Pulse-shaper-assisted phase optimization of an ultrabroadband spectral comb

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Synthesis of ultrashort single-cycle optical pulses requires an over-an-octave-wide coherent spectrum. In the past, broadband collinear Raman generation in molecular gases has been used to produce mutually coherent equidistant frequency sidebands spanning several octaves of optical bandwidths [1]. It has been argued that these sidebands can be used to synthesize optical pulses as short as a fraction of a femtosecond (fs) [2]. The Raman technique relies on adiabatic preparation of near-maximal molecular coherence by driving the molecular transition slightly off resonance so that a single molecular superposition state is excited. Molecular motion, in turn, modulates the driving laser frequencies and a very broad spectrum is generated, hence the term for this process “molecular modulation”. By phase locking, a pulse train with a time interval of the inverse of the Raman shift frequency is generated.

We apply the “molecular modulation” method to Raman active crystals and generate broadband spectral sidebands by focusing two 50-fs laser beams non-collinearly into a synthetic single-crystal diamond. The center wavelengths of the two pulses are 1200 nm and 1030 nm. The spectral sidebands come out at different angles and cover infrared, visible and ultraviolet spectral regions. We combine the sidebands into a collinear beam by using a spherical mirror and a prism [3].

Pulse shaper (Phazzler, FastLite) serves several purposes in our experiment. First it is used to measure and then compensate the phase of each sideband to be close to Fourier Transform Limited. Secondly, we use the pulse shaper for compensating the phase difference between the sidebands. At last, the pulse shaper can be used for measurement of the synthesized pulse.

The frequency combs that are generated in the crystal have some special properties—the sum frequency generation signal (SFG) between the AS 1 and AS 3 overlaps both in time and spectrum with the second harmonic generation (SHG) of AS 2 (left figure), which leads to beating when we vary the phase of AS 2 by the pulse shaper (left figure). This demonstrates the mutual coherence of the spectral sidebands, and shows the pulse shaper’s capability to control their phases in a precise and stable manner. When the phases across the 3 sidebands’ spectra are adjusted to all be equal, the resultant pulse duration is expected to be 11 fs (FWHM). Our limitation so far is set by the pulse shaper which operates at wavelengths range from 530 nm to 930 nm, so at most 6 sidebands can be used for pulse shaping; utilization of this full bandwidth will enable us to produce pulses as short as 4.8 fs. Significant advances beyond that point can be obtained when two (or more) pulse shapers covering adjacent spectral ranges are used.