Interfacing Collective Atomic Excitations and Single Photons

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We study the performance and limitations of a coherent interface between collective atomic states and single photons. A quantized spin-wave excitation of an atomic sample inside an optical resonator is prepared probabilistically, stored, and adiabatically converted on demand into a sub-Poissonian photonic excitation of the resonator mode. The measured peak single-quantum conversion efficiency of $\chi = 0.84(11)$ and its dependence on various parameters are well described by a simple model of the mode geometry and multilevel atomic structure, pointing the way towards implementing high-performance stationary single-photon sources.

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A quantum-coherent interface between light and a material structure that can store quantum states is a pivotal part of a system for processing quantum information [1]. In particular, a quantum memory that can be mapped onto photon number states in a single spatiotemporal mode could pave the way towards extended quantum networks [2,3] and all-optical quantum computing [4]. While light with sub-Poissonian fluctuations can be generated by a variety of single-quantum systems [5–7], a point emitter in free space is only weakly, and thus irreversibly, coupled to an electromagnetic continuum.

To achieve reversible coupling, the strength of the emitter-light interaction can be enhanced by means of an optical resonator, as demonstrated for quantum dots in the weak- [8,9], trapped ions in the intermediate- [10], and neutral atoms in the strong-coupling regime [11,12]. By controlling the position of a single atom trapped inside a very-high-finesse resonator, McKeever *et al.* have realized a high-quality deterministic single-photon source [12]. This source operates, in principle, in the reversible-coupling regime, although finite mirror losses presently make it difficult to obtain full reversibility in practice.

Alternatively, superradiant states of an atomic ensemble [13] exhibit enhanced coupling to a single electromagnetic mode. For three-level atoms with two stable ground states, these collective states can be viewed as quantized spin waves, where a spin-wave quantum (magnon) can be converted into a photon by the application of a phase-matched laser beam [3]. Such systems have been used to generate [14,15], store, and retrieve single photons [16,17], to generate simultaneous photon pairs [18,19], and to increase the single-photon production rate by feedback [20–22]. The strong-coupling regime between magnons and photons can be reached if the sample's optical depth (OD) exceeds unity. However, since the best reported failure rates for magnon-photon conversion in these free-space [14–18,20–24] and moderate-finesse-cavity [19,25] systems have been around 50%, which can be realized with OD ≤ 1, none of the ensemble systems so far has reached the strong, reversible-coupling regime.

In this Letter, we demonstrate for the first time the strong-coupling regime between collective spin-wave excitations and a single electromagnetic mode. This is evidenced by heralded single-photon generation with a single-quantum conversion $\chi = 0.84(11)$, at fourfold suppression of two-photon events. The atomic memory exhibits two Doppler lifetimes $\tau_s = 230$ ns and $\tau_l = 23$ μs that are associated with different magnon wavelengths $\lambda_s = 0.4$ μm and $\lambda_l = 23$ μm written into the sample.

Our apparatus consists of a 6.6 cm long, standing-wave optical resonator with a TEM_{00} waist $w_c = 110$ μm, finesse $F = 93(2)$, linewidth $\kappa/(2\pi) = 24.4(5)$ MHz, and free spectral range $\Delta\nu = 2.27$ GHz. The mirror transmissions M_1 and M_2 and the round-trip loss L near the cesium D_2 line wavelength $\lambda = 2\pi/k = 852$ nm are $M_1 = 1.18(2)\%$, $M_2 = 0.039(2)\%$, and $L = 5.5(1)\%$, respectively, such that a photon escapes from the resonator in the preferred direction with a probability of $T = 0.175(4)$. The light exiting from the cavity is polarization-analyzed and delivered via a single-mode optical fiber to a photon counting module. The overall detection probability for a photon prepared inside the resonator is $q = Tq_1q_2q_3 = 2.7(3)\%$, which includes photodiode quantum efficiency $q_1 = 0.40(4)$, interference filter transmission $q_2 = 0.609(2)$, and fiber coupling and other optical losses $q_3 = 0.65(4)$.

