

Temporal Dynamics of Photon Pairs Generated by an Atomic Ensemble

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The time dependence of nonclassical correlations is investigated for two fields (1,2) generated by an ensemble of cold Cesium atoms via the protocol of Duan et al. [Nature **414**, 413 (2001)]. The correlation function $R(t_1, t_2)$ for the ratio of cross to auto-correlations for the (1,2) fields at times (t_1, t_2) is found to have a maximum value $R^{\max} = 292 \pm 57$, which significantly violates the Cauchy-Schwarz inequality $R \leq 1$ for classical fields. Decoherence of quantum correlations is observed over $\tau_d \simeq 175$ ns, and is described by our model, as is a new scheme to mitigate this effect.

In recent years quantum measurement combined with conditional quantum evolution has emerged as a powerful paradigm for accomplishing diverse tasks in quantum information science [1, 2, 3, 4]. For example, Duan, Lukin, Cirac and Zoller (*DLCZ*) [4] have proposed a scheme for the realization of scalable quantum communication networks that relies upon entanglement created probabilistically between remotely located atomic ensembles. By utilizing successful measurements to condition subsequent steps in their protocol, *DLCZ* have developed a scheme that has built-in quantum memory, entanglement purification and resilience to realistic sources of noise, thereby enabling a quantum repeater architecture to overcome photon attenuation [5, 6].

Central to the *DLCZ* protocol is the ability to *write* and *read* collective spin excitations *into* and *out of* an atomic ensemble, with efficient conversion of discrete spin excitations to single-photon wavepackets. Observations of the resulting non-classical correlations between the optical fields generated from *writing* and *reading* such spin excitations have recently been reported by several groups, both at the single-photon level [7, 8, 9] as appropriate to the protocol of *DLCZ* and in a regime of large photon number $n \sim 10^3 - 10^7$ [10]. Generation and detection efficiencies have now been improved so that excitation stored within an atomic ensemble can be employed as a controllable source for single photons [8].

A critical aspect of such single-photon wavepackets is that they are emitted into well-defined spatio-temporal modes to enable quantum interference between emissions from separate ensembles (e.g., for entanglement based quantum cryptography [4]). However, with the exception of the verification of the time-delay implicit for the Raman processes employed [10], experiments to date have investigated neither the time or spatial dependence of quantum correlations for the emitted fields from the atomic ensemble. The high efficiencies achieved in Ref. [8] now enable such an investigation for the temporal properties of nonclassical correlations between emitted photon pairs, which we report in this Letter.

Specifically, we study the time dependence of quantum correlations for photons emitted from an ensemble of cold Cesium atoms, with photon pairs created sequentially by classically controlled *write* and *read* pulses. Large vio-

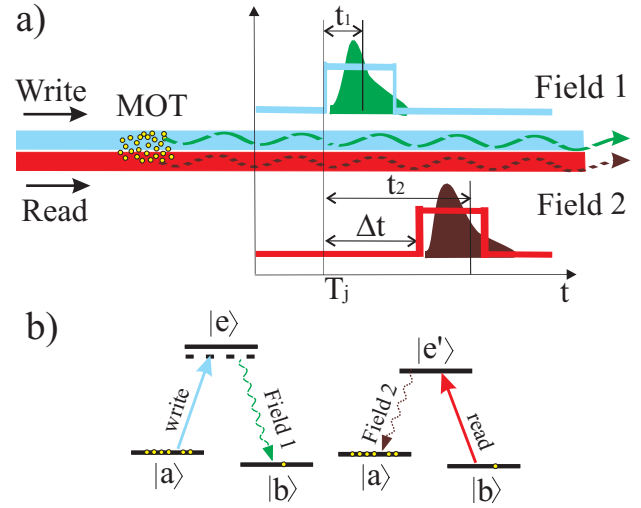


FIG. 1: (a) Schematic of experiment. *Write* and *read* pulses propagate into a cloud of cold Cs atoms (MOT) at times T_j and $T_j + \Delta t$ respectively, and generate the correlated output fields 1 and 2. Quantum correlations for these fields at times (t_1, t_2) are investigated by way of photoelectric detection. (b) The relevant atomic level scheme.

lations of the Cauchy-Schwartz inequality $R \leq 1$ for the ratio of cross to auto-correlations are observed for the generated fields, with $R^{\max} = 292 \pm 57 \not\leq 1$. By contrast, previous measurements have reported violations $R \not\leq 1$ only for detection events integrated over the entire durations of the *write* and *read* pulses ($R = 1.84 \pm 0.06$ in Ref. [7], $R = 1.34 \pm 0.05$ in Ref. [9], and $R = 53 \pm 2$ in Ref. [8]). We also map the decay of quantum correlations by varying the time delay between the *write* and *read* pulses, and find a decoherence time $\tau_d \simeq 175$ ns. We have developed a model to describe the decoherence and find good correspondence with our measurements. This model is utilized to analyze a new proposal that should extend the correlation times to beyond $10 \mu\text{s}$, which would allow for entanglement between atomic ensembles on the scale of several kilometers.

Our experimental procedure is illustrated in Figure 1. An initial *write* pulse at 852 nm creates a state of collective excitation in an ensemble of cold Cs atoms, as her-

aided by a photoelectric detection event from the Raman field 1. After a user-programmable delay Δt , a *read* pulse at 894 nm converts this atomic excitation into a field excitation of the Raman field 2. The (*write*, *read*) pulses are approximate coherent states with mean photon numbers $(10^3, 10^5)$, respectively, and are focussed into the MOT as TEM₀₀ Gaussian beams with orthogonal polarizations and beam waist $w_0 = 30 \mu\text{m}$. This scheme is implemented in an optically thick sample of four-level atoms, cooled and trapped in a magneto-optical trap (MOT) [11]. In particular, we utilize the ground hyperfine levels $6S_{1/2}, F = \{4; 3\}$ of atomic Cs (labelled $\{|a\rangle; |b\rangle\}$), and excited levels $\{6P_{3/2}, F = 4; 6P_{1/2}, F = 4\}$ of the D_2, D_1 lines at $\{852; 894\}$ nm (labelled $\{|e\rangle; |e'\rangle\}$).

Each attempt to generate a correlated pair of photons in the (1, 2) fields is preceded by shutting off the trapping light for 700 ns. The re-pumping light is left on for an additional 300 ns in order to empty the $|b\rangle$ state, thus preparing the atoms in $|a\rangle$. The j^{th} trial of a protocol is initiated at time T_j when a rectangular pulse from the *write* laser beam, 150 ns in duration (FWHM) and tuned 10 MHz below the $|a\rangle \rightarrow |e\rangle$ transition, induces spontaneous Raman scattering to level $|b\rangle$ via $|a\rangle \rightarrow |e\rangle \rightarrow |b\rangle$. The *write* pulse is sufficiently weak so that the probability to scatter one Raman photon into a forward propagating wavepacket is much less than unity for each pulse. Detection of one photon from field 1 results in a ‘‘spin’’ excitation to level $|b\rangle$, with this excitation distributed in a symmetrized, coherent manner throughout the sample of N atoms illuminated by the *write* beam [4]. Regardless of successful detection of a photon in field 1, we next address the atomic ensemble with a *read* pulse at a time $T_j + \Delta t$, where Δt is controlled by the user. The *read* light is a rectangular pulse, 120 ns in duration, tuned to resonance with the $|b\rangle \rightarrow |e'\rangle$ transition.

To investigate the photon statistics, we use four avalanche photodetectors, a pair for each field i , labelled as $D_{iA, iB}$, which are activated at $(T_j, T_j + \Delta t)$ with $i = (1, 2)$, respectively, for 200 ns for all experiments. The quantity $p_\tau(t_l, t_m)$ is defined as the joint probability for photoelectric detection from field l in the interval $[T_j + t_l, T_j + t_l + \tau]$ and for an event from field m in the interval $[T_j + t_m, T_j + t_m + \tau]$, where l and m equal 1 or 2. $p_\tau(t_l, t_m)$ is determined from the record of time-stamped detection events at $D_{1A, 1B}, D_{2A, 2B}$. In a similar fashion, $q_\tau(t_l, t_m)$ gives the joint probability for detection for fields (l, m) in the intervals $([T_j + t_l, T_j + t_l + \tau], [T_k + t_m, T_k + t_m + \tau])$ for two trials $k \neq j$.

