms [6]. The Rydberg blockade mechanism is in principle also applicable to create entanglement of collective excitations (spin waves), provided sufficiently small atomic ensembles are employed [5, 7]. This attractive capability could permit realizations of large scale, complex entangled matter-light systems. The basic requirement of large optical thickness of the atomic ensembles is, however, in conflict with the short range of the blockade radius. The challenge remains to achieve sufficient optical depth with small ensembles (<10 μm) using tight, densely populated optical lattices and/or optical cavities.

In this Letter we propose an alternative approach that alleviates the difficulties of the small sample-blockade mechanism and makes it possible to realize fast entanglement generation and distribution in large, free-space atomic ensembles. Rather than trying to prevent multiple excitations via the Rydberg blockade mechanism, our idea is to allow multiple Rydberg level excitations to self-interact and dephase. The interaction-induced phase shifts suppress the contribution of multiply excited states in phase matched optical retrieval. The dephasing mechanism therefore permits isolation and manipulation of individual spin wave excitations.

The strong interaction required to dephase multiple excitations is induced by mixing adjacent, opposite-parity Rydberg levels with a microwave field [8]. These levels experience resonant dipole-dipole interactions ($ns + n′p → n′p + ns$) that extend over the whole ensemble in contrast to the weaker, short range Van der Waals coupling due to non-resonant processes ($ns + ns → np + (n−1)p$).

We consider a cloud of cold alkali atoms. Since the procedure we propose is fast compared to atomic motional timescales in a cold ensemble, we assume that the positions of the atoms are fixed and postpone discussion of motional effects until later. The relevant atomic levels are sketched in Fig. 1, the ground level $|g⟩$, the first excited level $|e⟩$, the target Rydberg level $|s⟩ = |ns⟩$ and the Rydberg level used for the dephasing protocol $|p_j⟩ = |n′p_j⟩$, $j = 1/2, 3/2$ to be discussed below. We suppress the state magnetic quantum numbers $m$ assuming that optical pumping of the ground level produces laser excitation of only a selected Zeeman state of the target Rydberg level; the coupling of magnetic sublevels by Rydberg interactions will be included in the results presented below. While we consider a target $s$-orbital Rydberg level, it is possible to apply the formalism to $d$ levels as well.

Figure 1. Ground level atoms are two-photon excited to a Rydberg $s$-level, which is then mixed with a $p$-orbital by applying a microwave pulse of Rabi frequency $Ω$. The inset shows the effect of dipole-dipole interaction and microwave dressing of an atom pair. The atomic spin-wave is retrieved from the Rydberg state with a $π$-pulse resonant to a low-lying excited state.

Rydberg level excitation: The target Rydberg level $|ns⟩$ is laser excited through a two-photon resonant transition with large single photon detuning $Δ_1$. The effective excitation Rabi frequency is $Ω_{2ph} = Ω_1Δ_2/(2Δ_1)$ where $Ω_1$ and $Ω_2$ are the Rabi frequencies of the lasers (see Fig. 1).

The ensemble, defined by the region illuminated by the
waist of the excitation laser (typically 50-60 μm), contains \( N \gg 1 \) atoms. We may assume the atoms are initially independent since we consider (i) short laser pulses, whose bandwidth determines a Rydberg blockade radius \([1]\) that is much smaller than the size of the ensemble, and (ii) the pulse duration creates on average a single excitation, \( T \approx 1/(\sqrt{\Omega N \delta_{ph}}) \). Thus laser excitation produces the state \( |\Psi_0\rangle = \sum_{\alpha} c_{\alpha} |\alpha\rangle \), where \( |\alpha\rangle \) contains excitation with wavevector equal to the sum of the excitation laser wavevectors, \( k_0 = k_1 + k_2 \), and amplitude \( c_{\alpha} = \sqrt{\pi/\alpha!} \) associated with a Poissonian distribution of unit mean. The maximum efficiency of the single spin-wave preparation is \( |c_1|^2 = 1/\epsilon \). In the limit of weak excitation, \( |\alpha_{k_0}\rangle \approx (\zeta^k_{k_0})^n|0\rangle/\sqrt{\alpha!} \), where \( \zeta^k = (1/\sqrt{\alpha}) \sum e^{-ik \cdot r_i} |\mu\rangle \) is the spin-wave annihilation operator and the spin-wave vacuum \( |0\rangle \) has all the atoms in the ground state: \( |\Psi_0\rangle \) corresponds to a coherent state of spin waves.

