Coherent octave-spanning mid-infrared supercontinuum generated in As$_2$S$_3$-silica double-nanospike waveguide pumped by femtosecond Cr:ZnS laser

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Abstract: A more than 1.5 octave-spanning mid-infrared supercontinuum (1.2 to 3.6 μm) is generated by pumping a As$_2$S$_3$-silica "double-nanospike" waveguide via a femtosecond Cr:ZnS laser at 2.35 μm. The combination of the optimized group velocity dispersion and extremely high nonlinearity provided by the As$_2$S$_3$-silica hybrid waveguide enables a ~100 pJ level pump pulse energy threshold for octave-spanning spectral broadening at a repetition rate of 90 MHz. Numerical simulations show that the generated supercontinuum is highly coherent over the entire spanning wavelength range. The results are important for realization of a high repetition rate octave-spanning frequency comb in the mid-infrared spectral region.

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1. Introduction

Frequency combs in the mid-infrared (mid-IR) spectral region of 2-20 μm (500-5000 cm^{-1}) are highly desirable for high precision spectroscopy, because many molecules possess strong characteristic vibrational transitions in this domain ("molecular fingerprint region") [1]. A frequency comb with higher repetition rate is especially preferred for several applications (including direct frequency comb spectroscopy, optical waveform synthesis, calibration of astrophysical spectrometers, etc) since a larger spacing between comb components eases access to individual comb modes [2]. A self-stablized frequency comb requires the spectrum to span at least a full octave so that a self-beat note between the two comb modes – with frequencies *f* and 2*f* (doubled via second-harmonic generation) can be detected and locked. Efficient (i.e., low threshold energy) and coherent spectral broadening is therefore highly desirable, especially for the case of high repetition rate frequency comb generation, where the energy of a single pump pulse energy is limited by the average power available. This normally requires a stable mid-IR ultrafast laser as the pump source, combined with a highly nonlinear waveguide with suitable dispersion characteristics. Although several approaches to high repetition rate mid-IR frequency comb generation have previously been reported [3–7], octave-spanning spectral broadening has been rarely demonstrated [8–10].

With respect to the pump source, among all candidates, femtosecond solid state lasers are probably the most reliable, naturally low-noise and compact choices for frequency comb generation. Frequency combs based on Ti-sapphire and ytterbium femtosecond lasers in the near-IR wavelength range have demonstrated excellent stability and spectral power. Recently a 1 GHz coherent supercontinuum (SC) spanning from 600 to 1700 nm was demonstrated in a chip-based silicon nitride waveguide via pumping through an Yb:CALGO laser [11]. To extend the spectral broadening further into the mid-IR wavelength range, in the work reported here we have developed a laser based on Cr:ZnS. Indeed, chromium-doped chalcogenide (Cr:ZnS/Cr:ZnSe) based lasers, often called the "Ti-sapphires of the infrared" due to their very broad absorption and emission bands, have recently matured to provide femtosecond performance in all respects comparable with Ti:sapphire lasers [12–14]. As such, they represent ideal compact and stable sources for generation of high quality coherent octave-spanning supercontinua in the mid-IR.

Here we report generation of a more than one octave-spanning SC in the mid-IR spectral region by pumping a recently developed As_2S_3-silica double-nanospike waveguide with a femtosecond Cr:ZnS laser. The repetition rate of the SC is 90 MHz. The extremely high nonlinearity of the hybrid waveguide, together with the greatly enhanced coupling efficiency obtained through the double-nanospike structure and the high peak power and good pulse

