

On-Demand Superradiant Conversion of Atomic Spin Gratings into Single Photons with High Efficiency

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We create quantized spin gratings by single-photon detection and convert them on demand into photons with retrieval efficiencies exceeding 40% (80%) for single (a few) quanta. We show that the collective conversion process, proceeding via superradiant emission into a moderate-finesse optical resonator, requires phase matching. The storage time of $3 \mu\text{s}$ in the cold-atom sample, as well as the peak retrieval efficiency, are likely limited by Doppler decoherence of the entangled state.

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Atoms isolated in vacuum offer the longest storage times for quantum bits, with possible coherence times of several seconds [1]. However, massive particles are ill suited for transmitting quantum states in view of the particle velocity and vacuum requirements. In contrast, photons are ideal carriers of quantum information, but are not easily stored. The conversion of quantum states between atomic and photonic representations is thus the subject of much recent interest. Proposed applications include single-photon sources [2,3], quantum repeaters for cryptography and teleportation [4,5], and quantum computation with linear optics and detection [6].

In order to achieve coherent coupling between matter and light, the atomic system must appear opaque to the photon. For a single atom, this can be accomplished by strong coupling to an electromagnetic mode in an extremely high-finesse optical cavity [2], but the technical requirements are stringent. An alternative approach, not requiring a resonator, is to prepare an entangled state of a many-atom sample (Dicke state [7] or spin grating [4]) that couples collectively to a free-space electromagnetic mode [3,8–12]. To reach high fidelity in the conversion process, the optical depth $N\eta$ of the sample must be large [4], where N is the number of atoms and η is the mode's single-atom optical depth or cooperativity parameter. Pursuing one version of the original idea by Duan, Lukin, Cirac, and Zoller for a conditional single-photon source [4], we adopt a hybrid approach where an atomic ensemble interacts collectively with a moderate-finesse optical resonator operating in the weak-coupling regime for a single atom ($\eta \ll 1$), but in the strong-coupling regime for the sample ($N\eta \gg 1$). The resonator allows $N\eta \gg 1$ even for an optically thin atomic sample, facilitating near-unity fidelity in the conversion between atomic and photonic quantum states.

We demonstrate the conversion of quantized atomic-spin gratings into photons with peak efficiencies exceeding 40% (80%) for single (a few) quanta of excitation. The collective coupling of the spin grating to the cavity mode is evident in a superradiantly enhanced rate of photon emis-

sion, requires phase matching, and persists for as long as $3 \mu\text{s}$, likely limited by the atoms' thermal motion. This system has the potential for simultaneous holographic storage of many spin gratings that can be independently converted into photons on demand, and for suppressing Doppler broadening by trapping the atoms in the Lamb-Dicke regime, promising quantum-state storage times in excess of 1 s.

Three other groups [3,9–13] have recently made excellent progress generating delayed, correlated pairs of photons. Reference [11] reports peak conversion efficiencies in the few-excitation limit of 30% and decoherence times of $3 \mu\text{s}$ using a room-temperature vapor. In Ref. [3], photon pairs displaying very nonclassical statistics within a decoherence time of $0.2 \mu\text{s}$ are generated in a laser-cooled atomic sample. Reference [13] reports the preparation and readout of a quantum memory by single-photon emission from two regions of a laser-cooled gas. Very recently, another group has generated pairs of simultaneous, strongly correlated photons from laser-cooled atoms [14].

Quantum information can be stored in a single Λ -configuration atom as a superposition of two stable ground states $|0\rangle$ and $|1\rangle$. The stored information can be converted into a photon by spontaneous Raman emission

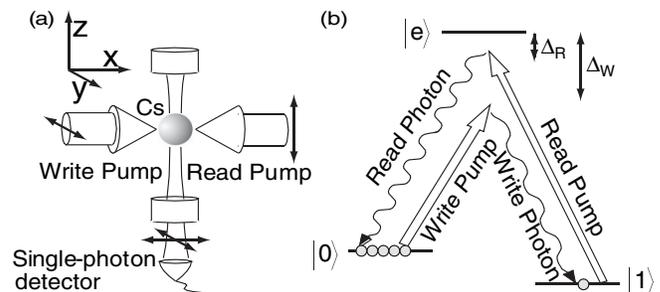


FIG. 1. (a) Experimental setup for the creation, storage, and on-demand retrieval of quantized spin gratings in an atomic sample by single-photon detection. Beam propagation directions, linear polarizations, and optical-cavity orientation are indicated. (b) Idealized atomic energy level diagram.