**Microwave-dressed Rydberg levels:** Atoms laser excited to the Rydberg state \( |s\rangle \) are assumed to have negligibly weak Van der Waals interactions. A microwave field couples the transition \( |s\rangle \rightarrow |p\rangle \), transferring population to the \( |p\rangle \) level and thus inducing a long-range resonant dipole-dipole interaction \([3]\) (see Fig. 1).

The interaction Hamiltonian for the atomic system is given by \( H_I = \sum_{\mu, \nu} V_{\mu}^\nu + \sum_{\mu < \nu} V_{\mu \nu}^d \); the first term contains the electric dipole interaction of each atom \( \mu \) with the microwave field: \( V_{\mu}^\nu = -d_{\mu} \cdot \hat{E}(t) (\mu \epsilon e^{-i\omega t} + \epsilon^* e^{i\omega t})/2 \), where \( d_{\mu} \) is the electric dipole moment of atom \( \mu \) and \( \{\epsilon, \omega, \hat{E}(t)\} \) are the polarization vector, angular frequency and time-dependent amplitude of the electric field. The Rabi frequency for the coupling between \( |s\rangle \) and \( |p\rangle \) is defined as \( \hbar \Omega(t) = \hat{E}(t) \cdot D \), where \( D \) is the reduced matrix element for the transition. The spatial phase of the microwave field is suppressed because the size of the sample is much smaller than its wavelength. The dipole-dipole interaction between atoms is given by

\[
\hat{V}_{\mu \nu}^{dd} = \frac{1}{4\pi\epsilon_0 R^3} \left[ \frac{1}{R^2} \left( \frac{3 (d_{\mu} \cdot \mathcal{R}) (d_{\nu} \cdot \mathcal{R})}{R^2} - d_{\mu} \cdot \mathcal{R} - 3 (d_{\nu} \cdot \mathcal{R}) (d_{\mu} \cdot \mathcal{R}) / R^2 \right) \right] \quad (1)
\]

where \( \mathcal{R} \) is the interatomic separation. The dipole-dipole matrix element for a given angular momentum channel may be written as \( C_{3\mu}/R^3 \) \([1] [3] [5]\) and may be calculated with a semiclassical approach \([10]\). We ignore non-resonant Van der Waals processes \( C_{6\mu}/R^6 \) and retain only the resonant couplings \([3] [11]\).

**Dephasing protocol:** Resonant dipole-dipole interactions cause phase shifts of polarized Rydberg atom pairs, triples, etc. The accumulated phase decouples these excitations from the phase matched radiation mode during the retrieval process. With a suitable protocol, we can take advantage of this dephasing to generate high quality single photons. A single channel model of the interaction is sufficient to demonstrate the physics of the protocol.

We consider a Ramsey-like 2π-pulse sequence: a single-atom \( \pi/2 \) microwave pulse that polarizes the Rydberg atoms is followed by an interval \( \Delta T \), during which resonant interactions occur and is terminated by a restoring \( 3\pi/2 \)-pulse. A many-body state containing a single Rydberg excitation undergoes a complete rotation. A given atom pair prepared in the target Rydberg level \( |s\rangle \) experiences the following transformation assuming the regime of strong dressing, \( \Omega \gg V_{dd} \): (a) the \( \pi/2 \)-pulse is responsible for the evolution \( |s\rangle |s\rangle \rightarrow (1/2)(|s\rangle |s\rangle - |p\rangle |p\rangle + i(|p\rangle |s\rangle + |s\rangle |p\rangle) \), (b) during the interval \( \Delta T \) the state transforms to \((1/2)(|s\rangle |s\rangle - |p\rangle |p\rangle + i e^{i\varphi} (|p\rangle |s\rangle + |s\rangle |p\rangle)\), where the phase \( \varphi = V_{dd} \Delta T / \hbar \), (c) the \( 3\pi/2 \)-pulse \(|s\rangle \rightarrow 1/\sqrt{2}(|s\rangle - |p\rangle) \) and \(|p\rangle \rightarrow 1/\sqrt{2}(|s\rangle - |p\rangle) \), completes the overall transformation that maps \( |s\rangle |s\rangle \) into

\[
|\chi(\varphi)\rangle = e^{i\varphi/2} \langle s|s\rangle \cos \left( \frac{\varphi}{2} \right) + i|p\rangle|p\rangle \sin \left( \frac{\varphi}{2} \right). \quad (2)
\]