quality of the Cr:ZnS laser, enables octave-spanning coherent SC generation with a threshold pulse energy of hundreds of pJ.
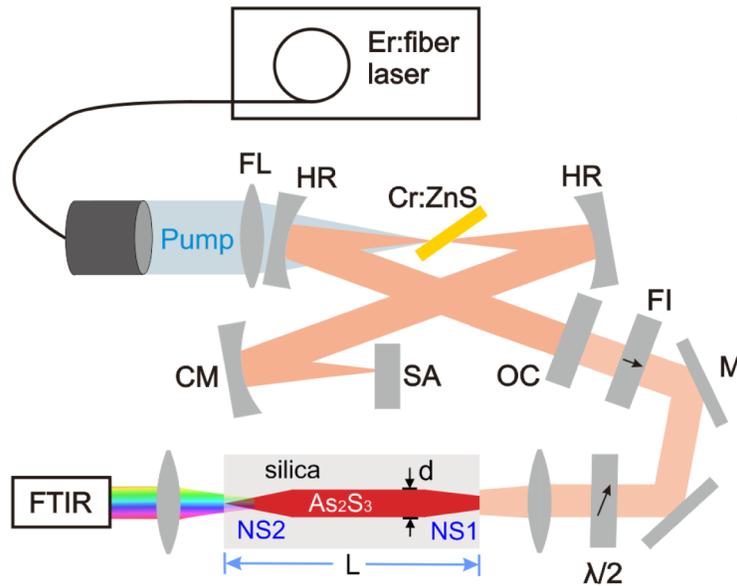

Fig. 1. Schematic view of the experimental setup. FL, focusing lens; HR, high reflective mirror; SA, saturable absorber (graphene-based saturable absorber mirror); CM, chirped mirror; OC, output coupler; FI, Faraday isolator; λ/2, half-lambda waveplate; NS, nanospike; FTIR, Fourier transform infrared spectrometer.

2. Experimental configuration

The experimental setup is shown in Fig. 1. The mode-locked Cr:ZnS laser was pumped by an Er-doped fiber laser emitting 5 W of output power at 1.61 μm. The pump radiation was focused by a 40-mm lens to a 30-μm radius waist inside the active crystal. The X-folded laser cavity with a round-trip length of about 3.3 m was astigmatically compensated by proper choice of the folding angles introduced by the highly reflective (HR) mirrors. Mode-locking was initiated and supported by a graphene-based saturable absorber (SA) mirror (multilayer graphene transferred to a surface of a dielectric high-reflector mirror, modulation depth of about 3%, details can be found in [13]). The cavity mode was additionally focused on to the graphene SR by ROC = 200 mm concave mirror to form a waist of about 80-μm radius. The normal dispersion of the 2.5 mm-thick passively-cooled Cr:ZnS active element was compensated by a single chirped mirror (CM), resulting in a slightly anomalous cavity round-trip net GDD (about –500 fs^2) and ensuring fundamental soliton mode-locking. In this regime of operation the pulse energy was limited to about 1.5 nJ due to the high third-order optical nonlinearity of the active medium [14]. In order to maximize the femtosecond pulse energy and avoid soliton breakup with subsequent pulse doubling, the active crystal was moved along the cavity axis 2-3 mm away from the point of the cavity mode waist. This resulted in a slightly larger mode size on the active crystal and thus decreased the energy fluence, allowing higher pulse energies to be achieved before soliton breakup occurred. Finally, we successfully extracted 360 mW of average output power from the laser oscillator at a pulse repetition frequency of 90 MHz, corresponding to a pulse energy of ~4 nJ. The laser output power was sufficiently stable, being characterized by about 0.7% power RMS over 1 hour. After several hours of operation at this power level a clearly visible CW component usually appeared in the pulse spectrum. Although less than 1% of the total output power was located in this CW component, the laser was subsequently routinely operated at ~2 nJ pulse energy so as to ensure a smooth supercontinuum spectrum. Figure 2 plots the optical spectrum and

