In recent years much effort has been directed toward generating quantum-mechanical states of the electromagnetic field with a well-defined number of light quanta (i.e., photon-number or Fock states). In addition to being of fundamental interest, these states represent an essential resource for the practical implementation of many ideas from quantum information science such as quantum communication \cite{1}. Over the past decade, tremendous progress has been made in generating single-photon states by using photon pairs in parametric downconverters \cite{2}, single emitters \cite{3,4}, and single atoms in high-finesse cavities \cite{5,6}. While parametric down-conversion techniques have recently been used to generate multi-photon states \cite{7}, it remains experimentally challenging to implement schemes that allow for simultaneous control over both photon number and spatio-temporal properties of the pulse.

In this Letter we describe a novel technique for generating pulses of light with controllable photon numbers, propagation direction, timing, and pulse shapes by exploiting long-lived coherent memory for photon states in an optically dense atomic medium \cite{8}. We experimentally demonstrate key elements of this technique in photon-counting experiments. This approach combines different aspects of earlier studies on “light storage” \cite{9,10} and Raman preparation and retrieval of atomic excitations \cite{11,12,13}. It is particularly important in the contexts of long-distance quantum communication \cite{14}, and EIT-based quantum nonlinear optics \cite{15,16,17}.

In our approach we first optically pump a large ensemble of N atoms with a three-state “lambda” configuration of atomic states (see Fig.1a) in the ground state \ket{g}. Spontaneous Raman scattering \cite{18} is induced by a weak, off-resonant laser beam with Rabi frequency \Omega_R and detuning \Delta_R, referred to as the write laser. This two-photon process flips an atomic spin into the metastable state \ket{s} while producing a correlated frequency-shifted photon (a so-called Stokes photon). Energy and momentum conservation ensure that for each Stokes photon emitted in certain direction there exists exactly one flipped spin quantum in a well-defined spin-wave mode. The number of spin wave quanta and the number of photons in the Stokes field thus exhibit strong correlations, analogous to the correlations between photons emitted in parametric down conversion \cite{19}. As a result, measurement of the Stokes photon number \nS ideally projects the spin-wave into a nonclassical collective state with \nS spin quanta \ket{S}. After a controllable delay time \tau_d (see Fig. 1b), the stored spin-wave can be coherently converted into a light pulse by applying a second near-resonant laser beam with Rabi frequency \Omega_R (retrieve laser), see Fig. 1a. The physical mechanism for this retrieval process involves EIT \cite{16,20,21,22,23} and is identical to that employed in previous experiments \cite{4,10}. The direction, delay time \tau_d, and rate of retrieval are determined by the direction, timing, and intensity of the retrieve laser, allowing control over the spatio-temporal properties of the retrieved pulse (referred to as the anti-Stokes pulse). Since the storage and retrieval processes ideally result in identical photon numbers in the Stokes and anti-Stokes pulses \cite{10}, this technique should allow preparation of an n-photon Fock state in the anti-Stokes pulse conditioned on detection of n Stokes photons.

The experimental apparatus (see Fig. 1c) is similar to that used in our earlier work \cite{11}. The primary experimental challenge lies in transmitting the few-photon Stokes and anti-Stokes pulses while simultaneously blocking the write and retrieve laser beams. A polarization beamsplitter separates the write (retrieve) laser from the Stokes (anti-Stokes) Raman light, and further filtering is provided by an etalon or an optically-pumped $^{87}$Rb cell in the write channel, and a $^{85}$Rb cell in the read channel; this combined filtering separates the write (retrieve) laser from the Stokes (anti-Stokes) Raman light to one part in $10^{9}$ ($10^{12}$). Experimentally, we take advantage of the long coherence time of the atomic memory ($\sim 3 \mu s$ in the present experiment, see Fig. 3) to create few-photon pulses with long coherence lengths ($\sim$ few $\mu$s)
that significantly exceed the time-resolution of the APDs (~ 50 ns). This allows us to directly count the photon number in each of the pulses and to directly measure the pulse shapes by averaging the time-resolved APD output over many experimental runs. Fig. 2a shows the average number of detected Stokes photons per unit time (photon flux) in the write channel as a function of time during the 1.6 μs-long write pulse. The magnitude of the photon flux (and hence the total number of photons in the pulse) is controlled by varying the excitation intensity. The shape of the Stokes pulse changes qualitatively as the total number of photons in the pulse exceeds unity: for pulses containing on average one photon or less, the flux is constant in time (more generally it follows the shape of the write laser), whereas for pulses containing more than one photon, the flux increases with time.

