Raman Scattering Beyond the Master Equation: Photon-Matter Correlations and Statistics

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Abstract: We present 1D and 3D models that take into account Stokes-photon–excitation pair correlations in Raman scattering, revealing nontrivial dependence of the photon statistics on linewidth, dispersion and collection angle.

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Spontaneous Raman scattering has traditionally been modeled using the master equation [1], in which excitation lifetimes are taken into account through coupling to an external bath. In this approach the excitation is created with a discrete energy, while its amplitude decays exponentially. Although this treatment models well the spectrum of the Raman gain, it neglects the correlations between spontaneous Stokes photon and excitation. Here we show that these correlations have a critical effect on the state of the photon and consequently on its quantum-state purity and the photon-statistics of the Raman scattering. We present 1D and 3D models of the Raman interaction that take finite excitation linewidths into account in the Hamiltonian. In addition to the dependence on linewidth, we also find that the degree of correlation depends strongly on collection geometry and medium dispersion, such that Stokes photons scattered counter-propagating from the pump are highly correlated with the material excitation. This work has implications for the quantum state purity of photons generated via Raman scattering in solid-state systems [2] as well as atomic vapors [3], and thence on the implementation of quantum protocols such as the Duan-Lukin-Cirac-Zoller protocol [4] and quantum memories [5].

The general Hamiltonian for Raman scattering processes [6] can be restricted to reflect only the process of Stokes scattering, and recast in terms of photon and excitation field operators as:

$$H_I^{\text{Stokes}}(t) = \int d^3\vec{r} \int d\Omega\, g(\Omega)\, \hat{E}_p^{(+)}(\vec{r},t)\hat{E}_s^{(-)}(\vec{r},t)\hat{B}^{(-)}(\Omega,\vec{r},t) + h.c., \tag{1}$$

where the subscripts p,s, refer to pump and Stokes, respectively. The $\hat{B}(\Omega,\vec{r},t)$ field-operator is associated with the excitation field at point $\vec{r}$ in the medium and energy $\hbar\Omega$. The function $g(\Omega)$ is the frequency dependence of the coupling strength that gives rise to the Raman scattering spectral gain. Under normal circumstances it takes the form of a Lorentzian lineshape. The inclusion of $g(\Omega)$ and the energy-dependence of the $\hat{B}(\Omega,\vec{r},t)$ operator is due to the interaction of the excitation created by the Raman scattering with the environment. The treatment of the interaction in these terms is the major difference between our approach and the master equation; the latter makes use of a non-Hermitian Hamiltonian to account for coupling to the environment, thus neglecting any information about the quantum state of the environment.

We further assume that the $\hat{B}(\Omega,\vec{r},t)$ field operators are associated with excitations that obey boson statistics; this is justified when the distance between interaction sites is on the order of or smaller than the wavelength of the light [7]. In addition, we treat these excitations as localized – a valid assumption within the timescales involved in our work (laser pulses $\sim$1 ps duration).

Beginning with a 1D model, corresponding to waveguided propagation of pump and Stokes pulses, the photon-excitation joint states created in the spontaneous Raman scattering process with finite excitation linewidth are

$$|\Psi\rangle_{1\text{D}} = \int_{-L/2}^{L/2} dz \int d\omega_s d\Omega\, f_{1\text{D}}(\omega_s,\Omega,z)\hat{a}_s^\dagger(\omega_s)\hat{B}^\dagger(\Omega,z)|0\rangle, \quad \text{where} \quad f_{1\text{D}}(\omega_s,\Omega,z) = N_{1\text{D}}E(\omega_s+\Omega)g(\Omega)e^{i(k_p-k_s)z}, \tag{2}$$

with normalization $N_{1\text{D}}$ and pump envelope $E(\omega_s+\Omega)$.

For scattering in bulk media, we employ 3D calculations, considering a Gaussian-profile pump beam and assuming that the Stokes photons are collected with a single-mode fiber, which we model as a projection onto another Gaussian profile. We find that in this case the joint amplitude is given by

$$f_{3\text{D}}(\omega_s,\Omega,z) = N'_{3\text{D}}\frac{|\vec{k}_p||\vec{k}_s|}{q_f^*(z)q_p(z)}f_{1\text{D}}(\omega_s,\Omega,z), \tag{3}$$

where $q_j(z) = z + iz_{R,j}$ is the complex beam parameter for pump or fiber mode with Rayleigh range $z_{R,j}$. Thus we find for free-space focused beams the joint state is similar to that of the 1D model, with a z-dependent correction. For media longer than the pump and/or fiber mode Rayleigh ranges, this correction apodizes the collection of

Stokes photons along the medium, suppressing collection of photons generated at the ends of the medium due to the spatial dependence of the overlap of pump and collection modes. This overlap is shown schematically in Fig. 1(a). For media smaller than the Rayleigh ranges ($z \ll z_{R,j}$ for both $j = p, f$) we recover the 1D result.