interferometric autocorrelation of the mode-locked laser pulses, showing the measured pulse duration and FWHM spectral bandwidth at this power level to be respectively 100 fs and 59 nm.
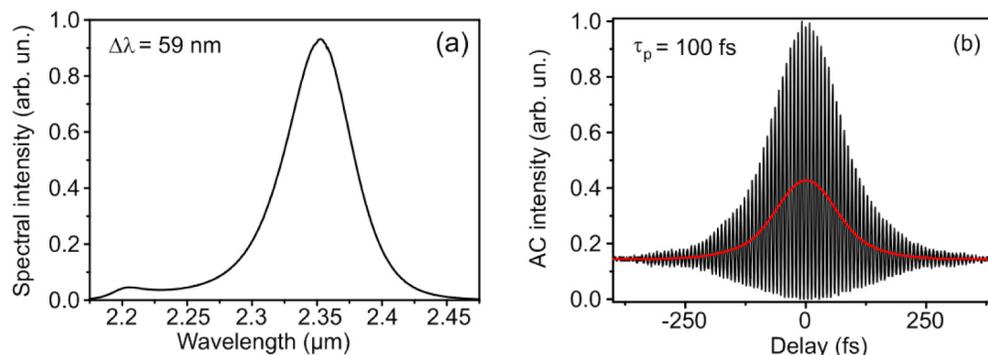

Fig. 2. (a) Typical optical spectrum and (b) interferometric autocorrelation of the mode-locked laser pulses from the femtosecond Cr:ZnS laser.

The highly nonlinear waveguides used had hybrid As_2S_3-silica double-nanospike structures. They were fabricated using the pressure-assisted melt-filling technique, which enables a μm-diameter chalcogenide wire to be created inside a narrow silica capillary fiber [15, 16]. The silica cladding provides a robust shield for the fragile chalcogenide microwire. Tight modal confinement in such narrow waveguides increases the effective nonlinearity (γ ~10 $W^{-1}m^{-1}$ for 1 μm core diameter), while varying the core diameter allows the group velocity dispersion (GVD) to be adjusted. To match the waveguide GVD with the pump wavelength, two different samples, with core diameters d = 3.2 and 1.3 μm, were used in the experiments. Figure 3(a) plots the numerically simulated GVD of the HE_{11} mode in these two samples. It can be seen that given the 2.35 μm pump wavelength (vertical green dashed line) the 3.2 μm sample has weak anomalous dispersion close to a zero dispersion point, whereas the 1.3 μm sample provides pronounced anomalous dispersion with two zero dispersion points on opposite sides of the pump wavelength.

In order to improve the coupling efficiency into such ultrahigh numerical aperture waveguides, a "double-nanospike" structure with inverse nanotapers integrated at both ends of the waveguide has been implemented [17], as shown in Fig. 1. Through adjusting the nanospike parameters at the fabrication stage, including the lengths and tip diameters of nanospikes (NS1 and NS2), the near-field mode radii and divergence angles for the input and output light can be tuned. In the current experiment, the lengths of the input NS (NS1) were ~350 and ~150 μm respectively for the 3.2 and 1.3 μm core diameter samples, the lengths of NS2 being ~300 μm in both cases. Previously the double-nanospike waveguide was pumped with pulses from an Er-doped femtosecond fiber laser, resulting in generation of an octave-spanning supercontinuum reaching to 2.5 μm [17]. In order to fully exploit the long wavelength transparency of the chalcogenide waveguide, as well as shifting the generated spectrum away from the absorption edge of the As_2S_3 glass so as to avoid thermal effects, it is beneficial to further red-shift the pump source. This makes a femtosecond Cr:ZnS laser, emitting at ~2.35 μm, a natural choice of pump source.