[see Fig. 1(b)], but the conversion is not useful unless the photon is emitted into a small solid angle. Directional photon emission can be achieved by storing the information as a Dicke state of a many-atom sample and taking advantage of superradiance [7]. Consider an atomic sample located inside an optical resonator and prepared in state $|0\rangle$ by optical pumping. When the sample Raman scatters a photon from an applied pump laser into the cavity [Fig. 1(a)], it is not possible to determine which of the N atoms has made the transition $|0\rangle \rightarrow |1\rangle$. The total wave function of the sample must be symmetrized with respect to which atom carries the excitation. Each of the N terms in this entangled state (the first excited Dicke state [7]) contains a phase factor $\exp(i(\vec{k}_{\text{wa}} - \vec{k}_{\text{we}}) \cdot \vec{x}_j)$ that depends on the spatial position \vec{x}_j of the atom j carrying the excitation and on the wave vectors of the absorbed and emitted write photons \vec{k}_{wa} and \vec{k}_{we} . The Raman emission process thus “writes” a collective quantum excitation, or spin grating [4], into the sample.

After a variable delay time, the atoms are then illuminated by a “read” pump beam driving the reverse Raman transition. This field mixes the metastable ground state $|1\rangle$ with the excited state $|e\rangle$ and converts the spin grating into an optical polarization grating whose spatial phase depends on the spatial phases of the spin grating and the read pump. For an appropriate geometry, the N terms of the Dicke state constructively interfere to provide an N -fold enhancement of the rate for emitting the second photon into the cavity compared to the single-particle emission rate. The atomic sample thus forms the quantum analogue of a phased array of antennas with highly directional emission. The phase-matching condition for constructive interference requires that the sample be restored to the initial state, or that zero net momentum be transferred to the atom in the combined write-read process. The read photon is therefore super-radiantly emitted into the direction $\vec{k}_{\text{re}} = -\vec{k}_{\text{we}} + \vec{k}_{\text{wa}} + \vec{k}_{\text{ra}}$, where \vec{k}_{ra} is the wave vector of the absorbed read pump photon.

The probability of successfully emitting a photon into the desired mode is determined by the collective coupling to the preferred mode compared to all other modes [4]. If we denote the single-atom Raman scattering rate into free space by Γ_{sc} and the cavity decay rate by κ , the majority of the present work is conducted in the limit $\kappa/\Gamma_{\text{sc}} > N\eta \gg 1$ of overdamped superradiance [15], wherein the atoms radiate irreversibly into the cavity mode at a rate $\Gamma_{\text{cav}} = N\eta\Gamma_{\text{sc}} < \kappa$, so that the emission is adequately described by rate equations [7]. Neglecting decoherence, the success probability $P_s = N\eta/(1 + N\eta)$ for conversion of the stored excitation into a photon in the cavity mode is limited by competition with other modes, and approaches unity for $N\eta \gg 1$. Compared to free-space systems, the cavity not only enhances the desired emission rate, but also can filter out nonresonant light scattered from the pump beams. Furthermore, it allows us to use small samples that are optically thin in every direction but the cavity axis, facil-

itating optical pumping and other manipulation of the atoms.

The near-confocal standing-wave resonator used in this experiment has finesse $F = 1000$, linewidth $\kappa/2\pi = 2$ MHz, and a TEM₀₀ mode waist size $w = 100$ μm . The wave number of the emitted photon is $k = 2\pi/\lambda$, where $\lambda = 852$ nm. The single-atom cooperativity is $\eta = 12F/(\pi k^2 w^2) = 6.9 \times 10^{-3}$. Photons emerging from the resonator are detected using single-photon counting modules (SPCM-AQR-12-FC from PerkinElmer) with an overall efficiency of $Q = 0.05$, limited by cavity mirror losses. All photon numbers quoted below are detected photon numbers multiplied by $1/Q$. A magneto-optical trap (MOT) provides $N \approx 10^4$ cold Cs atoms at a temperature between 10 and 30 μK within the TEM₀₀ mode volume. Thus we expect $N\eta \approx 70$ and a success probability $P_s = 98\%$ for the conversion of a single spin-grating excitation into a photon in a single transverse mode.