Following Refs. [7, 8], we introduce the time-dependent ratio $R_\tau(t_1, t_2)$ of cross-correlation to auto-correlation for the (1, 2) fields, where

$$R_\tau(t_1, t_2) \equiv \frac{[p_\tau(t_1, t_2)]^2}{p_\tau(t_1, t_1)p_\tau(t_2, t_2)}. \quad (1)$$

This ratio is constrained by the inequality $R_\tau(t_1, t_2) \leq 1$ for all fields for which the Glauber-Sudarshan phase-space function is well-behaved (i.e., *classical* fields) [7, 12]. Beyond enabling a characterization of the quan-

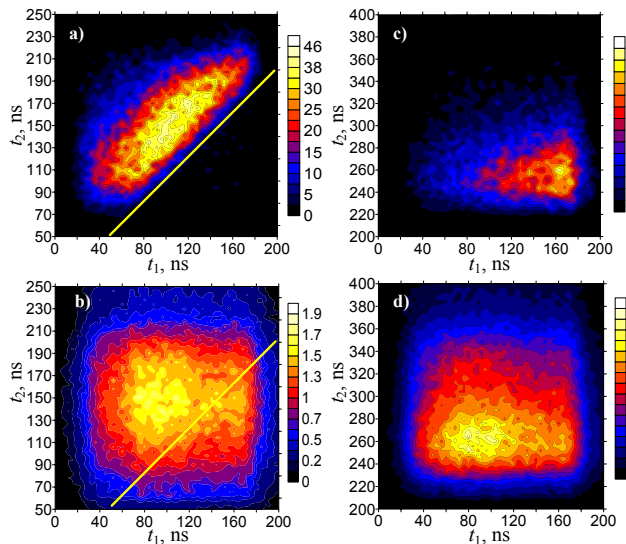


FIG. 2: Probability for joint detection from the fields (1, 2) at times (t_1, t_2) with origin at the beginning of the *write* pulse. (a) $p_\tau(t_1, t_2)$, and (b) $q_\tau(t_1, t_2)$ for overlapped *write* and *read* pulses, $\Delta t = 50$ ns, with the solid line corresponding to $t_2 = t_1$. (c) $p_\tau(t_1, t_2)$ and (d) $q_\tau(t_1, t_2)$ for consecutive *write* and *read* pulses, $\Delta t = 200$ ns. In all cases, the bin size $\tau = 4$ ns, and the joint probabilities p_τ and q_τ have been scaled by 10^9 .

tum character of the (1, 2) fields in a model independent fashion, measurements of $R_\tau(t_1, t_2)$ also allow inferences of the quantum state for collective excitations of single spins within the atomic ensemble.

The first step in the determination of $R_\tau(t_1, t_2)$ is the measurement of the joint probability $p_\tau(t_1, t_2)$ for the (1, 2) fields, and for comparison, $q_\tau(t_1, t_2)$ for independent trials. In our experiment, we focus on two cases: (I) nearly simultaneous application of *write* and *read* pulses with offset $\Delta t = 50$ ns less than the duration of either pulse, and (II) consecutive application of *write* and *read* pulses with $\Delta t = 200$ ns longer than the *write*, *read* durations. Results for $p_\tau(t_1, t_2)$ and $q_\tau(t_1, t_2)$ are presented in Fig. 2 as functions of the detection times (t_1, t_2) for the fields (1, 2). For both $\Delta t = 50$ and 200 ns, $p_\tau(t_1, t_2) \gg q_\tau(t_1, t_2)$, indicating the strong correlation between fields 1 and 2, with the maximal ratio $g_{1,2}^\tau(t_1, t_2) = p_\tau(t_1, t_2)/q_\tau(t_1, t_2) \gtrsim 30$, which is much greater than reported previously [7, 8, 9]. In Fig. 2, $\tau = 4$ ns, leading to statistical errors of about 8% for the largest values shown.

In case (I) for nearly simultaneous irradiation with *write* and *read* pulses, Fig. 2(a) shows that $p_\tau(t_1, t_2)$ peaks along the line $t_2 - t_1 = \delta t_{12} \simeq 50$ ns with a width $\Delta t_{12} \simeq 60$ ns, in correspondence to the delay δt_{12} and duration Δt_{12} for read-out associated with the transition $|b\rangle \rightarrow |e'\rangle \rightarrow |a\rangle$ given an initial transition $|a\rangle \rightarrow |e\rangle \rightarrow |b\rangle$ [10]. Apparently, the qualitative features of $p_\tau(t_1, t_2)$ depend only upon the time difference between photon detections in fields 1 and 2 (i.e., $p_\tau(t_1, t_2) \approx F(t_2 - t_1)$). In case (II) with the *read* pulse launched 200 ns after

