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Mid-IR supercontinuum in optical fibers drawn from low phonon energy glasses

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ABSTRACT

Optical fibers mid-infrared (mid-IR) supercontinuum (SC) generation for sources covering the 1–20 µm range are of great interest for many applications in optics, spectroscopy, sensing for environmental monitoring or medical diagnosis and treatment. We present here our work regarding two low phonon energy glasses families, leading to highly nonlinear optical fibers for SC generation: tellurites and seleno-telluride glasses. Tellurite fibers are suitable for working in the 1-5 µm range, when seleno-telluride ones are intended to the 2-16 µm range. For tellurites, we focus on the definition of glass pairs suitable for the drawing of step index fibers with a controlled chromatic dispersion for a femto-second (fs) pumping around 2 µm. In the case of chalcogenide glasses, we focus on the Ge-Se-Te ternary system, which offers the advantage of allowing the drawing of step index or micro-structured fibers avoiding the usage of toxic arsenic. Depending on the fiber geometry the management of the chromatic dispersion is quite different. Suspended core fibers allow to shift deeply the unique zero dispersion wavelength (ZDW) towards short wavelengths for fs pumping around 2-3 µm. For step index fibers, it is possible to design waveguides with no, one or two ZDW. Various pumping schemes are available between 3 and 9 µm, with a fs tunable source. As a result, SC generation experiments in these different fibers allow to reach wide spanning spectra, between 1 and more than 5 µm for tellurite fibers, and between 2 and more than 14 µm in the case of chalcogenides ones.

Keywords: Tellurite glasses, chalcogenide glasses, optical fibers, nonlinear properties, supercontinuum generation, infrared.

1. INTRODUCTION

Mid-infrared (mid-IR) light sources from 2 to 20 µm are of key importance for applications in molecular sensing, medicine, security and defense, and in free-space communication. The mid-IR spectrum covers important atmospheric windows, and numerous features of molecular gases, toxic and explosive agents, pollutants etc. Bright, coherent and broadband radiations bring numerous practical advantages for applications such as spectroscopy and frequency metrology, and this can be conveniently obtained by means of fiber-based supercontinuum light generated from femtosecond mode-locked lasers, thus surpassing thermal infrared sources and challenging quantum cascade lasers. In the last decade, strong efforts have been made to transfer robust supercontinuum (SC) fiber sources from near-infrared into the mid-IR spectral range, mainly driven by the purpose of covering the largest spectral region possible. Recent developments now focus on satisfying the user needs for mid-IR SC application such as compact and high repetition rate sources, single-mode behavior, all-fiber systems, high power spectral density, and coherent emission across the SC spectrum. The recent development of highly nonlinear optical fiber based on tellurite or chalcogenide glasses appears as an interesting alternative to other optical fibers, because of their wide IR transmission combined to their high nonlinearity. This implies to control the glasses compositions, the fiber drawing operations, and to manage the chromatic dispersion in order to reach adequate dispersion profiles in accordance with the used pumping source. The characteristics of the two glasses families of interest are quite different from this point of view. The lower dispersion of tellurites allows to use lower pumping wavelengths, typically in the 2-3 µm range, for which commercial laser sources are available, but with a lower IR extension, when the strong dispersion of chalcogenides often leads to a pumping at higher wavelengths, less convenient, but with a broad IR extension as a result. We present here our last achievements related with infrared supercontinuum generation with both tellurite fibers and chalcogenide fibers.

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2. SUPERCONTINUUM GENERATION IN TELLURITE FIBERS

2.1 Tellurite glasses

Tellurite glasses are a family of low phonon energy glasses, with a phonon energy for the Te-O bond around 700 cm\(^{-1}\) and an IR transparency which reaches 6 \(\mu\)m on bulk samples. We developed our step-index tellurite fibers for mid-IR SC studies through the built-in-casting and the rod-in tube techniques which are well described in the literature\(^{12-14}\). A compatible core/cladding glass pair was chosen based on their optical, thermal and mechanical compatibility (Table 1) for the build-in-casting process, and their suitable refractive index difference\(^{14}\). The compositions are respectively 80TeO\(_2\)-5ZnO-10Na\(_2\)O-5ZnF\(_2\) (TZNF, molar fraction) and 60TeO\(_2\)-5ZnO-20Na\(_2\)O-15GeO\(_2\) (TZNG).