In the limit \( \varphi \rightarrow 0 \), we recover the initially prepared atom pair excitation \(|s\rangle |s\rangle \) and more generally the survival amplitude for this state is \( e^{\varphi^2/2} \cos(\varphi/2) \). The accumulated phase will be different for each atom pair. As a consequence, the probability for emission of a photon pair in the phase matched mode is reduced through destructive interference of distinct atom pair contributions, as we argue below. A small number of atom pairs will experience Rydberg blockade \( \Omega \ll V_{dd} \) and while their effect is not detrimental it is anyway negligible.

We may write the many-body state after the Ramsey 2π-pulse as \(|\Psi\rangle = c_0|0\rangle + c_1|1_{k_0}\rangle + c_2|\Psi^{(2)}\rangle + O(c_3)\), by defining

\[
|\Psi^{(2)}\rangle = \sqrt{\frac{1}{N}} \sum_{\nu > \mu = 1} e^{i(k_0 \cdot (r_\mu + r_\nu))} |\chi(\varphi_{\mu \nu})\rangle_{\mu \nu}. \quad (3)
\]

Here \( \varphi_{\mu \nu} \) is the phase induced on the atom pair \((\mu, \nu)\), \( N = N(N - 1)/2 \) is the number of distinct pairs and \(|1_{k_0}\rangle = S_{k_0}^{(1)}|0\rangle \) is the one spin wave state.

**Optical readout procedure:** Rydberg excitations in the \(|s\rangle \) level can be optically retrieved by a laser of wavevector \( k_3 \), transferring the population to an intermediate state which decays to the ground level as shown in Fig. 1. The quality of single-photon emission is evaluated through measurement of the normalized correlation function, \( g^{(2)} = \langle \hat{a}_{k_0}^{\dagger} \hat{a}_{k_0} \hat{a}_{k_0} \hat{a}_{k_0}^{\dagger} \rangle / \langle \hat{a}_{k_0}^{\dagger} \hat{a}_{k_0} \rangle^2 \), in the phase matched direction, \( k_0 = k_0 - k_3 \). Here \( \hat{a}_{k_0}^\dagger \) is the annihilation operator for photons in the phase matched mode. Analysis of the decay process shows that we may write \( \hat{a}_{k_0}^\dagger = \sqrt{\eta} S_{k_0} + \sqrt{1 - \eta} \xi \), where the operator \( \xi \) describes vacuum fluctuations, and \( \eta \) is the coupling efficiency. Hence the correlation function becomes

\[
g^{(2)} = \langle \hat{S}_{k_0}^\dagger \hat{S}_{k_0}^\dagger \hat{S}_{k_0} \hat{S}_{k_0} \rangle / \langle \hat{S}_{k_0}^\dagger \hat{S}_{k_0} \rangle^2. \]

The phase-matched retrieval of Poissonian excitations corresponds to the value \( g^{(2)} = 1 \). For simplicity, we focus on the effects of single and double excitations and truncate the many
particle coherent state beyond this point: $c_α = 0$ for $α > 2$. The corresponding value of $g^{(2)}$ for the truncated coherent state is $g^{(2)}(0) = e/4$. The state $|Ψ⟩$ yields

$$g^{(2)} = \frac{4g^{(2)}(0)}{[1 + \frac{1}{N^2} \sum_{ν=1}^{N} \sum_{ν≠μ} e^{i \hat{ϕ}_{μν}} \cos (\frac{2π}{N})]}^2,$$

in the spin-wave approximation $(N - 1)/N \approx 1$.

This expression clearly shows that the probability for photon pair emission is a superposition of the atom pair contributions determined by the Ramsey protocol in (2): the broader the distribution of phases $ϕ_{μν}$ the more effective is the destructive interference of two-photon amplitudes.

Since the phases $ϕ_{μν}$ depend on time, we rewrite the correlation function of Eq. (4) as $g^{(2)}(t) = 4g^{(2)}(0)f(t)/(1 + h(t))^2$, which defines $f(t)$ and $h(t)$. At $t = 0$, it is trivial to verify that $f(t) = h(t) = 1$. For $t \rightarrow \infty$, assuming the atoms are randomly distributed in the ensemble, the accumulated phase behaves like a random variable: $(e^{iπ/2} \cos (\pi/2)) = 1/2$, which gives $f, h \rightarrow 1/4$. We therefore predict the asymptotic value of the correlation function after the Ramsey protocol to be $g^{(2)} \rightarrow g^{(2)}(0) 16/25$.