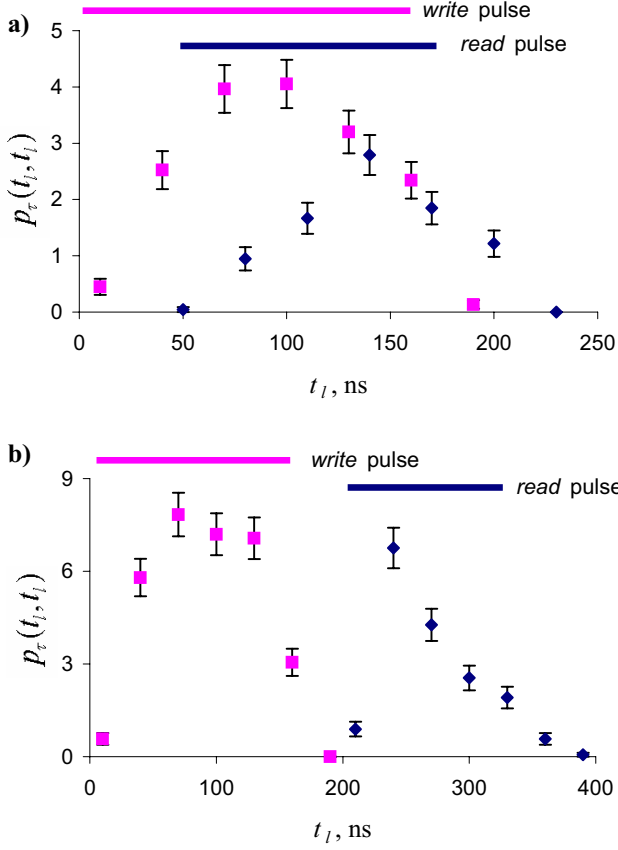


FIG. 3: Probability of joint detection $p_\tau(t_1, t_1)$ for field 1 (squares) and $p_\tau(t_2, t_2)$ for field 2 (diamonds) as functions of respective detection times t_1 and t_2 . Bin size $\tau = 30$ ns. (a) Overlapping *write* and *read* pulses, $\Delta t = 50$ ns; (b) Consecutive *write* and *read* pulses, $\Delta t = 200$ ns.

the *write* pulse, excitation is “stored” in the atomic ensemble until the readout. The production of correlated photon pairs should now be distributed along $t_2 \simeq \delta t_{12}$ with width $\simeq \Delta t_{12}$. Instead, as shown in Fig. 2(c), $p_\tau(t_1, t_2)$ peaks towards the end of the *write* pulse (i.e., $t_1 \gtrsim 100$ ns), and near the beginning of the *read* pulse (i.e., $200 \lesssim t_2 \lesssim 300$ ns). Early events for field 1 lead to fewer correlated events for field 2, as $p_\tau(t_1, t_2)$ decays rapidly beyond the line $t_2 - t_1 = \tau_d \simeq 175$ ns. The marked contrast between $p_\tau(t_1, t_2)$ for $\Delta t = 50$ and 200 ns results in a diminished ability for the conditional generation of single photons from excitation stored within the atomic ensemble [8] and, more generally, for the implementation of the *DLCZ* protocol for increasing Δt . The underlying mechanism is decoherence within the ensemble, as will be discussed.

Fig. 2(b, d) displays $q_\tau(t_1, t_2)$ for independent trials $j \neq k$. $q_\tau(t_1, t_2)$ is expected to be proportional to the product of intensities of the fields 1 and 2, in reasonable correspondence to the form shown in Fig. 2(b, d) for our roughly rectangular *write*, *read* pulses, but distinctively different from $p_\tau(t_1, t_2)$ in (a, c).

