Supercontinuum Generation in Barium Fluoride Using Bessel Beams

Krithika Dota, ^{1,2} Jayashree A. Dharmadhikari, ² Deepak Mathur, ^{1,2} and Aditya K. Dharmadhikari ¹

¹ Tata Institute of Fundamental Research, 1 Homi Bhabha Road, Mumbai 400 005, India ² Centre for Atomic and Molecular Physics, Manipal University, Manipal 576 104, India (Received August 20, 2013)

We investigate supercontinuum generation using femtosecond duration pulses in a 15 mm long barium fluoride crystal using an axicon-generated Bessel beam. Spectral profiles of the generated supercontinuum are measured using axicon lenses of different cone angles, and with two different incident beam diameters. Our measurements reveal that the spectral profile is, indeed, dependent on the distance between the crystal and the tip of axicon, and also on incident laser power. We observe both modulated and smooth spectral profiles as the axicon cone angle is changed. Our experimental observations manifest the variation of the input laser intensity along the propagation axis in the crystal. The spectrum is found to be smoothest when the crystal is kept in physical contact with the tip of the axicon. The spectral extent is broadest for the highest axicon cone angle.

DOI: 10.6122/CJP.52.431 PACS numbers: 42.25.Bs, 42.65.Jx, 52.38.Hb, 42.65.Sf

I. INTRODUCTION

Femtosecond supercontinuum generation is a visually spectacular effect and has been of interest for the past three decades [1, 2] and pioneering contributions have been made in the areas of femtosecond supercontinuum generation and filamentation nonlinear optics by Chin and coworkers [2]. The supercontinuum manifests itself in the light transmitted through a medium as a white disk surrounded by a distinct, concentric, rainbow-like conical emission, the low-divergence central part of which has come to be called the whitelight continuum or supercontinuum. There exist various mechanisms that seek to rationalize supercontinuum generation (SG): self-phase modulation [3]; ionization-enhanced SPM [3, 5], with additional contributions from the interplay of diffraction and instantaneous electronic Kerr nonlinearity [6]; stimulated Raman scattering; self-steepening; four-wave parametric processes; shock wave, group velocity dispersion, chromatic dispersion and band gap [7– 11]. All such studies have considered the use of a Gaussian profile of the laser beam that is focused using a spherical lens. A Bessel beam, on the other hand, has a non-diffracting property that makes it suitable for longer distance propagation of optical energy than is possible with Gaussian or near-Gaussian beams. This affords an obvious advantage: it allows the Bessel beam to maintain a tight focus over distances larger than the conventional Rayleigh range. Another property that is important for propagation of femtosecond laser pulses is that the beam is "self-healing" in the sense that if the beam is partially obstructed it can re-form at a different location further along its propagation path [12–15]. Bessel beams (or axicon beams) have attracted the attention of both theoreticians and experimentalists [16, 17] for a very long time but it is only relatively recently that the relative simplicity of using axicons to generate Bessel beams has begun to be practically appreciated, opening new opportunities for their application in parametric down conversion [18], waveguide writing [19], femtosecond laser filamentation [20], and stimulated Raman scattering [21]. Bessel beam filamentation in air and liquid has begun to be studied [22–26] and it has been demonstrated that the on-axis intensity distribution can be simply controlled by (i) changing the distance between the axicon and the sample under study, (ii) altering the bluntness of the axicon tip, and (iii) selecting the input beam size [27]. Theoretical simulations carried out to investigate dispersion-induced distortions of field distribution along longitudinal and transverse direction in a femtosecond laser beam generated using an axicon lens [28] have shown that dispersion increases laser pulse duration in the course of its propagation. A numerical study in K108 glass, focusing on supercontinuum conical emission, has predicted that continuum conical emission first decomposes into speckles, followed by splitting into narrow rings as the pulse energy is increased [29].

