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# Filamentation-induced spectral broadening and pulse shortening of infrared pulses in Tellurite glass



P. Béjot <sup>a</sup>, F. Billard <sup>a</sup>, C. Peureux <sup>a</sup>, T. Diard <sup>a</sup>, J. Picot-Clémente <sup>a</sup>, C. Strutynski <sup>a</sup>, P. Mathey <sup>a</sup>, O. Mouawad <sup>a</sup>, O. Faucher <sup>a</sup>, K. Nagasaka <sup>b</sup>, Y. Ohishi <sup>b</sup>, F. Smektala <sup>a,\*</sup>

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#### ABSTRACT

Filamentation of infrared femtosecond pulses in Tellurite glass is reported, leading to the generation of a supercontinuum generation spanning from the visible up to 4  $\mu$ m. The angular distribution of the supercontinuum shows clear evidence of conical waves generation, in particular, in the visible region. Moreover, taking advantage of the spatio-temporal self-focusing effect occurring in the Tellurite glass, a twofold pulse shortening is demonstrated. Tellurite glass appears as a very convenient, versatile and promising medium for femtosecond nonlinear optics in the infrared region.

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#### 1. Introduction

Supercontinuum generation in the mid-infrared region (MIR) is actually a main topic of research in nonlinear optics because of its potential applications such as detection of biological, toxic or pollutant chemicals species, metrology, tomography or active hyperspectral imaging. The production of supercontinuum sources can be basically divided in two distinct categories. The first one is based on the use of optical fibers, originally in silica [1-8]. The main drawback of silica fibers is their transparency range that stops around 2.2 µm [9,10], preventing an efficient generation of mid-infrared supercontinua. Consequently, attention has been devoted to design new types of glasses with transparency belonging to the MIR region. Materials such as tellurite, chalcogenide, heavy oxides or fluoride glasses have been proposed as potential alternatives to silica because they all exhibit broader transmission windows and for most of them higher nonlinear optical properties [11]. Novel optical fibers from these glasses yield to supercontinua spanning from the ultraviolet into the mid-infrared [12–16]. Despite the extreme bandwidth achieved in optical fibers, the use of external waveguide limits intrinsically the peak power and is alignment sensitive. Alternatively, supercontinuum

 $\hbox{\it E-mail address: } Frederic. Smektala@u-bourgogne. fr~(F.~Smektala).$ 

can be produced by filamentation in transparent media. Mid-infrared supercontinua have been produced in bulks [17–21] and even in liquids [22] and in gas [23]. Here, we focused our attention on a low-cost and easy to produce tellurite-based bulk which can be further drawn into optical fiber, because of its wide transmittance window, chemical durability and high optical nonlinearity, in order to get a better understanding of its nonlinear behavior.

With a similar bulk tellurite glass material less than 1 cm in thickness, a supercontinuum generation spanning from the visible to 6 µm was previously reported [21]. Nevertheless, as already reported in different bulk materials [20,24,25], one drawback of supercontinua produced by filamentation is the spatial homogeneity of the spectrum. Indeed, during the nonlinear propagation, conical waves are spontaneously produced. Such waves are interpreted as the manifestation of the nonsolitary stationary solutions to the nonlinear Schrödinger equation and arise from the nonlinear dynamics of the pulse in Kerr dispersive media [26–29]. The formation of such waves during the filamentation process makes the generated supercontinuum not uniform neither spatially nor angularly. In this context, it is of prime importance to characterize angularly the generated supercontinuum for its use in further applications. Here, we propose a full angular characterization of the supercontinuum generation as a complement to these previous results already published in Tellurite glasses. Moreover, we show that the nonlinear propagation in such a glass can lead to a pulse duration compression up to a factor of 2.

<sup>&</sup>lt;sup>a</sup> Laboratoire Interdisciplinaire Carnot de Bourgogne, UMR 6303 CNRS-Université de Bourgogne Franche-Comté, 9 Av. A. Savary, BP 47870, F-21078 DIJON Cedex, France

<sup>&</sup>lt;sup>b</sup> Research Center for Advanced Photon Technology, Toyota Technological Institute, 2-12-1 Hisakata, Tempaku, Nagoya 468-8511, Japan

<sup>\*</sup> Corresponding author.

