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Spectral red-shifting of multi-frequency Raman orders

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ABSTRACT

In this paper, the spectral characteristics of the anti-Stokes orders generated by multi-frequency Raman generation are investigated. In particular, we observe that the orders can be red-shifted from the expected Raman transition frequency. The experimental observations indicate that Stark shifting rather than four-wave mixing is responsible for the red-shift. Current numerical models are insufficient to reproduce the experimental observations of red-shifting of the anti-Stokes orders.

1. Introduction

Multi-frequency Raman generation (MRG) efficiently generates a large number of Raman orders spanning the spectral region from the infrared to the ultraviolet. MRG is essentially cascaded stimulated Raman scattering that has been greatly enhanced by using two pump beams that have a frequency separation given by the Raman transition [1]. Most MRG research is concerned with making as broad a spectrum as possible to then make as short a pulse as possible by coherently adding all the orders [2]. It has been shown in the impulsive regime, where the bandwidth of the pump spans the Raman transition, that the number of Raman orders generated by a subsequent probe pulse is maximized by minimizing the Group Velocity Dispersion (GVD) [3]. In the other extreme, where the Raman coherence is driven adiabatically by two single-mode fields, the process is again optimized when the characteristic length for stimulated Raman generation and the phase slip length match [4]. In earlier work, we minimized the GVD to optimize MRG in the transient regime, where the pump duration is on the order of the coherence time of the Raman transition [5]. However, when the MRG was optimized, we observed that the individual spectra of the anti-Stokes orders would often be double peaked, with a narrow peak at the expected nth order Raman frequency and a broader, less intense peak to the red or lower frequency side of the Raman peak [5]. These red-shifted shoulders also appear in the transient pumping data shown by Sali et al. [6] but they were not discussed in the paper. These shoulders have only appeared in the MRG spectrum produced in the transient regime where the pump bandwidths match the Raman transition bandwidth. In the impulsive regime, the MRG is not measured directly with the pump beams because the ultra short pump pulses also generate a broad continuum due to self-phase modulation (SPM). With adiabatic pumping, the narrow spectral bandwidth of the orders has not been measured. However, it was shown in the adiabatic case that MRG is optimized when the pumping is detuned from the exact Raman resonance. It was also noted that there was an asymmetry between positive and negative detuning, in that many more anti-Stokes orders were generated for positive detuning, that is, when the two pump wavelengths were closer together than the Raman transition. The authors noted that for positive $\Delta\omega$, the transition is Stark shifted into resonance [4].

In this paper, we explore two possible causes for the red-shifted shoulders. One possible explanation is competition with non-resonant four-wave mixing. If our pump beams were slightly detuned, the anti-Stokes orders could be separated by the difference of the pump frequencies as well as by the natural Raman separation. If this were the case, we would expect four-wave mixing to be equally strong on both the blue and red side of the transition, depending on the detuning of the pump pulses. Also, we would expect that four-wave mixing should be much less efficient than the resonant stimulated Raman process. Another explanation could be that the Raman levels are Stark shifted to the red. In this case, the frequency separation of the Raman transition would be

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red-shifted at the peak intensity of the pulses due to the Stark-shift. Thus the MRG process can only be resonant when the pump frequency separation is reduced to match the shifted Raman transition. As a result, the MRG process would be more efficient with a red-detuned pump frequency separation.

2. Experimental method

Because four-wave mixing is not resonant with the transition, the coherence time scale should be much shorter than the Raman process. In order to determine the effect of four-wave mixing, we varied the instantaneous frequency separation of the two pumps, while keeping the average frequency separation set to the Raman transition and recorded the resulting MRG spectra. Our experimental system allowed this to be easily done. Even in the transient regime, the pulse durations are short enough to cause SPM and so the pump pulses must be stretched in time [6]. Because the intense pump pulses are generated with chirped pulse amplification (CPA), the pulses can be easily stretched by decreasing or increasing the separation of the gratings in the pulse compressor away from the optimum compression distance. The pumps are then predominantly linearly chirped, either positively or negatively. If both pump pulses have the same linear chirp, then the instantaneous frequency separation of the two pulses is constant throughout the stretched pulse duration. This instantaneous frequency can easily be tuned by varying the time between the pulses while the average frequency separation remains the difference of the two pulse spectra. This idea is shown in Fig. 1, where we depict both pulses having the same positive linear frequency chirp and the higher frequency pulse is delayed with respect to the lower frequency pulse. The center frequencies of the two pulses are separated by the Raman transition frequency, ν_{Raman}. The separation of the instantaneous frequencies, $\Delta\nu_{\mathrm{instant}}$, is then less than ν_{Raman}. We measured the cross-correlation of the two pulses using sum frequency in a Barium Borate (BBO) crystal to determine the zero time, where the instantaneous and average frequency separation match.

