Post-Compression of High-Energy Pulses at 5 µm Wavelength

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Due to the lack of suitable gain media for direct amplification, high-energy few-cycle pulses at longer wavelengths, *i.e.* larger than 4 μm, are typically generated in optically parametric chirped pulse amplifiers (OPCPA) [1]. Pertinent applications in strong-field and attosecond physics like hard X-ray generation [2] or high harmonic generation (HHG) [3] heavily rely on shortest possible pulse durations for best performance. As processes in strong-field physics can be driven much more efficiently with pulses in the two-cycle regime or below, efforts have been spent on generating even shorter optical pulses by nonlinear post-compression in bulk materials [4]. To this end, nonlinear compression at 5 μm was already demonstrated with 53 fs pulse duration using zinc selenide for SPM broadening and CaF_2 for subsequent post-compression. As ZnSe exhibits nearly vanishing GVD at 5 μm, no self-compression appears possible and a spatio-temporal analysis of the post-compressed pulses was not even attempted [5].

Here we present soliton self-compression of high-energy pulses at 4.9 μm in zinc sulfide which has the zero dispersion wavelength at 3.6 μm. Apart from a 4 mm thick polycrystalline ZnS plate, the self-compression setup only contains a two-lens telescope. The input pulses with 2.5 mJ energy are provided by the idler of a midwave-IR OPCPA [1]. The broadening of the 80 fs input pulses leads to a spectral extension from 3.5 μm to 6.0 μm ($1/e^2$ level), which is sufficient to support the generation of 28 fs pulses. The self-compression scheme is demonstrated with a reduction of pulse durations to 37 fs which corresponds to only two optical cycles at 5 μm wavelength. The loss in the nonlinear compression process is only 0.2 yielding 2.0 mJ energy pulses in the 1 kHz pulse train.

The input intensity at the ZnS is tailored to avoid the occurrence of small-scale filamentation, resulting in an almost undisturbed beam profile. Furthermore, the self-compressed two-cycle pulses exhibit an outstanding spatio-temporal homogeneity over the entire beam profile, which distinguishes them from other bulk compression schemes. The characteristic V-parameter remains larger than 0.85 within the $1/e^2$ intensity range of the beam profile and its weighting with intensity gives a remarkable averaged overlap of 0.972 and 0.975 in x- and y-direction, respectively.

This high spatio-temporal homogeneity stems from the transformation of a Gaussian input profile into a spatial Townes soliton, resulting in a peak power of 45 GW, which constitutes a record value for kHz pulse trains beyond 4 μm wavelength. The combined action of self-focusing and subsequent diffraction takes part behind the ZnS sample and leads to homogenization of spectral content across the beam during formation of the Townes profile. While these Townes profiles are difficult to observe in the visible or near-infrared spectral range, we would like to point out that the diffraction operator in the wave equation scales with wavelength. Given the additional slight reduction of the nonlinear refractive index n_2 at 5 μm wavelength, we estimate that the Townes soliton can host 10 times higher pulse energies than at 0.8 µm.

We additionally explore the limiting effect of multiphoton absorption in ZnS and record generated high harmonics up to 8^{th} order. These harmonics support a >2-octave supercontinuum spectrum in the visible and short-wave IR spectral region with sizable 0.1 mJ pulse energy. Furthermore, the nonlinear refractive index of polycrystalline ZnS at 5 μm has been derived from our measurements to $n_2 = 3.6\times10^{-19}$ m^2/W which appears in reasonable agreement with values in the literature.

Using the demonstrated 37 fs pulses as driver for hard X-ray generation [2], substantially shorter X-ray pulses can be expected than obtainable with the midwave-IR OPCPA driver output pulses [1], promising substantially improved temporal resolution of mid-IR-pump X-ray-probe studies.

References

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