18p-D5-1

Multi-frequency Raman Generation with Chirped Pumping

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1. Introduction

Multi-frequency Raman Generation (MRG) is a promising technique to generate single femtosecond pulses with sufficient energy to carry out nonlinear optics experiments.[1] In this technique, two intense pulses having a frequency separation that corresponds to a Raman transition strongly drive the transition creating a number of Stokes and anti-Stokes orders through a cascaded stimulated Raman process. To date, using MRG pulse trains of 1.6 fs pulses have been achieved in the long pulse, adiabatic regime [2]. The adiabatic regime has the disadvantage of producing a very long train of pulses and so each individual pulse has a correspondingly low intensity. The temporal length of the pulse train corresponds to the inverse of the spectral width of the driving pulses and so the pulse intensity could be greatly enhanced by pumping with two short pulses. Short pulse pumping unfortunately leads to the competing nonlinear effect of self-phase modulation. To avoid self-phase modulation, transient MRG uses broadband pump pulses that are linearly chirped to longer pulse durations. [3,4] Pumping this way leads to each Raman order also being chirped. To achieve transform limited pulse duration in the transient regime, each order will then need to be optimally compressed before phasing all the orders together. The goal of our research is to achieve a large number of Raman orders, which are each spectrally broad with a well-determined spectral phase.

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 5 nm bandwidth. The peak frequency separation equals the 23.25THz Raman frequency. We lengthen the pulses to 1.5 ps by not completely recompressing the amplified chirped pulses. We have measured the Raman spectra for both positively and negatively chirped pulses for various time delays between the two pump pulses. By changing the time delay between the two, chirped pulses, the instantaneous frequency separation of the pulses is varied. When the pump pulses are positively chirped, red-shifted orders appear next to the anti-Stokes Raman orders and in some cases have more power than the Raman orders.[5]

3. Discussion

Red shifted shoulders also appear in reported MRG spectra for hydrogen and methane under similar conditions, but were not discussed in the paper.[3] Large spectral peaks also appear on the red sides of the anti-Stokes orders with a positively chirped impulsive pumping scheme in lead tungstate[6]. The authors of this work consider the extra peaks to come from four wave mixing. We too thought that the extra peaks were a result of four-wave mixing. In our transient experiments, we could arrange to have the two peak frequencies match the Raman transition, but have the instantaneous frequency separation tune through the transition. When the instantaneous frequency separation is greater than the Raman transition, the Raman orders still displayed a red-shifted shoulder rather than the expected blue shift. As the instantaneous frequency separation was reduced, the shoulder pushed to lower frequencies but then stopped shifting with further red shifting of the pump frequency. Even more surprising is that with negatively chirped pulses the orders remained peaked at the Raman transition frequency for all time delays. These observations of red shifted spectral peaks then do not match what is expected if the shoulders are coming from four-wave mixing.

4. Conclusions

The Raman orders can be significantly broadened by the red-shifted shoulders and if they can be properly phased they would lead to even shorter pulse trains and so higher intensity pulses. Currently there is no theory of multi-frequency Raman generation that can explain the observed red shifting of the orders with positively chirped pulses If the goal of MRG is to generate very intense, ultrashort pulses we need to understand these MRG spectra so we can properly make use of them.

Acknowledgements

The author is grateful for funding from the Natural Sciences and Engineering Research Council.

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