The states $|0\rangle$ and $|1\rangle$ are hyperfine levels $|F, m_F\rangle$ of the Cs ground state $6^2S_{1/2}$, and the intermediate state $|e\rangle$ belongs to the $6^2P_{3/2}$ manifold. We choose $|0\rangle = |4, 4\rangle$, and either transitions for which the hyperfine quantum number F changes (“hyperfine storage”), $|1\rangle = |3, 3\rangle$, or transitions for which only the magnetic quantum number m_F changes (“magnetic storage”), $|1\rangle = |4, 3\rangle$.

To prepare the atoms for a write-read sequence, the magnetic field of the MOT is switched off 5 ms in advance to reduce Zeeman broadening. The atoms are optically pumped into the initial state $|0\rangle = |4, 4\rangle$, with the quantization axis set along \hat{y} by a uniform magnetic field of 0.1 mT [Fig. 1(a)]. For hyperfine (magnetic) storage, the \hat{y} -polarized (\hat{z} -polarized) write pump transfers atoms to the state $|1\rangle = |3, 3\rangle$ ($|1\rangle = |4, 3\rangle$) by the emission of σ^+ -polarized (π -polarized) photons. The \hat{z} -polarized (\hat{y} -polarized) read pump allows a collectively enhanced transition back to $|0\rangle$ via cavity emission of a π -polarized (σ^- -polarized) photon. For hyperfine storage, we easily observe background count rates below 10 kHz. For magnetic storage, the pumps are near resonant with the cavity, resulting in 1 MHz background count rates due to superradiant Rayleigh scattering from atoms in $|4, 4\rangle$.

In the hyperfine scheme, the frequencies of the write and read pumps are set such that the write and read photons are emitted into the same transverse but two distinct longitudinal cavity modes, separated by five free spectral ranges or 5×2 GHz = 10 GHz, close to the Cs ground-state hyperfine splitting of 9.2 GHz. The write and read beams with peak powers of 30 and 2 mW are detuned by $\Delta_w/2\pi \approx -900$ MHz and $\Delta_r/2\pi < -100$ MHz from the transitions $6^2S_{1/2}$, $F = 4$ to $6^2P_{3/2}$, $F' = 4$ and $6^2S_{1/2}$, $F = 3$ to $6^2P_{3/2}$, $F' = 4$, respectively. To ensure good spatial mode matching, the two orthogonally polarized beams are transmitted through the same single-mode optical fiber, expanded to waist size 1.7 mm, split by a polarizing beam splitter, and directed onto the atoms from opposite directions.

Figure 2 demonstrates the writing of a hyperfine spin grating into the ensemble (thick line) and the conversion of the excitation into read photons in the cavity mode after a programmable delay of up to several μs by switching on the read pump (thin lines). The curves indicate the average time dependence of the emitted photons. No collective read emission is detected unless the write pump is applied first. In this data set, the average number of write photons scattered into the cavity is $M_w = 10$, while the inset demonstrates efficient readout with recovered fraction $R = 0.57 \pm 0.15$, at an average stored excitation number much smaller than 1, $M_w = 0.06$.

The read photons are emitted 30 times more promptly ($0.3 \mu\text{s}$ typical pulse widths) than the measured single-particle optical pumping time of $\tau = 11 \mu\text{s}$ for hyperfine storage. This ratio is in agreement with the expectation that the collective emission rate by the grating is $N\eta$ times enhanced over the single-particle rate. In the magnetic sublevel scheme at large read pump intensities such that $N\eta\Gamma_{\text{sc}} \geq \kappa$, we observe lengthening of the read emission pulse duration, indicating that Rabi flopping is becoming important in the collective atom-photon coupling. The description of the excitation conversion process is more appropriately described as a dark-state rotation in this limit [16].