The experimental MRG system is the same as previously described except we now employ a hollow fiber with a diameter of 150 μm and length of 0.5 m [7]. The fiber is filled with sulfur hexafluoride, SF_6. A dual-wavelength CPA delivers the two pulses, the pump and first Stokes, that have peak wavelengths of 780 and 830 nm, respectively and bandwidths of 5 nm each [8]. This peak wavelength separation corresponds to the 23.25 THz A_{1g} vibrational Raman transition of the SF_6. The two colors are compressed with a three-grating compressor. The back mirror in the dispersion line of the pump pulse is on a translation stage to vary the time

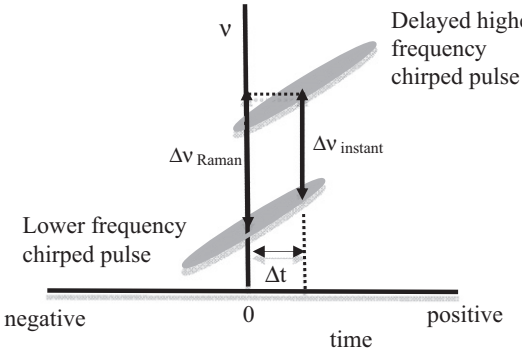

Fig. 1. Depiction of linear chirped pump pulses showing that the instantaneous frequency difference $\Delta\nu_{\mathrm{instant}}$ between the two pulses is dependent on time delay, Δt, between the pulses. The center frequencies of the two pulses are separated by the Raman transition frequency, $\Delta\nu_{\mathrm{Raman}}$.

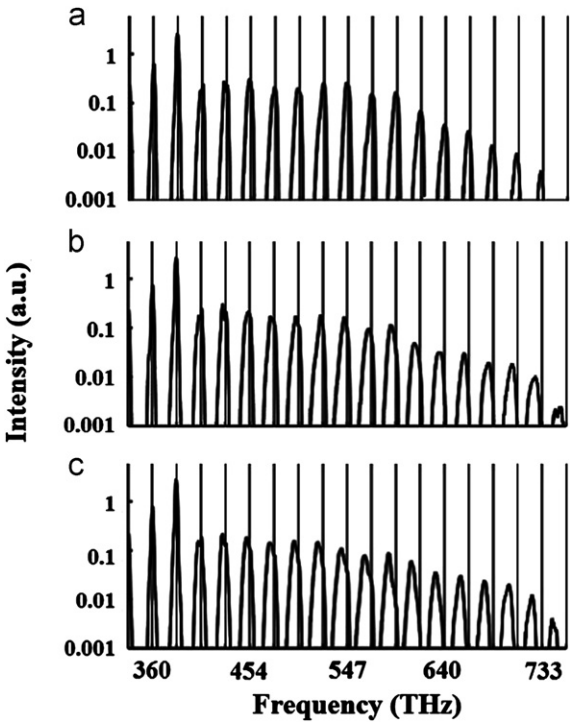

Fig. 2. Results of varying the time delay for positively chirped pulses, where the pump pulse leads the Stokes pulse by (a) 0.33, (b) 0, (c) −0.33 ps, respectively. The vertical lines are separated by the 23.25 THz Raman separation.

delay between the two pulses. We measured the MRG spectra as a function of time delay between the two chirped pump pulses.

3. Results

In the first set of experiments, we positively chirped the pulses. The SF_6 pressure was 170 KPa above atmosphere. The total energy in the two pulses before the fiber was 2.4 mJ. Fig. 2 shows the results of varying the time delay between the pump and Stokes pulses, which had pulse durations of 1.5 and 1.3 ps, respectively, giving a chirp rate of 1.7 THz/ps. The pump pulse leads the Stokes pulse by (a) 0.33, (b) 0 and (c) −0.33 ps, which correspond to an instantaneous frequency separation of 23.7, 23.3, 22.8 THz, respectively. The first few anti-Stokes orders show spectral content at the Raman shifted frequencies as well as the red-shifted shoulders. The higher orders only show the shifted spectra. When the pump frequencies are separated by more than the Raman transition, where we expect four-wave mixing to cause a blue shifted shoulder, the MRG orders appear peaked at the Raman transition with no shoulders. When the pulses are timed together and so the frequency separation is resonant with the Raman transition, the higher orders are red-shifted so that they are separated by a frequency of 22.9 THz. When the pulses are timed shifted to have an instantaneous separation of 22.8 THz, the MRG orders are separated by just 22.6 THz. Also very surprising is that the intensity of the highest anti-Stokes order actually increases with a negative time delay between the pulses even though the combined pump intensity would be less.

A second set of experiments was done with the same wavelengths and bandwidths, but with negatively chirped pulses. The total energy of the pulses was 1.8 mJ before the fiber. The results are shown in Fig. 3, for stretched pulse durations of 0.9 ps. The SF_6 pressure was 100 KPa above atmosphere. The pump pulse leads the Stokes pulse by (a) −0.33, (b) 0 and (c) +0.33 ps, corresponding to an instantaneous frequency separation of 24, 23.3, 22.5 THz, respectively. As can be seen in Fig. 3, for negatively chirped pulses, the anti-Stokes orders remain peaked at the