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Multi-octave supercontinuum generation from mid-infrared filamentation in a bulk crystal

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Abstract. We present supercontinuum generation pumped by femtosecond mid-infrared pulses in a bulk homogeneous material. The spectrum extends from 450 nm into the mid-infrared, and carries high spectral energy density (3 pJ/nm–10 nJ/nm). The supercontinuum has high shot-to-shot reproducibility and preserves the carrier-to-envelope phase. Our result paves the way for compact supercontinuum sources with unprecedented bandwidth.

Bright, broadband and coherent light sources are required, for example, in optical coherence tomography [1], time-resolved spectroscopy [2] and microscopy [3]. Such supercontinua are also a step towards arbitrary multi-color waveform synthesis, which offers simultaneous probing and control [4] of electronic, vibrational and rotational motion, or synthesizing single cycle waveforms at arbitrary wavelengths [5]. These applications place widely varying demands on the characteristics of a supercontinuum source. Novel optical fibers can yield supercontinua spanning the ultraviolet into the mid-infrared [6–8]. However, a requirement for shot-to-shot repeatability and a smooth spectrum limits the allowable propagation length, restricting the bandwidth [7]. Spectral broadening in bulk materials, in contrast, has thus far not achieved bandwidths comparable to those from fibers. In particular, long wavelength extensions of the spectrum are inefficient and require extreme conditions [9–11]. This suggests the necessity of increasing the pump wavelength to extend the long wavelength edge of the supercontinuum, but raises the question of whether a corresponding red shift of the short wavelength edge will occur.

Here, we present a supercontinuum produced by filamentation of femtosecond mid-infrared pulses in bulk yttrium aluminium garnet (YAG) crystal. The spectrum spans 450–4500 nm, the broadest ever produced by filamentation in bulk. We verify carrier-envelope phase stability and shot-to-shot repeatability. Simulations indicate an absence of complex pulse splitting and self-compression of the pump pulse by a factor of 10.

The pump pulses were produced with an optical parametric chirped pulse amplifier (OPCPA), which delivered carrier-envelope phase stable 85 fs pulses at 3100 nm center wavelength and with a repetition rate of 160 kHz [12]. The setup for generating and measuring the supercontinuum is shown in Fig. 1(a). Pulses of 6.9 μJ energy were collimated to 4 mm diameter and were focused by a 75 mm focal length CaF_2 lens to a $1/e^2$ diameter of 50 μm. The peak power was 76 MW which is approximately three times the critical power for YAG. The 2 mm-thick YAG plate was placed in the focal region, producing a clearly visible filament. We observed no irregular or interference patterns typically indicative of multifilamentation. At a distance of 54 mm after the exit plane of the YAG plate, we acquired an angularly-resolved $\{\theta, \lambda\}$ spectrum which provided our connection to the theory and

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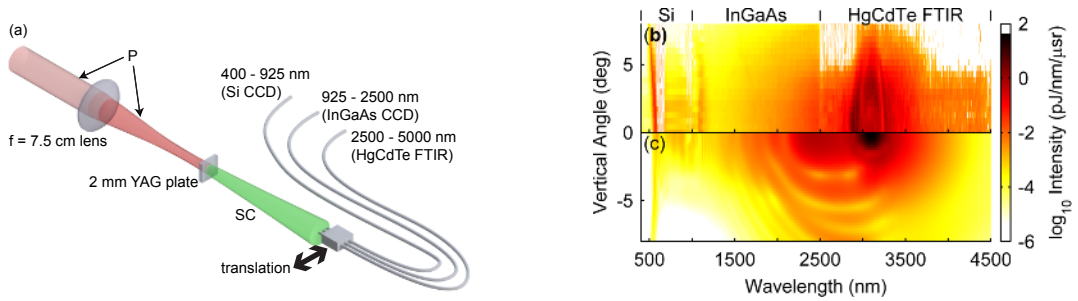

Fig. 1. (a) Schematic of the generation and measurement setup; the mid-infrared pump beam (P, red) is focused by a 75 mm focal length CaF_2 lens. The supercontinuum (SC, green) is generated in a 2 mm thick YAG plate and the $\{\theta, \lambda\}$ distribution is recorded by scanning three closely spaced fibres across the emerging beam. (b) Measurement and (c) simulation of the far field $\{\theta, \lambda\}$ representation in absolute units; the detector ranges are shown above the figure.

numerical simulations. Three adjacent optical fibres, each aligned parallel to the optical axis, delivered light to three spectrometers: a silicon charged-coupled device (CCD) spectrometer (Ocean Optics HR4000), an InGaAs CCD spectrometer (Ocean Optics NIR256), and a Fourier transform infrared (FTIR) spectrometer (Oriel MIR8025) with HgCdTe detector. The three fibres were scanned across the beam as a single unit, acquiring spectra at each position. We applied spectral response calibrations, and the data from each spectrometer were scaled to achieve consistency in their regions of overlap (925 nm and 2500 nm) and with the measured pulse energy.

