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 vital 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 spatio-temporal 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 [2, 4, 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- [5, 6], 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 \( F=1 \) [14, 16], store and retrieve single photons \( F=1,2 \) [18, 19], to generate simultaneous-photon pairs \( F'=2 \) [17, 25], and to increase the single-photon production rate by feedback [21, 22, 23]. The strong-coupling regime between magnons and photons can be reached if the sample’s optical depth OD exceeds unity. However, since the failure rate for magnon-photon conversion in these free-space \( F=1,2 \) [15, 16, 17, 18, 20, 21, 22, 23] or moderate-finesse cavity \( F=2 \) [24, 23] systems has been around 50% or higher, which can be realized with \( OD \leq 1 \), 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 efficiency of \( \chi = 0.84(11) \), at a seven-fold suppression of two-photon events. The atomic memory exhibits two Doppler lifetimes, \( \tau_s = 230 \text{ ns} \) and \( \tau_l = 23 \mu \text{ s} \), that are associated with different magnon wavelengths \( \lambda_s = 0.4 \mu \text{ m} \) and \( \lambda_l = 23 \mu \text{ m} \) written into the sample.

Our apparatus consists of a 6.6 cm long, standing-wave optical resonator with a TEM00 waist \( w_c = 110 \mu \text{ m} \), finesse \( F = 93(2) \), linewidth \( \kappa/(2\pi) = 24.4(5) \text{ MHz} \), and free spectral range \( \Delta\nu = 2.27 \text{ GHz} \). The mirror transmissions \( M_1, M_2 \) and round-trip loss \( L \) near the cesium \( D_2 \) line wavelength \( \lambda = 2\pi/k = 852 \text{ nm} \) 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 \( q = T q_1 q_2 q_3 = 2.7(3)\% \), which

\[ \text{FIG. 1: (a) Setup for the conditional generation of single photons using a sample of laser-cooled Cs atoms inside an optical resonator. (b) Level scheme for the system with hyperfine and magnetic sublevels } |F,m_F\rangle. \text{ The atomic sample is initially prepared in } |g\rangle \text{ by optical pumping.} \]
includes photodiode quantum efficiency $q_1=0.40(4)$, interference filter transmission $q_2=0.609(2)$, and fiber coupling and other optical losses $q_3=0.65(4)$. The large cavity losses in our system arise from Cs deposition. For the conditional autocorrelation measurement described at the end of the paper, we cleaned the mirrors. This decreased our losses to $L=0.30(15)\%$, increased the cavity finesse to $F=420(40)$ and the escape probability to $T=0.78(8)$, and improved our overall detection probability to $q=20(3)\%$.

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 $N\eta=0.1$ and $N\eta=200$. Here $\eta = \eta_0 \left| c_r \right|^2$ is the single-atom optical depth (cooperativity parameter) for the read transition with reduced dipole matrix element $c_r=3/4$ (see Fig. 1), $\eta_0 = 24 F / (\pi k^2 w_r^2)$ is the optical depth for an atom located at a cavity antinode on a transition with unity matrix element, and $N$ is the effective number of atoms at this location that produces the same optical depth as the extended sample. The single-atom, single-photon Rabi frequency $2\gamma$ is given by $\gamma = 4g^2/\kappa^2$, where $\Gamma = 2\pi \times 5.2$ MHz and $\kappa$ 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 magnetic field direction. 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. 1b). The write and read pump beams, derived from independent, frequency-stabilized lasers, have a waist size $w_r=300$ $\mu$m, enclose a small angle $\theta \approx 2^\circ$ with the cavity axis, and are linearly polarized along the bias field (Fig. 1a). 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, 24]. At some later time, the quantized spin wave generated in the write process 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, 24]. 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.