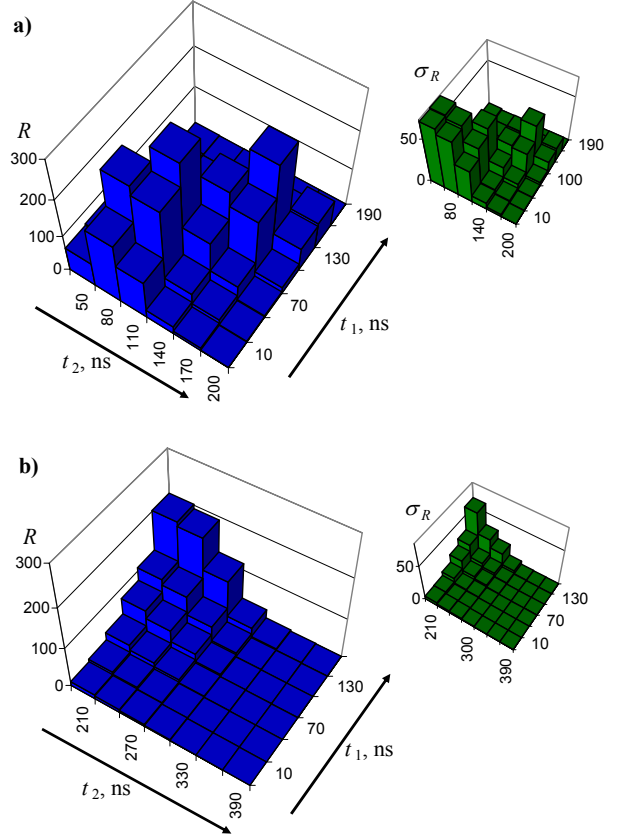


FIG. 4: The experimentally derived ratio $R_\tau(t_1, t_2)$ [Eq. 1] as a function of detection times (t_1, t_2) for the (1, 2) fields, with $R_\tau \leq 1$ for classical fields. The left column gives $R_\tau(t_1, t_2)$ for $\Delta t = 50$ ns (top) and $\Delta t = 200$ ns (bottom), while the right column gives the associated statistical uncertainties. Bin size $\tau = 30$ ns.

To deduce $R_\tau(t_1, t_2)$ from Eq. 1, we next determine the joint detection probabilities $p_\tau(t_1, t_1)$ for field 1 and $p_\tau(t_2, t_2)$ for field 2 from the same record of photoelectric events as for Fig. 2 (a, c). Since the rate of coincidences for auto-correlations is roughly 10^2 times smaller than for cross-correlations for the (1, 2) fields, we increase the bin size τ to 30 ns to accumulate enough events to reduce the statistical errors to acceptable levels. Fig. 3 shows the resulting time dependencies of $p_\tau(t_1, t_1)$ and $p_\tau(t_2, t_2)$ for cases (I, II). While the shape of $p_\tau(t_1, t_1)$ associated with the *write* pulse does not change with Δt , the profile of $p_\tau(t_2, t_2)$ from the *read* pulse is affected and exhibits a rise time that is ~ 3 times shorter for $\Delta t = 200$ ns than for $\Delta t = 50$ ns. This prompt rise in (b) is consistent with the observation that stored excitation is efficiently addressed at the beginning of the *read* pulse for non-overlapping *write*, *read* pulses, while the longer rise time in (a) results from continuous excitation and retrieval of atoms from the state $|b\rangle$ for the overlapping case.

We employ the data in Figs. 2, 3 together with Eq. 1 to construct the ratio $R_\tau(t_1, t_2)$, with the result presented in Fig. 4 [13]. Not unexpectedly, the trends

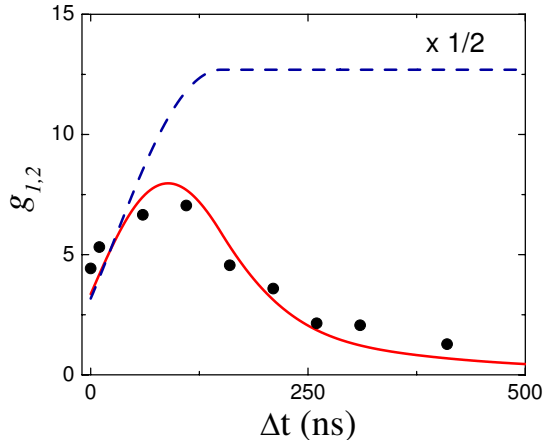


FIG. 5: Coherence time assessment. Experimentally acquired $g_{1,2}$ (black dots), theoretical description of the current experiment with $K = 1.1$ MHz (solid line), and the theoretical prediction for a spin polarized $m_F = 0$ MOT (dotted line).

for $R_\tau(t_1, t_2)$ closely resemble those of the joint probability $p_\tau(t_1, t_2)$ for correlated pair generation previously discussed. As for the violation of the Cauchy-Schwarz inequality $R_\tau(t_1, t_2) \leq 1$ for classical fields [7, 12], we observe maximal violations with $R_\tau^{\max} = 292 \pm 57$ for $\Delta t = 50$ ns and $R_\tau^{\max} = 202 \pm 60$ for $\Delta t = 200$ ns ($R_\tau = 198 \pm 33$ in the neighboring bin). The relatively large errors in $R_\tau(t_1, t_2)$ arise predominantly from the uncertainties in $p_\tau(t_1, t_1)$ and $p_\tau(t_2, t_2)$ (Fig. 3) [14].