The result of this measurement, presented in Figure 1(b), shows that the long wavelength edge decays smoothly, reaching the noise level at 4500 nm, whilst the short wavelengths extend to 450 nm. The highest spectral energy density is found around the mid-infrared pump wavelength with a maximum angularly-integrated spectral density of 10 nJ/nm. The smallest energy content is observed in the 750–1000 nm range at a few pJ/nm; note that these energies are still sufficient for most absorption spectroscopy experiments.

We tested the shot-to-shot reproducibility of the waveform using an f–$2f$ interferometer, with which we acquired the spectral interference pattern formed by the supercontinuum and its second harmonic. The phase of the fringes is sensitive to the carrier-envelope phase of the supercontinuum pulses, as well as their intensity fluctuations [13]. Figure 2(a) shows a 15 s subset of a dataset recorded over a 10 minute interval. Over the full 10 minutes, the fringe phase has a standard deviation of 259 mrad [12].

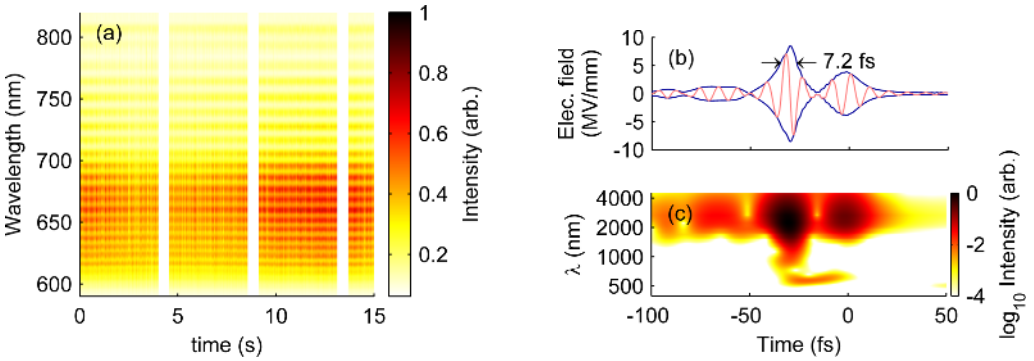

Fig. 2. (a) Spectral interference fringes produced by f–$2f$ interference between the long and short wavelength edges of the spectrometer over time. (b) Temporal envelope (dark blue) and field oscillations (light red) and (b) spectrogram of the on-axis simulated pulse emerging from the YAG plate; note the logarithmic intensity colour and wavelength scale in the spectrogram.

We performed a full 3D simulation, with the result shown in Fig. 1(c). The uncertainty in the medium parameters — specifically the material dispersion, Kerr index and ionization rates — over such a large spectrum render quantitative comparison challenging. Nonetheless we find that the main spectral features are qualitatively reproduced. The simulation confirmed an extended self-channeling behavior characteristic of filamentation. The beam undergoes several self-focusing events and does not diffract over a distance greater than the Rayleigh range of 1.17 mm.

We used the simulation to examine the output temporal on-axis profile, shown in Fig. 2(b). The pulse duration is reduced by more than a factor of 10 (the satellite peaks are 50% lower in peak intensity) and the main peak has near-single-cycle duration. Pulse compression is known to occur in filamentation [14] but this is the first time that numerics exhibit such a marked compression in a condensed medium. We attribute this mostly to the weak anomalous dispersion at mid-infrared wavelengths, combined with the higher order of the multiphoton ionisation processes (with respect e.g. to 800 nm pump pulses) that limit the maximum intensity and enable stronger nonlinear reshaping dynamics. A spectrogram of the output pulse, shown in Fig. 2(b), shows that the short wavelength components, below 1500 nm, emerge nearly simultaneously with the main peak. Furthermore the overall time-frequency distribution is simple, containing only a few distinct peaks. Therefore, the numerics suggest bulk supercontinuum as a simple means of producing near-single-cycle and carrier-envelope-phase stable pulses using mid-infrared pump lasers.

In summary, we demonstrated the first stable multi-octave supercontinuum from filamentation of mid-infrared femtosecond pulses in bulk material. The measured spectrum spans 450 nm to 4500 nm, with a spectral energy density of 2 pJ/nm to 10 nJ/nm. The smoothness of the spectrum indicates absence of complex pulse splitting and a carrier-envelope phase measurement indicates shot-to-shot reproducibility over a significant spectral range. We obtained an angularly-resolved far-field spectrum with the main features captured by full 3D simulations. By demonstrating a simple and robust method for coherently extending the spectrum of an amplified femtosecond mid-infrared pulse down to the short wavelength edge of the visible, we provide a powerful platform for the next generation of ultrafast applications.

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