As the conversion efficiency $\chi$ of a single stored magnon into a photon in the cavity approaches unity, small fractional uncertainties in $\chi$ result in large uncertainties in the failure rate $1-\chi$. However, the interesting physics of entangled atomic states coupling to photons that rules the matter-light interface hinges on understanding the failure rate. Thus, we explore how to accurately estimate $\chi$ by studying the directly measurable conditional retrieval efficiency $R_c = \langle (n_w n_t) - \langle n_w \rangle\langle n_t \rangle \rangle / \langle n_w \rangle$, and unconditional retrieval efficiency $R_u = \langle n_t \rangle / \langle n_w \rangle$. Here $n_w$ and $n_t$ are the write and read photon numbers in a given time interval, respectively, referenced to within the resonator. Note that neither measure $R_c, R_u$ is a priori an accurate estimator of the single-quantum conversion efficiency $\chi$. The conditional quantity $R_c$ is insensitive to read backgrounds, but requires accurate calibration of detection efficiency, and systematically differs from $\chi$ both at low and high $\langle n_w \rangle$ [24]. $R_c$ can be measured at larger $\langle n_w \rangle$ and 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 [24].

Fig. 2 shows the conditional and unconditional retrieval efficiencies $R_c, R_u$ versus average write photon number $\langle n_w \rangle$ inside the lower-finesse 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 $\langle n_w \rangle$ is due to read backgrounds, while the drop in $R_c$ is due to write backgrounds, that are not accompanied by a spin wave. The increase of $R_c$ with $\langle n_w \rangle$ is due to double excitations. An accurate value for the single-quantum conversion efficiency $\chi$ can be extracted from the measured data by means of a model that includes uncorrelated constant write and read backgrounds, independently measured to be $b_w = 0.0028(4)$ and $b_r = 0.0074(9)$ when referenced to

![Fig. 2: Conditional ($R_c$, solid circles) and unconditional ($R_u$, open squares) retrieval with model predictions, versus intra-cavity write photon number ($n_w$), at a write-read delay of 80ns. The single-quantum conversion efficiency $\chi$ can also be obtained as the y-axis intercept of the linear fit to $R_c$ (solid black line). Inset: Non-classical write-read correlation $g_{wr} > 2$ with model (solid line) and theoretical limit $g_{wr} \leq 1/\langle n_w \rangle$ (dashed line).](image-url)
the cavity. This model predicts \( \langle n_w \rangle R_w = \chi (\langle n_w \rangle - b_w) + b_w \) and \( \langle n_w \rangle R_c = \chi (\langle n_w \rangle - b_w) [1 + (g_{w w} - 1) (\langle n_w \rangle - b_w)] \). The measured write second-order autocorrelation function \( g_{w w}^{\text{meas}} = 2.4(2) \) differs from the expected value \( g_{w w} = 2 \), likely due to observed fluctuations in write pulse intensity. A fit of \( R_c, R_w \) to the model, with the conversion \( \chi \) as the only fitting parameter, 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 \( \chi_w = 0.85(2) \) extracted from the unconditional retrieval efficiency. \( \chi_w \), being independent of detection efficiency, is more precise. Since \( b_w, b_r \ll 1 \), the magnon-photon conversion \( \chi \) can also be estimated as the \( y \) intercept of the linear fit \( R_c = \chi (1 + (g_{w w} - 1) (\langle n_w \rangle)) \). 

The inset to Fig. 2 shows the write-read cross correlation \( g_{w r} = \langle n_w n_r \rangle / (\langle n_w \rangle \langle n_r \rangle) \) versus \( \langle n_w \rangle \), as well as the predicted dependence with no free parameters. In the region \( \langle n_w \rangle > 0.05 \), where \( g_{w r} \) approaches its fundamental limit \( g_{w r} \ll 1 / \langle n_w \rangle \), backgrounds are negligible, and the unconditional recovery \( R_u \) is also a good estimate of \( \chi \). In the figures which follow, we estimate \( \chi \) as \( R_u / (1 + \langle n_w \rangle) \), and use it to examine the physical limitations on 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 \( 2.4 \). In the on-resonance (dark-state rotation) regime \( 11, 12 \), the limit \( \chi_0 \) is due to the stronger suppression of free-space scattering (by a factor \( (N \eta)^{-2} \)) compared to the suppression of cavity emission (factor \( (N \eta)^{-1} \)). In either case, large optical depth is key to a good interface.