To further verify the collective nature of the excitation storage, we examine the phase-matching conditions. First, the recovery efficiency R was measured as a function of a small angle between the two pump beams (see Fig. 3), which are approximated as plane waves. If the pumps deviate from counterpropagation by a small angle in the \hat{x} - \hat{y} plane, an impulse perpendicular to the cavity axis is imparted to the atoms. This momentum can be partially canceled even if the write and read photons are both emitted into the same cavity mode, since the cavity modes have an angular spread of $\phi = \lambda/(\pi w)$ given by each

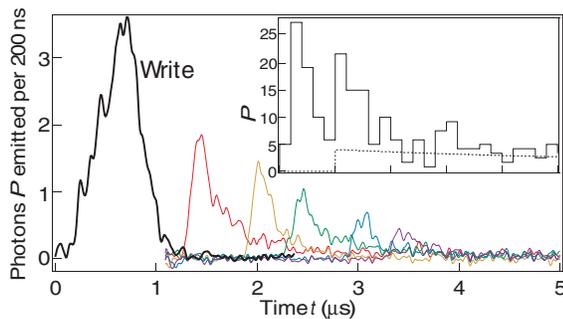


FIG. 2 (color online). Conversion of quantized spin grating into photons. The emission probability P per 200 ns (detected photon number multiplied by $Q^{-1} = 20$) is shown versus time t . When the write pump is applied for 600 ns, the sample scatters photons into the cavity (thick black line). Application of the read pump after various time delays results in collective emission (thin lines). The inset shows $P(t)$ at a mean emitted write photon number of $M_w = 0.06$ and a retrieval efficiency of $(57 \pm 15)\%$. The dashed line indicates the read background.

mode's far-field diffraction pattern. A simple overlap integral then predicts a $1/e^2$ width in R versus angle of $\Delta\phi = \phi/\sqrt{2} = 1.9 \text{ mrad}$ for the TEM_{00} mode. The measured efficiency displays a sharp peak of $1/e^2$ size $0.9(1) \text{ mrad}$, on top of a broader pedestal of half width half maximum of 12 mrad . The observed line shape results from emission into multiple (5 or more) nearly degenerate transverse modes, as is expected for our near-confocal cavity with 1 MHz transverse-mode spacing, corresponding to about one-fourth the write laser width.

For antiparallel pump beams, phase matching requires the read and write photons to be emitted into the same transverse cavity mode. The read pump frequency and cavity length determine the mode for read photon emission. To test transverse-mode matching, the write pump frequency is scanned, varying the write emission mode (inset of Fig. 3). We observe maximum retrieval when the read photons are scattered into the same transverse mode as the write photons. If the retrieval were not collective, the efficiency would be independent of the transverse mode into which the write photons were emitted.

The thermal motion of the atoms introduces velocity-dependent phase factors, resulting in decoherence of the superradiant state. In Fig. 4(a), the fractional recovery R in the hyperfine (magnetic) storage data decays from peak values of 30% (80%) with a characteristic time scale of $3.4 \mu\text{s}$ ($1.7 \mu\text{s}$). This time scale agrees well with the Doppler time τ_D for the four-photon write-read process or, equivalently, the time for the atoms to move by a spin-grating wavelength, with values $1.6 \leq \tau_D \leq 2.7 \mu\text{s}$ for typical temperatures $10 \leq T \leq 30 \mu\text{K}$.

This Doppler decoherence is also believed to limit the peak recovery efficiency, since the time to write and read the spin-grating excitation can be a significant fraction of the Doppler time τ_D . To demonstrate this, R is plotted for both configurations versus the single-particle scattering rate Γ_{sc} due to the read pump in Fig. 4(b). The increase

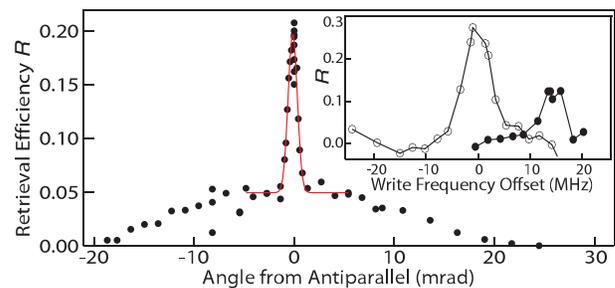


FIG. 3 (color online). Tests of phase matching. The retrieval efficiency R is plotted versus angle between the write and read pump beams. A Gaussian fit (solid line) to the central peak indicates a $1/e^2$ width of $0.9(1) \text{ mrad}$. In the inset, retrieval efficiency is plotted versus write pump frequency for two different read pump frequencies, so that the transverse cavity mode for which read emission is resonant differs by 15 MHz between the two curves. Each curve is taken with the cavity frequency and read pump frequency fixed.