<table>
<thead>
<tr>
<th></th>
<th>(n) (633 nm)</th>
<th>(T_g) (°C)</th>
<th>(\alpha) (\times 10^{-6} \cdot K^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>TZNF</td>
<td>2.037</td>
<td>280</td>
<td>16.7</td>
</tr>
<tr>
<td>TZNG</td>
<td>1.905</td>
<td>272</td>
<td>13.0</td>
</tr>
</tbody>
</table>

Table 1. Main properties of the core and cladding glasses used for build-in-casting.

The synthesis and quenching of the glass preforms are performed in a glove box under dry atmosphere to avoid any water contaminations from the environment and thus reducing the amount of residual OH groups in the glass matrix. To further reduce the presence of hydroxyl compounds and limit the mid-IR absorption of the fiber, zinc fluoride is added to the core composition for dehydration of the bulk glass. At the end of this process, we obtain a 45 mm long and 16 mm large cylindrical preform inside which it is easy to differentiate the two different compositions (figure 1)\(^{12}\).

![Figure 1](image1.png)

(a) (b)

Figure 1. Pictures of the cross-section (a) and side-section (b) of the step-index preform synthetized with the built-in-casting method.

2.2 Tellurite fibers

One part of the step-index glass preform obtained thereby is heated above the vitreous transition temperature \(T_g\) in order to be drawn into a large core step-index fibre for loss measurements (figure 2).

![Figure 2](image2.png)

Figure 2. Large-core (TZNF/TZNG) fiber losses measured by means of the cutback method.
The fiber exhibits background losses below 1 dB/m between 1 and 2.8 μm. Thanks to our dehydration process, the glass OH content is very low, namely the water-related absorption band between 3 and 4 μm is almost eliminated, before losses increase when reaching the multiphonon absorption edge around 4 μm. Even at such wavelength, the fiber losses still remain moderate when considering cm-long fiber segments for SC studies in the following. The second part of the step-index glass preform is drawn into capillaries with an outer diameter of 800 μm and then used for the next rod-in-tube stage. A second cladding glass preform used as a tube is made following the same protocol as previously described and drilled mechanically. The step-index rod is then inserted into the hole of the second cladding preform and another drawing stage is performed to obtain small-core step-index fibers (see Figure 3).

Figure 3. Cross-section of the small core step-index fibre under optical binocular with (a) a×20 objective and (b) a×100 objective.

By monitoring the total diameter of the fibre during the latter drawing, it is possible to estimate quite accurately the core size of the resulting fiber. Typically, core diameters varying between 3 and 6 μm are obtained when the outer fiber diameter is roughly between 120 and 240 μm.

2.3 Supercontinuum generation in tellurite fiber.

The wavelength-dependent refractive index curve for each glass can be determined with different interpolation schemes based on two-pole Sellmeier equations fitting near-IR measurements on bulk materials\cite{14,15}. Then properties of the fundamental guided mode are investigated by numerically solving the dispersion equation for cylindrical step-index waveguides. Figures 4 reports the calculated dispersion curves and effective mode area variations as a function of wavelength\cite{12}. We observe that our fiber design provides distinct dispersive regimes by simply varying the core diameter from 3 to 4 μm. An all-normal dispersion profile and a two zero dispersion wavelength (ZDW) profile are obtained. We find that anomalous dispersion only exists for core diameters $\phi \geq 3.25$ μm. The first ZDW can be easily tuned between 1.92 and 2.35 μm, while a second ZDW is present for $\phi < 3.8$ μm, and shifted from 2.4 to 4.3 μm. The flexibility of GVD properties beyond 2 μm combined with strong optical confinement allow us to investigate an overview of relevant nonlinear dynamics of SC generation.

Figure 4. Wavelength-dependent curves of dispersion parameter $D$ and effective mode area $A_{\text{eff}}$ of the fundamental guided mode in distinct step-index tellurite fibers with respective core diameters $\phi = 3, 3.25, 3.5, 3.75,$ and 4 μm.

For experimental SC demonstrations we used as a pump source an optical parametric amplifier (OPA) allowing to change the pump wavelength between 1.5 and 4 μm. We made a study as a function the fiber core diameter, the pump
wavelength and the fiber length. We achieved an infrared spectral bandwidth of over 4000 nm spanning from 1.3 µm to 5.3 µm when a 9-cm-long segment of 3.5-µm core tellurite fiber was pumped at 2400 nm (figure 5). This is, to the best of our knowledge, the broadest supercontinuum in the mid-infrared obtained in tellurite fibers. The enhancement in spectral broadening compared to recent publications is mainly attributed to the low and rather flat dispersion (even with multiple ZDWs) associated with an optimized pump wavelength. The long-wavelength edge of the SC is here limited by the intrinsic transmission window of the tellurite glass. The achieved output average power is 150 mW with ~45% power proportion in the important transparent 3 to 5 µm atmospheric window. Our results make step-index tellurite fibers suitable candidates for the next generation of nonlinear photonic devices in the important spectral region from 2 to 5 µm, in particular to take advantage of the recent development of ultrashort pulse lasers sources beyond 2 µm\textsuperscript{16}.