![Figure 2. Decay of the correlation function $g^{(2)}$ in the phase matched mode versus interaction time $ΔT$ for a single Ramsey $2\pi$-pulse cycle in the strong dressing limit. Principal quantum numbers: $n = n' = 60$ (Blue squares), $n = n' = 79$ (Red diamonds) and $n = n' = 100$ (Green circles). Black dashed line represents the asymptotic limit $g^{(2)}(0) 16/25$. Inset: effect of repeated cycles for $n = 100$ with $ΔT = 1μs$ and $Ω = 10^{3}s^{-1}$; first cycle $n' = 100$, $j = 1/2$, second cycle $n' = 99$, $j = 1/2$, third cycle $n' = 100$, $j = 3/2$, fourth cycle $n' = 99$, $j = 3/2$. Dark regions correspond to the duration of each 2$π$ microwave pulse cycle.](image)

This asymptotic value agrees with simulations of a multichannel theory which also shows a stronger subpoissonian dip at short times, Fig. 2. As an example we take Rb comparing the time dependence for the Rydberg levels with principal quantum numbers $n = (60, 79, 100)$. We consider $N = 100$ atoms randomly distributed in a cubical box with side $L = 60μm$. The correlation function in Eq. (4) is found by solving the two-body Schrodinger equation for each atom pair. The fine-structure transitions $|ns⟩-|np_{1/2, 3/2⟩}$ have (16,36) available coupled channels involving different combinations of the magnetic quantum numbers, respectively. While the asymptotic value of $g^{(2)}$ is independent of $n$, we observe that the rate of dephasing, as measured by the temporal position of the minimum, scales in inverse proportion to the strength of the interaction $V_{dd} \propto n^4$.

**Single photon generation:** Although a single Ramsey $2π$-pulse cycle produces subpoissonian emission statistics asymptotically, the more pronounced and fast initial dephasing of $g^{(2)}$ suggests an improved protocol in which i) the value of $ΔT$ optimizes the transient dephasing, and ii) repetitions of the Ramsey cycle further reduce $g^{(2)}$ by the replacement

$$e^{i\hat{ϕ}_{μν}} \cos (\frac{1}{2}\hat{ϕ}_{μν}) \rightarrow \prod_{q=1}^{R} e^{i\hat{ϕ}_{μν}} \cos (\frac{1}{2}\hat{ϕ}_{μν})$$

in Eq. (4). In each of the repeated cycles, it is essential that the microwave field couples the $|ns⟩$ target level to a different and unpopulated $|np_{j}⟩$ level otherwise the coherence established with the target level invalidates Eq. (4). As each cycle represents a single-particle $2π$-pulse the single photon contribution is unaffected throughout.

For $n \gg 1$, the channels $|ns⟩ \leftrightarrow |np_{j}⟩$ and $|ns⟩ \leftrightarrow |n' p_{j}⟩$ have similar interaction strengths and may each be employed in the repeated Ramsey protocol. In Fig. 2 inset, we show a sample evolution of $g^{(2)}$ for a sequence of $R = 4$ Ramsey cycles with $n = 100$ and $j = 1/2, 3/2$. We plot the function $e^{-T/τ}$ where $τ = ΔT + 2π/Ω$ is the duration of a single cycle. The comparison shows that the decay rate of $g^{(2)}$ is approximately exponential and high quality single excitations are generated in several $μs$, well within the lifetime of this Rydberg state. For comparison, the DLCZ protocol [12, 13], which has a typical trial period $~1μs$, and excitation probability $~10^{-3}$, generates single excitations in a time of order 1 ms [14].

**Entanglement of Rydberg spin waves:** We have shown that the repeated Ramsey protocol allows the fast creation of a single spin wave, stored in the Rydberg level $|ns⟩$, coupled to the phase matched mode. An independent spin wave associated with the orbital $|n's⟩$ can also be generated provided the dipole-dipole interaction does not cause interference of the dephasing protocols for the levels $n$ and $n'$. For $n \gg 1$, the dipole coupling between Rydberg levels $ns$ and $n'p$ decays rapidly as the energy difference increases: for example, for $n = 100$ the interaction strength between adjacent 100s and 99$p_{j}$ or
100p\textsubscript{j} orbitals is 100 times larger than that for 100s or 98p\textsubscript{j}. These single spin waves may also be transported through the Rydberg spectrum by using single-particle microwave \(\pi\)-pulses: the phase matching condition is not affected provided the Rydberg transition wavelength is much larger than the ensemble size.

