Supercontinuum generation in chalcogenide-silica step-index fibers

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Abstract: We explore the use of a highly nonlinear chalcogenide-silica waveguide for supercontinuum generation in the near infrared. The structure was fabricated by a pressure-assisted melt-filling of a silica capillary fiber (1.6 µm bore diameter) with Ga₄Ge₂₁Sb₁₀S₆₅ glass. It was designed to have zero group velocity dispersion (for HE₁₁ core mode) at 1550 nm. Pumping a 1 cm length with 60 fs pulses from an erbium-doped fiber laser results in the generation of octave-spanning supercontinuum light for pulse energies of only 60 pJ. Good agreement is obtained between the experimental results and theoretical predictions based on numerical solutions of the generalized nonlinear Schrödinger equation. The pressure-assisted melt-filling approach makes it possible to realize highly nonlinear devices with unusual combinations of materials. For example, we show numerically that a 1 cm long As₂S₃:silica step-index fiber with a core diameter of 1 µm, pumped by 60 fs pulses at 1550 nm, would generate a broadband supercontinuum out to 4 µm.

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References and links

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20. The minimum effective mode area was determined by searching for the core radius that yields the smallest effective area at a fixed core index.

1. Introduction

Supercontinuum (SC) light can be generated in suitably designed optical fibers using a variety of different sources including CW fiber lasers [1], Q-switched microchip lasers [2], ps fiber lasers [3] and high repetition rate lasers delivering sub-100 fs pulses [4]. Efficient SC generation relies crucially on an appropriate spectral dependence of the group velocity dispersion (GVD), and typically involves the interplay of self-phase modulation, four-wave mixing, the Raman effect, solitons, soliton fission and the soliton self-frequency shift. For example, when sub-fs laser pulses of a few nJ are launched into a fiber with a zero dispersion wavelength close to the laser wavelength, massive spectral broadening occurs within a few cm or less [5].

Photonic crystal fibers (PCF) are particularly suited to SC generation because their GVD spectrum and zero dispersion wavelengths (ZDWs) can be engineered over a very wide range [6,7]. The PCFs used in most cases consist of a strand of fused silica containing an array of microscopic hollow channels around a central glass core. Although the window of transparency of silica and lead silicate glasses begins to close off beyond ~1.9 µm, SC light has been generated out to 3000 nm, though at greatly reduced brightness [8,9]. If SC generation is to be extended further into the mid-IR, other core materials must be used. One promising candidate is chalcogenide glass, which combines a substantial nonlinear coefficient (more than 100 times larger than in silica) with a window of transparency that can extend out...
to 20 µm or beyond. Step index [10] and microstructured [11] fibers, as well as fiber tapers [12] have been fabricated using a variety of different chalcogenide glasses. A drawback of these materials is that they are difficult to produce in large quantities, usually toxic, mechanically less stable than silica and often suffer environmental degradation. It is also quite difficult to draw high quality microstructured fiber due to the steep temperature dependence of the viscosity [13].

Here we report SC generation from 60 fs pulses at 1550 nm in a dispersion-tuned chalcogenide-silica step-index fiber fabricated by pressure-assisted melt-filling [14–16] (Fig. 1a). The fiber was only 1 cm long and had a core diameter of 1.6 µm. Pulse energies below 100 pJ were sufficient to generate a broad SC extending from 980 nm (limited by the chalcogenide absorption edge) to ~2000 nm. The experimental spectra agree well with the results of a full numerical model based on the generalized nonlinear Schrödinger equation. Finally we present modeling results that suggest that SC light can be generated out to 4000 nm in As$_2$S$_3$ filled capillary with a core diameter of 1200 nm, at a pulse intensity (60 fs, 1550 nm) of 120 GW/cm$^2$ only.

2. Fiber design

![Fig. 1](image)

Fig. 1. (a) Schematic of the hybrid chalcogenide-silica step index waveguide. (b) Scanning-electron micrograph of the sample used in the experiments (core: Ga$_4$Ge$_{21}$Sb$_{10}$S$_{65}$; cladding: silica; core diameter: 1.6 µm). The sample was cleaved at around 500°C, so the chalcogenide surface is smooth (see text). (c) Side image of a sample with a continuous chalcogenide strand (core diameter 10 µm, outer fiber diameter 200 µm).

