Coherent Raman generation with wavefront shaping

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ABSTRACT

Molecular modulation in Raman-crystals provides a pathway towards generation of subfemtosecond broadband pulses of light in the UV-VIS-NIR range. By adding a transverse wavefront shaping of the input beams we expand the capabilities of molecular modulation. We obtain very different effects depending on how each beam is shaped.

For example, we can add topology to the input beams (pump, Stokes or both), by introducing orbital angular momentum (OAM) with a phase-only wavefront modulation. This topology will then transfer to the Raman sidebands, generated through molecular modulation, according to phase-matching conditions and a certain mathematical law (see Fig. 1 A). This property of Raman sidebands helps to study the pure quantum nature of OAM in nonlinear interactions.

Moreover, we can extend the total spectral bandwidth of generated Raman sidebands with an adaptive wavefront correction of pump or Stokes beams (see Fig. 1 B). This may help to generate a shorter and more energetic ultrashort pulse. Such pulses are essential for the end goal of studying high-power, nonlinear interactions and helping us to obtain a deeper understanding of femtosecond chemistry.

Fig. 1 A: beam profiles and obtained Raman sidebands with (bottom) and without (top) OAM. Sidebands generated using OAM beams have clear “vortex” structure and carry OAM. B: beam profiles and obtained Raman sidebands with (bottom) and without (top) adaptive wavefront correction. Spectra generated with the corrected beams is extended to the blue side of the visible range. Arrows AS1-AS8 show anti-Stokes sidebands of the order of 1-8.

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