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Pump polarization-state preservation of picosecond generated white-light supercontinuum

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Abstract: Supercontinuum (SC) generation is among the most interesting nonlinear optical effects lately discovered, due to the complexity of the mechanisms responsible for its generation. This phenomenon, first demonstrated by Alfano and Shapiro using picosecond pulses in condensed phase, has found novel applications in optical pulse compression, time-resolved spectroscopy and material characterization among many others. Here, we demonstrate that picosecond generated SC white-light in water preserves the polarization state: linear, elliptical and circular of the pump source. Moreover, we were able to determine the SC polarization rotation direction in the circular case. With the generation of pulsed circularly polarized SC, new studies and applications are envisaged in the biological, medical and pharmaceutical field. Amino acids, involved in the origin of life, and other chiral structures represent an attractive target for this type of study.

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References and links
The development of ultra-short pulses high power lasers has helped to gain a better understanding of nonlinear optical effects in different media [1-3]. It has also allowed the study of new ultra-fast phenomena in different fields during the last two decades [3]. For instance, supercontinuum (SC) generation is among the most interesting and highly explored nonlinear optical effects due to the complexity of the mechanisms responsible for its generation. Several of its applications in remote-sensing [4,5], optical pulse compression [6,7], time-resolved spectroscopy and material characterization [8-11] have been already investigated. SC generation is characterized by a remarkable spectral broadening of short pulses propagating through nonlinear optical materials spanning from near UV to NIR [12].

The SC phenomenon was first demonstrated by Alfano and Shapiro using picosecond pulses through liquids and solids [13]. Their experimental observations led to the conclusion that supercontinuum is mainly generated by self-phase modulation (SPM). A pulse that experience self-focusing, thus generating a long filament that emits white-light in the forward direction with a small divergence, was proposed as the physical mechanism for SC generation [14-16]. Filament formation is the result of a balance between the Kerr self-focusing and the defocusing effect induced by the free electrons in the generated plasma [17]. Nowadays, however, it is known that SC generation strongly depends on the input pulse width, and the predominantly responsible processes for its generation are: self-phase modulation (SPM), induced-phase modulation (IPM), crossed-phase modulation (XPM), stimulated Raman...
scattering (SRS) and four-wave mixing (FWM) [8]. In the femtosecond regime, SPM (associated with self-focusing), IPM and XPM are recognized as the main nonlinear effects responsible for SC generation [18]. On the other hand, when using picosecond or longer pulses, the dominant mechanisms are SRS and FWM [19], which produces new spectral components with their corresponding asymmetry. Nevertheless, SPM cannot be totally discarded from SC generation in the picosecond regime since it is the responsible for SRS [19].

Recently, it has been demonstrated that SC is a coherent source with similar spatial convergence as the input laser, i.e. SC can be considered an ultra-fast white-light laser [20]. Moreover, it has been established that the pump polarization direction determines the SC output energy and polarization direction [21]. Experimental studies in condensed phase, regarding SC generation dependence with incident polarization, have shown that for linear polarization the SC generation is stronger than for circular [22]. In addition, it has been proven that the ellipticity of the SC follows the incident linear polarization component, independently of the sample [13, 22]. However, a detailed study of the supercontinuum polarization state dependence with the polarization of picosecond pulses has not been explored yet.

Herein, we report the experimental study of the polarization state dependence of supercontinuum generation with the input polarization state. The SC was produced in water using the fundamental of a 25 ps (FWHM), Nd:YAG laser working at 10 Hz. We show that picosecond-induced SC indeed preserves the polarization state of the incident beam. Within this temporal regime, SC generation mechanisms have been elucidated based on polarization dependence. Worth noticing is the fact that the generation of circularly polarized SC will open a new corridor of significant applications in the study of chiral molecules (such as the amino acids present in the origin of life) and chiral structures of interest in the biological, medical and pharmaceutical field.

2. Experimental section

Figure 1 depicts the experimental setup for the SC generation. As a pumping source, we used the fundamental wavelength (1064 nm) of a regenerative amplified mode-locking Nd:YAG laser (EKSPLA), 25 ps (FWHM), operating at a 10 Hz repetition rate. The beam was focused with an achromatic convergent lens (L1) of 25 cm focal length into a 10 cm long quartz cylindrical cell, containing de-ionized triple distilled water (n₀ = 1.333, n₂ = 4.1×10⁻¹⁶ cm²/W) [23].