The forms for $p_\tau(t_1, t_2)$ and $R_\tau(t_1, t_2)$ for the cases $\Delta t = 50$ and 200 ns imply a decoherence process operative on a time scale $\tau_d \sim 175$ ns. To investigate this decay, we have performed a separate set of experiments with the delay Δt varied $0 \leq \Delta t \leq 400$ ns. For each Δt we determine the normalized correlation function $g_{1,2}^\tau$ from the ratio of integrated coincidence counts to singles counts over the entire detection window (i.e., $\tau = 200$ ns), with the results presented in Fig. 5.

In Fig. 5, the initial growth of $g_{1,2}^\tau$ for small Δt is due to the finite time required to produce sequentially photons in the (1, 2) fields, which is already evident in Fig. 2. More troublesome is the rapid decay of $g_{1,2}^\tau$. A likely candidate responsible for this decay is Larmor precession among the various Zeeman states of the $F = 3, 4$ hyperfine levels of the $6S_{1/2}$ ground level.

To investigate this possibility, we have extended the treatment of Ref. [15] to include the process involving the *read* beam as well as the full set of Zeeman states for the $F = 3, 4$ hyperfine levels. The sample of Cs atoms is assumed to be initially unpolarized and distributed over the same range of magnetic fields as for the MOT. With *write* and *read* pulses that approximate those used in

our experiment, we obtain an expression for the probability $\tilde{p}_\tau(t_1, t_2)$ to generate a pair of photons at times (t_1, t_2) for fields 1 and 2 as a function of the offset Δt . By summing contributions for all (t_1, t_2) over the detection windows, we arrive at the joint probability $\tilde{p}_{1,2}(\Delta t)$ that we compare to the measured $g_{1,2}(\Delta t)$ by way of a single overall scaling parameter for all Δt , as the rate of single counts in fields (1, 2) is measured not to depend on Δt (to within 20%). The result is the solid curve in Fig. 5 that evidently adequately describes the impact of Larmor precession on our experiment. The form of $\tilde{p}_{1,2}(\Delta t)$ strongly depends upon the inhomogeneity of Zeeman splitting across the MOT, which is described by the parameter $K = \mu_B g_{F_g} L b / h$, where L is the MOT diameter, b is the gradient of the magnetic field for the MOT, and g_{F_g} is the Landé factor. The curve in Fig. 5 is the theoretical result for an initially unpolarized sample with $K = 1.1$ MHz as for our experiment (i.e., $L \approx 3.6$ mm and $b \approx 8.4$ G/cm).

An obvious remedy for this dephasing is to eliminate the magnetic field altogether, as by transferring the sample to a dipole-force trap [11]. Alternatively, we have developed a scheme that should allow for long coherence times even in the presence of the quadrupole field of the MOT by utilizing only magnetic-field insensitive states. The *write*, *read* beams are polarized σ_\pm and are aligned along the z -axis of the MOT, which provides the quantization axis. Atoms within the approximately cylindrical volume illuminated by these beams are initially spin polarized into $F = 3, m_F = 0$ [16]. The (1, 2) fields are selected to be σ_\pm , which results in spin excitation stored in $F = 4, m_F = 0$. The prediction of our model for this new protocol for the same experimental conditions as before but now with an initially spin polarized sample is shown as the dashed curve in Figure 5, resulting in an increase of more than $3\times$ in $g_{1,2}^\tau$, and significantly extending the decoherence time to more than $\tau_d \sim 10\mu\text{s}$.

In conclusion, we have reported the first observations of the temporal dependence of the joint probability $p_\tau(t_1, t_2)$ for the generation of correlated photon pairs from an atomic ensemble, which is critical for the protocol of Ref. [4]. Our measurements of $p_\tau(t_1, t_2)$ are an initial attempt to determine the structure of the underlying two-photon wavepacket [17]. The nonclassical character of the emitted (1, 2) fields has been tracked by way of time dependence of the ratio $R_\tau(t_1, t_2)$, with $R_\tau^{\max} = 292 \pm 57 \not\leq 1$. Decoherence due to Larmor precession has been characterized and identified as a principal limitation of the current experiment. A new scheme for effectively eliminating this decay process has been proposed and analyzed, and could be important for the experimental realization of scalable quantum networks [4] as well as for an improved source for single photons [8].

This work is supported by ARDA, the Caltech MURI Center for Quantum Networks, and the NSF.

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