Stimulated Raman scattering microscopy by spectral focusing and fiber-generated soliton as Stokes pulse

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We demonstrate stimulated Raman microscopy with broadband pump and Stokes pulses, using spectral focusing to attain spectral resolution and to rapidly acquire spectra within a spectral window determined by the bandwidth of the Stokes pulse, we use the redshifted soliton generated in a photonic crystal fiber, which allows for simple shifting of the accessible spectral window. © 2011 Optical Society of America


Stimulated Raman scattering (SRS) microscopy [1–4] is—along with coherent anti-Stokes Raman scattering (CARS) microscopy [5,6]—at present the most acclaimed method of vibrational microscopy. The predominant setup for SRS microscopy uses transform-limited picosecond pump and Stokes pulses of a spectral width roughly equal to the linewidth of the Raman-active vibrations of interest. The work detailed in the present Letter derives from [7–9], which demonstrated that CARS microscopy can be performed using equally chirped broadband pump and Stokes pulses to improve spectral resolution, the so-called spectral focusing scheme, and from [10], in which CARS microspectroscopy was demonstrated using a redshifted soliton generated in a photonic crystal fiber (PCF) as the Stokes pulse. The extension of spectral focusing to SRS microscopy warrants a closer study, because the detection schemes in CARS and SRS are different; whereas CARS microscopy detects the emitted anti-Stokes intensity, SRS microscopy measures the stimulated Raman loss (SRL) of the pump or the stimulated Raman gain (SRG) of the Stokes. This Letter has a threefold aim: (i) to demonstrate SRS microscopy by spectral focusing, (ii) to show that a redshifted soliton, generated in a PCF is viable as the Stokes pulse in an SRG measurement, and (iii) to demonstrate that the potential frequency and time jitter of the redshifted soliton can be suppressed in the spectral focusing scheme.

To calculate the SRG with equally chirped pump and Stokes pulses, we use the methods of [11]. We assume pump and Stokes fields of the form

\[ E_P(t) = E_{P0}(t)e^{-i\omega_p t - i\alpha\delta} + \text{c.c.}, \]

\[ E_S(t - \Delta t) = E_{S0}(t - \Delta t)e^{-i\omega_S(t - \Delta t) - i\alpha\delta} + \text{c.c.} \]

\[ E_P, E_S \] are the complex field envelopes, \( E_{P0}, E_{S0} \) are the absolute field envelopes, and \( \alpha \) is the common chirp parameter. We model the sample as a damped oscillator with frequency \( \Omega \) and dephasing rate \( \Gamma \) and assume that we are far from electronic resonance, which gives the following response function:

\[ S_{\text{SRG}}^{(3)}(t_3, t_2, t_1) \propto \left(-\frac{i}{\hbar}\right)^3 \delta(t_3)e^{i\Omega_2 - \Gamma_2}\delta(t_1). \]

Introducing the detuning \( \Delta = \Omega - \omega_p(t) + \omega_S(t) = \Omega - \omega_p + \omega_S - 2\alpha\Delta t \), the induced polarization is

\[ P_{\text{SRG}}^{(3)}(t) = \iint E_P(t - t_3 - t_2 - t_1)E_S(t - \Delta t - t_2 - t_1) \times E_P(t - t_1)S_{\text{SRG}}^{(3)}(t_3, t_2, t_1)dt_1 dt_2 dt_3 + \text{c.c.} \]

\[ = E_{P0}(t)e^{-i\omega_p(t - \Delta t) - i\alpha\delta} \]

\[ \times \int dt_2 e^{i\Delta \omega_2 - \Gamma_2}E_{P0}(t - t_2)E_{S0}(t - \Delta t - t_2) + \text{c.c.}. \]