The laser output was isolated using a Faraday rotator, and a half-wave plate was used to adjust the polarization state, resulting in a maximum of ~1 nJ pulse energy reaching the waveguide. The pump pulse was coupled into the waveguide through an AR-coated aspheric lens made of chalcogenide glass, the coupling being adjusted with the aid of an IR camera. Another lens of the same type was used to collimate the SC light emitted by the waveguide. The collimated light was then delivered to the input of the Fourier transform infrared spectrometer (FTIR) used for the spectral measurements.
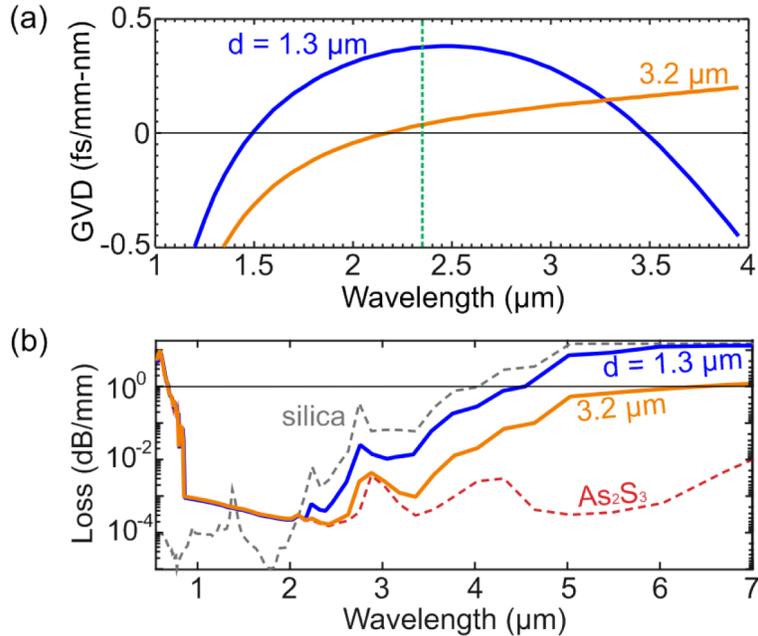
Fig. 3. (a) Simulated GVD curves and (b) propagation loss (solid curves) of the fundamental mode for the two double-nanospike As$_2$S$_3$-silica waveguides used in the experiment, calculated using the measured material losses for silica and As$_2$S$_3$ (grey and red dashed lines).

3. Supercontinuum generation

Figures 4(a) and 4(d) show respectively the measured SC spectra at several coupled pulse energies, generated in the 3.2-µm-core-diameter sample (5 mm long) and the 1.3-µm-core-diameter sample (3 mm long). Octave-spanning supercontinua were obtained in both cases at a launched pulse energy of ~100 pJ. The broadest supercontinuum, extending from ~1.2 to ~3.6 µm (–30 dB level), was measured for the 3.2 µm-core waveguide at a maximum launched pulse energy of ~200 pJ (corresponds to 3.5 kW peak power). For the 1.3 µm-core sample at 70 pJ launched pulse energy, a dispersive wave at 4.7 µm was observed in addition to the octave-spanning spectrum from 1.65 to 3.4 µm.

   The SC spectral evolution along the waveguides were modeled by numerically solving the generalized nonlinear Schrödinger equation in the frequency-domain [18]. The model included the simulated dispersion curve (Fig. 3(a)), third-order nonlinearity, the Raman effect, and the propagation loss of the fundamental mode of the waveguide (Fig. 3(b)). The modeled spectra at the output face of both waveguides are plotted in Figs. 4(b) and 4(e), and the corresponding spectral evolution over the fiber length is shown in Figs. 4(c) and 4(f). In both cases the simulated spectra agree very well with experiment. For the 3.2 µm case, initially symmetric spectral broadening is caused by self-phase modulation, then at ~3.1 mm propagation distance, strong dispersive waves are generated at ~1.2 µm. For the 1.3 µm diameter waveguide, the nonlinearity is much stronger, so that soliton fission happens after less than 1 mm of propagation. At this point the higher-order solitons (with order N ~5) breaks up into fundamental solitons and becomes phase-matched to dispersive waves at both longer (~5.5 µm) and shorter wavelengths (~700 nm, estimated from simulation, not observed due to the limited spectral range of the FTIR). The strong Raman shift further red-shifts the solitons, resulting in the successive emission of several dispersive waves. The high propagation loss at longer wavelengths, caused by the absorption in the silica cladding, strongly attenuates wavelengths longer than ~4.5 µm (see Fig. 3(b)). In contrast, for the 3.2

