Photon-Pair State Engineering in Raman-Mediated Four-Wave Mixing

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Abstract: Towards delayed-choice generation of single photons in pure quantum states, we measure and model the purity of Stokes photons scattered from sapphire, measuring a maximum purity of 0.99±0.03 and high quantum correlation with anti-Stokes photons.

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Single photons in pure quantum states are an important resource for quantum applications in communication, cryptography, and computation, but despite major progress in the field, realization of a source of high-purity, deterministic single photons remains a challenge. One approach to implementing delayed-choice single photon generation is the Duan-Lukin-Cirac-Zoller (DLCZ) protocol [1], wherein delayed anti-Stokes photons are generated via spontaneous Raman scattering. In this process a pump photon inelastically scatters, creating a Stokes photon together with a material excitation, which can be converted deterministically into an anti-Stokes photon within the excitation lifetime. Although this protocol has been demonstrated in various media, the purity of the photonic quantum states generated through this process has only been partially explored [2]. In solid-state media, the material excitation may take the form of a phonon mode; in a simple picture, the larger the ratio is between the bandwidth of the pump photon and the linewidth of the phonon mode, the higher the purity of the emitted photons will be. We present data of the purity of Stokes photons as a function of material thickness and pump bandwidth for c-axis single-crystal sapphire (α-Al_2O_3), which presents low fluorescent background [3] and is amenable to waveguide geometries. These results are qualitatively consistent with a simplified theoretical model based on the material dispersion relation, phonon frequency and phonon linewidth.

In this experiment ∼100 fs pulses at 80 MHz repetition rate from a modelocked Ti:sapphire laser are focused into a sapphire crystal in free space. The Stokes-scattered photons heralding the phonon mode at 746.6 cm^{-1}, with linewidth of 11.0 cm^{-1} [6], are collected in single mode fiber after spectral filtering of the pump photons. The far detuning of this phonon mode allows for the use of a wide range of pump bandwidths, controlled by a dispersive prism $4f$ pulse shaper. After polarization state filtering, Stokes photons are detected by two avalanche photodiode (APD) single photon detectors in the Hanbury Brown-Twiss interferometric configuration shown in Figure 1(a). The output of each APD is sent to a time-stamping coincidence counter, allowing for measurement of the second-order Stokes autocorrelation function

$$g^{(2)}_{S,S}(t_1,t_2) = \frac{\langle : \hat{n}_1(t_1)\hat{n}_2(t_2) : \rangle}{\langle \hat{n}_1(t_1)\rangle\langle \hat{n}_2(t_2)\rangle}, \tag{1}$$

expressed in terms of time-dependent photon number operators $\hat{n}_i = \hat{a}_i^{\dagger}(t)\hat{a}_i(t)$ in arms 1 and 2 of the interferometer at times t_1 and t_2, where $\langle :: \rangle$ indicates normal ordering. Normalized at time zero, this second order autocorrelation can be shown to be identical to $1+P$, where P is the Stokes photon state purity [4,5].

At a pump bandwidth of approximately 8 nm, we observe purities of 0.99±0.03 and 0.87±0.04, for crystal thicknesses of 1 mm and 8 mm, respectively. For a pump bandwidth of 1.2 nm and 8 mm crystal length, the purity decreases to 0.61±0.06. Uncertainties in purity are statistical, calculated from recorded coincidence histograms by adding in quadrature the Poissonian uncertainty in coincidences at time $\Delta t = t_2 - t_1 = 0$ and averaged coincidences at $\Delta t \neq 0$. The coincidence histogram for the 1 mm crystal thickness at 8 nm bandwidth is shown in Figure 2.

These results are in qualitative agreement with a simplified theoretical model. This model takes into account energy conservation given a certain phonon frequency and linewidth, and group velocity dispersion (GVD) given by the dispersion relation in sapphire [7]. Assuming collinear propagation, we find the GVD of the crystal induces coupling between the Stokes photon frequency and momentum, leading to a degradation of Stokes photon state purity for larger crystal thickness and larger pump bandwidth, in agreement with our experimental data.