An ensemble containing between 10^3 and 10^6 laser-cooled ^{133}Cs atoms is prepared along the cavity axis, corresponding to an adjustable optical depth between OD $= N\eta = 0.1$ and $N\eta = 200$. Here $\eta = 24F|c_r|^2/(\pi k^2 w_c^2)$ is the single-atom optical depth (cooperativity parameter) for the read transition with reduced dipole matrix element $c_r = \sqrt{3/4}$ [see Fig. 1(b)] for an atom located at a cavity antinode, and N is the effective number of such atoms that produce the same optical depth as the extended sample. The single-atom vacuum Rabi frequency $2g$ is given by $\eta = 4g^2/(\kappa\Gamma)$, where $\Gamma = 2\pi \times 5.2$ MHz and κ are the atomic and cavity full linewidths, respectively.

Starting with a magneto-optical trap (MOT), we turn off the magnetic quadrupole field, apply a 1.8 G bias field perpendicular to the resonator, and optically pump the atoms into a single hyperfine and magnetic sublevel $|g\rangle$ with two laser beams propagating along the bias field. The relevant atomic levels are the electronic ground states $|g\rangle = |6S_{1/2}; F = 3, m_F = 3\rangle$, $|f\rangle = |6S_{1/2}; 4, 3\rangle$ and excited states $|e\rangle = |6P_{3/2}; 4, 3\rangle$ and $|d\rangle = |6P_{3/2}; 3, 3\rangle$ [Fig. 1(b)]. The write and read pump beams, derived from independent, frequency-stabilized lasers, have a waist size $w_p = 300$ μm, enclose a small angle $\theta \approx 2°$ with the cavity axis, and are linearly polarized along the bias field [Fig. 1(a)]. The write pump is applied for 60 ns with a detuning of $\Delta_w/(2\pi) = -40$ MHz from the $|g\rangle \rightarrow |e\rangle$ transition at a typical intensity of 70 mW/cm^2. With some small probability, a "write" photon is generated inside the resonator by spontaneous Raman scattering on the $|g\rangle \rightarrow |e\rangle \rightarrow |f\rangle$ transition to which a resonator TEM$_{00}$ mode is tuned [3,25]. The detection of this write photon heralds the creation of a quantized spin wave inside the ensemble. At some later time, the generated magnon is strongly (superradiantly) coupled to the cavity if the Raman emission $|f\rangle \rightarrow |d\rangle \rightarrow |g\rangle$ from a phase-matched read pump beam restores the sample's initial momentum distribution [3,13,25]. The read pump is ramped on in 100 ns, with a peak intensity of up to 7 W/cm^2. It is detuned by $\Delta_r/(2\pi) = 60$ MHz relative to the $|f\rangle \rightarrow |d\rangle$ transition, such that the "read" photon is emitted into another TEM$_{00}$ resonator mode. The write-read process is repeated for 2 ms (up to 800 times) per MOT cycle of 100 ms.

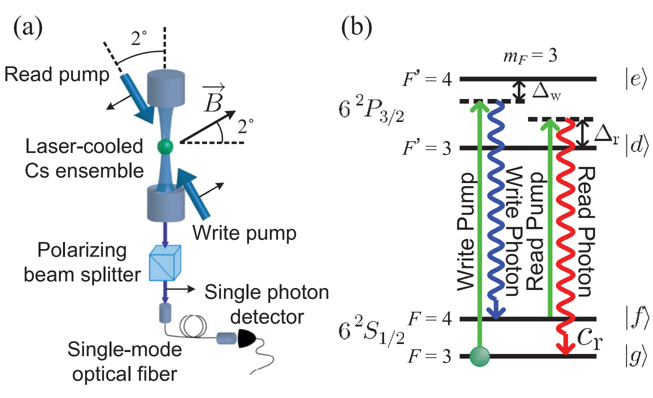

FIG. 1 (color online). (a) Setup for the conditional generation of single photons using a sample of laser-cooled Cs atoms inside an optical resonator. (b) Level scheme for the system with hyperfine and magnetic sublevels $|F, m_F\rangle$. The atomic sample is initially prepared in $|g\rangle$ by optical pumping.

