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Spectral-temporal composition matters when cascading supercontinua into the mid-infrared

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Abstract: Supercontinuum generation in chalcogenide fibers is a promising technology for broadband spatially coherent sources in the mid-infrared, but it suffers from discouraging commercial prospects, mainly due to a lack of suitable pump lasers. Here, a promising approach is experimentally demonstrated using an amplified 1.55 μm diode laser to generate a pump continuum up to 4.4 μm in cascaded silica and fluoride fibers. We present experimental evidence and numerical simulations confirming that the spectral-temporal composition of the pump continuum is critical for continued broadening in a chalcogenide fiber. The fundamental physical question is concerned with the long-wavelength components of the pump spectrum, which may consist of either solitons or dispersive waves. In demonstrating this we present a commercially viable fiber-cascading configuration to generate a mid-infrared supercontinuum up to 7 μm in commercial chalcogenide fibers.

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References and links
1. Introduction

Supercontinuum generation (SCG) in chalcogenide glass fibers have attracted a great deal of interest due to their nonlinear properties and wide mid-infrared (MIR) transmission band across the molecular fingerprint region to above 14 µm [1–5]. Together with the high beam quality and intensity of a fiber-delivered beam SCG in chalcogenide fibers has opened the prospects for new powerful MIR analytical tools for broadband fundamental vibrational spectroscopy [6], optical coherence tomography [7], and hyperspectral microscopy [8,9].

Currently, the most mature MIR fiber SC technology is based on fluoride glasses, such as ZrF₄-BaF₂-LaF₃-AlF₃-NaF (ZBLAN), owing to their high-power capabilities and excellent transmission from the UV to about 4.5 µm [10,11]. Due to these unique properties commercial ZBLAN SC lasers are now emerging [12–14], and in this regard cascaded SCG has been one of the key enabling technologies for efficient ZBLAN supercontinuum sources, because it has enabled the use of matured technologies such as semiconductor laser diodes, silica fibers, and fiber amplifiers [12,14,15]. However, the use of ZBLAN fibers limits the spectrum to 4.5 µm due to strong multiphonon absorption in the host material [11]. Extending the cascading scheme to chalcogenide fibers presents an attractive alternative to direct pumping schemes. However, the dynamics and efficiency of such a double-cascading scheme is highly dependent on the material and wave-guiding properties of the fibers. In general, efficient SCG towards longer wavelengths requires pumping in the anomalous dispersion regime near the fiber zero-dispersion wavelength (ZDW), where soliton dynamics is dominating [16,17]. Standard step-index silica fibers exhibit anomalous dispersion from around 1.3 µm all the way to the silica multiphonon absorption edge near 2.5 µm. Consequently, pumping a step-index silica fiber with a laser diode or erbium-doped silica fiber laser at 1.55 µm, inherently results in a SC where the long-wavelength edge is composed of solitons. Such an SC has been found to be an excellent pump source for continued broadening in ZBLAN fibers through soliton self-frequency shifting (SSFS), because it extends above the ZDW of typical step-index ZBLAN fibers at around 1.6 µm [11]. Using this scheme efficient SCG in ZBLAN fibers has been demonstrated using various combinations of seed lasers and optical amplifiers [12,14,15,18]. However, unlike silica the dispersion of step-index ZBLAN fibers is less well known due to the more complex composition of heavy-metals, and in fact these fibers may have a second ZDW before the multiphonon absorption edge. The presence of a second ZDW completely changes the spectral-temporal dynamics of the long-wavelength part of the spectrum. As solitons shift towards the second ZDW the SSFS is halted due to an increasing transfer of energy to dispersive waves (DW) that are phase-matched to a resonant wavelength in the normal dispersion regime across the ZDW. To conserve both energy and momentum the soliton frequency must shift in the opposite direction of the DW, thus counteracting the SSFS – an effect referred to as spectral recoil [17,19–21]. The average power of such a long-wavelength spectrum consisting of DWs can be equivalent or even higher than one consisting exclusively of solitons [21], which raises the fundamental question whether both spectra could be used for continued broadening in a chalcogenide fiber. To date, this has remained an open question since there have been no experimental results with coupling dispersive waves, or cascading a ZBLAN SC into a chalcogenide fiber. A ZBLAN-chalcogenide cascading scheme was studied numerically by Kubat et al. using a scalar approach to demonstrate the possibility of generating a spectrum from 0.9 to 9.0 µm by pumping a 5 µm diameter suspended-core...
As$_2$Se$_3$ fiber with a ZBLAN SC consisting solely of solitons [22]. However, these simulations should be considered a mere proof-of-concept since they omitted higher-order modes and the coupling of power to both polarization states, which strongly limits the spectral broadening [23]. Gattass et al. experimentally demonstrated SCG from 1.9 to 4.8 \( \mu \text{m} \) by launching a 2.4-2.5 \( \mu \text{m} \) silica continuum into two meters of As$_2$S$_3$ step-index fiber with 10 \( \mu \text{m} \) core diameter [24]. In their experiment the spectral broadening was limited by a strong absorption peak at 4 \( \mu \text{m} \) and the reduced efficiency of normal dispersion pumping in a large-core chalcogenide fiber.