![Experimental schematic used to generate and analyze the SC generation (details in the text).](image)

Typical irradiances, between 1.4×10¹² W/cm² and 3.0×10¹² W/cm² (5 mJ to 10 mJ), were employed for SC generation. The beam waist (w₀) was estimated to be ca. 100 μm at the focal point. Separation of the SC from the laser pump was achieved through an infrared filter.
Due to the conical divergence of the supercontinuum, the beam was collimated with an achromatic lens (L2) of focal distance f = 10 cm. A quarter waveplate (WP) was introduced before L1 to vary the incident laser polarization state (linear, elliptical and circular). The circularly polarized SC was converted back into linear by employing a 6.8 μm nematic liquid crystal cell (LCC) filled with E7 [24], and oriented at an incident angle α = 59.5° with respect to the cell normal. The LCC was tilted to induce a phase difference of λ/4 at 646 nm.

The polarization of the SC was determined measuring the extinction ratio of the full spectrum as a function of the analyzer angle. We used a broadband calcite polarizer (P) placed after L2 as analyzer. The SC was attenuated by neutral density filters (F). An achromatic positive lens (L3) of focal distance f = 7 cm was utilized to couple the SC into an optical fiber of diameter φ = 50 μm. The optical fiber was attached to a USB-2000 Ocean Optics spectrometer. A personalized National Instruments LabView™ platform program was written for the simultaneous control of P sweeping angle (through a stepper motor) and real-time reading of full SC spectra.

3. Results and discussion

Under the conditions described in the experimental section, we were able to generate SC of approximately 300 nm that covered almost the whole visible range (from 450 to 750 nm). Figure 2(a) shows the measured SC normalized spectra pumping with linear, elliptical and circular polarization. It is noticeable the presence of two strong bands at 541 nm and 645 nm for all incident polarizations, and, for the linear case, an enhanced emission between approximately 560 nm and 630 nm. Pumping with elliptical and circular polarization, two new weak bands appear at approximately 475 nm and 700 nm. The relative amplitudes (A541/A645) of the strong bands, vary with incident polarization and become more pronounced for linear input. As a matter of fact, the two strong bands show comparable amplitude when pumping with circular polarization. This information indicates that SC is mainly generated by SRS and FWM [19]. In addition, between the strong bands maxima, the spectral separation (∆ν = 2,700 cm⁻¹) corresponds to SRS in water [25].

On the other hand, when SC was generated with a linear polarization, the observed enhancement, between 560 and 630 nm, is attributed to positive refractive index changes in water, resultant from SPM [23]. Although in the pico-second regime and in an isotropic liquid, SPM is not regarded as the dominant effect, it cannot be totally ignored working at high irradiances, i.e. > 1.5×10¹² W/cm² [23]. In the SC spectra, the separation between the four main bands as well as their asymmetry indicates that SRS determines the generation of SC and not FWM [19]. Indeed, as a result of parametric generation using picosecond pulses, spectral broadening has never been observed in isotropic liquids such as water but in fibers using multimode phase-matching [26,27].

![Fig. 2. (a). Normalized SC spectra for different input polarization states: linear (solid line), elliptical (dotted line), circular (dashed line). (b). SC output energy as a function of input energy of linearly (●) and circularly (●) polarized light.](image-url)
In order to gain a better understanding of the mechanisms responsible for the SC generation, we performed measurements of the integrated SC output energy vs. input energy for linear and circular polarization. In Fig. 2(b), it can be observed that the SC output energy is greater for linear polarization than that of circular. Besides, it initiates at a lower input energy. Figure 2(b) depicts a near linear dependence for linear polarization, and a sectioned (different dependence within different energy ranges) dependence with circular. In the latter, it seems that a linear increment up to 10 mJ takes place. Beyond this input energy a plateau is achieved up to ca. 13 mJ. Thereafter, it starts increasing until the maximum input energy available. The ratio of the slopes (linear/circular) is equal to 1.47, i.e. approximately 1.5, considering the plots only below 10 mJ. This value accurately matches with the ratio of refractive index change ($\delta n_{\text{linear}}/\delta n_{\text{circular}}$) for a nonresonant electronic nonlinearity [1].

