Photon-Phonon Pair Correlations in Sapphire

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Abstract: We measure the quantum-state purity of Raman-scattered photons from sapphire, achieving a purity of 1.00 ± 0.03 and quantitative agreement with a new theoretical model of photon-phonon correlations that includes dispersion and finite excitation lifetime.

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Raman scattering is an important and popular resource for a host of proposed quantum applications [1, 2], yet the correlations between photon–excitation pairs, which may influence the purity of the emitted photons, have only been partially explored [3]. We present a phenomenological model for the photon–excitation joint state, including effects due to finite excitation linewidth and material dispersion. We empirically test this model on photon–phonon pairs produced in the well-suited and novel photonic medium of single-crystal sapphire, which presents low fluorescent background [4] and amenability to waveguide and fiber geometries [5]. Investigating photon–phonon correlations through simulation and experiment, we find quantitative agreement between experimental data and model predictions in the absence of any fitting parameters.

The process we wish to describe is depicted in Figure 1(a). We consider a laser pump beam of field amplitude $E(\omega_p)$ propagating in the z direction incident on a Raman-active medium of effective length L, which undergoes collinear three-wave mixing, producing a Stokes-scattered photon of frequency ω_s and an excitation of frequency Ω. We assume far off-resonant scattering such that we can adiabatically eliminate the effects of intermediate states in the interaction and write the effective interaction Hamiltonian:

$$H_{int} = \eta \int_{-L/2}^{L/2} dz \int d\omega_p d\omega_s d\Omega \, E(\omega_p) e^{i(n_p \frac{\omega_p}{c} z - \omega_p t)} g(\Omega) \hat{a}_s^\dagger(\omega_s) e^{-i(n_s \frac{\omega_s}{c} z - \omega_s t)} \hat{B}^\dagger(\Omega, z) e^{i\Omega t} + \text{c.c.}, \tag{1}$$

where η is a coupling coefficient, $n_{p,s}$ are the indices of refraction for pump and Stokes photons, respectively, $g(\Omega)$ the lineshape of the excitation, $\hat{a}_s^\dagger(\omega_s)$ the creation operator for a Stokes photon, and $\hat{B}^\dagger(\Omega, z)$ the creation operator for an excitation at point z along the interaction medium. For long interaction times, in this perturbative approach we write the joint photon–excitation state as

$$|\Psi\rangle = \int_{-L/2}^{L/2} dz \int d\omega_s d\Omega \, f(\omega_s, \Omega, z) \hat{a}_s^\dagger(\omega_s) \hat{B}^\dagger(\Omega, z) |vac\rangle, \tag{2}$$

revealing the joint spectral amplitude to be $f(\omega_s, \Omega, z) = \alpha E(\omega_s + \Omega) g(\Omega) e^{i(n_p \omega_p / c - n_s \omega_s / c) z}$, for normalization constant α. This amplitude captures the spectral correlations between Stokes photon and excitation, including those arising from the excitation linewidth and group velocity dispersion (GVD) in the medium.

In this experiment ∼100 fs pulses at 80 MHz repetition rate from a modelocked Ti:sapphire laser are focused into single-crystal c-axis sapphire (α-Al_2O_3) in free space. Sapphire is an attractive choice for this study due to its non-negligible dispersion and phonon linewidth that is nearly single frequency compared to full pump bandwidth (∼ 5000 GHz) but on the same order as our narrowest pump bandwidth (∼ 50 GHz). 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 a Hanbury Brown-Twiss interferometric configuration. The output of each APD is sent to a time-stamping coincidence counter, allowing for measurement of the second-order Stokes autocorrelation function $g^{(2)}_{S,S}(t_1, t_2) = \frac{\langle :\hat{n}_1(t_1)\hat{n}_2(t_2): \rangle}{\langle \hat{n}_1(t_1) \rangle \langle \hat{n}_2(t_2) \rangle}$, 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 [7]. This measured state purity of Stokes photons acts as an indicator of the degree of photon–phonon correlation, allowing for comparison with our model.