Naturally, the choice of chalcogenide fiber affects the efficiency and dynamics of the SC cascade. The most commonly reported chalcogenide glasses used in optical fibers are As$_2$S$_3$ [24–29] and As$_2$Se$_3$ [4,30–32], where As$_2$Se$_3$ has both a stronger nonlinearity and longer MIR transmission edge, albeit at the cost of a longer material ZDW of \( \sim 7 \) \( \mu \text{m} \) compared to \( \sim 5 \) \( \mu \text{m} \) in As$_2$S$_3$ [1–3]. Consequently, efforts have been made to reduce the ZDW of chalcogenide fibers by utilizing microstructured design, such as suspended-core fibers [31,32]. Such single-glass microstructured fibers allow for further engineering of the waveguide dispersion, while maintaining relatively simple fabrication, making them an attractive choice for commercial SCG fibers [1,31].

In this letter we demonstrate the importance of the ZBLAN SC spectral-temporal composition in cascaded silica, ZBLAN, and chalcogenide fibers for extended MIR SC spectral coverage. We present experimental evidence, supported by numerical simulations, of fundamentally different spectral-temporal SCG dynamics in two similar ZBLAN fibers and its impact on spectral broadening in a chalcogenide fiber.

2. Experimental setup

The experimental setup for cascaded SCG is shown in Fig. 1. The seed was based on a 1.55 \( \mu \text{m} \) semiconductor laser diode delivering 3 ns pulses at 40 kHz repetition rate, which generated around 400 mW SC from 1.5 to 2.2 \( \mu \text{m} \) in standard silica single-mode fiber (SMF) to generate a continuum from 1.5 to 2.2 \( \mu \text{m} \) (blue curve). This SC was coupled to around 3 m Tm-doped fiber pumped by a 0.79 \( \mu \text{m} \) laser to amplify and extend the continuum from 1.8 to 2.8 \( \mu \text{m} \) (purple curve). Aspheric lenses L1-L5 was used to couple the spectrum in and out of the ZBLAN/chalcogenide fibers. The long-wavelength part of the full ZBLAN spectrum (red curve) was filtered out with a 3.5 \( \mu \text{m} \) long pass filter (LP35) and coupled into the chalcogenide fiber. The output was collimated and analysed with an FTIR and PbSe camera. A 4.5 \( \mu \text{m} \) long pass filter (LP45) was inserted when measuring the power and spectrum generated at longer wavelengths.
compared to an erbium-doped fiber amplifier was made to extend the long-wavelength edge in the ZBLAN fiber [14,15]. The amplified spectrum was then coupled to one of the two tested ZBLAN fibers and then further into a chalcogenide fiber by Black-diamond aspheric lens telescopes. Coupling to the small chalcogenide fiber core was confirmed with the aid of a PbSe camera, and the resulting spectrum was measured using a Fourier transform infrared (FTIR) spectrometer. Measurements with comparable output power rather than pump power were chosen to account for changes in coupling to the suspended-core fiber. Measurements of the intermediate silica and fluoride spectra were performed using a scanning grating spectrometer.

The chalcogenide fiber used in the experiments was a commercial As_{38}Se_{62} suspended-core fiber from Perfos with a 4.5 μm core diameter, ZDW at 3.5 μm, and low loss in the 3.2-8.0 μm window [30]. The two ZBLAN fibers (A and B) were acquired from Fiberlabs Inc. ZBLAN A had a core diameter of 6.4 ± 0.1 μm, ZBLAN B had a core diameter of 6.9 ± 0.7 μm, and both fibers had a reported NA of 0.265. The dispersion of the fibers was measured using white-light interferometry from ~1.1-4.6 μm revealing two ZDWS in both fibers. The first ZDW was around 1.51 μm in both fibers, however, due to the smaller core ZBLAN A had a shorter second ZDW at 3.56 μm, and due to the larger core variation of ZBLAN B the second ZDW was found to vary from 4.37 to 4.43 μm. This difference in ZDW between the two fibers is essential to the experiment because it means that in ZBLAN A the long-wavelength edge of the spectrum consist of DWs generated beyond the second ZDW in the normal dispersion regime, whereas in ZBLAN B it consist entirely of solitons, as illustrated in Fig. 2(a) and (b), respectively.

