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Multiple-μJ mid-IR supercontinuum generation in quadratic nonlinear crystals

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***Abstract*—Pumping a quadratic nonlinear crystal in the mid-IR we observe octave-spanning mid-IR supercontinua. A self-acting cascaded process leads to the formation of a self-defocusing nonlinearity, allowing formation of filament-free octave-spanning supercontinua in the 2.0-7.0 μm range with 10s of μJ pulse energies, much higher than filament-based techniques. This allows to use the supercontinuum as ultra-broadband excitation pulses in nonlinear optical applications.**

HIGH-POWER mid-IR supercontinuum (SC) sources are needed to overcome the limited bandwidth from OPA-type schemes, for, e.g., femtosecond pump-probe spectroscopy of the fundamental frequencies of vibrational modes. They rely mainly on self-phase modulation (SPM) spectral broadening in bulk glasses or crystals [1]–[3], eventually limited by the onset of multiple filamentation due to the self-focusing nature of the SPM effect, and thus to few-μJ energies [**?**], [4]–[6]. Multi-color mixing in gases gives even more extreme bandwidths, but also much less mid-IR energy [7]–[10]. The challenge is to keep a high bandwidth and increase the power spectral density (PSD) so the SC source can be used as pump instead of probe.

Here we use a quadratic nonlinear crystal cut for strongly phase-mismatched second-harmonic generation (SHG), which allows us to flip the sign of the SPM effect so it becomes self-defocusing. This gives filamentation-free octave-spanning SC generation with much higher energies, as recently demonstrated in the near-IR [**?**], [11], [12]. Here we show that by pumping a mid-IR crystal, lithium thioindate ($LiInS_2$, LIS), in the mid-IR close to its zero dispersion wavelength (ZDW), we get octave spanning SC generation with 10s of μJ pulse energies [13].

The LIS crystal was cut with $\theta = 90°$ and $\phi = 0$. This gives a maximum quadratic nonlinearity $d_{\rm eff} = d_{33} = 16$ pm/V and makes the SHG strongly phase-mismatched ($\Delta kL \gg 2\pi$) leading to a self-defocusing cascading SPM-like nonlinearity $n_{2,\rm casc} \propto -d_{\rm eff}^2/\Delta k$ [13]. We pumped with 85 fs 50 μJ pulses in the 3.0-3.9 μm range from a commercial OPA and DFG system. The nearly transform-limited pulses were loosely focused (0.27 mm FWHM) in the crystal. The output was measured with a spectrometer that used long-pass filters to selectively cover the 1-7 μm range. A typical power sweep is shown in Fig. 1 for $\lambda_0 = 3.60$ μm, and the onset of an octave-spanning SC was found at 150 GW/cm^2. For the maximum intensity (using all the available pump energy) a flat SC is seen, extending beyond 6 μm. The SC process is fuelled by the cascading-induced self-defocusing SPM nonlinearity, self-

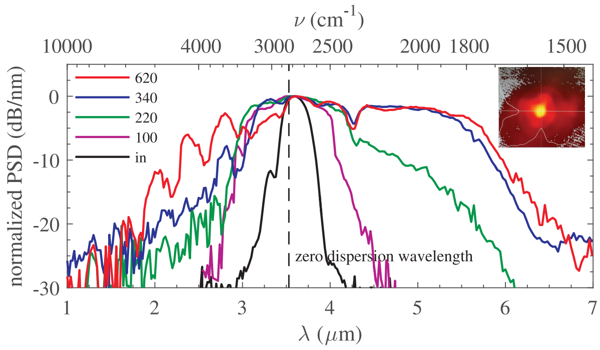

Fig. 1. SC generation in 15 mm LIS for $\lambda_0 = 3.60$ μm and sweeping the pump intensity (indicated in GW/cm^2). Inset: SC transverse mode profile.

steepening, soliton and dispersive wave formation below and above the ZDW, respectively. Importantly, most of the pump energy is retained in the SC. The inset shows the Gaussian-like transverse profile of the SC as measured with a mid-IR sensitive camera, indicating that the technique is filament free. Thus, expanding the pump spot size allows for using much higher pulse energies. We used the SC source as an excitation pulse in 2D mid-IR spectroscopy extended from the setup in [10] and in sum-frequency generation spectroscopy [14], where we were able to record spectra over a significantly broader bandwidth than with an OPA-based pump. This shows that the SC source has a high enough coherence, PSD and bandwidth to be used to pump a wide range of vibrational modes, and is therefore a vital next step in bright mid-IR SC sources. The technique can also be used in other mid-IR crystals as long as a defocusing nonlinearity can be realized [15], allowing various possibilities for pumping close to the ZDW in the short-wavelength mid-IR range, with the generated spectra extending potentially beyond 10 μm with multiple-μJ energies.

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