A protocol to entangle two independent spin waves in levels \(n\) and \(n' = n + 1\) is sketched in Fig. 3. We define the operators \(|n\rangle\) and \(|np_{1/2}\rangle\) respectively, as level \((n+1)s\) levels to \(|np_{1/2}\rangle\) and \(|np_{3/2}\rangle\) respectively, transforming \(\hat{S}^\dagger_{n+1}\hat{S}^\dagger_{n}|0\rangle\rightarrow (\hat{S}^\dagger_{n+1}\hat{S}^\dagger_{n}|0\rangle - \hat{P}^\dagger_{n/2}\hat{P}^\dagger_{n/2}|0\rangle + i(\hat{S}^\dagger_{n+1}\hat{P}^\dagger_{n/2}|0\rangle + \hat{P}^\dagger_{n/2}\hat{S}^\dagger_{n}|0\rangle))/2\). The resonant dipole-dipole interactions couple the \(s\) and \(p\) orbitals thus inducing different phase shifts for each atom pair. The system evolves into the many body state \(|0\rangle + c_{1,0}/2(\hat{S}^\dagger_{n+1} + \hat{S}^\dagger_{n} + i\hat{P}^\dagger_{n/2} + i\hat{P}^\dagger_{n/2})(|0\rangle + c_{1,0}^2)|\Phi\rangle\) with

\[
|\Phi\rangle = \frac{1}{2} \left[ \hat{S}^\dagger_{n+1}\hat{S}^\dagger_{n}|0\rangle - \hat{P}^\dagger_{n/2}\hat{P}^\dagger_{n/2}|0\rangle + i \frac{i}{N} \sum_{\mu,\nu} |\xi_{\mu\nu}\rangle \right].
\]

The motional dephasing of optical ground-Rydberg co-

erences has been a serious problem in exploiting Ryd-

berg atom interactions [4]. While atom trapping would in

principle alleviate this effect, so far no effective Ryd-

berg atom confinement schemes have been demonstrated,

although there are promising works in that direction [16].

For a cold MOT of Rb the average atomic velocity \(v \sim 0.1\)

m/s, while the spin wave grating period for two-photon

excitation is only \(\Lambda \sim 1\) \(\mu m\), giving a coherence time

\(\Lambda/(2\pi v) \sim 2\) \(\mu\text{s}\). In order to overcome this limitation, we

propose the four-photon excitation scheme shown in Fig.

3(b) for atomic Rb. In this case the four wavevector

mismatch can be made equal to zero and the corresponding

spin wave period diverges, thereby eliminating motional

decoherence. Since all the transitions involved possess

strong dipole moments, the Rabi frequency can easily

exceed several MHz with available laser powers. An opti-

cal depth of 10 is achievable with a gas of density \(10^{12}\)

cm\(^{-3}\) and diameter 50 \(\mu m\), sufficient for photon retrieval

in the phase matched mode [17]. The maximum efficiency

of single spin-wave preparation, and hence single-photon

generation, is given by \(1/e\).

In conclusion, we propose techniques for the fast cre-

ation of single quantum excitations and entanglement of

such excitations in large atomic ensembles, suitable for

efficient light-matter state transfer. The protocols we

propose are based purely on the dephasing of multiple

excitations due to resonant \(1/r^3\) dipole-dipole interac-

tions induced by microwave coupling of opposite parity

Rydberg states. In the future, it will be interesting to in-

vestigate an intermediate regime between blockade and

dehasing that optimizes efficiency, speed and error prob-

ability for laboratory implementation.

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Figure 3. a) Atomic levels involved in entanglement of two spin-wave excitations via a protocol described in the text. b) Four-photon excitation of Rydberg spin-waves in atomic Rb, with \((\lambda_1, \lambda_2, \lambda_3, \lambda_4) = (795, 1475, 2294, 1005)\) nm. Collinear and off-axis geometries lead to spin waves of period 50 \(\mu m\), and \(\infty\), respectively.
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