We cannot directly measure the joint amplitudes in Eqs. (2) and (3); however, since they describe the entanglement between Stokes photon and excitation, the degree of entanglement plays a significant role on the photon statistics, specifically on the second-order coherence autocorrelation function $g^{(2)}(0)$ [8]. In particular, when the Stokes-photon wavefuntion is not correlated with the material excitation, $g^{(2)}(0) = 2$ and the photon quantum state is pure. Correlation between the photon and the excitation results in reduced $g^{(2)}(0)$ value, down to a lower limit of $g^{(2)}(0) = 1$ for an infinitely-correlated state. In this case the photon is in an infinitely mixed state.

In this work we consider single-crystal, c-cut bulk sapphire as the Raman medium. Using a Hanbury-Brown–Twiss interferometer (Fig. 1(b)), we measure the second-order Stokes autocorrelation function at various pump pulse bandwidths. We compare the experimental results to the ones calculated with our model. As can be seen in Fig. 1(c), the value of $g^{(2)}(0)$ increases with bandwidth, as we expect from our 3D model. Despite the general agreement between the experiment and model on the trend, the experimental $g^{(2)}(0)$ are consistently lower than the ones predicted by the model—we attribute this to fluorescent photons from electronic excitation of crystal impurities that are collected and occur in conjunction with the Raman scattering process.

We repeat the above, this time measuring the autocorrelation function of the backward-scattered Raman photons; in this case the joint amplitude is evaluated to be $f^{\leftarrow}_{3D}(\omega_s, \Omega, z) = -N'_{3D} \frac{|\vec{k}_p||\vec{k}_s|}{q^*_f(z) q_p(z)} f^{\leftarrow}_{1D}(\omega_s, \Omega, z)$, where $f^{\leftarrow}_{1D}(\omega_s, \Omega, z) = N'_{1D} E(\omega_s + \Omega) g(\Omega) e^{i(k_p + k_s)z}$. Although the spectrum of the backward-scattered photons is identical to that of the forward-scattered, we find (see Fig. 1(c)) that the $g^{(2)}(0)$ values are much lower and close to the lowest limit of 1, indicating high correlations between these photons and the excitations in the matter. Physically, these correlations are between the arrival time of the photon at the detector and the point in the crystal where the interaction took place (hence the location of the excitation).

We have also generalized the above treatment to any off-axis collection angle of Stokes photons. As we would expect based on the findings above, our preliminary results indicate a strong dependence of the photon statistics on the collection angle. These results have important implications for the use of Raman scattering generally in quantum applications. This work is supported by NSF Grant Nos. 1521110, 1640968, 1806572, and DMR-1747426.

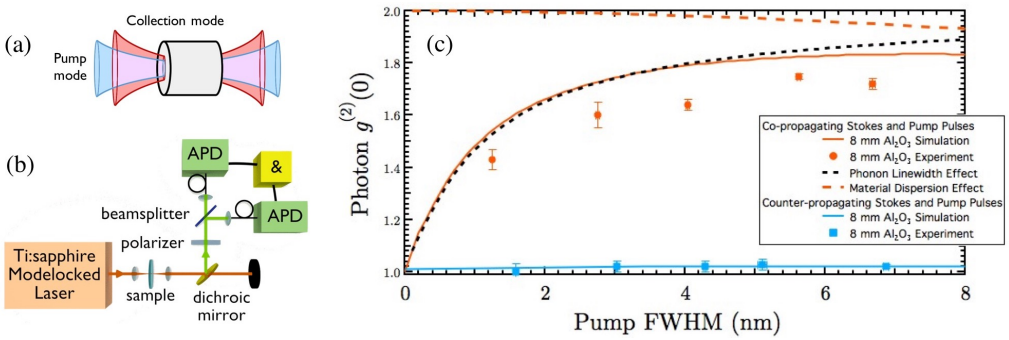

Fig. 1. (a) Schematic of pump and collection modes. (b) Hanbury-Brown–Twiss interferometer used to measure Stokes photon $g^{(2)}(0)$. (c) Simulated and measured Stokes $g^{(2)}(0)$ in co- (orange) and counter-propagating (blue) collection geometries, and contributions from excitation lineshape (black, dotted) and dispersion (orange, dashed).

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