As the magnon-photon conversion efficiency χ approaches unity, small fractional uncertainties in χ result in large uncertainties in the failure rate $1 - \chi$. The rich physics of strong coupling between collective excitations and light hinges on an understanding of the failure rate. Thus, we explore how to accurately estimate χ, the conversion efficiency of a (perfectly prepared) single magnon into a photon, from directly measurable quantities such as the conditional retrieval efficiency $R_c = (\langle wr\rangle - \langle w\rangle \times \langle r\rangle)/\langle w\rangle$ and unconditional retrieval efficiency $R_u = \langle r\rangle/\langle w\rangle$. Here w and r are the write and read photon numbers in a given time interval with averages $n_w \equiv \langle w\rangle$ and $n_r \equiv \langle r\rangle$, respectively, referenced to within the resonator, and the subtracted term in R_c accounts for accidental write-read coincidences. Note that neither measure R_c nor R_u is *a priori* an accurate estimator of χ. The conditional quantity R_c is insensitive to read backgrounds but requires accurate calibration of detection efficiency and systematically differs from χ both at low and high n_w [24]. R_u provides better statistics, since it does not rely on correlated events, but is sensitive to read backgrounds which must be independently measured, e.g., by breaking the phase-matching condition [25].

Figure 2 shows the conditional and unconditional retrieval efficiencies R_c and R_u, respectively, versus average write photon number n_w inside the resonator at fixed optical depth $N\eta = 10$. A carefully calibrated 17(4)% correction due to detector afterpulsing has been applied to R_c. The rise in R_u at small n_w is due to read backgrounds (read pump scatter light), while the drop in R_c is due to

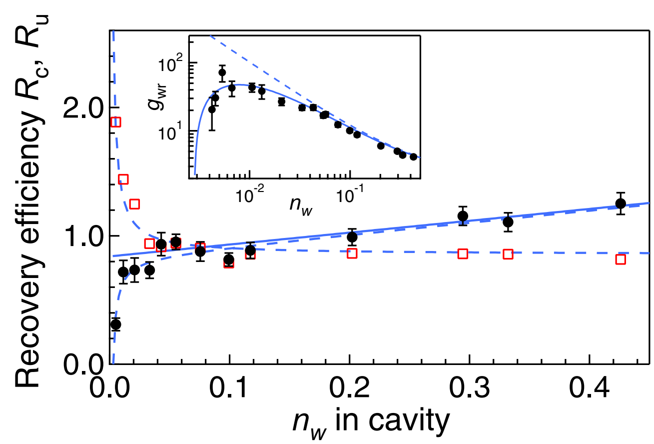

FIG. 2 (color online). Conditional (R_c, solid circles) and unconditional (R_u, open squares) retrieval with model predictions versus intracavity write photon number n_w at a write-read delay of 80 ns. The single-quantum conversion efficiency χ can also be obtained as the y-axis intercept of the linear fit to R_c (solid line). Inset: Nonclassical write-read correlation $g_{wr} > 2$ with model (solid line) and theoretical limit $g_{wr} \lesssim 1/n_w$ (dashed line).

write backgrounds (detector dark counts) that represent a false write signal not accompanied by a spin wave. The increase of R_c with n_w is due to double excitations.

The fundamental, n_w-independent quantity χ can be accurately extracted from the measured data by means of a model that includes the separately measured constant write and read backgrounds [$b_w = 0.0028(4)$ and $b_r = 0.0074(9)$, respectively, when referenced to inside the cavity] that are uncorrelated with the signal. Then $m_w \equiv n_w - b_w$ is the number of "real" magnons that can be converted into read photons, and b_r represents false read events. This model predicts $R_u = n_r/n_w = (\chi m_w + b_r)/n_w$. Similarly, $R_c = \chi m_w[1 + (g_{ww} - 1)m_w]/n_w$, where the term with the second-order write autocorrelation function g_{ww} corresponds to enhanced conditional retrieval if the magnons are bunched ($g_{ww} > 1$). A fit of R_c and R_u to the model, with the conversion χ and g_{ww} as the only fitting parameters, yields a good match between data and model and good agreement between the value $\chi_c = 0.84(11)$ extracted from the conditional and the value $\chi_u = 0.85(2)$ extracted from the unconditional retrieval. The fit yields $g_{ww}^{\rm fit} = 2.1(2)$, in reasonable agreement with the directly measured value $g_{ww}^{\rm meas} = 2.4(2)$ and the expected value $g_{ww}^{\rm th} = 2$ for the bosonic magnon creation process [14]. Since $b_w \ll 1$, the magnon-photon conversion χ can also be estimated as the y intercept of the linear fit $R_c = \chi[1 + (g_{ww} - 1)n_w]$.