The step index fibers investigated here consist of a fused silica cladding (refractive index 1.44 at 1550 nm) and a cylindrical core of Ga$_4$Ge$_{21}$Sb$_{10}$S$_{65}$ glass [10,17,18], which has a bulk ZDW at ~5 µm and a refractive index of ~2.25. The calculated GVD of the HE$_{11}$ guided mode (at a wavelength of 1550 nm) is plotted against core diameter in Fig. 2a. It is zero for a core diameter of 1.7 µm. In the experiments the core diameter was 1.6 µm (Fig. 1b), which moves the ZDW to a wavelength shorter than 1550 nm. The pump light then falls in the anomalous dispersion region (Fig. 2a), which is favorable for SC generation.
The large core-cladding refractive index difference strongly confines the light to the core, making effective use of the high chalcogenide nonlinearity ($2 \times 10^{-18}$ m$^2$/W, compared to $2 \times 10^{-20}$ m$^2$/W for silica). The effective area $A_{\text{eff}}$ [19] of the HE$_{11}$ mode is only 1.5 times larger than in an air-clad chalcogenide strand (inset of Fig. 1b) [20]. If other core materials with lower refractive index are used (e.g., Schott SF6 [21], tellurite or Ge-doped silica glass) this ratio is much less favorable (Fig. 2b). Finally we note that the micro-Raman spectrum of the chalcogenide glass is identical before and after pumping into the silica capillary, suggesting that the bulk properties of the glass are not affected by melt-filling [14].

3. Fabrication

Using pressure-assisted melt-filling, molten chalcogenide glass ($T_g = 315^\circ$C) was forced into the silica capillary fibers at 665$^\circ$C and 100 bar. The upper ends of the capillaries were fused shut, and they were placed vertically, resting on the chalcogenide melt. The temperature was chosen to avoid evaporation of the chalcogenide while providing sufficiently low viscosity. Filling lengths of a few cm were reached after 30 minutes (Fig. 1c). The maximum strand length is limited by the length of the hot zone and the filling time. After filling, the samples are cooled down while keeping the pressure constant to prevent bubble formation in the chalcogenide core resulting from glass decomposition and reboiling [14]. When the filled fiber was cleaved at room temperature the chalcogenide surface appeared rather rough and the
core developed periodic cracks along its entire length, a phenomenon that we attribute to mechanical stress caused by the large difference in thermal expansion between chalcogenide and silica. By hot-cleaving at a temperature of ~500°C, however, we found that the surface roughness was greatly reduced (see Fig. 1b) and the cracking in the core eliminated. Despite the large mismatch in thermal expansion coefficients, which would cause the core to pull away from the silica cladding, no evidence of core-cladding gaps was seen, perhaps because the samples are cooled under high pressure.

4. Optical setup

Samples ~1 cm long were sufficient for observation of efficient SC generation, and since no tapered pigtail was necessary, the GVD was constant along the whole sample. This meant that no pre-chirping of the pump pulse was required as is the case for fiber tapers [12]. Pulses from an erbium-doped mode-locked fiber laser (60 fs, 100 MHz repetition rate, 1.2 nJ, bandwidth 1470 to 1630 nm) were launched into the fiber using an aspheric lens with focal length 8.0 mm (Fig. 3a). Launch efficiencies of 35% were achieved with piezo-controlled translation stages. Using a microscope objective (40 x, NA 0.65), the transmitted light was projected on to an iris diaphragm so as to block any unwanted cladding light. A multimode fiber delivered the signal to an optical spectrum analyzer (OSA) or an infrared CCD spectrometer.

5. Experimental results

Figure 3b shows the measured SC spectra at increasing pulse energies, the numbers next to each graph indicating the pulse energy in pJ in the fiber, i.e., after taking into account a coupling efficiency of 35%. The grey curve represents the pump laser spectrum. Strong spectral broadening occurs on both sides of the laser spectrum, reaching a bandwidth of ~1000 nm at a pulse energy of 21 pJ, the range 1200 to 1730 nm lying above the −10 dB level. The maximum broadening is achieved at a pulse energy of 60 pJ (blue curve), ranging from 980 to ~2000 nm. The long wavelength edge of the SC at 60 pJ cuts off at 10 μW/nm, the measurement being limited by the spectrometer sensitivity (Fig. 3c).

