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Control of supercontinuum generation with polarization of incident laser pulses

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ABSTRACT Supercontinuum generation is dependent on the polarization state of the incident laser. The polarization of the generated supercontinuum is the same as that of the incident laser. The magnitude of the generated supercontinuum depends on the polarization of the incident laser and increases as the polarization changes from circular to linear, irrespective of the nature of the sample, be it isotropic, anisotropic, or chiral. In all samples, the polarization dependence indicates a preference for the linear component of the incident laser beam. The anisotropic sample shows an additional difference in the generated supercontinuum for the two perpendicular directions of the incident laser polarization.

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1 Introduction

In recent years, the development of stable ultrashort pulsed laser sources with high peak powers has resulted in a lot of interest in the area of the extreme nonlinear phenomenon of supercontinuum generation (SG) [1–15]. SG occurs when the spectral bandwidth of the laser pulse increases drastically as it passes through a medium and remains coherent. The process of SG using short laser pulses ($\leq 10^{-12}$ s) has found extensive use in spectroscopy, ultrafast pulse generation, compression, and amplification. There are also situations in which SG can be a nuisance, such as in coherent optical communications, where it distorts channel signals [16]. One simple solution to such a complication is to reduce the intensity of the incident laser so that it is below the threshold for SG. However, it is usually not possible to do so as the minimum intensity necessary for signal transmission through a useful fiber length is often enough to result in SG [17]. Thus, it is important have a scheme to control SG without compromising the peak intensity of the incident laser pulse.

To control the SG process, a clear understanding of its mechanism is necessary. Although a lot of research has gone into understanding SG, a comprehensive theory is yet to emerge. SG is said to arise from several nonlinear optical effects, such as self- and cross-phase modulation, four-wave mixing (FWM), stimulated Raman scattering, multi photon excitation (MPE), and many others [1, 2, 5, 7–11, 13]. In the condensed phase, one study also evokes the band gap of the material as a necessary condition for SG [15]. Our earlier study [17] of the intensity dependence of the SG process over two orders of magnitude of the incident laser pulse has shown that the rate of suppression of SG is intensity dependent. The ellipticity dependence of the threshold and of the efficiency of SG in isotropic samples, as discussed in [17], offer a simple way of controlling SG. Our systematic polarization-dependent experiments, as presented here, show that SG is highly suppressed when circularly polarized light is used. A recent theory has examined such polarization dependence of nonlinear processes, linked it to a difference in the self-focusing of light in different polarization states, and proposed a model based on propagational differences between the linear and circular polarizations [18]. It was theoretically predicted in 1966 that the ionization potential for circularly polarized light is much higher than that for linearly polarized light [19]. However, the fact that differences in linear physical properties with light polarization can correspond to differences in higher order nonlinear processes with multiply competing mechanisms is intriguing.

Our previous experiments were performed on a 10-Hz sub-picosecond amplified laser system [17], which had poor shot-to-shot stability. Careful shot-to-shot measurements with pulse discrimination and rejection were required to maximize the signal-to-noise ratio so that the effect of polarization on SG and the threshold levels could be measured. In this paper, we present the results from our kHz amplified 50-fs laser system, which has a noise level below 2%. This allows much more stable and accurate data collection with averaging capabilities. We have chosen a range of samples from liquid and solid states, depending on their characteristics of being isotropic or anisotropic, and have made a comparison between the isotropic and anisotropic cases. We also present preliminary SG results for a chiral molecule 2R-butanol that were collected with the 10-Hz laser. In spite of the lower signal-to-noise ratio available from the 10-Hz laser, such preliminary data show that circular polarization-induced suppression continues to hold. Further investigations on chiral samples are currently in progress with the kHz laser system, which produces improved signal-to-noise data.

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The most important part in this paper, however, is our observation that the ellipticity dependence of SG follows the linear polarization component and is independent of the nature of the sample. This could happen as a result of the complex nature of the nonlinear processes involved in SG.