μm diameter waveguide, the propagation loss is negligible (less than 1 dB/mm) out to ~6.5 μm because of tighter modal confinement in the core.
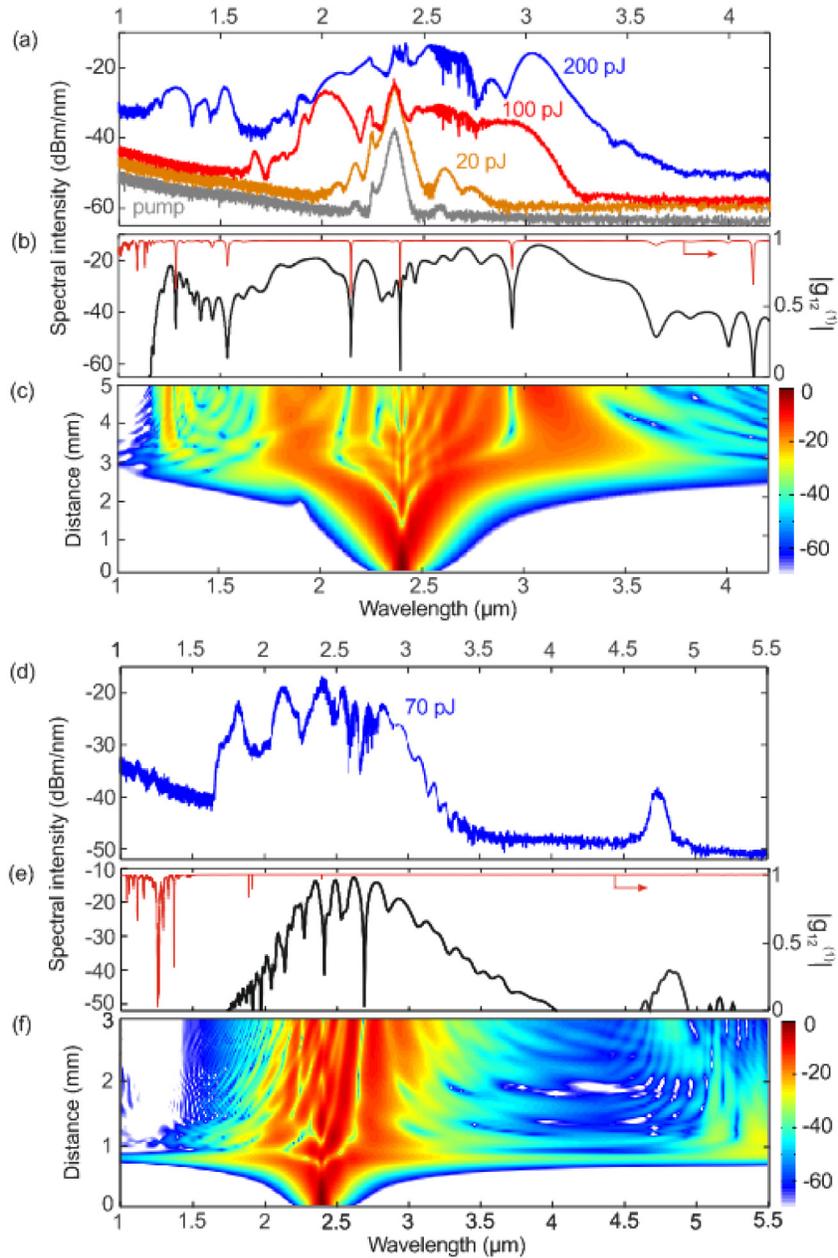

Fig. 4. (a) Measured SC spectra generated by the As_2S_3-silica double-nanospike waveguide at different pump pulse energies for the 5-mm-long waveguide with 3.2 μm core diameter. (b) Simulated SC spectrum at the output face of the waveguides. (c) Simulated SC spectral evolution along the waveguide. (d)(e)(f), Results for 3-mm-long waveguide with 1.3 μm core diameter. The dark-red solid curves in (b) and (e) plot the calculated degree of coherence of the SC spectrum at the output face of the waveguides.