![Optical Setup](image-url)

![SC Spectra](image-url)

Fig. 3. (a) Schematic of the optical setup. The optical micrographs on the right-hand side show the near-field mode patterns at (A) 1310 nm and (B) 1550 nm. (b) SC spectra of the sample. Over the range 1 to 1.75 μm an OSA was used (minimum detectable spectral power density 10^{-6} μW/nm). Beyond 1.75 μm (the grey-shaded region) an Ocean Optics (OO) CCD spectrometer was used (minimum detectable spectral power density 10^{-2} μW/nm). The grey curve is the laser spectrum and the colored curves are the output spectra at different launched pulse energies in pJ (the numbers adjacent to each curve) taking account of the 35% coupling efficiency.)
Using a beam profiler and narrow-band optical filters at 1310 and 1550 nm (right-hand side of Fig. 3a), we confirmed that the SC light is always generated in the HE$_{11}$ mode, i.e., not in any higher order mode.

### 6. Simulations

Numerical simulations were performed by solving the generalized nonlinear Schrödinger equation [22]:

$$\frac{\partial A(z, \tau)}{\partial z} = DA(z, \tau) - i\left(\gamma(\omega_k) + i\gamma_1 \frac{\partial}{\partial \tau}\right) \times \left(A(z, \tau) \int_{-\infty}^{\infty} R(t') |A(z, \tau)|^2 dt'\right)$$  \hspace{1cm} (1)

by the split-step Fourier method. Equation (1) describes the change in pulse envelope $A(z, \tau)$ during propagation, $\tau = t - z/v_g$ being the time in a reference frame moving at the group velocity of the pulse, $t$ the physical time and $v_g$ the group velocity. The operator $D$ includes the linear dispersion of the device and is applied in the spectral domain. The nonlinearity is represented by $\gamma(\omega_0) = \gamma_0 + \gamma_1 (\omega - \omega_0)$ with $\gamma_0 = 7.2 \text{ W}^{-1}\text{m}^{-1}$ and $\gamma_1 = 7.68 \times 10^{-15} \text{ sW}^{-1}\text{m}^{-1}$.

Since the Raman response function $R(t)$ for Ga$_4$Ge$_{21}$Sb$_{10}$S$_{65}$ glass is unknown, we used the response of As$_2$S$_3$ glass, with a Raman period of 15.5 fs and a lifetime of 230.5 fs [23,24].

![Fig. 4. (a) Numerically modeled output spectra at different positions (spectral intensity in dB (scale bar on the right)). The dashed black line indicates the zero dispersion wavelength of the HE$_{11}$ mode at 1550 nm. The upper diagram shows the output spectrum at 1 and 10 cm (ND: normal dispersion, AD: anomalous dispersion regime).](image)

Figure 4 shows the spectral evolution of a 14 pJ pulse (duration 59 fs) propagating along a 1 cm long device. The pump wavelength (1550 nm) is close to the ZDW of the HE$_{11}$ core mode. Launching light close to the ZDW generates two spectral sidelobes by self-phase modulation. The separation of these lobes increases with distance, spreading out into the normal and anomalous dispersion regions. The growth of the bandwidth halts at ~4 mm, the lobe in the anomalous GVD region developing into a soliton with energy 6.6 pJ, center...
wavelength 1848 nm and order $N = 1.55$. This pulse transforms into a $N = 2$ soliton [25] and sheds energy to 1180 nm, contributing to the strong spectral band centered at 1170 nm. Note the spectral forking that occurs at ~0.6 cm and ~1200 nm when the soliton (at 1848 nm) overlaps temporally with linear radiation in the normal dispersion region. During subsequent propagation the trailing edge of the linear radiation is trapped by the soliton, leading to the generation of two spectrally separated bands [26].

Propagation beyond 1 cm does not broaden or change the spectrum significantly, apart from a gradual Raman-related shifting of the soliton to longer wavelengths. This is illustrated in the upper plot in Fig. 3, showing the spectrum after 1 cm and 10 cm. Although the soliton has shifted to 2020 nm, the short wavelength edge has not changed. Our simulations show that a device length of < 1 cm is sufficient to generate efficient SC. Longer sample lengths merely reduce the transmission, especially at longer wavelengths, without enhancing the SC width.