In earlier work carried out in our laboratory with Gaussian beams of ~ 45 fs duration [30] we have demonstrated very efficient whitelight generation in BaF₂ and have reported results of systematic studies carried out under varying experimental conditions, such as laser energy, pulse duration, polarization, and external focusing [31]. The rate of change in electron densities within BaF₂ have also been obtained from such studies [32]. We have experimentally shown excercising control over the filamentation dynamics within a large BaF₂ crystal [33], and estimated six-photon absorption cross-section [34]. All the above mentioned work was carried out using Gaussian beam focused by a spherical lens. In the following we report results of experiments that we have conducted on supercontinuum generation using axicon lenses. Despite the above-noted resurgence of interest in axicon-related studies, there continues to be a dearth of experimental data on axicon-generated supercontinuum generation in condensed media. In recent work [35] we carried out experiments that revealed the following aspects of axicon based filamentation: i) a higher incident laser energy is required for a filament as the axicon cone angles increases; (ii) the filament size is independent of whether an axicon or a spherical lens is used; and (iii) the distance between the tip of the axicon and the BaF₂ crystal is a crucial parameter that quantifies changes in periodicity of focusing-refocusing events within the crystal. In the following we extend these studies and report on the systematics of supercontinuum generation using femtosecond duration pulses in a 15 mm long barium fluoride crystal focused by an axicon-generated Bessel beam, including measurements of the spectral profile of the supercontinuum we generate using axicon lenses of different cone angles, using two different incident beam diameters. We present our results in the form of both images as well as spectra.

II. EXPERIMENTAL METHOD

Figure 1 is a schematic depiction of our experimental set-up used to investigate supercontinuum generation in a BaF₂ crystal (15 mm long) using axicon lenses of different

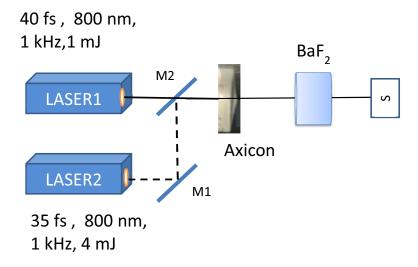


FIG. 1: (Color online) Schematic representation of our experimental set-up. M1, M2: dielectric mirrors; S: fibre-coupled spectrometer.

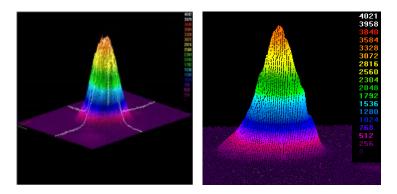
cone angles.

We utilized two seperate Ti-sapphire lasers (Laser 1: 800 nm wavelength, 1 kHz repetition rate, with 7 mm diameter beam, 1 mJ energy and 40 fs pulse duration; Laser 2: 800 nm wavelength, 1 kHz repetition rate, with 10 mm diameter beam, 4 mJ energy and 35 fs pulse duration).

The beam from one or the other of these lasers was made incident on an axicon lens (we used different lenses with cone angles varying from 178° to 170°). Figure 2 shows the beam profile of the two lasers along with the beam profile of the Bessel beam generated using a 178° axicon; the profile has been measured using a CCD-based beam profiler. The Bessel beam profile is found to be identical for two incident laser beams (Laser 1 and Laser 2); we show only the beam profile obtained using Laser 2 in Fig. 2. The Bessel beam thus characterized was propagated through BaF₂ such that supercontinuum could be generated and measured. We employed a fibre-coupled spectrometer (Ocean Optics USB 2000) to quantify the spectral extent of the supercontinuum emerging from the crystal. Far-field images of the whitelight and conical emission obtained under different conditions were captured using a CCD camera coupled to a computer.

III. RESULTS AND DISCUSSION

We probed supercontinuum generation using axicon lenses with different cone angles in BaF_2 with both the lasers (7 mm and 10 mm beam diameter). We made two types of measurements: one in which the incident input energy was kept constant and the distance between the axicon and the crystal was varied, and another in which the latter distance was



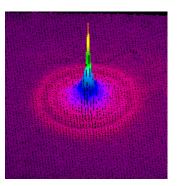


FIG. 2: (Color online) Beam profiles of Laser 1 (left, top panel) and Laser 2 (right, top panel). The lower panel shows the beam profile of the Bessel-like beam generated 9 cm from the tip of an axicon with 178° cone angle.

fixed and the input energy was varied. Figure 3 (top panel) shows typical supercontinuum spectra of the central portion of the transmitted beam; this was obtained as a function of distance using an axicon with 170° cone angle at a fixed input energy of 90 μ J. As the distance between the axicon and the crystal increases (5.5 cm to 9.5 cm), there is an increase in spectral width, particularly towards the blue side of the spectrum. Moreover, the axicon-generated supercontinuum spectrum begins to exhibit distinct peaks: the spectrum appears to be highly modulated, in contrast to the smooth continuum that is generally obtained using a spherical lens [36]. In the context of Gaussian laser beams, modulated axial supercontinuum has also been observed in sapphire [37] and water (along with the stimulated Raman scattering) [21]. Modulations are also observed in the spectrum when the supercontinuum is generated from a hollow fibre; these are usually attributed to a laser pulse undergoing self-phase modulation in the leaky modes (higher order modes) of the hollow fibre [38, 39]. In case of Bessel beam propagation a similar analogy may be made to rationalize the modulations in the spectra that we observe. Recently using a

short length sapphire crystal Majus and Dubietis [40] have demonstrated modulation-free supercontinuum using an axicon. This is attributed to the fact that the 3 mm crystal experiences no variation in the intensity of the central spot. We show an entirely different method to generate modulation free supercontinuum in a long sample using low cone angle axicon and low incident energy.

The middle panel of Fig. 3 depicts the supercontinuum spectra we obtained using a 175° cone angle axicon. Even though the extent of the supercontinuum is somewhat reduced, it is interesting to note the absence of modulations in the spectrum. Similarly in the case of 178° cone angle axicon the modulations are found to be completely absent. The intensity variation in the central spot is constant over a distance much larger than the crystal size. To ascertain this fact we have measured supercontinuum yield as a function of distance from the tip of the axicon. Figure 4 shows how the total supercontinuum yield depends on incident laser beam diameter and on axicon cone angle. Here the yield was taken to be the area under the supercontinuum spectrum over the wavelength range 600-900 nm (normalized, in each case, to the peak at 812 nm). It is clear that supercontinuum yield is constant as the crystal is translated along the laser propagation direction. Moreover, it appears that the axicon-generated supercontinuum depends on the input beam diameter that decides the depth of focus. For the 1 cm beam the depth of focus is larger than for the 7 mm beam. As is seen, for 1 cm beam diameter with 175° cone angle, we observe nearly constant supercontinuum generation up to a distance of 10 cm. The yield of supercontinuum generation is more in the case of the 7 mm beam compared to the 10 mm beam even though the incident energy is less.

We now discuss results obtained when the axicon is touching the crystal. Here, the supercontinuum is measured as a function of incident energy. Figure 5 shows the supercontinuum spectra obtained using cone angles of 170°, 175°, and 178°, with the barium fluoride crystal being kept in physical contact with the tip of the axicon in each case. The incident energy was varied from 27 to 340 μ J. At a fixed energy of 200 μ J the spectra are compared for the three axicons when the crystal was touching the tip of the axicon. The supercontinuum extent is seen to be distinctly wider in the case of 170° cone angle axicon compared to what is obtained with the other two axicon cone angles. Our results (Figs. 3, 5) are consistent with earlier observations in water [21]. Two transition regions were identified in this case: one is the "smooth" case in which the water sample touched the axicon tip and the second "sharp" case in which the sample is at some distance from the axicon tip. In the former, a transition occurred from a linear to a nonlinear regime in smooth fashion whereas in the latter the transition occurred abruptly. The sharp regime has been attributed to collapse of the Bessel beam initiated by a Kerr-driven (four wave mixing) process [41] which gives rise to localized enhancement of peak laser intensity that, in turn, results in relaxation of the condition for transverse momentum conservation.

An image of the generated supercontinuum is shown in Fig. 6 in which the left panel shows the image when a 170° cone angle axicon was used. Here, the central disc is the supercontinuum and the surrounding rings are the conical emission. Contrast this with the measured spectra of the central disc shown in Figs. 3 and 4.

In summary, we have carried out a systematic investigation of supercontinuum gen-

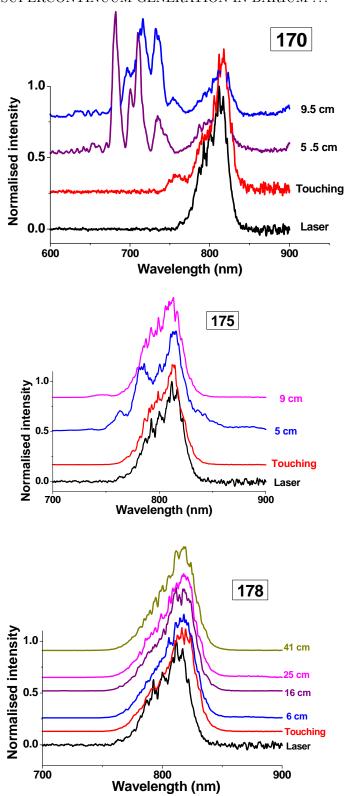


FIG. 3: (Color online) Supercontinuum generation at different distances between the BaF_2 crystal and the axicon tip, for cone angles of 170° , 175° , and 178° .

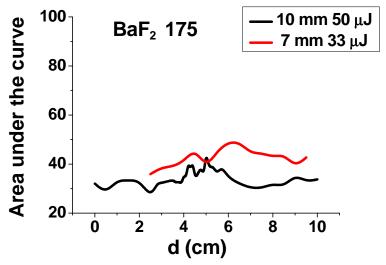


FIG. 4: (Color online) Supercontinuum yield at different distances (d) from the axicon tip obtained with beams from Laser 1 (red) and Laser 2 (black) for an axicon cone angle of 175°.

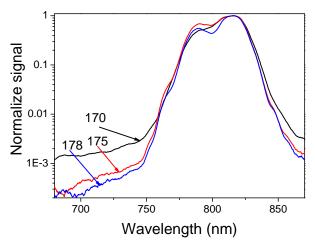


FIG. 5: (Color online) Whitelight spectra obtained using three different axicon cone angles at a fixed incident laser energy of 200 μ J with the BaF₂ crystal touching the axicon tip. Note the smoothness of the spectra (see text).

eration in a crystal of barium fluoride with femtosecond duration pulses using axicon lenses with different cone angles. Our results are presented in terms of images as well as spectra. With higher cone angle we observed modulations in the supercontinuum spectra. The input beam size is also found to have relevance in the yield of supercontinuum generation. By placing the barium fluoride crystal in physical contact with the tip of the axicon, we observed a smooth spectrum, with spectral extent being broader for higher cone angles.

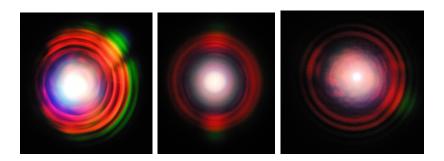


FIG. 6: (Color online) Far-field images of the supercontinuum and conical emission obtained using cone angles of 170° (left image), 175° (centre image), and 178° (right image).

IV. ACKNOWLEDGMENTS

JAD thanks the Department of Science and Technology for assistance under the Women Scientists Scheme while DM acknowledges generous support as a J C Bose National Fellow.

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