3. Spectral-temporal analysis and numerical simulations

The fundamentally different spectral-temporal dynamics was observed experimentally from the evolution of the long-pass filtered ZBLAN spectrum with increased pump power, as seen in Fig. 3(a) and 3(b). The figure shows how the DW peak in ZBLAN A remains centered around 4.1 μm while growing with increasing power, whereas in ZBLAN B the edge of the spectrum shifts continuously due to SSFS. Red-shifted DWs and spectral recoiling has been thoroughly investigated in fibers with negative dispersion slope going from the anomalous dispersion regime towards the second ZDW, which is the case for both ZBLAN A and B. Energy transfer to the DWs increase with propagation length, i.e., as the soliton approaches the ZDW and the overlap between the DW and the soliton becomes stronger. However, while the energy of the solitons remain localized in time the DWs are spread in a tail travelling ahead of the solitons [19,20]. To our knowledge, such a pump source consisting of DWs has
never been used in a cascading scheme before. While it may seem intuitive that DWs should be inferior to solitons as a pump source, in this case the second ZDW of ZBLAN B coincides with the absorption edge of the fiber, which may reduce the efficiency. In our experiments we investigated this by coupling the 3.5 \( \mu \text{m} \) long-pass filtered part of the ZBLAN continua into \( \sim 167.5 \text{ mm} \) suspended-core fiber. This way either exclusively DWs or solitons were launched into the anomalous dispersion regime of the suspended-core fiber.

In order to confirm the correctness of the experimental results a series of SC simulations were performed in an attempt to reproduce the experimental observations. Pulse propagation was simulated in MATLAB using the generalized nonlinear Schrödinger equation rewritten in the interaction picture formulation and integrated using a fourth order Runge-Kutta method \([22,33,34]\). The precision of the integration was improved by using an adaptive step size routine. The model assumes single-mode and single polarization only, and noise was modelled using the one-photon-per-mode scheme. For convenience we limited the pulse duration to 20 ps, and to match the experimental results in the silica fiber we used a diode laser peak power of 10 kW and a length of 10 m. The dispersion was modelled using a simplified dispersion relation from the specifications of Corning SMF28. The loss was determined empirically by incorporating appropriate loss edges modelled as Gaussian high- and low-pass filters and a Gaussian OH-absorption at 1.3 \( \mu \text{m} \) to give a good match to the experimental results. The Tm-doped silica fiber model included additional absorption and emission bands for the Tm transitions modelled as broad Gaussian loss and gain peaks centred at 1.6 \( \mu \text{m} \) and 1.9 \( \mu \text{m} \), respectively. Before further propagating the Tm-amplified spectrum the time-window was wrapped around, so the short-wavelength edge was shifted to the end of the window to allow for additional temporal broadening in the ZBLAN fiber without changing simulation parameters. Such a shift had no impact on the simulations, since the time axis is relative. A fiber length of 4.25 m with a gain of 4.13 dB/m was used to match experimental data. Because the spectral-temporal dynamics in the ZBLAN fibers are essential to the experiments, they were modelled much more accurately using the measured dispersion. The loss and numerical aperture (NA) were provided by the manufacturer (NA = 0.265 assumed to be constant). The wavelength dependent effective area and nonlinear parameter were calculated using COMSOL Multiphysics, incorporating the given fiber parameters, and coefficients for the Kerr effect and delayed Raman response found in the literature \([17,34,35]\). The spectrum was multiplied with a factor of 0.9 before further propagation to reduce broadening in the ZBLAN fibers to better match experimental spectra. The results of the silica and ZBLAN simulations are shown in Fig. 4.
Figure 4 shows the normalized output spectra from the different silica and ZBLAN fibers, while the remaining windows (b-e) show the corresponding intensity-normalized spectral-temporal composition at the output of the fibers. The expected dynamics of red-shifted DW generation (d) and continuous SSFS (e) is evident from the spectrograms. It is also evident that a small portion of the short-wavelength spectrum has wrapped around in the time-window, however, this part of the spectrum is relatively weak and was filtered out before propagation in the suspended-core fiber. A length of 15 m of ZBLAN fiber was needed in order to push the DW peak of ZBLAN A close to the experimentally observed peak at 4.15 μm. This discrepancy may be due to measurement uncertainty of the third-order dispersion and underestimation of the material and confinement losses at the long-wavelength edge.

For the chalcogenide fiber the full dispersion was obtained using COMSOL based on the refractive index of AMTIR glass [33]. The nonlinear operator was again calculated using COMSOL, incorporating the modelled Kerr coefficient from Romanova et al. [36] and the
delayed Raman response measured by Ung and Skorobogatiy [37]. Estimation of fiber loss was performed as in Kubat et al. [33]. Before propagation in the chalcogenide fiber the ZBLAN spectra were long-pass filtered and multiplied by a dampening factor of 0.21 to match experimental observations. This corresponds to a coupling loss of $-6.8$ dB which is reasonable for this fiber material and geometry. The ZBLAN A spectrum was then equalized with the ZBLAN B spectrum in terms of summed average power to make the two comparable.