2 Experimental

In our experimental setup (Fig. 1) we used linearly polarized incident pulses from a commercial chirped-pulse multipass amplified Ti:Sapphire laser (Odin, Quantronix Inc.) operating at 50 fs, 800 nm, and a 1-kHz repetition rate. Using an optical wedged flat, 4% of the 1-mJ/pulse laser pulses were focused with a 50-cm lens into a 1-cm cuvette of sample that was placed about 6 cm before the focus. With this arrangement a spot-size of 0.6 mm at the cuvette gave an intensity on the order 10^{10} W/cm^2. This produced a stable supercontinuum from the liquids in the cuvette or the solid sapphire sample. A quarter-wave plate was used to vary the incident laser polarization from linear through elliptical to circular and vice-versa. Though the peak laser powers (> 100 MW) used in these experiments were above the self-focusing thresholds for the samples used, the large spot-size at the sample surface ensured that SG did not occur until the self-focusing process brought down the spot-size inside the sample to the threshold intensities. We made sure that there was no SG from the cuvette alone or in air at the focal point of the laser without the cuvette.

We found that the output polarization of the generated supercontinuum was the same as the input laser polarization for both the linear and circular cases, by using a Glan-Laser polarizer with an effective range of 400–1000 nm. We also used the Glan-Laser polarizer to measure the linear component of the incident laser beam at various positions of the $\lambda/4$ plate without any sample. We collected the entire supercontinuum using lenses, either into a monochromator (Acton Reseach Inc.) coupled to a CCD (Roper-Scientific Inc.) or into a pulse photodiode, PD (ThorLab DET-210), with the residual fundamental minimized by spatial filters. For PD detection, we cut off the fundamental and the IR light completely with the help of two 720-nm cut-off short-pass filter. While monochromator detection is polarization sensitive due to the gratings, the response of the PD has no such dependence. We did not make any polarization-related measurements through the monochromator.

After establishing the overall spectral characteristics of the SG from different samples, the total supercontinuum was measured as a function of the incident laser polarization, which was varied with the help of a first-order $\lambda/4$ plate. We also used a $\lambda/2$ plate to ascertain the effect of anisotropy on SG for sapphire. There was no such effect on the isotropic sample of acetone.

In the case of 2R-butanol, for which the experiments were carried out on a 10-Hz laser system, a dual photodiode scheme was used, one to measure the SG and the other to measure the pulse-to-pulse fluctuations of the incident laser pulse. This scheme enabled us to distill out useful data over the error-bars for the 10-Hz sub-picosecond amplified laser system, which has large pulse-to-pulse fluctuations. Care was taken to keep the intensity of the incident laser at the 1-mm quartz cuvette at 10^{10}W/cm^2 when the ellipticity-dependent measurements were taken with the help of a first-order $\lambda/4$ plate.

3 Results and discussion

The supercontinuum generated from several samples that we used, such as water, acetone, methanol, sapphire, and fused silica, had similar overall spectral characteristics and we represent a couple of them in Fig. 2 with respect to the incident amplified laser beam spectrum. It is evident from Fig. 2 that the overall spectral content of the generated supercontinuum was wider than our detection capabilities with our CCD-coupled monochromator. We checked that the spectral content of supercontinuum did not change with changes in the incident laser polarization. Since spectral measurements with monochromators are polarization dependent, we did not use them for any quantitative purpose. We chose optically transparent samples to avoid self-absorption of SG by the generation medium itself. The polarization dependence of SG was obtained by making integrated measurements with the