The inset in Fig. 2 shows the write-read cross correlation $g_{wr} = \langle wr\rangle/(n_w n_r)$ versus n_w, as well as the predicted dependence with no free parameters (solid line). In the region $n_w > 0.05$ of negligible backgrounds, g_{wr} approaches its fundamental limit $g_{wr} \lesssim 1/n_w$. The large value of g_{wr} corresponds to strongly nonclassical write-read correlations—a necessary condition for sub-Poissonian noise of the read photons. To verify the single-photon character of the read field conditioned on having detected a write photon, we measure the conditional second-order read autocorrelation function $g_{rr|w}$ with two detectors. At $n_w = 0.15(3)$, we find $g_{rr|w} = 0.27(21) < 1$, clearly demonstrating that the source produces single photons. While the result agrees with the expected value $g_{rr|w} \approx g_{ww} n_w = 0.3$ for this value of n_w, the error bar for this time-consuming three-photon measurement remains relatively large due to the low detection efficiency stemming from cavity losses. After completing the experiments described below, we cleaned the deposited cesium off of the mirrors, which reduced the cavity losses (and the effect of detector dark counts) by a factor of 7 and extended the high-recovery region of Fig. 2 down to $n_w = 0.005$. We then measure $g_{rr|w} = 0.15(8)$ at $n_w = 0.007$.

The analysis of Fig. 2 shows that the quantity of fundamental interest, the single-magnon conversion χ, in the region of negligible write backgrounds ($0.04 \leq n_w \leq 0.4$), is well approximated by $\chi \approx R_c/(1 + n_w)$. (Here we make use of $g_{ww} \approx g_{ww}^{\rm th} = 2$ and $n_w \approx m_w$.) In the following, we evaluate this expression measured at fixed write photon number n_w to examine the magnon-photon interface.

The most fundamental limit on the conversion process $\chi_0 = N\eta/(N\eta + 1)$ arises from the competition between the sample's collective coupling to the cavity mode and single-atom emission into free space. In the off-resonant (collective-scattering) regime, this limit originates from the collective enhancement of the read rate by a factor $N\eta$ relative to the single-atom free-space scattering rate [25]. In the on-resonance (dark-state rotation) regime [3,11,12], the limit χ_0 is due to the stronger suppression of free-space scattering [by a factor of $(N\eta)^{-2}$] compared to the suppression of cavity emission [factor of $(N\eta)^{-1}$]. In either case, large optical depth is key to a good interface.

The existence of other excited states in cesium results in additional decoherence mechanisms, such as off-resonant scattering. More relevant in our case are (spatially varying) light shifts due to other excited states that decrease linearly, rather than quadratically, with the excited-state energy splitting. Such light shifts dephase the spin grating and reduce the magnon-photon conversion by $\chi_{ls} = 1 - 2s^4\phi_r^2$ to lowest order in the ratio $s = w_c/w_p \ll 1$. Here ϕ_r is the average light-shift-induced phase accumulated by an atom on the pump beam axis during the read process, and w_c (w_p) is the cavity (read pump) waist. Note that χ_{ls} does not depend on the read pump intensity I_r, since both the light shift and the read rate are proportional to I_r.

Figure 3 shows that this dephasing dramatically changes the dependence of conversion efficiency on optical depth $N\eta$. While the conversion efficiency χ_0 for a three-level atom approaches unity for large $N\eta$ (dashed line), the increase in read photon emission time in the dark-state rotation regime (by a factor of $N\eta$) for atoms with multiple excited states increases the dephasing χ_{ls} and reduces the conversion. The predicted conversion $\chi_0\chi_{ls}$ including all