The observed evolution of the Stokes pulses can be understood qualitatively by considering the mutual growth of the photon field and spin excitation: the first flipped spin *stimulates* subsequent spin excitations which are accompanied by increased probability of Stokes photon emission. This process is governed by the collective Raman scattering rate \( \xi = \eta \Omega W^2 \gamma / \Delta_{W}^2 \), which is equal to the product of the optical depth \( \eta \) and the single atom scattering rate \( \Omega W^2 \gamma / \Delta_{W}^2 \), where \( \gamma \) is the decay rate of |\( e \rangle \). For short excitation times \( t \), we consider the evolution of several Hermite-Gaussian modes of Stokes radia-
For larger (smaller) retrieve laser intensity, the excitation is released faster (slower), while the amplitude changes in such a way that the total number of anti-Stokes photons is always equal to the number of spin-wave excitations. In practice, decay of the spin coherence during the delay time $\tau_d$ and finite optical depth flatten and broaden the anti-Stokes pulse, reducing the total number of anti-Stokes photons which can be retrieved within the coherence time of the atomic memory, as indicated by theoretical calculations (Fig. 2b) based on Ref. [23]. The detailed comparison between theory and experiment in Fig. 2 suggests that the bandwidth of the generated anti-Stokes pulse is close to being Fourier-transform limited, while the transverse profile effectively corresponds to only a few spatial modes.

To quantify the correlations, we consider the normalized photon shot noise level $\text{PSN}_{th} = \bar{n}_S + \bar{n}_{AS}$, which represents the maximum degree of correlations possible for classical states [24]. In the experiment, great care is taken to eliminate systematic sources of errors, in particular APD dead-time effects. To this end we experimentally determine photon shot noise for each channel by using a 50-50 beamsplitter and two APDs per detection channel (see Fig. 1c) which allows us to accurately determine the measured $\text{PSN}_{meas} = \text{var}(AS1-AS2) + \text{var}(S1-S2)$ value for each experiment. For correlation measurements we typically choose the excitation intensities such that the average number of photons in each channel is on the order of or smaller than unity. Under such conditions the measured PSN approaches the expected, theoretical value of PSN.

To quantify the correlations, we consider the normalized variance $V = \text{var}((n_{AS} - n_S))/\text{PSN}_{meas}$, which is one for classically correlated pulses and zero for pulses exhibiting perfect number correlations. Using this method, we measure $V = 0.942 \pm 0.006$ for the data shown in Fig. 3 at delay time $\tau_d = 0$ [25].

![Fig. 3](image.png)

**Fig. 3:** (Color Online) Observation of nonclassical correlations. Normalized variance $V$ (blue) and mean number of anti-Stokes photons (red) versus delay time $\tau_d$. The open and closed symbols represent two experimental runs with similar experimental parameters. The dotted line is an exponential fit (characteristic time $\sim 3\mu s$) to the mean number of anti-Stokes photons. The solid line is the result of a theoretical model including the effects of loss, background, and several spatial modes on the Stokes and anti-Stokes channels [26].

that within the spin coherence time, it is possible to control the timing between preparation and retrieval, while preserving nonclassical correlations.

It is important to note that at $\tau_d = 0$ the observed value $V = 0.942 \pm 0.006$ is far from the ideal value of $V = 0$. One source of error is the finite retrieval efficiency, which is limited by two factors. Due to the atomic memory decoherence rate $\gamma_c$, the finite retrieval time $\tau_r$ always results in a finite loss probability $p \approx \gamma_c \tau_r$. Moreover, even as $\gamma_c \rightarrow 0$ the retrieval efficiency is limited by the finite optical depth $\eta$ of the ensemble, which yields an error scaling as $p \sim 1/\sqrt{\eta}$. The anti-Stokes pulses in Fig. 3 have widths on the order of the measured decoherence time, so the atomic excitation decays before it is fully retrieved. The measured maximum retrieval efficiency at $\tau_d = 0$ corresponds to about 0.3. In addition to finite retrieval efficiency, other factors reduce correlations, including losses in the detection system, background photons, APD afterpulsing effects, and imperfect mode matching.