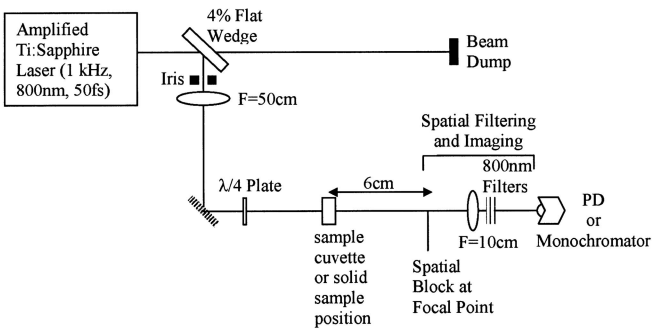

FIGURE 1 Experimental SG setup showing both of the data collection techniques: spectra measurements and total SG intensity measurements through the photodiode PD. A focussing lens ($F = 50$ cm) generated the supercontinuum, which was collected through a $F = 10$ cm lens. The cuvette was placed 6 cm before the focus. The polarization was changed with the help of the $\lambda/4$ plate. For the polarization-component measurements of the incident beam, a Glan-Laser polarizer replaced the sample to act as an analyzer, while for the anisotropic experimental measurements, the $\lambda/4$ plate was replaced by a $\lambda/2$ plate

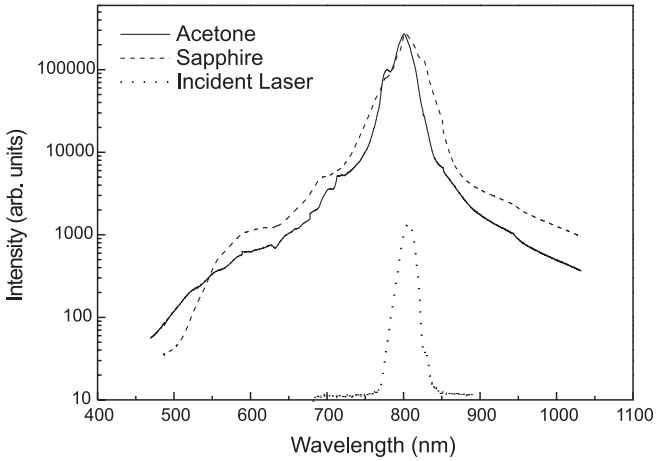

FIGURE 2 Continuum generated in acetone and sapphire at identical input intensities from the 50-fs amplified Ti:Sapphire laser. The incident laser beam had a much smaller spectral content

help of the pulsed PD. In the presence of two 720-nm IR-cut filters, we did not detect any signal from the fundamental laser beam into the PD in the absence of any sample. This ensures that we measured only the UV and visible parts of the supercontinuum light.

We saw a strong modulation in the SG signal on the PD, even for an isotropic sample such as acetone, as the polarization of the incident laser beam was changed by rotating the angle (θ) of the $\lambda/4$ plate (Fig. 3). Similar results were obtained for other isotropic samples, like fused silica and water, in agreement with our previous work [17] of the ellipticity dependence of SG. We also found that a chosen linear component of the incident laser beam followed the same pattern (Fig. 3) as its ellipticity was rotated with the help of a $\lambda/4$ plate. For this measurement, the 800-nm spatial filters and the IR-cut filters were removed. A Glan-Laser polarizer was first aligned in place of the sample to completely transmit the linear component of the incident laser polarization in the absence of the $\lambda/4$ plate (Fig. 1). Next the $\lambda/4$ plate was reinserted before the Glan-Laser polarizer to perform the measurements of the linear polarization component of the laser as a function of the ellipticity of the incident laser beam.

We also observed a similar polarization dependence of SG for the anisotropic sample of sapphire (Fig. 4). There was an additional small effect of anisotropy (an order of magnitude lower than that for the linear-to-circular measurements), which was more evident when the incident linear polarization was rotated with the help of a $\lambda/2$ plate in place of the $\lambda/4$ plate (Fig. 1). These results are also shown on the same plot in Fig. 4.

