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Broadband photon-counting Raman spectroscopy in short optical waveguides

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We present a method of directly measuring the spontaneous Raman scattering in optical waveguides in an alignment-free setup. Using a pulsed laser, liquid-crystal-on-silicon spatial light modulator and single-photon detector, we create a broadband photon-counting Raman spectrometer. The temperature and polarization dependence are characterized in an As2S3 amorphous glass fiber for a Stokes detuning range of 1 to 9 THz from the pump frequency. We fit our experimental data with a theoretical model and extract the Raman-gain spectrum and compare to free-space measurements of bulk As2S3. The sensitivity of the method in principle allows direct characterization of chip-scale nanophotonic devices. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4767220]

Raman scattering is a well-known phenomenon that can be exploited for amplification in optical fiber telecommunication but is considered a nuisance in quantum communication networks. For example, there is significant interest in using amorphous glass waveguides for quantum applications including correlated photon-pair generation,1–3 entanglement generation,4,5 and the frequency conversion of single photons.6,7 However, spontaneous Raman scattering (SpRS) is a detrimental noise source in such devices.2 Partial suppression of SpRS has been demonstrated using cryogenic cooling8–11 and almost complete mitigation has been achieved in an As2S3 waveguide by placing the single photon channels in a known minimum of the Raman spectrum.12 Knowledge of the SpRS spectrum enables the choice of lowest noise configuration for the pump and photon channels,13 but at the single-photon level, SpRS cannot be detected using a standard spectrum analyzer.

Spontaneous and stimulated Raman scattering have been measured in order to characterize both Raman noise and material gain spectra. The spectral distribution of SpRS can be measured in bulk samples using the 90° scattering method,14–17 where a free-space beam is incident on the sample and the SpRS component that is emitted perpendicular to the pump is collected by free-space optics and measured with a spectrograph. SpRS can also be measured in optical fibers but requires a pulsed laser and long fiber lengths to achieve a measurable signal strength.18–20 Direct measurements of stimulated Raman scattering have been performed using nonlinear pump-probe techniques.21–25 All of the techniques described require high powers incompatible with the damage threshold of many on-chip devices and, in the nonlinear case, the addition of either a highly tunable or ultra-broad bandwidth probe. Recently, photon-counting techniques have been demonstrated to measure weak SpRS signals for very small detunings and low temperatures in silica fibers.26,27 However, no method applicable to measuring SpRS directly over a broad bandwidth in chip-scale waveguides has been presented.

In this letter, we show that it is possible to create a broadband photon-counting spectrometer, using commercially available integrated components in an alignment-free setup. The method, outlined in Fig. 1, uses a pulsed laser to excite SpRS in the waveguide, a tunable bandpass filter to resolve the spectral distribution, and a single photon detector to provide the sensitivity needed to measure short devices. We demonstrate this by measuring the temperature and polarization dependence of the SpRS spectrum in an As2S3 chalcogenide glass fiber.28 By fitting the experimental data, we extract the Raman-gain spectrum and find good agreement with previous measurements.15,17 In principle, the single photon sensitivity allows SpRS to be measured directly in chip-scale photonic devices, where 90° scattering and nonlinear techniques are not applicable.

We implement our method using the experimental setup shown in Fig. 2. As a first demonstration, we used a short fiber for ease of immersion in cryogenic liquid, but the technique is equally applicable to planar waveguides. The laser generated 10 ps pulses centered at 1535 nm with a 0.3 nm bandwidth at a repetition rate of 10 MHz. An isolator protected the laser from back-reflections and a variable attenuator (ATT) with a polarization controller conditioned the