These correlations between Stokes and anti-Stokes pulses allow for the conditional preparation of the anti-Stokes pulse with intensity fluctuations that are suppressed compared with classical light. In order to quantify the performance of this technique, we measured the second-order intensity correlation function $g^{(2)}(AS)$ and mean number of photons $\bar{n}_{AS}$ for the anti-Stokes pulse conditioned on the detection of $n_S$ photons in the Stokes channel (see Fig. 4). For classical states of light, $g^{(2)} \geq 1$, whereas an ideal Fock state with $n$ photons has $g^{(2)} = 1 - 1/n$. Note that the mean number of anti-Stokes photons grows linearly with $n_S$, while $g^{(2)}(AS)$ drops below unity, indicating the non-classical character of the anti-Stokes photon states. In the presence of back-
ground counts, \( g_{nS}^{(2)}(AS) \) does not increase monotonically with \( n_S \), but instead exhibits a minimum at \( n_S = 2 \). The Mandel Q parameter \( Q^S \) can be calculated using \( Q^S_{nS} = \frac{\bar{n}_{AS}^{(2)} \cdot (AS) - 1}{\bar{n}_{AS}^{(2)}(AS) - 1} \); from the measurements we determine \( Q^S_{nS=2} = -0.09 \pm 0.03 \) for conditionally generated states with \( n_S = 2 \) (\( Q \geq 0 \) for classical states and \( Q = -1 \) for Fock states).

The observed reduction in the intensity fluctuations is imperfect due to background and losses in both the preparation and the retrieval detection channels. These can be accounted for in a theoretical model that yields reasonable agreement with experimental observations (solid curve in Fig. 4). Using this model corrected for loss and background on the retrieval channel (dotted line in Fig. 4), we estimate \((\bar{n}_{AS=2}^{AS}, Q^S_{nS=2}^{AS})\) to be approximately \((2.5, -0.85)\).

Although the corrected parameters are closer to the ideal limit, they still do not correspond to a perfect Fock state. This is due to loss and background in the preparation channel, which prevent measurement of the exact number of created spin excitations. In principle, the conditional state preparation can be made insensitive to overall Stokes detection efficiency \( \alpha_S \) by working in the regime of a very weak excitation \( [14] \); however, Stokes channel background counts \( n_{BG}^{AS} \) prevent one from reaching this regime in practice. A qualitative condition for high quality Fock state generation, \( \zeta \equiv n_{BG}^{AS} (1 - \alpha_S) / n_S \alpha_S \ll 1 \), is only marginally fulfilled in our experiments (\( \zeta \sim 0.3 \)), accounting for the imperfectly prepared atomic states. Refinements in the Stokes detection system and better transverse mode selection should permit conditional Fock state generation with much greater purity, thereby providing a basic building block for long-distance quantum communication \([14]\).

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[25] For the experimental conditions of Fig. 3, we estimate that dead-time effects reduce the measured \( n_{AS} \) by 2.5% from its actual value and increase \( V \) by less than 0.2% from its actual value (well within the ~0.6% error bars).
[26] Based on experimental measurements, the overall detection efficiency \( \alpha \) and number of background photons \( n_{BG} \) used in the model are \( \alpha = 0.07 \), \( n_{BG} = 0.3 \) \( \alpha_{AS} = 0.21 \), \( n_{BG}^{AS} = 0.12 \) on the Stokes (anti-Stokes) channel, with a retrieval efficiency characteristic decay time of 3 µs and 4 transverse spatial modes assumed.
[27] Parameters used in the model (estimated from experi-
mental measurements): $\alpha_S = 0.35, n_S^{BG} = 0.27$ ($\alpha_{AS} = 0.1, n_{AS}^{BG} = 0.12$) on the Stokes (anti-Stokes) channel.