As expected, there was no difference in SG between right and left circular polarizations for either of the above samples. For chiral samples, however, such a difference is expected, since chiral molecules have magnetic moments that result in a difference in absorption for left and right circularly polarized light. This is due to the combined polarization contributions from the electric and magnetic transition moments of a molecule of particular handedness [20]. Linear optical activity is a well-understood phenomenon and is a widely applicable tool for structural determination studies. Our initial experimental results on the 2R-butanol molecule (Fig. 5) seem to indicate an additional optical activity in terms of a difference between left and right circular polarizations. However, this optical activity is difficult to ascertain from the data collected with the 10-Hz laser system [17], which has a very low signal-to-noise ratio in spite of the shot-to-shot data acquisition (large error-bars in Fig. 5). The optical activity, which can be quantified as the difference in SG from the two different directions of a circularly polarized laser, is barely above the signal-to-noise levels in the data presented in Fig. 5. Further experiments are in progress with our kHz experimental laser system, which has a higher signal-to-noise ratio. Despite these

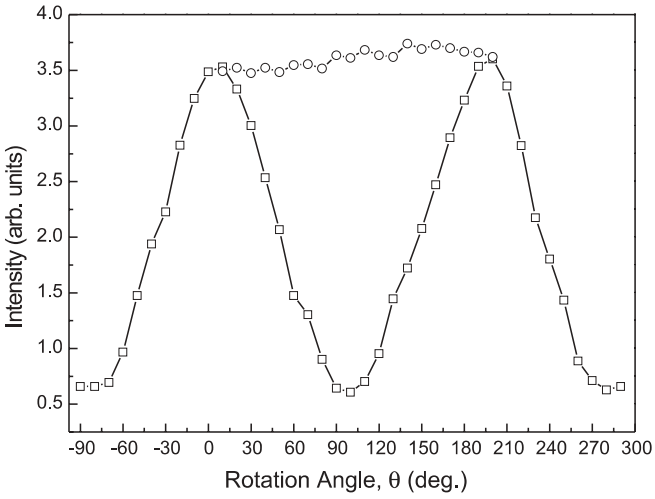

FIGURE 4 Ellipticity dependence of the SG in the anisotropic sample of sapphire as a function of the incident laser polarization (*open squares*), which also showed more efficient SG with linear polarization. The difference between vertical and horizontal polarizations was quantified with the help of a $\lambda/2$ plate rotation *(open circles)* in place of the $\lambda/4$ plate

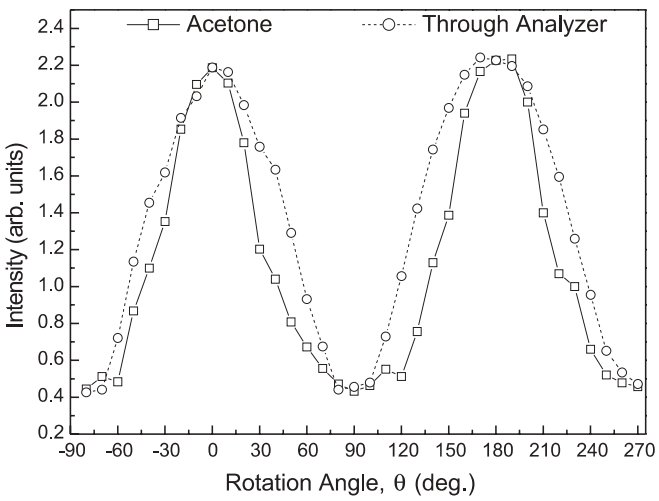

FIGURE 3 Ellipticity dependence of the SG in acetone as a function of the incident laser polarization (*open squares*) compared with the linear laser component of the incident laser beam through the Glan-Laser polarizer (which acts as analyzer), which undergoes the same ellipticity changes without any sample (*open circles*)

