Temporal characteristics of single order from Multi-frequency Raman Generation

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1. Introduction

Multi-frequency Raman generation (MRG) is a method to generate a broad spectrum. Long trains of single femtosecond pulses have been generated with this nonlinear technique, in the adiabatic regime[1]. When pumped in the transient regime, the pump pulses are frequency chirped to avoid continuum generation from self-phase modulation so that each individual order will have to be separately compressed and then each order phased together to generate ultrashort pulses in a short pulse train. Towards this goal we are measuring the pulse profile of the first anti-Stokes order with Frequency Resolved Optical gating (FROG) and compressing this order using a simple prism compressor. We have previously observed that peaks can appear in the MRG spectra to the red side of the Raman orders [2,3]. These extra peaks correspond to a two-photon Rabi frequency shift. Our current investigation of the temporal profile of the single order will help determine the two-photon Rabi shift in the transient regime. We want to also determine if the extra frequency bandwidth can lead to better pulse compression of a single Raman order, which would then yield a shorter pulse train when all compressed Raman orders are phased together.

2. Experiment

We pump the vibrational Raman transition of sulphur hexafluouride with two pulses from a two-colour, Ti:sapphire amplified laser system, each having ~3 nm bandwidth. The peak frequency separation equals the 23.25THz Raman frequency. We lengthen the pulses to ~1 ps by not completely recompressing the amplified chirped pulses. We can tune the instantaneous frequency separation by varying the pulse delay between the two, chirped pulses. When the frequency separation is greater than the Raman transition we don't observe the red shoulder. As we go through resonance and further decrease the frequency separation, the shoulder increases in amplitude. We measure the amplitude and phase of the first anti-Stokes order with cross-FROG, using the pump pulse as the reference pulse. We then put the Raman order through a prism pair compressor and measure the autocorrelation of the compressed pulse.

2. Discussion of Results

The FROG reconstruction shows that the red shoulder is a pulse delayed from the Raman peak. The Raman peak has the same linear chirp as the pump pulse but the red-shifted shoulder is an unchirped pulse. The delay between the pulses increases from 200 fs to 500 fs when the frequency separation from the pump increases from resonance at 23.25 THz down to 22.75 THz, but then the delay decreases back to 200 fs at a pump separation of 22.25 THz.

The autocorrelation of the compressed order confirms the FROG results. When the compressor is set to remove the linear chirp equal to the pump chirp a single, near transform limited 400fs pulse is measured when the red shoulder peak is absent. As the delay between the pump pulses is reduced, the autocorrelation becomes three peaked indicating a double pulse. In this case, with the added dispersion the pulses are now ~1ps apart.

We have shown that for shorter pump pulses, the red shifted shoulder becomes broad and the higher orders no longer have the Raman order[3]. In the future we want to measure the temporal characteristics of higher orders where the Raman peak is eliminated and determine whether the red shoulders remain unchirped. If so, this would eliminate the need to first compress the orders before simply timing the individual order to achieve short pulse trains of single femtosecond pulses. Further work is needed to understand why the shoulders are unchirped.

3. Conclusions

Our work shows that multi-frequency Raman generation in the transient regime is complicated by two-photon Rabi frequency shifting. To date Rabi frequency shifting has only been investigated in the adiabatic regime. This work starts the investigation of the frequency shift in the transient regime.

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References

[1] A.V. Sokolov, et al. Phys. Rev. Lett. **87**, (2001) 033402
[2] Z. Cui, et. al., Opt. Commun. **288**, (2013) 118
[3] H. Yan, et al., Appl. Sci, **4**, (2014) 390