The result is very similar to the result for the case where \( \alpha = 0 \) and \( \Delta = \Omega - \omega_p + \omega_S \), which we will call the “equivalent transform-limited case.” The only difference is that the \( e^{-i\alpha\delta(t - \Delta t)^2} \) prefactor to the integral is absent in the equivalent transform-limited case. This prefactor is of no consequence as the SRG, detected as the change in the Stokes intensity induced by the pump pulse,

\[ \text{SRG} = \int dt |P_{\text{SRG}}^{(3)}(t) + E_S(t)|^2 - \int dt |E_S(t)|^2, \]

is indifferent to a common phase on \( P_{\text{SRG}}^{(3)}(t) \) and \( E_S(t) \). The SRL is given by the negative of the SRG. We also calculate the CARS signal \( I_{\text{CARS}} \), in which case

\[ S_{\text{CARS}}^{(3)}(t_3, t_2, t_1) \propto \left(-\frac{i}{\hbar}\right)^3 \delta(t_3)e^{i\Omega_2 - \Gamma_2}\delta(t_1). \]

\[ P_{\text{CARS}}^{(3)}(t) = \iint E_P(t - t_3 - t_2 - t_1)E_S(t - \Delta t - t_2 - t_1) \times E_P(t - t_1)S_{\text{CARS}}^{(3)}(t_3, t_2, t_1)dt_1 dt_2 dt_3 + \text{c.c.} \]

\[ = E_{P0}(t)e^{-i(2\omega_p - \omega_S + 2\alpha\Delta t) - i\alpha\delta} \]

\[ \times \int dt_2 e^{i\Delta \omega_2 - \Gamma_2}E_{P0}(t - t_2)E_{S0}(t - \Delta t - t_2) + \text{c.c.}. \]

Then, the CARS signal,

\[ I_{\text{CARS}} = \int dt |P_{\text{CARS}}^{(3)}(t)|^2, \]

is the same as in the equivalent transform-limited case. The SRG, SRL, and CARS in the spectral focusing scheme
thus give the same signal as in the equivalent transform-limited case, from which conclusions concerning signal generation and resolution can be directly transferred to the spectral focusing case. This equivalence exists for single-frequency detection; for multiplex detection, a difference would subsist. The important difference is that the spectral focusing scheme has the possibility of changing $\Delta$ simply by changing $\Delta t$, as seen from the initial definition of $\Delta$, allowing rapid acquisition of a spectrum.

The experimental setup is sketched in Fig. 1. The pulses from the laser (Amplitude Systems pulse, 50 MHz, 1036 nm, 185 fs) act as pump pulse. In the nonlinear PCF [NL-2.0-745-02, Blaze Photonics, group-velocity dispersion $\beta_2 = -0.082 \text{ps}^2/\text{m}$, nonlinearity parameter $\gamma = 0.079 \text{W} \cdot \text{m}^{-1}$ at $\lambda = 1036 \text{nm}$], a fundamental soliton is formed, which redshifts due to the soliton self-frequency shift [12, 13] and acts as a Stokes pulse. The pump and Stokes pulses are negatively chirped by grating pairs (300 lines/mm) and are recombined on an interference filter and sent to the microscope. An image is obtained by raster scanning the sample. An acousto-optical modulator (AOM) imposes a modulation on the pump (1 MHz, modulation depth 70%). For the SRG measurement, the Stokes pulse is detected on an amplified InGaAs photodiode and the SRG signal is detected by a lock-in integrator, locked to the AOM. For the CARS measurement, the CARS signal is detected by an avalanche photodiode.