Beyond 14 mJ, it looks as if the increment respects the same tendency as below 10 mJ, with similar slope ratio as well. These results are in agreement with L. D. Barron’s study on polarization effects in stimulated Raman scattering and related phenomena [28]. In his work, he determined a threshold power ratio of 4:6, for linear and circularly polarized incident laser radiation, and pointed out that other effects such as optical orientation and self-focusing could be more relevant in determining the polarization dependence of the power threshold in liquids. The author also predicted linearly polarized self-trapped filaments with same azimuth when pumping with linearly polarized light, and elliptically polarized filaments with arbitrary azimuth using a circularly polarized incident radiation. The latter is a consequence of the lack of definition of the azimuth in circularly polarized beams. His calculations were performed considering the polarization directly from the molecular scattering process and assuming that spontaneous Raman photons stimulate further emission of similar photons [29]. Therefore, knowing that stimulated Raman photons preserve the polarization of their spontaneous originators, and having determined that SC generation using 25 ps pump pulses is mainly originated by SRS, pump polarization-state preservation of SC within this temporal regimen can be anticipated.

In Fig. 3, we show the contour plots depicting the output intensity of the generated SC as a function of the rotation angle ($\theta$) of the analyzer (P) and the SC wavelength. The measurements were carried out for different input polarizations: linear (a), elliptical (b) (only showed for an ellipse with axes ratio $\approx \frac{5}{2}$) and circular (c). The $\sin^2(\theta)$ behavior of the normalized intensity for linear and elliptical incident polarization (for all wavelengths), and the invariant spectral intensity for circular, indicates that the input polarization state is preserved by the SC for all three states.

![Fig. 3. SC intensity contour plots as a function of the rotation angle ($\theta$) of the analyzer (P) and emission wavelength for linear (a), elliptical (b) and circular (c) pump polarization.](image-url)
To better illustrate the spectral transmittance dependence through the analyzer, Fig. 4 shows the experimental and theoretical fitting of the transmittance curves at 646 nm (similar behavior was found for all wavelengths). The full spectral transmittance through the analyzer decreases to zero at approximately $\theta = 180^\circ$ for linearly polarized SC. However, for elliptical polarization it achieves a minimum transmittance of ca. 0.35, and for circular it remains constant. Besides, we found that the SC polarization progressively changes from linear to circular passing through different elliptical states. The shift to smaller angles of the elliptical curve with respect to the linear is an indication of the direction of polarization transition from linear to circular. For smaller pump ellipticity (more circular) a smaller ellipticity was also observed on the SC, thus, a greater shift to smaller angles and a reduced peak-to-valley amplitude of the normalized intensity was obtained (data not showed).

In addition to what Alfano and Shapiro [13] reported, we have found that using circularly polarized input one can generate circularly polarized SC, and that its rotation direction is similar to that of the pump beam. This statement is explained in detail after demonstrating that SC is indeed circularly polarized and not just unpolarized white-light continuum.

In order to confirm circularly polarized SC, we used the LCC described in the experimental section to introduce a phase difference $\Delta \phi$ of $\pi/2 (\lambda/4)$ at 646 nm. For this wavelength, we chose an angle of incidence ($\alpha$) with respect to the LCC normal to selectively induce the effect in this high intensity peak. For $\alpha_{646} = 59.5^\circ$, a path length of 7.84 $\mu$m, corresponding to a $\Delta \phi_{646} = 11\pi/2$, reconverted this wavelength back to linear polarization. The contour plots, shown in Figs. 5(a) and 5(b), demonstrate that the polarization state of the generated SC with circularly polarized pump is circular. The reconverted SC spectrum using the LCC shows a clear $\sin^2(\theta)$ dependence [Fig. 5(b)] around 646 nm. Other spectral regions presenting minima can be also visualized at different $\alpha$ as a consequence of the wavelength dependence of $\Delta \phi(\lambda)$. The absence of Fabry-Pérot interferences fringes due to the presence of the LCC was discarded by removing the analyzer P [30]. The SC spectrum was not altered by the LCC.