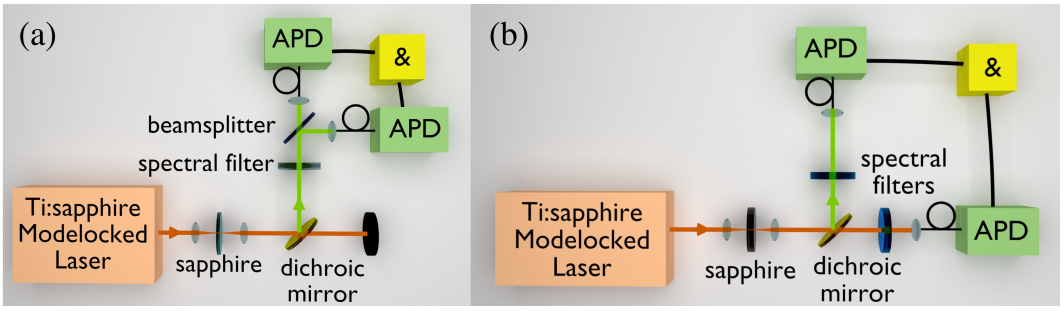

Fig. 1. Experimental setup for (a) $g^{(2)}_{S,S}$ autocorrelation measurement of Stokes photons, to determine photon state purity, and (b) $g^{(2)}_{S,AS}$ crosscorrelation measurement of Stokes and anti-Stokes photons, to determine the degree of quantum correlation. For both measurements, avalanche photodiodes (APDs) connect to a time-stamping coincidence counter (&).

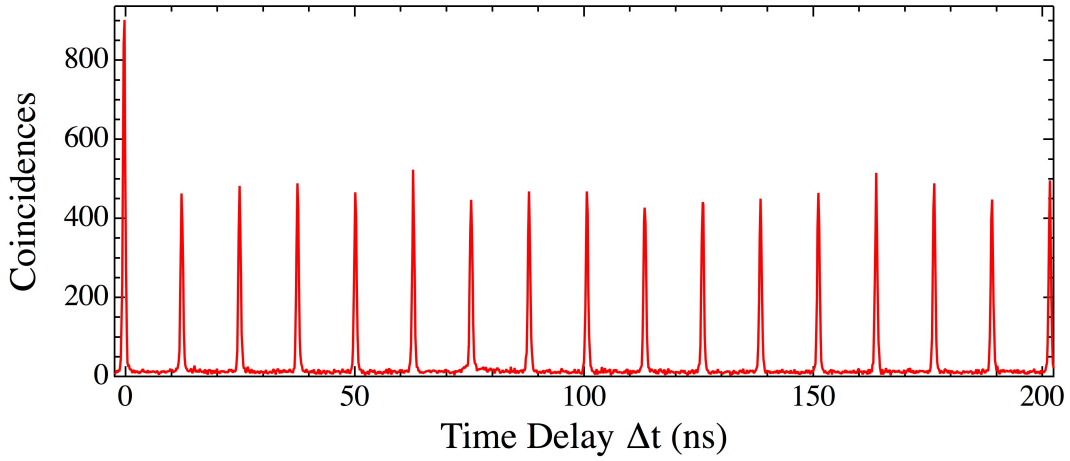

Fig. 2. Second-order autocorrelation coincidence histogram for Stokes photons scattered from a 1 mm sapphire crystal.

We also measure the second-order coherence crosscorrelation between Stokes and anti-Stokes photons, defined in the same way as Equation (1), where arms 1 and 2 refer instead to the Stokes and anti-Stokes collection arms (see experimental setup shown in Figure 1(b)). We find a value of 340±10 for 8 mm crystal thickness, indicating a high degree of quantum correlation between Stokes and anti-Stokes photons. This and the near-unity measured purities lay the groundwork for implementation of the DLCZ protocol in dispersive media and for efficient, delayed-choice production of pure single photons. This work is supported in part by NSF Grant Nos. 1521110 and 1640968.

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