In the imaging experiment, we attempted to match the pulse durations to the typical dephasing time of the sample. The transform-limited pump has temporal and spectral widths FWHM of 185 fs and 8.5 nm and is Gaussian. For the transform-limited Stokes, the numbers are 70 fs and 20 nm and the profile is hyperbolic-secant squared. The grating pairs imposed second-order spectral phases $\Phi_2^S = \Phi_2 = -0.130 \text{ps}^2$ on both pulses, leading to a pump duration of FWHM = 1.9 ps and a Stokes duration of FWHM = 3.8 ps, as verified by intensity autocorrelation and cross-correlation. The corresponding chirp parameter is $\alpha = -3.8 \times 10^{-6} \text{fs}^{-2}$. As a sample, we used 20 $\mu$m polystyrene beads in index-matching oil sandwiched between two microscope cover slides. The oil has no Raman-active vibrational resonances in the 1000 cm$^{-1}$ region that we are studying. In Fig. 2, we present the resulting SRG and CARS images. In Fig. 2(b), we set $\Delta t = 0$ fs and choose $\lambda_S$ such as to give $\lambda_p - \lambda_S = 1003 \text{cm}^{-1}$, the frequency of the strongest peak in the polystyrene Raman spectrum. As expected, SRG is observed in the beads while no SRG is seen in the oil. Detuning $\Delta t$ by $\pm 350$ fs, as in Figs. 2(a) and 2(c), corresponds to changing $\Delta$ by $-2a \Delta t = \pm 14 \text{cm}^{-1}$. And as seen from the figures, this causes the SRG in the beads to disappear. It must be noted that the overlap between pump and Stokes is still significant at $\Delta t = \pm 350$ fs, because the FWHM of the cross-correlation is several ps. This details that spectral focusing takes place. We present the CARS images of the same sample under the same conditions in Figs. 2(d)–2(f), which display a fairly good contrast but still suffer from a nonnegligible nonresonant background from the oil as well as from the bead. The images at $\pm 350$ fs are slightly different due to the asymmetry in the CARS spectrum introduced by the interference between the resonant and nonresonant CARS signal. As is well known, SRG does not suffer from nonresonant contribution.

On the same sample, we investigate the spectroscopic capabilities of our setup. The raw data are presented in Fig. 3. The laser focus was placed inside a bead [position I in Fig. 3(a)] or in the oil [position II in Fig. 3(a)] and a CARS and an SRG spectrum were acquired in each place by recording the signal as function of $\Delta t$. Figures 3(c) and 3(e) show the resulting CARS spectra, from which it is apparent that the CARS signal has a large nonresonant contribution, in a bead as well as in the oil, but the two polystyrene peaks at 1003 and 1034 cm$^{-1}$ can be clearly seen. On the other hand, the thus obtained SRG spectrum in Fig. 3(d) in a bead displays the two Raman bands as distinct peaks with zero background. When compared with the Raman spectrum, Fig. 3(b), one finds

![Fig. 1. (Color online) Sketch of the experimental setup: dashed lines signify beams propagating in a lower plane and dotted lines are beams propagating in a higher plane. BS, beam splitter; PCF, photonic crystal fiber; AOM, acousto-optical modulator; LP, long-pass filter; SP, short-pass filter; PD, photodiode; APD, avalanche photodiode. Inset, pulse sequence and energy-level diagram for the SRS.](image)

![Fig. 2. Images with the chirp optimized for spectral resolution. (a)–(c) SRG and (d)–(f) CARS images of polystyrene beads in index-matching oil for different $\Delta t$. Pixel dwell time 20 ms, $\lambda_S = 1156$ nm, $P_p = 14 \text{ mW}$, $P_S = 1 \text{ mW}$. The scale bar is 10 $\mu$m.](image)
The scheme is immediately compatible with any femtosecond laser with wavelength to the red of the zero-dispersion wavelength of the PCF, including femtosecond lasers operating at higher repetition rates, which could increase the available average Stokes power to compensate for the intrinsically limited soliton pulse energy [14]. Because the soliton is transform limited when it leaves the PCF, there are no adverse effects from higher order chirp, which can potentially arise if using a PCF-generated dispersive wave as the Stokes pulse. The presented light source can thus be made to fulfill all requirements for high-speed SRS imaging and microspectroscopy, if all the most recent developments of SRS imaging [1–4] as well as the optimizations discussed above are incorporated. In addition, the presented scheme allows for rapid acquisition of spectra over a wide spectral range.

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References