To the extent that the atomic system is not a simple three-level system, additional sources of magnon decoherence, such as non-resonant scattering from other excited states, reduce the conversion efficiency. More relevant in the present case are spatially inhomogeneous light shifts due to other excited states that decrease linearly, rather than quadratically, with the excited-state energy splittings. Such light shifts dephase the spin gratings, and reduce the magnon-photon conversion by \( \chi_{ls} = (1 - 2 s^2 \phi_r^2) \) to lowest order in the ratio \( s = w_c / w_p \ll 1 \). Here \( \phi_r \) 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 \( \chi_{ls} \) depends upon read pump size \( w_p \), but not the read pump intensity \( I_r \), since both light shift and read rate are proportional to \( I_r \). As such, as the pump waist is increased at fixed read intensity, the resulting failure rate should decrease as the inverse square of the pump power.

Fig. 3 shows that this dephasing effect dramatically changes the dependence of the conversion efficiency on optical depth \( N \eta \). While the conversion efficiency \( \chi_0 \) for a three-level-atom approaches unity for large optical depth \( N \eta \) (dashed line), the increase in read-photon emission time in the dark-state rotation regime (by a factor \( N \eta \)) for atoms with multiple excited states increases the dephasing \( \chi_{ls} \), and reduces the conversion. The predicted conversion \( \chi_0 \chi_{ls} \) including all 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 the decoherence of the magnon caused by the atoms’ thermal motion during the 120 ns storage time. For the small angle \( \theta \approx 2^\circ \) between running-wave pump beams and cavity standing wave, the write photon emission process creates a superposition of
two spin waves of very different wavelengths. Backward emission corresponds to a short wavelength $\lambda_s \approx \lambda/2 = 0.4\ \mu m$, and is highly Doppler-sensitive, while forward emission with $\lambda_l = \lambda/(2\sin(\theta/2)) = 23\ \mu m$ is nearly Doppler free. The recovery versus storage time $\tau$ at $N \eta = 10$ (Fig. 4) shows the two corresponding Gaussian time constants $\tau_s = 240\ ns$ and $\tau_l = 23\ \mu s$.

The long-time conversion is limited to 25%, because each individual spin-wave component alone can only be recovered with 50% probability due to the mismatch between the standing wave cavity mode and the running-wave magnon. The highest observed conversion efficiency in Fig. 4 of $\chi = 0.95(13)$ is higher than for the inset or Fig. 2. The data for Fig. 4 was taken after carefully realigning the bias magnetic field along the quantization axis defined by the pump beam polarizations, while the inset and Fig. 2 were taken before realignment. This suggests that spin precession due to imperfect magnetic field alignment could also reduce the conversion efficiency. The result $\chi = 0.95$ was obtained for a single write-photon value $\langle n_w \rangle = 0.27(3)$, so we conservatively quote $\chi = 0.84$ obtained from the fit to the data versus $n_w$ with un-optimized fields in Fig. 2.

Using the lower loss ($F = 420$) cavity with clean mirrors to minimize sensitivity to detector afterpulsing and improve the data collection rate, we measure the read autocorrelation function $g_{rr}(\tau)$ conditioned on having detected a write photon. Due to the seven times higher detection efficiency, the detector dark count rate does not appreciably lower the recovery down to $n_w = 0.005$ in this configuration. For $\langle n_w \rangle = 0.007$ we obtain $g_{rr}(\tau) = 0.15(8)$ at an optical depth $N \eta = 10$, clearly demonstrating the sub-Poissonian nature of the source.

In summary, we have realized an interface between spin-wave quanta and narrowband single photons with a performance near 90%, representing the first experimental demonstration of strong coupling between collective spin-wave excitations and photons. Several proposed mechanisms appear to adequately explain the remaining failure rate of the magnon-photon interface, and indicate the path to future improvements. If the conditional single-photon source described here were operated as a single-shot source by applying the read beam only when a write photon was detected, it already would have almost comparable performance to recently demonstrated feedback-enhanced sources [21, 22, 23]. At $n_w = 0.007$ our source would unconditionally deliver photons with probability 0.6% at $g_{rr} = 0.15$, to be compared to 5.4% at $g_{rr} = 0.41$ for 150 trials [21], or 2.5% at $g_{rr} = 0.3$ for 12 trials. It should be straightforward to implement feedback as demonstrated in Refs. [21, 22, 23] into our setup. If the Doppler effect can be eliminated by confining the atoms in a far-detuned optical lattice, the resulting substantial increase in magnon storage time would allow the 150 trials necessary to implement an unconditional source with near-unity single-photon probability.

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