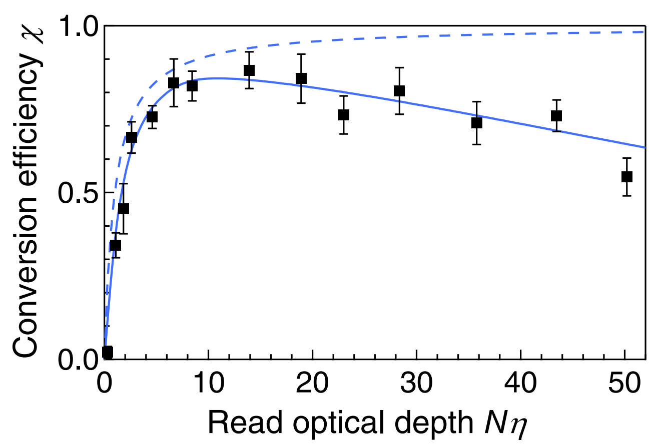

FIG. 3 (color online). Magnon-photon conversion efficiency χ versus read optical depth $N\eta$ at a write-read delay of 120 ns. The optical depth is extracted from the write scattering rate and known intensities and detunings. The dashed line shows the predicted conversion χ_0 for a three-level system; the solid line is the prediction from a model including dephasing from additional excited states.

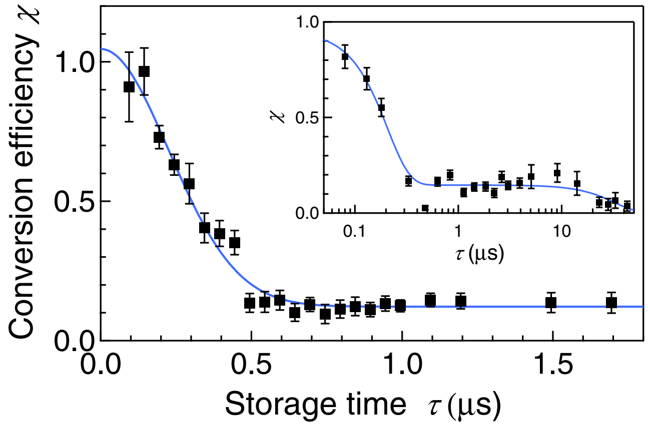

FIG. 4 (color online). Conditional single-photon conversion efficiency χ versus the delay time between write and read pulses τ. The two time scales, as apparent in the inset, are due to the superposition of a short- and a long-wavelength magnon in the standing-wave resonator.

atomic excited hyperfine states produces the correct functional form, as well as the position and peak value of the recovery efficiency, at a waist ratio of $s^{-1} = w_p/w_c = 3$, in good agreement with the measured value of 3.0(4).

The prediction in Fig. 3 also includes a small conversion reduction due to magnon decoherence caused by the atoms' thermal motion during the 120 ns storage time. For the small angle $\theta \approx 2°$ between running-wave pump beams and the cavity standing wave, the write process creates a superposition of two spin waves of very different wavelengths. Backward emission corresponds to a short wavelength $\lambda_s \approx \lambda/2 = 0.4$ μm and is highly Doppler-sensitive, while forward emission with $\lambda_l = \lambda/[2\sin(\theta/2)] = 23$ μm is nearly Doppler-free. The recovery versus storage time τ at $N\eta = 10$ [Fig. 4] shows the two corresponding Gaussian time constants $\tau_s = 240$ ns and $\tau_l = 23$ μs.

The long-time conversion is limited to 25%, because each individual spin-wave component alone can be recovered only with 50% probability due to the mismatch between the standing-wave cavity mode and the running-wave magnon. The highest observed conversion in Fig. 4 of $\chi = R_c/(1 + n_w) = 0.95(13)$, obtained for a write photon number $n_w = 0.27(3)$, is higher than for the inset in Fig. 2. The data for Fig. 4 were taken after carefully realigning the bias magnetic field. This suggests that spin precession due to imperfect magnetic-field alignment could also reduce the conversion efficiency. Since we did not measure $g_{rr|w}$ under the optimized conditions, we conservatively quote $\chi = 0.84$ obtained from Fig. 2.

In summary, we have demonstrated strong coupling between a single magnon and a single photon. Several proposed mechanisms appear to adequately explain the remaining failure rate of the magnon-photon interface and indicate the path to future improvements. Given the low resonator finesse of $F \approx 100$, the resonator's output coupling can easily be improved to over 99%. If the Doppler effect can be eliminated by confining the atoms in a far-detuned optical lattice, the resulting increase in magnon storage time combined with feedback [20–22] will allow the implementation of an unconditional source with near-unity single-photon probability.

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