By pumping a Raman transition with two strong pumps a broad spectrum of many Raman orders can be generated. When the pump duration is in the transient regime, extra peaks can appear in the multi-frequency Raman generated (MRG) spectra to the red side of the Raman orders. In earlier work, we showed that these extra peaks correspond to a two-photon Rabi frequency shift. Currently we are investigating the temporal structure of the first anti-Stokes Raman order with and without the red shifted peak.

The Raman medium is sulphur hexafluoride, with a Raman transition at 23.25 THz. We measured the phase and amplitude of the electric field of the first anti-Stokes order using Frequency Resolved Optical Gating (FROG) as a function of time delay between the two chirped pump pulses. By varying the delay between the chirped pulses, the instantaneous frequency separation of the pulses is tuned through the Raman transition. The FROG traces and corresponding amplitudes and phases of the electric fields are shown in Figure 1 for different frequency separations (THz): (a) 23.75, (b) 23.25, (c) 22.75, (d) 22.25.

As the pump frequency separation is red shifted, the red peak grows. The peak occurring at the Raman frequency has the same frequency chirp as the pump pulses, but the red-shifted shoulder appears to be unchirped and delayed from the Raman peak, with the delay varying from 200 fs in Figure 1b and 500 fs (Figure 1c). As the two pulses overlap in time and frequency, we have confirmed these predictions by simulating the FROG traces using one linearly chirped anti-Stokes pulse with a delayed unchirped pulse of the same duration with a 1.52 THz red-shift. We have also used a prism compressor to compress the first anti-Stokes order when there is no red-shifted peak and note that when we change the timing of the pumps, a three peak autocorrelation is measured and the time delay between the pulses grows to 1 ps.