Figure 5. Experimental SC spectrum obtained for a 9 cm tellurite fiber length and a 3.5 µm core diameter pumped at 2.4 µm with a constant incoming power of 400 mW.

3. SUPERCONTINUUM GENERATION IN CHALCOGENIDE FIBERS

3.1 Chalcogenide glasses

Chalcogenide glasses are a family of low phonon energy glasses, with a phonon energy varying between 350 cm\textsuperscript{-1} and 200 cm\textsuperscript{-1} from sulfide to telluride glasses, and an IR transparency which reaches respectively 10 µm to more than 20 µm on bulk samples. In the present work, the studied compositions belonging to the Ge-Se-Te ternary system for both core and cladding glasses, more precisely on compositions belonging to the pseudo binary GeSe\textsubscript{4}– GeTe\textsubscript{4}, on the Se rich side\textsuperscript{17}. The thermal and optical properties of bulks and fibers have been investigated as well as the purification process, in order to obtain low-losses optical fibers. The cladding and core compositions are Ge\textsubscript{20}Se\textsubscript{60}Te\textsubscript{20} (87.5% GeSe\textsubscript{4} – 12.5% GeTe\textsubscript{4}) and Ge\textsubscript{20}Se\textsubscript{60}Te\textsubscript{20} (75% GeSe\textsubscript{4} – 25%GeTe\textsubscript{4}) respectively (table 2). In the following, these two compositions are indifferently called GST glasses.

<table>
<thead>
<tr>
<th>Composition</th>
<th>T\textsubscript{g} (°C)</th>
<th>Drawing temperature (°C)\textsuperscript{a}</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ge\textsubscript{20}Se\textsubscript{60}Te\textsubscript{20} (core)</td>
<td>162</td>
<td>430</td>
</tr>
<tr>
<td>Ge\textsubscript{20}Se\textsubscript{70}Te\textsubscript{10} (clad)</td>
<td>159</td>
<td>430</td>
</tr>
</tbody>
</table>

The starting elements are 5N bulky Ge and Te from Umicore, Se is 5N pellets from Alfa Aesar. After weighing of stoichiometric quantities, all three elements are inserted into silica ampoules which are evacuated to 1 x 10\textsuperscript{-5} mbar for several hours. Under such a secondary vacuum, Te and Se containing ampoules were heated up to 430°C and 230°C, respectively, in order to remove TeO\textsubscript{2} and SeO\textsubscript{2} superficial oxides. Germanium was heated up to 600°C to remove free water adsorbed on the raw material surface. Then, the precursors are gathered in the same ampoule in which 50 ppm (wt) of Al and 1000 ppm (wt) of TeCl\textsubscript{4} are added for further purification. Al is used as an oxygen getter and TeCl\textsubscript{4} as a hydrogen getter. In the next step, the glass is distilled under dynamic and static vacuum to remove the impurities.
Finally, after sealing the synthesis ampoule, the glass batch is inserted into a rocking furnace for the refining process at 850°C for 10 hours, and then quenched and annealed at T_g – 5°C for 12h.

3.2 Chalcogenide fibers.

Form the previous glass synthesis, single material fibers made from both the core and cladding glasses are drawn in order to measure the material losses by the cut-back technique (figure 6). We also fabricated suspended core fibers by means of the mechanical drilling method on initial rods from core glass, with a 16 mm outer diameter: after annealing, a glass rod is drilled to obtain three holes of typically 1 mm diameter and 30 mm length, thus leading to the air-suspended structure with three fine struts supporting the suspended core. The resulting perform is then drawn into fiber under a nitrogen gas pressure to control the hole size. Step-index fibers are fabricated by means of the rod-in-tube method. First, a 8-cm-long rod with 16 mm outer diameter is stretched down to 900 µm glass cane which is then inserted in a drilled clad perform, thus providing the suitable preform to draw step-index fiber with 100-350 µm outer diameter (i.e., corresponding to a core diameter varying from 6 to 20 µm).