It is also worth noting that neither optical damage to the As_2S_3 core, nor any spectral instabilities, were observed at these pump energies, indicating that the SC spectrum could

potentially be broadened to even longer wavelengths or work at higher repetition rates by increasing the pump power and optimizing the nanospike design (i.e. the length and tip diameter of NS1) for specific laser beam parameters.

For the purpose of frequency comb generation, the coherence of the SC spectrum was numerically simulated by applying random quantum noise (one photon per mode) to the pump pulses and estimating the modulus of the complex degree of first-order coherence defined by [19]:

$$\left| g_{12}^{(1)}(\lambda) \right| = \left| \frac{\left\langle A_1^*(\lambda) A_2(\lambda) \right\rangle}{\left[\left\langle \left| A_1(\lambda) \right|^2 \right\rangle \left\langle \left| A_2(\lambda) \right|^2 \right\rangle \right]^{1/2}} \right| \tag{1}$$

where A_1 and A_2 denote the complex spectral envelopes of independently generated SC pairs. In the simulation, 20 SC shots were used, and the results are plotted as the dark-red solid lines in Figs. 4(b) and 4(e). As expected for a supercontinuum driven by soliton fission, the degree of coherence is fairly close to unity over the entire spectral range.

4. Discussion

The low threshold energy of SC generation is a consequence of several factors: the extremely high nonlinearity of the As_2S_3-silica hybrid waveguide, the wavelength-shifted zero-dispersion wavelength due to the small effective mode area, and the coupling efficiency (~20%) boosted by the nanospike structure. As a direct comparison, we measured the SC generated using a commercially available single-mode As_2S_3 fiber (IRFlex IRF-S-5 with effective mode area 28 μm^2, γ ~0.25 $W^{-1}m^{-1}$, numerical aperture 0.3 and cut-off wavelength 1.99 μm) pumped by the same Cr:ZnS laser. The results are shown in Fig. 5. Due to the rather large fiber core diameter, a coupling efficiency of ~60% was measured for a launched pulse energy of 1.23 nJ. It can be seen that even with six times higher pulse energy, the SC bandwidth is only half as wide (less than one octave) as that generated in the nanospike waveguide.
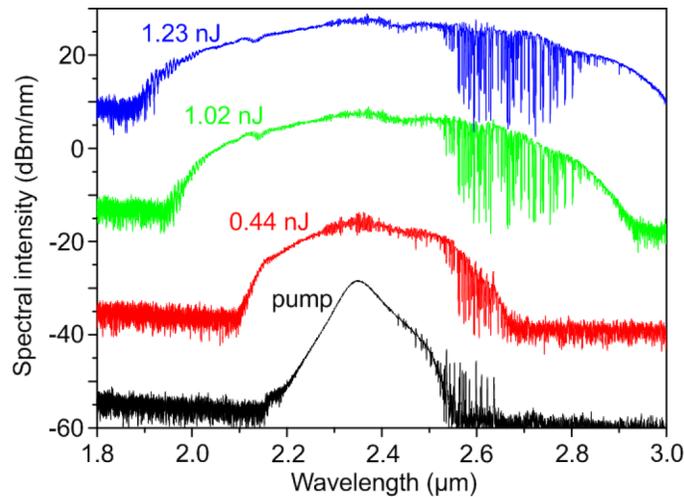

Fig. 5. Measured SC spectrum from a commercial As_2S_3 fiber pumped by the same Cr:ZnS femtosecond laser.

5. Conclusions

Broadband supercontinua can be generated in silica-cladding As_2S_3-core double-nanospike waveguides by pumping with 100 fs pulses at 2.35 μm from a mode-locked Cr:ZnS laser. For example, a more than one octave-spanning supercontinuum extending from ~1.2 to ~3.6 μm (–30 dB level) can be generated using only 200 pJ pump pulse energy. The results reported here could be important for the realization of the first solid-state-laser-based frequency comb in the mid-IR spectral range. Most recently, a GHz repetition rate Cr:ZnS solid-state femtosecond laser has been reported [20], making the present result particularly promising for extending this frequency comb system to higher repetition rates.