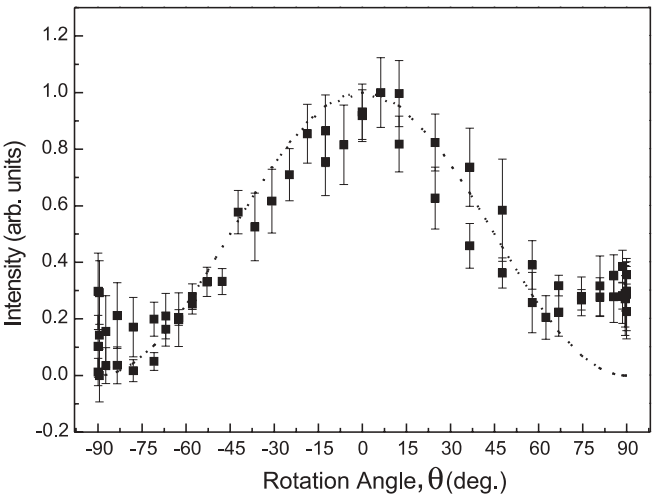

FIGURE 5 Ellipticity dependence of the chiral molecule 2R-butanol, which also showed more efficient SG with linear polarization. The *dotted line* is a symmetric cosine function plotted as a guide to the eye, and indicates slight evidence of optical rotation due to the chirality of 2R-butanol. However, it is barely larger than the error bars of the experimental data for definite conclusions to be made

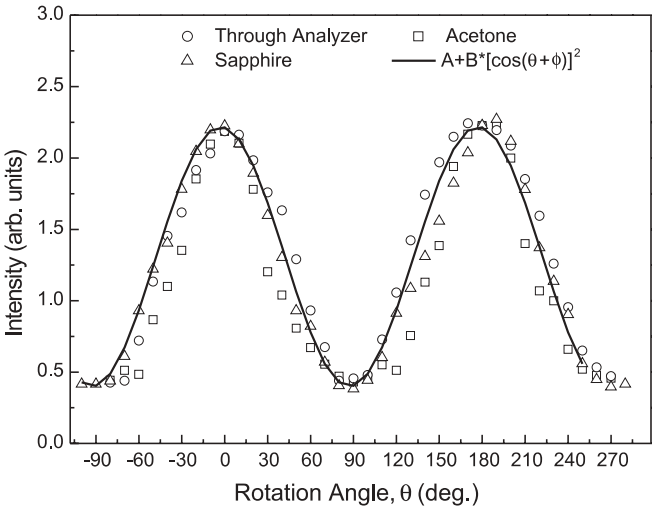

FIGURE 6 The overall ellipticity dependencies of the isotropic sample of acetone, the anisotropic sample of sapphire, and the linear component of the incident laser polarization are shown to be fit effectively by a cosine-squared relationship

difficulties, the result that SG is higher for linear polarization compared with either of the two circular polarizations is very clear in Fig. 5.

The results presented here indicate that, irrespective of the nature of the sample (isotropic, anisotropic, chiral, etc.), an important effect of increasing the ellipticity of the incident laser polarization with increasing ellipticity is in the suppression of SG. This is also identical to the linear polarization component available in the incident laser beam as it changes its ellipticity (Fig. 3). This linear polarization dependence follows a cosine-squared relationship that is in accordance with the complex component analysis of elliptically polarized light [21]. Following these arguments, we show that a cosine-squared function in Fig. 6 is able to fit all of the ellipticity dependence of the SG data. The anisotropic sample (sapphire) showed a small deviation from this cosine-squared analysis. As discussed in the previous paragraph, we expect a deviation from this analysis for the chiral sample (2R-butanol), but we cannot conclude this with certainty from the available data.

4 Conclusion

We have shown that the major effect of polarization on the SG process is the suppression of the supercontinuum as the incident laser polarization changes from linear to circular. This is true for the isotropic (acetone), anisotropic (sapphire), and chiral (2R-butanol) samples studied. The polarization dependence indicates a preference for the linear component of the incident laser beam. The anisotropic sample shows an additional difference in the SG for the two perpendicular directions of polarization of the incident laser beam, but this effect is an order of magnitude lower than the effect of the linear-to-circular ellipticity dependence. Although there is a hint of chirality in our preliminary data on 2R-butanol, further experiments are necessary to quantify it and this work is currently in progress.

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