more than 50 dB isolation. We achieved a 100 GHz wide flexibility pass-band with 90 dB of out-of-band noise suppression using a liquid-crystal-on-silicon (LCoS) spatial light modulator (Finisar Waveshaper) in combination with a tunable C/L band BPF. The SpRS spectrum was mapped by moving the filter pass-band to different detunings and counting the generated photons. The component loss varied over the full detuning range and was corrected for at each detuning. The Waveshaper was characterized using an amplified spontaneous emission (ASE) source before it was inserted into the setup. To measure the BPF, a 99%/1% coupler was placed between the Waveshaper and the BPF. A C/L band source of broad-band ASE was connected to the 1% input and the tunable BPF at the 99% output. A second coupler was connected after the BPF, with output 1 directed to a single-photon detector (SPD, Id-Quantique ID210) via an ATT and output 2 connected to an optical spectrum analyser (OSA). By blocking the SPD and opening the ASE source, the C+L BPF was manually tuned and the loss (including couplers) was measured for each detuning without breaking any fiber connections. The quantum efficiency of the detector at each detuning was also taken into account. From this, a frequency-dependent transfer function was calculated for the experimental setup.

The probability of scattering a photon from the pump to the Stokes field, within our measurement bandwidth $\Delta \nu = 100$ GHz, was less than 0.01 SpRS photons per pump pulse, inferred from the photon count rate, therefore it was appropriate to count photons using a SPD. This corresponded to a coupled peak power at the fiber of 1.6 W, assuming symmetric input-output coupling loss. The detectors were triggered in synchrony with the pulsed pump at 10 MHz and set to a detection efficiency of 10%, effective gate width of 1 ns, and a dead-time of 20 $\mu$s. The SPD characteristic dark count rate was approximately 50 Hz, when triggered at 10 MHz. To check for pump leakage, an ATT with comparable length pigtailed was temporarily inserted into the setup in place of the As$_2$S$_3$ fiber. The attenuation was set to match the coupling loss and no increase in the noise count rate was seen over the intrinsic detector dark counts, proving pump leakage to be negligible.

In SpRS, pump photons interact with the material through the polarizability of the molecular lattice and emit/absorb a phonon lowering/increasing the photon energy in the Stokes/anti-Stokes case. The experiment operated in the regime of pure spontaneous Raman scattering, as there were initially no photons in the Stokes mode to induce the Raman scattering process. We also considered photons generated by spontaneous four-wave-mixing (SFWM) in the fiber for both Stokes and anti-Stokes detuning. The dispersion parameter $D$ was calculated to be $-405.8$ ps km$^{-1}$ nm$^{-1}$ at 1535 nm corresponding to a calculated SFWM bandwidth of 0.8 THz. Our SpRS measurements start from 1 THz, therefore we consider the SFWM contribution to be small up to 2 THz and negligible beyond that detuning. Note that we define the bandwidth to be the half-width at half maximum.

To test our method, we characterize the SpRS in a As$_2$S$_3$ fiber including both the temperature and polarization dependence. The measurements for the component polarized parallel to the pump at 293 K and 77 K are plotted in Fig. 3.
as red circles and blue triangles, respectively. For near detunings, cooling the fiber to 77 K reduces the SpRS by more than 4 dB. As the frequency detuning from the pump is increased, changes in temperature \( T \) have less of an impact, with the SpRS rate for both temperatures converging. These effects can be explained by the dependence of the SpRS rate on the phonon population. SpRS can be described as the emission or absorption of a phonon by a pump photon. The Stokes case includes both spontaneous and stimulated emission of a phonon, where the stimulated component depends on the phonon number, with a distribution described by Bose-Einstein statistics

\[
n_{BE} = \frac{1}{e^{\nu/k_B T} - 1},
\]

with \( \nu \) the detuning from the pump frequency. This is not to be confused with the stimulated Raman scattering process enhanced by the presence of photons in the scattered mode. The Stokes intensity is therefore proportional to \( n_{BE} + 1 \). From Eq. (1), it follows that the low energy phonon states are more likely to be occupied, enhancing the photon count rate closer to the pump as observed in Fig. 3 for both temperatures. The SpRS component proportional to \( n_{BE} \) is dependent on \( T \) and is strongest close to the pump. This dependence of the SpRS rate on \( T \) weakens for increased detuning as the ratio between the phonon energy \( \hbar \nu \) and thermal energy \( k_B T \) increases and \( n_{BE} \) becomes less dominant. We note that even at zero temperature Stokes scattering occurs via spontaneous phonon emission.