![Figure 6. a: Infrared attenuation spectra of single material fibers made of core and cladding glasses; b: Profile of a chalcogenide step-index fiber with 10-µm core diameter captured by means of a scanning electron microscope (SEM).](image)

3.3 Supercontinuum generation in chalcogenide fibers.

Refractive index values of core and clad glass compositions are calculated based on the Clausius Mossoti relation and the use of refractive index measurements reported in literature on binary systems, namely GeSe_4 and GeTe_2 glasses. These obtained values can be well fitted by two-poles Sellmeier equations. The refractive index difference is found to be Δn = 8.9x10^{-2} at 5 µm. Based on a simple step-index profile, we studied the corresponding dispersive properties of the fundamental mode propagating in our chalcogenide step index fibers with different core diameters (figure 7a). An uncertainty of ±10^{-2} on the refractive index difference (Δn) between core and cladding is taken into account to roughly estimate the impact of potential discrepancies between our numerical predictions and the genuine refractive indices of the fabricated glasses, hence we plot three dispersion curves for each core diameter. For a core diameter ranging from 6 to 14 µm, we observe that our fiber can exhibit distinct dispersive properties such as an all-normal dispersion (ANDi) profile or even a two-zero dispersion wavelength (ZDW) dispersion profile. By simply tuning the core diameter during fiber drawing, we are able to manage the group-velocity dispersion (GVD) of the fundamental guided mode ranging from all-normal dispersion (Φ_{core} = 6 µm) to multi-ZDW GVD profile (Φ_{core} = 10 µm). A standard single-ZDW dispersion profile is then found for larger core diameters (Φ_{core} = 14 µm). Consequently, various nonlinear propagation regimes can be explored to optimize SC generation. For suspended core fibers, we performed numerical simulations of modal properties based on the fiber geometry derived from the SEM analysis and by means of commercial software using a fully vectorial finite-element model (figure 7b) to show the wavelength-dependence of dispersion parameter D for the fundamental guided mode for distinct values of the core diameter.
Figure 7. a: Calculated wavelength-dependent dispersion $D$ of the fundamental guided mode in our step-index fibers with varying core diameter $\phi = 6$, 10, and 14 $\mu$m. The solid black line indicates the chromatic dispersion from the core glass (Ge$_{20}$Se$_{60}$Te$_{20}$). Additional dashed curves give an estimate of possible dispersion variations induced by the uncertainty on refractive index difference for our glass pair; b: Calculated wavelength-dependent dispersion $D$ of the fundamental guided mode in our suspended core fibers with varying core diameter $\phi = 3$, 6, 9, and 12.7 $\mu$m. The solid black line indicates the chromatic dispersion from the core glass (Ge$_{20}$Se$_{60}$Te$_{20}$).

For supercontinuum generation experiments, we use a non-collinear optical parametric amplifier (NOPA) followed by a difference frequency generation (DFG) module, delivering fs pulses between 1 and more than 10 $\mu$m. The present flexibility in the design of GVD properties for step-index fibers combined with strong optical confinement and our tunable fs pumping allows us to investigate an overview of relevant configuration of SC generation. We made a study as a function the fiber core diameter, the pump wavelength and the fiber length. The largest SC spectrum here spans from 2 up to nearly 14 $\mu$m (-30 dB bandwidth) for a 10 $\mu$m core step index fiber, pumped at 7.7 $\mu$m (figure 8a)$^{17}$, with a flat dispersion behavior. In the case of suspended core fibers, the largest SC spectrum roughly spans from 2 to 11.5 $\mu$m, and is obtained for the pump fixed at 5.5 $\mu$m corresponding to the anomalous dispersion of the fiber, close to its ZDW (figure 8b).

Figure 8: a: Experimental SC spectrum obtained with 10-µm-core chalcogenide step-index fiber pumped at 7.7 $\mu$m; b: Experimental SC spectrum obtained with a suspended core chalcogenide fiber pumped at 5.5 $\mu$m.

4. CONCLUSION

We demonstrate the experimental development tellurite and chalcogenide fibers with low residual losses. Distinct fiber structures such as step-index or suspended-core profiles are designed and then drawn to evidence easy-accessible management of group-velocity dispersion properties. SC generation experiments are carried out, and our results clearly confirm the potential of these new fibers for nonlinear optics in the mid-IR. More particularly, a SC spectrum spanning from 1.3 to 5.3 $\mu$m is obtained with a tellurite step index fiber when a SC spanning from 2 to 14 $\mu$m is obtained by using a 40-mm-long segment of 10-µm-core step-index fiber made of the GST glass pair.
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