4. Results and discussion

As a result, coupling solitons from ZBLAN B provided almost 1.5 $\mu$m increased spectral broadening ($10$ dB bandwidth) compared to the DWs from ZBLAN A, as seen in Fig. 5(a) and 5(b) for comparable output power levels of $4.46$ mW and $4.48$ mW, respectively. The spectra were corrected by blackbody calibration and the data set was limited on the short-wavelength side to 2.8 $\mu$m due to the detection limit of the FTIR.

Further insight into the spectral-temporal dynamics was gained by simulating both pump cases. Figure 6(a) shows a comparison between the experimental and simulated spectra for the ZBLAN A pump case. It can be seen from the input (b) and output (c) spectrograms that solitons, seen as temporally narrow horizontal lines between $-25$ ps and $10$ ps, slowly develop from the DW radiation. However, a large part of the spectrum exhibit very little spectral broadening due to the low-amplitude nature of the DWs. Our simulations show that a much longer fiber is needed for the spectrum to develop, which makes the chalcogenide fiber losses a limiting factor. In the ZBLAN B pump case, spectral broadening in the chalcogenide fiber is assumed to be largely dominated by soliton fission and subsequent SSFS due to the large increase in the nonlinear Kerr coefficient from $n_2 = 2.2 \times 10^{-20} \text{m}^2 \text{W}^{-1}$ in ZBLAN [11,34] to $n_2 = 5.72 \times 10^{-18} \text{m}^2 \text{W}^{-1}$ in As$_2$Se$_3$ [36] and the greatly reduced effective modal area. This causes a large increase in the order of the injected solitons, which consequently breaks-up within a very short length of fiber into a large amount of fundamental solitons that may continue to red-shift and collide [38]. This was also corroborated by the simulations, which shows a large number of red-shifting solitons, as seen in Fig. 7.
In both cases the absence of blue-shifted DWs in our experiments is explained as a combination of reduced sensitivity of the FTIR spectrometer used in our measurements and a large OH loss peak at 2.9 μm due to contamination and diffusion of water vapor from the atmosphere. The output power and long-wavelength edge of the spectrum was limited by the typical large losses associated with coupling to a small-core chalcogenide fiber, including the
22% Fresnel reflections, and by optically induced damage to the fiber facet, which occurred above 53 mW average pump power. Damage was observed at fairly low pump powers due to the thermal isolation of the core and the very fragile suspending struts. Still this type of fiber was chosen over more robust designs due to the ZDW at 3.5 μm, which was ideal for this experiment. The bandwidth limit presented here thus represents the limitation of the chalcogenide fiber, rather than the cascading scheme itself. In fact, a step-index chalcogenide fiber with a ZDW of 3.2 μm was recently demonstrated [5], which should allow for further power scaling. Even so, with the ZBLAN B pump and using a maximum pump power of 51.4 mW before the in-coupling lens a continuum up to 7.0 μm with a total output power of 6.5 mW and 1.5 mW above 4.5 μm was obtained after collimation, showing good qualitative agreement with the simulated spectrum as seen in Fig. 8. These results are comparable to the results of Møller et al., who obtained 15.6 mW total power and 4.7 mW above 5.0 μm in the same type of fiber through direct pumping with 320 fs pulses at 4.4 μm from a parametric amplifier [30].

![Fig. 8. Brodest experimental spectrum (solid line) from soliton cascaded SCG with a total output power of 6.5 mW and 1.5 mW above 4.5 μm, and the corresponding simulated spectrum (dashed line). The inset shows the near-field image of the output beam superimposed on a scanning electron microscope image of the suspended-core region from Perfos.](image)

5. Conclusion

In conclusion, we have demonstrated experimentally that careful control over fiber dispersion to obtain solitons at the long-wavelength edge of the pump spectrum is crucial for efficient cascaded SCG. Supported by numerical simulations our results reveal that an SC consisting of DWs is not suitable for extending the spectrum further into the MIR in short chalcogenide fibers, even though the power spectral density of the long-wavelength edge may be higher than in a corresponding soliton SC. To achieve similar results pumping with dispersive waves would require longer lengths of fiber and higher average power, which means that the efficiency of the DW generation in a DW pump fiber must be significantly higher than the efficiency of SSFS in the soliton pump fiber to give any merit to such a scheme. In demonstrating this we have presented a commercially viable scheme for chalcogenide fiber-based MIR SC sources using a double-cascading configuration with a cheap 1.55 μm laser generating an SC in standard silica fiber, amplifying and coupling this into a fluoride fiber with carefully designed dispersion, and then finally coupling this into a chalcogenide fiber with a suitable low ZDW to generate a continuum up to 7 μm with 6.5 mW total output power and 1.5 mW above 4.5 μm.

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