Polarization effects are observable in the Raman scattering spectrum even in the absence of material anisotropy. In an isotropic medium, there are isotropic and anisotropic components corresponding to the two free parameters of the Raman response tensor.\(^{13,32} \) Figure 4 shows the SpRS component polarized parallel (green diamonds) and perpendicular (brown squares) to the pump, measured at room temperature. These orthogonal polarization components include different contributions from the isotropic and anisotropic Raman response and therefore have different shaped spectra, as previously observed for As\(_2\)S\(_3\) glass.\(^{15} \) The SpRS photon count rate\(^{13} \) is expressed as

\[
R_{SpRS}(\nu, T) = C\eta(\nu_s)\Delta\nu P_0 L \left[ 1 + n_{BE} \right] g_{gain}(\nu),
\]

where \( C \) is the Raman coupling coefficient, \( \eta(\nu_s) \) is a frequency dependent transfer function for the experimental setup, \( \nu \) is the detuning from the pump, \( \Delta\nu \) is the fixed measurement bandwidth, \( P_0 \) is the coupled peak power, \( L \) is the effective fiber length, \( \nu_s \) is the Stokes frequency, and \( g_{gain}(\nu) \) is the Raman-gain spectrum which we note is the Fourier transform of the Raman response functions.\(^{33} \) The Raman-gain includes the polarization components parallel and perpendicular to the pump \( g_{gain}(\nu) = g_{||} + g_{\perp} \). By fitting our measurement using Eq. (2), we extracted \( g_{gain}(\nu) \) and compared this with data from measurements of a bulk sample of As\(_2\)S\(_3\) glass. The normalized gain for the parallel polarization \( g_{||} \) extracted from our SpRS measurement and the gain measured in a bulk sample are shown in Fig. 5 as red circles and a black dashed line, respectively. The two spectra are in good agreement, further confirming the accuracy of our method. Our measurement was limited by our C/L band BPF and therefore did not extend beyond 9 THz from the pump. For small detunings, the Bose-Einstein enhancement (1) will increase approximately proportional to \( \nu^2 \), but it has previously been observed that the SpRS rate goes to zero at the pump.\(^{8,15,17} \) Therefore to lowest order, the gain (which is an odd function of \( \nu \)) must vary as \( \nu^2 \) close to the pump. The SpRS was not measured closer to the pump as the signal was contaminated with SFWM and pump leakage.

The extracted Raman-gain \( g_{gain}(\nu) \) can be used to calculate the anti-Stokes SpRS for the As\(_2\)S\(_3\) fiber, shown in Fig. 6. In contrast to the Stokes case the anti-Stokes intensity is proportional only to \( n_{BE} \). We note that a low-Raman
window exists at 7.4 THz detuning, for both the Stokes and anti-Stokes emission, resulting from a characteristic dip in the phonon density of states. The ability to characterize the Raman scattering is useful in quantum communication applications, where for example, the in-band noise of a single photon channel operating near a bright laser can be reduced by moving it to a known SpRS minimum. This could apply to photon-pair generation via SFWM, frequency conversion via Bragg-scattering four-wave-mixing, or a classical information signal transmitted adjacent to the quantum channel. Our technique could also be used to measure the increasingly popular silicon nitride platform, which has high nonlinearity and attractive low loss properties. Another material of interest is amorphous silicon, which is free from the two-photon-absorption inherent to its crystalline counterpart.

Possible improvements to our method include increasing the measurement range of 9 THz, currently limited by our C/L band BPF and C-band pump. This could be implemented using a tunable source, for example an optical parametric oscillator, to pump at a shorter wavelength and extend the maximum detuning range. For integrated devices where broad SFWM would contaminate the SpRS signal, the back scattered SpRS could be measured and used to predict the forward Stokes and anti-Stokes SpRS noise. The SFWM contribution could also be identified within the forward scattering by including a second detector at the anti-Stokes wavelength to measure coincidence counts, with the coincident photons originating from the FWM.

In conclusion, we have demonstrated a method for direct broadband photon-counting Raman spectroscopy in an integrated and alignment-free setup. 90° scattering methods require bulk samples, and nonlinear methods need long device lengths. Our method overcomes this with broadband single photon sensitivity, making it possible to characterize Raman scattering in short optical waveguides.

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