![Fig. 5. (a) Output spectrum of a 1 cm long hybrid device for different core radii (core material: $\text{As}_2\text{S}_3$, cladding: silica. Peak pulse intensity 120 GW/cm$^2$, center wavelength 1550 nm, pulse duration 59 fs). Note that the pulse energy will increase approximately as the square of the core diameter and is ~70 pJ at a diameter of 1100 nm. The spectral intensity is given in dB (scale bar on the right). The dashed black line indicates the zero dispersion wavelength of the fundamental core mode. Inset: schematic of the arsenic hybrid waveguide. (b) Material loss and loss of fundamental core mode for a core diameter of 1 µm. Grey area indicate regime in which the loss of the chalcogenide glass dominates the modal attenuation.](image)

### 7. Discussion & outlook

It is interesting to ask whether a broader SC could be produced using a different chalcogenide glass system. After examining several alternatives, we have found that $\text{As}_2\text{S}_3$ is an excellent choice. Using its known optical properties (refractive index ~2.44 at 1550 nm, bulk absorption ~0.97 dB/m at 1550 nm, $n_2$ ~2.6 × 10$^{-18}$ m$^2$/W, TPA coefficient ~6.2 × 10$^{-4}$ cm/GW, $T_g$ ~175°C) and dispersion, the spectra after 1 cm of propagation were calculated using Eq. (1) and plotted versus core radius in Fig. 5a. The pulse duration and the peak intensity at an experimentally realistic level (below the damage threshold [13]) are 59 fs and 120 GW/cm$^2$, resulting in a different maximum pulse energy for each core size. The dashed black curves indicate the ZDWs of the $\text{HE}_{11}$ mode, which largely determine the width of the spectrum. For a core size smaller than 300 nm the nonlinear coefficient is very high (50,000 W$^{-1}$km$^{-1}$ at a core radius of 200 nm) but normal GVD prevents effective broadening. As the core radius is increased, an anomalous dispersion region develops close to the pump wavelength, leading to a strong broadening. The most intense broadening occurs at core radii between 480 and 620 nm. For example, at a radius of 580 nm the spectrum extends from 685 nm out to 4 µm. At
this point the mid-IR light is generated by soliton-driven dispersive wave generation across the second ZDW (this requires negative $\beta_3$). Spectral broadening further into the IR is prevented because the mode spreads out into the silica cladding where the material loss is very high. Figure 5b compares the absorption of silica [27] and the $\text{As}_2\text{S}_3$ glass [28] with the calculated loss of the guided HE$_{11}$ mode. When the core size is further increased, the solitons are unable to reach the second ZDW and the mid-IR extension shrinks again.

In conclusion, silica capillary fibers can be successfully filled with chalcogenide glass so that the zero dispersion wavelength for the HE$_{11}$ mode lies close to 1550 nm. Experiments on a 1 cm long fiber filled with Ga$_4$Ge$_2$Sb$_{10}$S$_{65}$ glass show supercontinuum light from 1.1 $\mu$m to 2 $\mu$m with spectral intensities between 10 nW/nm and 1 $\mu$W/nm, for 60 fs pump pulse energies of only 60 pJ. Despite high attenuation in the silica cladding in the infrared beyond 2 $\mu$m, simulations show that an $\text{As}_2\text{S}_3$ filled fiber (core diameter 1.1 $\mu$m) would produce a supercontinuum spectrum from 700 nm to 4 $\mu$m when pumped at 1550 nm by 60 fs pulses of only 70 pJ energy. The pressure-filling technique uniquely allows chalcogenide glasses to be integrated into silica fibers, leading to highly nonlinear devices with very strong optical confinement, making them interesting candidates for all-optical switching. The silica cladding provides a robust sheath for the mechanically less stable chalcogenide glass and protects it from atmospheric degradation. Compared to silica fibers and silica PCFs, these hybrid structures have windows of transmission that can extend into the IR while providing very large nonlinearities, suggesting that they may be useful for generating supercontinuum light in the mid-IR. A hybrid fiber with a rare-earth-doped chalcogenide core may be useful for optical amplification, and incorporation of more exotic glasses may permit realization of fibers that respond to external magnetic fields.