#### 2. Spectral broadening and conical emission in Tellurite glass

The experimental setup is depicted in Fig. 1. The laser source is a chirped pulse amplified Ti:Sapphire system which delivers pulses centered at 790 nm with 3 mJ energy per pulse (100 fs pulse duration) at 1 kHz repetition rate. About 300 µJ, 65 fs near-infrared pulses (between 1.1-2.6 µm) are generated by directing the beam to a non-collinear optical parametric amplifier (NOPA). The beam diameter at the output of the NOPA is approximatively 1 cm (at  $1/e^2$ ) and the spatial mode quality factor ( $M^2$ ) is less than 1.3. In order to separate the signal (horizontally polarized) and idler (vertically polarized) beams, a LP-1500 nm longwave-pass filter (Spectrogon) and a polarizer were placed after the NOPA. The generated infrared beam is then focused with a 15 cm focal length off-axis parabolic mirror into a 1.5 cm long Tellurite glass sample. For the present study, we investigated the following glass composition: 80TeO<sub>2</sub>-10ZnO-10Na<sub>2</sub>O (molar %). The glass sample was fabricated by the conventional melt-quenching technique, starting from high purity 5 N raw materials: TeO<sub>2</sub>, ZnO and Na<sub>2</sub>CO<sub>3</sub>. The mixed batch was melted in a platinum crucible at 850 °C, during 2 h, in a furnace mounted on a water-free glove box, under dry air ([H<sub>2</sub>O] < 1 ppm vol). The glass melt was then quenched into a brass mold preheated at 220 °C, subsequently annealed at the vitreous transition temperature (Tg) for 8 h and finally slowly cooled down to room temperature. The sample was then polished to optical quality. In order to characterize the angular distribution of the output spectrum, an optical fiber (either in fused silica or in chalcogenide, depending on the spectral region to be measured) is placed on a rotation stage. Depending on the measured wavelength, the collected signal is then redirected into a spectrometer or a monochromator. The visible part of the spectrum (195-890 nm) is measured with the help of a USB4000 spectrometer (Ocean Optics) while the near-infrared part (890-2520 nm) is recorded with a NIRQUEST spectrometer (Ocean Optics). The midinfrared region (between 2.3 µm and 6 µm) is analyzed with the help of a home-made monochromator. For wavelengths greater than 3 µm, a high-pass filter is placed before the nitrogen cooled HgCdTe MCT detector (Hamamatsu) in order to avoid the spurious signal coming from the second-order diffraction of lower wavelengths. Finally, it was checked that the fiber used to collect the supercontinuum was not responsible for the spectral broadening of the pulse. It was done by inserting different optical densities between the Tellurite sample and the fiber used to collect the signal. Since the spectral shape did not depend on the power injected in the fiber, it excludes potential nonlinear effects that could occur in the latter to be responsible for the spectral broadening.

The connections between the different parts of the spectrum were done by equalizing the spectra recorded by each spectrometer in the overlapping detection range. Moreover, since the recorded signal decreases as the angle increases, the integration time is increased accordingly in order to keep a good detection

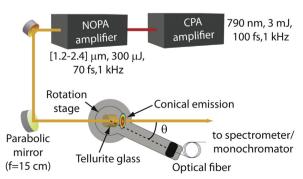
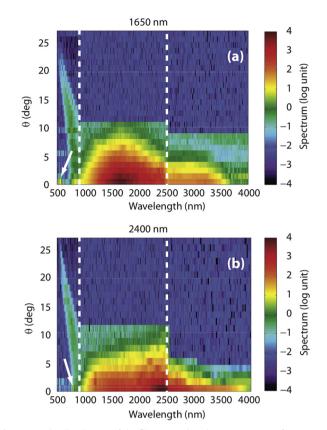


Fig. 1. Experimental setup for the angular characterization of the supercontinuum.



**Fig. 2.** Angular distribution of the filament-induced supercontinuum for a 1.65  $\mu$ m (a) and 2.4  $\mu$ m (b) pump wavelength. The white arrows highlight the third harmonic generated during the filamentation process. The dashed white lines delimit the spectral regions of the 3 different spectrometers used during the experiment.

dynamics over the whole angular measurements. The connection between each angle is then done by dividing the recorded signal by the integration time. The angular distributions of the supercontinuum obtained for a 1.65  $\mu$ m (normal dispersion regime) and for a 2.4  $\mu$ m (abnormal dispersion regime) pulse are shown in Fig. 2(a) and (b). For both cases, the energy was set to 30  $\mu$ J. The input energy has been chosen so as to maximize the supercontinuum generation while avoiding a permanent damage in the sample. The on-axis spectrum for the 1.65  $\mu$ m (2.4  $\mu$ m) pump pulse lies between [0.7–3.5  $\mu$ m] ([0.75–4  $\mu$ m]). Moreover, third harmonic is produced during the filamentation process as evidenced in Fig. 2. For both pump central wavelengths, a strong conical wave is produced in the short wavelength region [0.5–0.8  $\mu$ m]

In order to capture the full spatial and temporal dynamics of the pulse experiencing filamentation in Tellurite glass, numerical simulations were performed. Assuming a cylindrical symmetry around the propagation axis, the equation driving the propagation of a linearly polarized electric field envelope  $\varepsilon$  reads in the reciprocal space (the symbol tilde stands for the Fourier transform) [30]

$$\begin{split} \partial_z \tilde{\epsilon} &= i (k_z - \frac{\omega}{v_g}) \tilde{\epsilon} + \frac{\omega}{c^2 k_z} \left[ i \omega \left( n_2 | \epsilon|^2 \epsilon + \Delta n_r \epsilon \right) - \frac{e^2}{2 \epsilon_0 m_e} \zeta(\omega) \rho(\epsilon) \right] \\ &- L(\epsilon) - \frac{\alpha(\omega)}{2} \tilde{\epsilon} \end{split}$$

with  $n_2(\Delta n_r)$  the electronic (vibrational) nonlinear refractive index,  $v_g$  the group velocity, e ( $m_e$ ) the charge (effective mass) of the electron,  $k_z = \sqrt{k^2(\omega) - k_\perp^2}$  with  $k(\omega)$  the wave vector and  $k_\perp$  its transversal component,  $\zeta(\omega) = (\nu_{en} + i\omega)/(\nu_{en}^2 + \omega^2)$ , where  $\nu_{en}$  is the

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