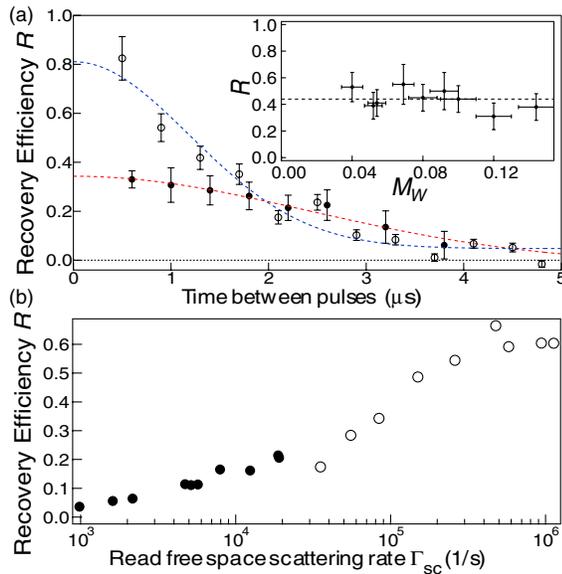


FIG. 4 (color online). Retrieval efficiency R of hyperfine (solid dots, $M_w = 1$) or magnetic (open circles, $M_w = 120$) excitations. M_w is the detected write photon number multiplied by the inverse detection efficiency $Q^{-1} = 20$. (a) Gaussian fits (dashed curves) to R vs storage time give time constants of $3.4(9)$ μs and $1.7(2)$ μs . The inset shows average retrieval $\bar{R} = 0.44(3)$ for one stored quantum, $M_w \ll 1$. (b) Retrieval improves with increasing read pump power (scattering rate).

in conversion R with pump power suggests that the hyperfine configuration's peak recovery efficiency is limited by the finite scattering rate. We observe larger peak conversion efficiency R for magnetic than for hyperfine storage because of the larger read scattering rates near atomic resonance. In the limit of much less than one stored quantum on average, $M_w \ll 1$, the mean observed recovery efficiency for hyperfine storage, taken for a variety of experimental parameters, is $R = 0.44(3)$ [Fig. 4(a), inset], while for magnetic storage we have observed recovery efficiencies of $R = 0.8(1)$ at an average emitted write photon number of $M_w = 20$. Magnetic storage can be extended to the regime of $M_w \ll 1$ in the future by applying a larger bias magnetic field to lift the degeneracy of the magnetic sublevels, and thereby significantly reduce the superradiant Rayleigh backgrounds. We expect that a longer coherence time will further increase the spin-grating-to-photon conversion efficiency to $R > 95\%$ for single-quantum hyperfine or magnetic excitations.

Doppler decoherence can be virtually eliminated by confining the atoms to the Lamb-Dicke regime [17] using a far-detuned optical lattice. Magnetic field inhomogeneity is then the leading source of decoherence, at coherence times of 10 μs for hyperfine and 70 μs for magnetic storage in our present setup. Magnetic-field-insensitive transitions offer the potential to extend the coherence time to seconds.

We have demonstrated a mechanism for storage of quantized excitations, and their conversion into photons,

that operates at high efficiency in the limit of less than one stored quantum on average. The implementation of a practical single-photon source by this technique, however, will require certain changes to the experiment. The low photon detection efficiency Q limits the rate of single-photon production, and can be improved by using lower-loss cavity mirrors. Correlation measurements will also be required to demonstrate nonclassical photon statistics, a feat currently hindered both by low detection efficiency and by the photon emission into multiple transverse cavity modes. Future efforts will therefore include the use of a single-mode optical resonator with losses dominated by mirror transmission.

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