With the aim of certifying that the polarization of SC is indeed circular for all wavelengths after being generated using circular polarized pump, we created a theoretical phase-mask (TPM) to simulate the induced phase difference by LCC for all spectral components. The TPM is defined as $\Delta \phi(\lambda) = \left[ (k \Delta n(\lambda) c) / \lambda \right]$, being $k = 2\pi/\lambda$ the wave vector,
the cell thickness (7.84 μm) and $\Delta n(\lambda)$ the refractive index change dependence of E7 with respect to the wavelength [31]. Then, by using $I(\theta(\lambda)) = \cos^2(\theta(\lambda))$, and defining the angle of the electric-field with respect to the analyzer main axis, $\theta(\lambda) = \theta \pm \Delta \phi(\lambda)$, we were able to obtain the contour plot of SC. $\theta$ is the angle of the electric-field with respect to the analyzer in the absence of any additional phase difference. An experimental circularly polarized SC, traveling through the TPM, was taken into account for the calculation. The computational treatment was performed employing a program that takes into consideration $\Delta \phi(\lambda)$ in the expression of the intensity transmitted through the analyzer.

Figure 5 shows the contour plots, (c) and (d), depicting the output intensity of the theoretical recovered SC passing through a $\lambda/4$ for 646 nm, introducing a $\Delta \phi(\lambda) = \pi/2$ and $\Delta \phi(\lambda) = -\pi/2$ respectively, as a function of $\theta(\lambda)$. The similarity between the experimental and theoretically reproduced contour plots certainly probes the circular polarization origin of the whole SC. In addition, the angle of the fringes (white arrows), which is related to the sign of $\Delta \phi(\lambda)$, reveals the direction of rotation of the SC (left-handed and right-handed circularly polarized light). The direction of the dark fringes indicates the rotation direction of SC, positive for clockwise [(a) and (c)] and negative for counter-clockwise [(b) and (d)]. SC rotation follows the pump polarization rotation. To our knowledge, this is the first time that circularly polarized SC has been observed. Perhaps, because most of the efforts in the characterization of SC have been concentrated on femtosecond pulses, in which SPM is the main mechanism for SC generation [18]. Conversely, by pumping with picosecond circularly polarized light). The direction of the dark fringes indicates the rotation direction of SC, positive for clockwise [(a) and (c)] and negative for counter-clockwise [(b) and (d)]. SC rotation follows the pump polarization rotation. To our knowledge, this is the first time that circularly polarized SC has been observed. Perhaps, because most of the efforts in the characterization of SC have been concentrated on femtosecond pulses, in which SPM is the main mechanism for SC generation [18]. Conversely, by pumping with picosecond circularly
polarized pulses, the supercontinuum generation maintains the incident polarization state due to its origin in SRS [8].

In Fig. 6, we present the actual pictures of the SC reflected on a white screen and their corresponding split spectrum for all three different incident polarizations, using a 1,800 lines/mm grating. A remarkable observation is the fact that while linear polarization generates a fully white beam, circular and elliptical polarizations induce more of an RGB (red-green-blue) type of spectrum. From the pictures, it can be noticed a discontinuity of the split spectrum in the circular and elliptical polarization, while using a linear polarization state, more colors appear in the spectrum. Besides, the wavelength distribution over the spot is more homogeneous in the linear polarization case than in the other two incident polarization states (green-blue ring on the periphery). This profile agrees with the explanation given for SC generated with linear polarization: a higher contribution from SPM was observed in the range of 560 and 630 nm.

![Fig. 6. Photographs of SC under linear (left), elliptical (center) and circular (right) polarization excitation conditions.](image)

4. Conclusion

In summary, we have demonstrated that picosecond generated SC in water preserves the polarization state of the input radiation, and that circularly polarized SC rotates in the same direction of the pump. In addition, we have confirmed that the origin of SC generation within this temporal regime is dominated by SRS, not discarding the contribution from SPM. With the generation of circularly polarized SC, new applications in the study of chiral molecules and structure are envisaged in the biological, medical and pharmaceutical field.

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