Molecular modulation with temporal and spatial shaping of laser fields
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Abstract
We develop a coherent Raman technique, termed molecular modulation, which allows ultrafast laser pulse shaping and non-sinusoidal field synthesis. The central feature of this technique is the preparation of an ensemble of molecules in a coherent superposition state. Experimentally, the molecular-modulation light source is characterized by a bandwidth spanning infrared, visible, and ultraviolet spectral regions, generating bursts of light synchronized with respect to molecular oscillations (Fig. 1). Controlled spectral and temporal shaping of the resultant waveform will allow arbitrary ultrafast, potentially non-sinusoidal, field synthesis. Additionally, we explore the possibility of optimized broadband generation via pump beam shaping. We add another dimension to the laser field engineering by using spatial light modulators to shape the transverse beam profiles, and move toward production of space- and time-tailored sub-cycle optical fields.

Figure 1: (a) Molecular modulation / broadband Raman generation in PbWO₄ driven by ~50 fs near-IR pump and Stokes pulses. The input beams are crossed at an angle, such that at the output the Stokes and anti-Stokes components naturally spread into a fan of multi-color beams [1]. (b) The pulse synthesized from pump, Stokes and anti-Stokes 1 through 4 sidebands (obtained in a different experiment, using a Raman transition in single-crystal diamond), as deduced from interferometric cross-correlation frequency resolved optical gating (ix-FROG) envelopes [2]. (c) Experimental setup for optimized Raman generation with a feedback-based wavefront shaping algorithm; detailed description can be found in Ref. [3]. Here yet another experimental configuration is used, with the Raman transition driven by chirped pulse replicas. (d) Average power of sidebands with (blue columns) and without (red columns) wavefront shaping [3].

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References: