Orientation-dependent depolarization of supercontinuum in BaF_2 crystal*

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We present a systematic investigation of the depolarization properties of a supercontinuum accompanied with femtosecond laser filamentation in barium fluoride (BaF_2) crystal. It is found that the depolarization of the supercontinuum depends strongly on the crystal orientations with respect to the incident laser polarization. At most crystal orientations, the depolarization of the supercontinuum rises with the increase of the input laser energies and finally saturates. While at 45°, the depolarization of the supercontinuum is not changed and keeps nearly negligible with the increase of the input laser energies. These peculiar depolarization properties of the supercontinuum can be ascribed to the orientation dependence of the cross-polarized wave (XPW) generation and ionization-induced plasma scattering in the BaF_2 crystal.

Keywords: femtosecond laser filamentation, supercontinuum, depolarization

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1. Introduction

One of the most spectacular and visual effects generated by the nonlinear propagation of intense femtosecond laser pulses in a transparent medium is dramatic broadening of the spectrum, termed as supercontinuum generation.[1,2] The wavelength of the supercontinuum ranges from the UV to the mid-infrared.[3–5] Therefore, the supercontinuum has a wide range of potential applications in various research fields, such as femtosecond time-resolved spectroscopy,[6] optical pulse compression,[7] optical parametric amplification,[8] and broadband spectrum light detecting and ranging (LIDAR).[9] The physical mechanism of supercontinuum generation in transparent media could be understood in the framework of femtosecond filamentation[2] which stems from the dynamic balance between the self-focusing induced by the optical Kerr effect and the defocusing induced by the strong-field generated plasma.[10] With this picture, many effects, such as the self-phase modulation (SPM),[11,12] self-steepening,[13,14] space–time focusing,[15] four-wave mixing,[16,17] ionization-enhanced SPM,[18] and group velocity dispersion,[19] can contribute to the supercontinuum generation. By now, many works have been paid to generate wider supercontinuum spectrum.[19–21] According to the standard scenario of filamentation, it is generally believed that the spectral extent of the supercontinuum is limited by the maximum intensity in the filament and broader supercontinuum spectra can be generated with a wider band gap material.[20,21] Meanwhile, the chromatic dispersion of the medium is also believed to be a major contributor to the spectral extent of the supercontinuum.[19]

Apart from concentrating on generating broader supercontinuum spectra, much attention has also been paid to the polarization properties of the supercontinuum due to its important role in spectroscopic applications. Usually, when the supercontinuum is generated in isotropic amorphous media, the polarization is believed to be the same as that of the incident laser.[22–24] However, when intense laser pulses propagate in BK7 glass, it was found that the polarization of the generated supercontinuum could be different from that of the incident laser,[25] which was described as the depolarization of the supercontinuum. In that work, the depolarization of the supercontinuum was attributed to the scattering of the generated plasma which was introduced in Ref. [26]. This viewpoint has also been adopted by some other groups.[2,27] In contrast, Yu *et al.* recently found that the polarization of the supercontinuum could also be changed by the initial polarization perturbation induced by the focus lens and this depolarization could be magnified by the cross-phase modulation of the nonlinear third-order polarization.[28] Meanwhile, relevant studies have also been extended to crystalline media.[27,29] Compared to the isotropic amorphous media, these materials possess orientation-dependent linear and nonlinear properties.[30] Nevertheless, it is expected that the depolarization of the su-

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percontinuum could be observed due to the scattering of the generated plasma, even when the incident laser is polarized along the crystal axis for which both of the birefringence effect and the cross-phase modulation of two perpendicular components of the incident light are absent. However, under such a situation, Buchvarov *et al.* did not observe the depolarization and thus excluded its mechanism of scattering of the generated plasma.[31] Therefore, whether the generated plasma plays an indispensable role in the depolarization of the supercontinuum is still under controversial and more experimental investigations on the polarization properties of the supercontinuum are quite necessary.

In this paper, we study the depolarization properties of the supercontinuum accompanied with femtosecond laser filamentation in the cubic BaF_2 crystal at various input laser energies and crystal orientations. It is found that the depolarization of the supercontinuum is strongly dependent on the crystal orientations. At the same time, the depolarization of the supercontinuum changes with the increase of the input laser energies due to the scattering of the generated plasma and finally saturates due to the intensity clamping effect at most crystal orientations. Moreover, a linearly polarized supercontinuum which has the same polarization of the incident laser can be obtained even at high input laser energies when the crystal orientation is chosen as 45° in BaF_2 crystal. Our experimental results reveal the critical role of the ionization-induced plasma in the depolarization properties of the supercontinuum accompanied with femtosecond laser filamentation in cubic crystals.

2. Experimental setup

Our experimental setup is depicted in Fig. 1. The laser system (Coherent, Legend HE+) consists of a broadband femtosecond oscillator and a regenerative amplifier which delivers 35 fs pulses with a maximal output energy of 5.0 mJ, a central wavelength of 800 nm, and a repetition rate of 1 kHz. A beam splitter is used to reflect a part of the laser pulse energy which is then finely adjusted by means of a broadband achromatic half-wave plate followed by a Glan–Taylor polarizer 1. The polarization direction of polarizer 1 is along the x axis. The first focusing lens ensures that the peak intensity is high enough to generate filament in the sample. Near the focus of the lens ($f = 50$ cm), a 1.5 mm thick BaF_2 crystal sample is mounted on a translation stage which can move along the direction perpendicular to the laser pulse propagation direction. The BaF_2 crystal is [001] cut. β is the angle between the x axis and the crystallographic axis [100]. Just after the sample, a second focusing lens is used to collimate the light before passing through the polarizer 2. Once the supercontinuum is generated in the medium, the polarizer 2 could be applied to select the parallel or perpendicular components of the whole supercontinuum spectra before entering an energy meter. The depolarization could be quantified by the extinction ratio (ER), which is the ratio of the intensity $I_\perp$ (measured when the polarizer 2 is at the perpendicular position to the polarizer 1) to the intensity $I_\parallel$ (measured when the polarizer 2 is at the parallel position to the polarizer 1). The ER is measured to be less than 10^{-5} without the sample when the polarizer 2 is perpendicular to the polarizer 1.

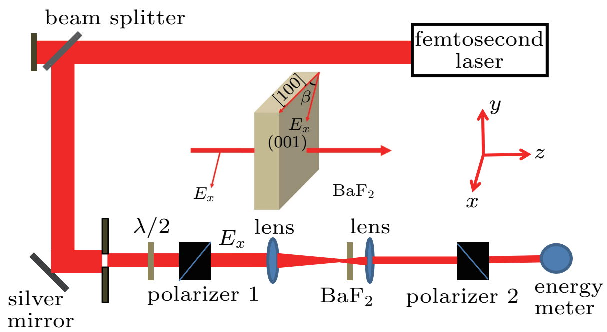

Fig. 1. A schemetic view of our experimental setup.

3. Results and discussions

Since we aim at studying the relationship between the depolarization property and the crystal orientation, it is quite necessary to calibrate the crystal orientation in advance. In our work, the crystal orientation of BaF_2 has been precisely calibrated with the cross-polarized wave (XPW) generation process.[32] After the crystal angle β is calibrated, the ERs with the changes of the input laser energies and under various crystal orientations are measured and the results are shown in Fig. 2. It is obvious that the ERs are dependent on the input laser energies and the crystal orientations. With the increase of the input laser energies, the ERs begin to increase and saturate finally at most crystal orientation angles, such as 0°, 10°, 20°, and 35°. However, at 45°, the ERs are almost not changed with the increase of the input laser energies and the ERs are about zero all the time. In the following, we will show that these peculiar depolarization properties can be ascribed to the orientation dependence of the cross-polarized wave generation and strong-filed-ionization induced plasma scattering in the BaF_2 crystal.

Firstly, we focus on the case of 0° in Fig. 2. Note that there is no XPW generation in the BaF_2 crystal under this condition.[32] When the input laser energy is low (< 6 μJ), the ER indeed nearly equals zero due to the negligible XPW generation. Once the filament is generated when the incident laser pulse energy is beyond 12 μJ, it can be seen that the ER increases dramatically and finally saturates. It is worth to note that a similar phenomenon has been observed in BK7[25] and the scattering of the plasma generated by the strong field ionization in the crystal degrades the laser polarization. With the increase of the incident laser energy, the plasma density will increase and finally saturate due to the intensity clamping effect during filamentation in the sample,[10,33] leading to the enhancement and saturation of the laser depolarization. Therefore, in our experiments, the depolarization of the supercontinuum at 0° may also be ascribed to the scattering of the generated plasma and the saturation of the ER may be due to the saturation of the plasma density.

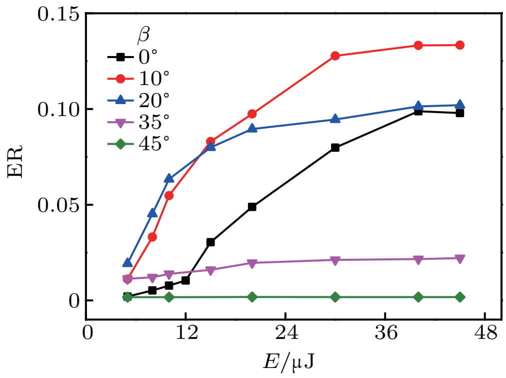

Fig. 2. ERs of the supercontinuum generated in BaF_2 crystal at different input laser energies E and under different crystal orientations β.

In order to further confirm that the density of the generated plasma is indeed saturated, we have also measured the spectra of the supercontinuum at 0°. Different from the experimental setup shown in Fig. 1, the second polarizer is replaced by an integrating sphere which is attached to two fiber-optic-coupled spectrometers QE65 Pro (Ocean Optics, wavelength range 350–1100 nm) and Maya2000 Pro (Ocean Optics, wavelength range 190–410 nm) with two fibers (Ocean Optics, QP-600-2-vis-nir and QP-600-2-XSR). The integration time employed in the measurements is 100 ms for QE6500 Pro and 10 s for Maya2000 Pro. The measured spectra are shown in Fig. 3. For lower energies, such as 5 μJ and 10 μJ in Fig. 3(a), the spectra intensity rises with the increase of the incident energy. Note that a logarithmic coordinate is used in this figure. At the same time, the spectra are broadened towards lower and higher frequencies symmetrically. As the input energy increases, i.e., from 20 μJ to 45 μJ in Fig. 3(a), the spectra intensity also rises and the spectra become gradually asymmetrical with a more significant broadening on the blue side than that on the red side. These findings can be explained as follows. When the incident pulse energies are low, i.e., 5 μJ and 10 μJ in our experiments, the spectra broadening is mainly due to the self-phase modulation and the plasma generation from multiphoton ionization can be neglected. The self-phase modulation leads to a symmetrically broadening of the spectra towards the blue-shift side and red-shift side, as shown in Fig. 3(a). In contrast, at high incident pulse energies, i.e., from 20 μJ to 45 μJ in our experiments, the plasma generation is expected to become the dominant effect for spectra broadening, since it has a much higher power dependence on the peak intensity. Moreover, the plasma generated only causes a blue-shift of the spectra. Therefore, the supercontinuum spectra become asymmetrical with a larger shift on the blue side than the red side. Attention should be paid to Fig. 3(b), at lower input laser energies such as 5 μJ and 10 μJ, the spectra intensity below 400 nm is nearly zero because the filament is not generated. At higher input laser energies, the spectra intensity below 400 nm becomes significant. At the same time, the cut-off of the spectra seems to be blue shifted with the increase of the laser energies until 40 μJ. At 45 μJ, the cut-off is no longer blue shifted, which means that the peak intensity is clamped in the sample.[33] This intensity clamping phenomenon is the results of dynamic balance between the self-focusing induced by the optical Kerr effect and the defocusing by the self-generated plasma, and indicates the saturation of the plasma density in the sample. Therefore, both the appearance of the supercontinuum spectra cut-off in Fig. 3 and the saturation of the depolarization of the supercontinuum shown in Fig. 2 suggest the saturation of the plasma density in the sample.

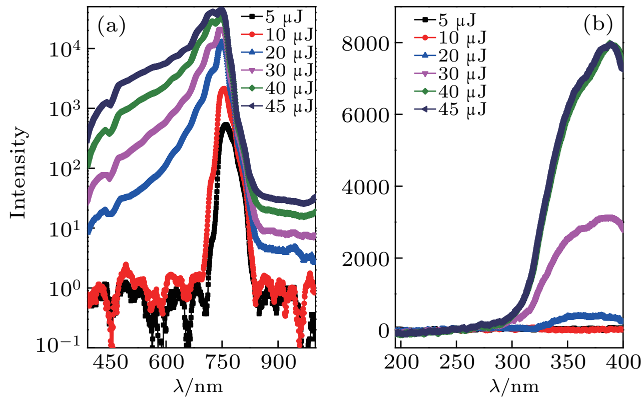

Fig. 3. The spectra of the supercontinuum generated in BaF_2 crystal at 0° with different input laser pulse energies. The spectra in (a) and (b) are measured with fiber optic spectrometers QE6500 and Maya2000, respectively.

Secondly, we pay attention to the situation at another special orientation angle, i.e., 45°. Note that the XPW efficiencies at 45° and 0° are very similar,[32] however, our work finds that the ER (green line in Fig. 2) at 45° is very different from that at 0° and remains nearly negligible all the time with the increase of the input laser energies. This finding suggests that, besides XPW generation, other angular dependent nonlinear effects should also play a significant role in determining the depolarization property.

In fact, the rate of strong field ionization in crystal is angle-dependent and according to the tunneling ionization theory in solids, the ionization rate can be written as[34]

$$W(I) \propto \exp\left(-\frac{(2\Delta)^{3/2}\sqrt{m^*}}{3e\hbar E}\right), \tag{1}$$

where E is the laser electric field amplitude, Δ is the band gap, and m^* is the effective mass of the electron and hole. In

BaF_2 crystal, the band gap Δ equals to 9.1 eV. According to formula (1), one sees that the tunnel ionization in solids depends exponentially on the effective mass. The effective mass under different crystal orientations is calculated using density functional theory (DFT) by Vienna *ab-inito* simulation package (VASP)[35] and the calculated results are shown in Fig. 4. The effective mass exhibits a modulation period of $\pi/2$ due to the fourfold symmetry of the BaF_2 crystal and is maximal at 45° in a period. Due to the angular dependence of the effective mass, the tunnel ionization rate in solids is also angle-dependent, which has been found to lead to a lot of interesting phenomena in crystals.[36–38] In our experiment, when the incident laser energy is constant, the larger the effective mass is, the less plasma will be generated.[35,36] Thus, the amount of generated plasma at 45° is much less than that at any other degrees and remains at a low level. Meanwhile, the XPW generation effect can be neglected at 45°. As a result, the ER keeps almost unchanged with the increase of the input laser energies at 45° and nearly equals zero all the time. For this special property, a linearly polarized supercontinuum can be obtained when linearly polarized intense laser pulses propagate in BaF_2 crystal at an orientation angle of 45°. This polarization property of the supercontinuum is very useful for femtosecond time-resolved spectroscopy.

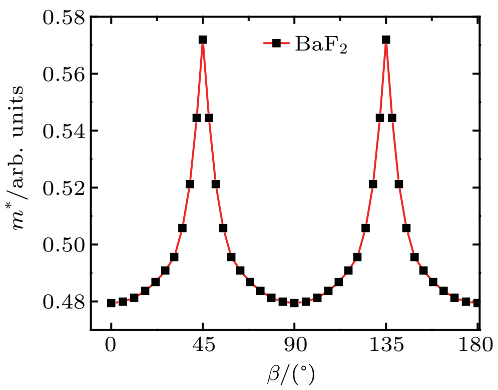

Fig. 4. Angular dependence of effective mass in BaF_2 crystal.

Here we would like to mention again the behavior of ER for the case of 0°, at which XPW generation is also absent. However, in this case, the ER curve changes dramatically with the incident laser energy, which can be ascribed to the most pronounced plasma scattering effect related to the smallest effective mass at 0°. Specifically, when the energy is below the critical energy ($\sim$ 12 μJ) of filamentation, the ERs at 0° are always small due to lack of XPW generation and low plasma density. However, when the input laser energy is above 12 μJ, the ERs increase dramatically because of the much dense plasma within the filament. This will lead to the appearance of an energy threshold of the ER as shown in Fig. 2 (black line). The pronounced difference of ER curves between 0° and 45° provides an additional evidence that the plasma generated by the strong field ionization plays a critical role on the depolarization property of the supercontinuum.

Thirdly, at other crystal orientation angles such as 10°, 20°, and 35°, both the XPW generation and the plasma scattering effects have to be taken into account, and the relative contribution to the depolarization is dependent on both the laser energy and orientation angle. For example, when the input laser energy is low ($<$ 12 μJ), the ionization-induced plasma density in the sample is very low and the XPW generation may be the main effect which leads to the depolarization of the supercontinuum. Therefore, the ER at 20° is much larger than that at other degrees, as shown in Fig. 2. When the input laser energy is further increased ($>$ 12 μJ), compared with the situation of 20° (blue line in Fig. 2), the ER at 10° (red line in Fig. 2) increases quickly and becomes larger. This may be due to the exponential dependence of the ionization rate on the effective mass, thus the plasma generated at 10° is much more than that at 20°, which leads to a much larger saturated ER. For the same reasons, the small ER at 35° under different input laser energies may also be attributed to the less XPW generation and plasma scattering than other orientation angles such as 10° and 20°.

4. Conclusion

We have investigated the depolarization properties of a femtosecond-laser-induced supercontinuum generated in BaF_2 crystal at different incident laser energies and under various crystal orientations with respect to the laser polarization. In our experiments, it is shown that the depolarization of the supercontinuum rises with the increase of the input laser energies and finally saturates at most crystal orientations. In contrast, a linearly polarized supercontinuum which has the same polarization of the incident laser can be obtained even at high input laser energies when the crystal orientation is 45°. These orientation-dependent depolarization of the supercontinnum can be attributed to the interplay of the orientation-dependent XPW generation and strong-field-ionization induced plasma scattering in BaF_2 crystal. Our work reveals the critical role of the generated plasma in the depolarization properties of the supercontinuum and provides a more comprehensive understanding of the depolarization of the supercontinuum during femtosecond laser filamentation in transparent crystals.

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