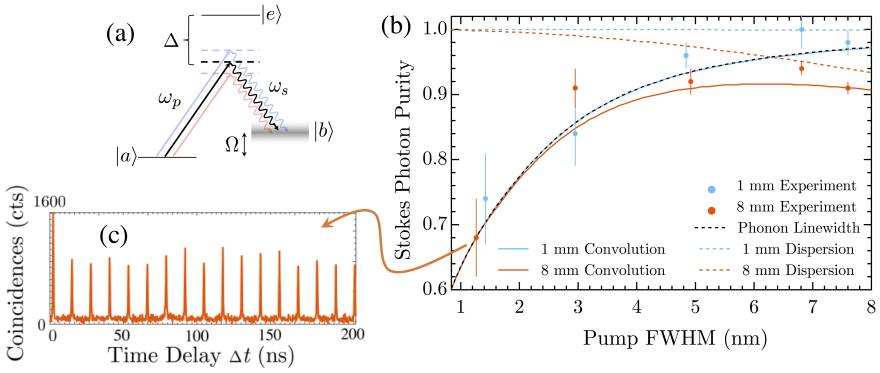

Fig. 1. (a) Raman level structure with ground state $|a\rangle$, pump photon ω_p, excitation Ω, far detuned (Δ) state $|e\rangle$, collective state $|b\rangle$, and Stokes photon ω_s. (b) Predicted (solid lines) and experimental (points) Stokes photon purities for 1 mm (blue) and 8 mm (orange) sapphire crystal lengths, and the isolated effects of finite phonon linewidth (black dashed line) and dispersion (blue and orange dashed lines). (c) Each datum corresponds to a second-order autocorrelation histogram.

In Figure 1(b) we present the results of these measurements for 1 and 8 mm sapphire crystal lengths at varying pump bandwidths. Solid lines show the predictions of the model. Dashed lines show only the effects of the phonon linewidth (black dashed line) and dispersion (blue and orange dashed lines). Notably, the Stokes photon purity for the 1 mm crystal length is determined almost completely by the effect of the phonon linewidth, and features negligible degradation of purity due to dispersion over the experimental range. We observe quantitative agreement between our model and the experimental data, along with a measurement of unity purity $P = 1.00 \pm 0.03$ for the 1 mm crystal length at 6.81 nm pump FWHM. We note that the predictions of our model in this figure are calculated without any fitting parameters.

Our model reveals a degradation of Stokes photon state purity for smaller pump bandwidths due to the excitation linewidth: in the limit of a monochromatic pump, the linewidth of the excitation allows for different color Stokes photons, each correlated with an excitation frequency through energy conservation. For larger bandwidths, the excitation is effectively at a single frequency compared to the pump and thus energy entanglement is diminished; however, we find a separate effect then arises due to GVD in the medium. Photon–excitation correlations increase proportional to the medium length (in Fig. 1(b), note the purity difference between 1 and 8 mm thicknesses at large bandwidth); this is due to GVD inducing temporal walk-off between Stokes and pump pulses. The degree of walk-off is determined by the spatial location of photon–excitation pair production, with each Stokes photon correlated with an excitation created at the same location. For smaller lengths the group delay between Stokes and pump is diminished, and with it this photon–excitation entanglement due to GVD. These competing effects lead to a maximum Stokes photon state purity at finite bandwidth.

Our one-dimensional treatment is applicable in waveguide or fiber geometries, or in bulk media when the interaction length is less than the Rayleigh range of the free-space focused pump beam. Work on a three-dimensional treatment is ongoing. We also measure the second-order crosscorrelation between Stokes and anti-Stokes photons, defined in the same way as $g^{(2)}_{S,S}(t_1,t_2)$, where arms 1 and 2 refer instead to the Stokes and anti-Stokes collection arms. 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 work is supported in part by NSF Grant Nos. 1521110 and 1640968.

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