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Versatile supercontinuum generation by using $\chi^{(2)}$ and $\chi^{(3)}$ nonlinearities in PPLN crystal for direct multiplex CARS measurement

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ABSTRACT

In this study, we report on experiments of spatio-temporal nonlinear frequency conversion in a periodically poled Lithium Niobate (PPLN) crystal designed for second-harmonic generation (SHG). We demonstrated a novel supercontinuum source based on the mixing of second and third-order nonlinearities. We could adjust the ($\chi^{(2)}$, $\chi^{(3)}$) nonlinearities by controlling the input laser polarization orientation, the pulse duration and the PPLN crystal temperature. We obtained an ultra-broadband spectrum, ranging from visible to infrared domains, by pumping a 20-mm-long PPLN crystal with a 3 ps pulse at 1030 nm. This broadband pulse was used to achieve direct multiplex Coherent Anti-Stokes Raman Scattering (M-CARS) imaging, without the need for any optical delay line to temporally synchronize the pump and the Stokes waves. Simultaneous vibrational signatures ranging from -3200 cm^{-1} to -500 cm^{-1} were obtained. Several filters were placed on the broadband supercontinuum path, to shape the output spectrum between 1030 nm and 1650 nm, before sending it into the microscope. The output spectral analysis allows for the demonstration of multimodal imaging, by using SHG, M-CARS and multiphoton fluorescence processes.

Keywords: Nonlinear optics, Nonlinear crystal, Supercontinuum, Multiplex CARS.

1. INTRODUCTION

The CARS technique is a nonlinear process, which combines the effects of Raman scattering and four-wave-mixing (FWM). A pump, a probe and a Stokes (monochromatic) waves interact with a sample, generating an anti-Stokes wave that contains information about molecular vibrations. To excite several vibrational modes, a monochromatic "Stokes" wave can be replaced by a polychromatic beam [1-2]. This so-called multiplex-CARS (M-CARS) method requires the development of supercontinuum (SC) laser sources. Indeed, many SC sources have been obtained by using photonic crystal fibers (PCFs) exploiting their third-order nonlinearities. In that case, the input laser was split into two parts: a pump beam and a Stokes beam (which was generated by exploiting Raman scattering in a PCF). These beams were temporally re-synchronized on the sample by the help of an optical delay line placed on the pump beam path. One of the

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challenges in M-CARS is to simplify the experimental setup by removing the optical delay line: this can be achieved by replacing PCFs with nonlinear crystals.

SC generation, obtained either with crystals or with optical fibers, is a complex process that involves several nonlinear effects (Self-phase modulation, four wave-mixing, soliton propagation, cascading effect, …). SC is typically obtained by pumping a nonlinear medium with short optical pulses (with durations ranging from several hundred of femtoseconds to several hundred of picoseconds). In this study, we will investigate the generation of an ultra-broadband spectrum in PPLN crystal, pumped with a picosecond laser. We will demonstrate that although the PPLN crystal which is commonly used for SHG, it can also provide efficient SC generation. Moreover, we show that the shape of the SC could be modified in real time, by adjusting the second and the third order nonlinearity amounts. The resulting SC is exploited as the Stokes beam in our M-CARS measurements. It should be underlined that the SC Stokes beam was directly synchronized with the pump beam, thanks to the short length of the PPLN crystal (20 mm).

2. EXPERIMENTAL SETUP

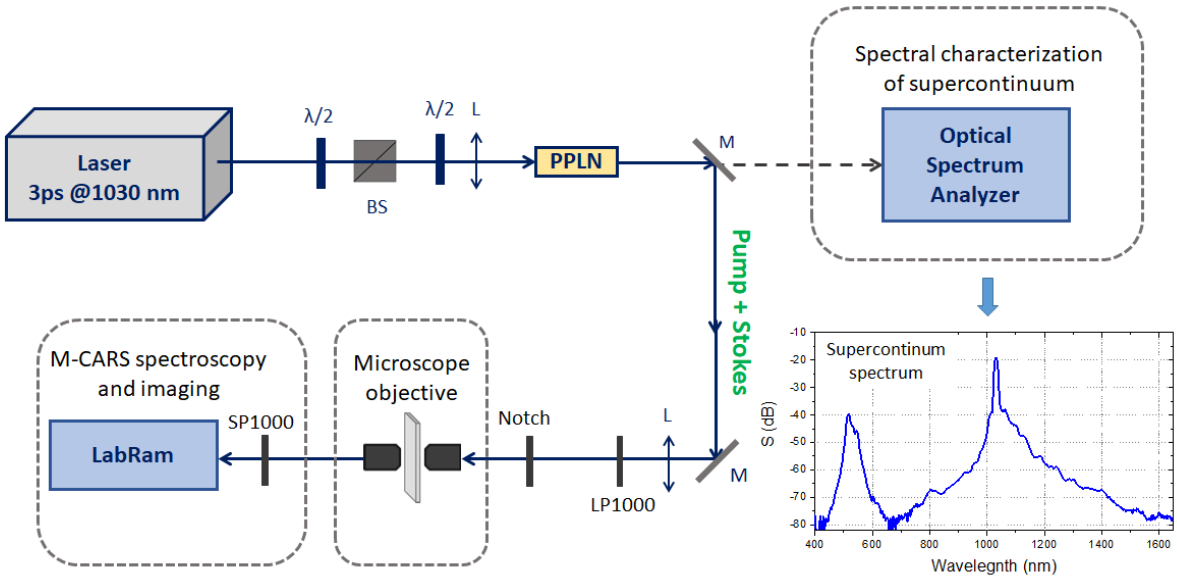

Figure 1: Experimental setup used to generate SC in a PPLN crystal and the M-CARS measurement. λ/2: half-wave plate; BS: polarizing beam splitter; L: lens; M: mirror; LP1000: long pass filter (>1000 nm); SP1000: short pass filter (<1000 nm); Inset: example of a supercontinuum obtained with our setup.

Figure 1 shows our experimental setup. To pump the PPLN crystal, we used a 30 kHz laser at 1030 nm, delivering pulses of tunable duration. Two half-wave plates and a polarizing beam splitter were added before the crystal, to control the optical energy and the beam polarization. The output beam was split into two pathways. The first one was used to analyze the SC spectrum with an optical spectrum analyzer. The second beam was delivered via different filters to a microspectrometer, to obtain the M-CARS spectrum and the images of the samples.

3. RESULTS

3.1 Supercontinuum generation

The PPLN crystal is known for its high second harmonic generation efficiency. However, recent studies have shown that, a broadband spectrum can be generated especially in the phase-mismatched conditions, with a temperature higher than the phase-matching temperature. This broadband spectrum is due to the modulation instability and the cascading effects

in a PPLN crystal exploiting the second order nonlinearity $\chi^{(2)}$ [3] (see figure 2). A rotation of the linear input polarization can drastically reduce the quadratic process which reshapes the supercontinuum in the visible and infrared domains. Because of its high third-order nonlinear susceptibility, the PPLN crystal can modify the initial spectrum of the pump beam and produce symmetric broadening between 800 nm and 1600 nm (see figure 1). Thus, the nonlinear process at the origin of the supercontinuum is mainly due to the self-phase modulation for an input polarization direction oriented close to 90° with respect to the first experiment. An intermediate position of the polarization direction (between 0 and 90°) can introduce an interplay between second and third-order nonlinearities and provides an easy control of the spectrum profile (see figure 2). The visible part of the supercontinuum ranging from 450 nm to 620 nm is due to quadratic conversion of the infrared spectrum by means of a remaining second harmonic generation within a type I process obtained with a large phase mismatch configuration.

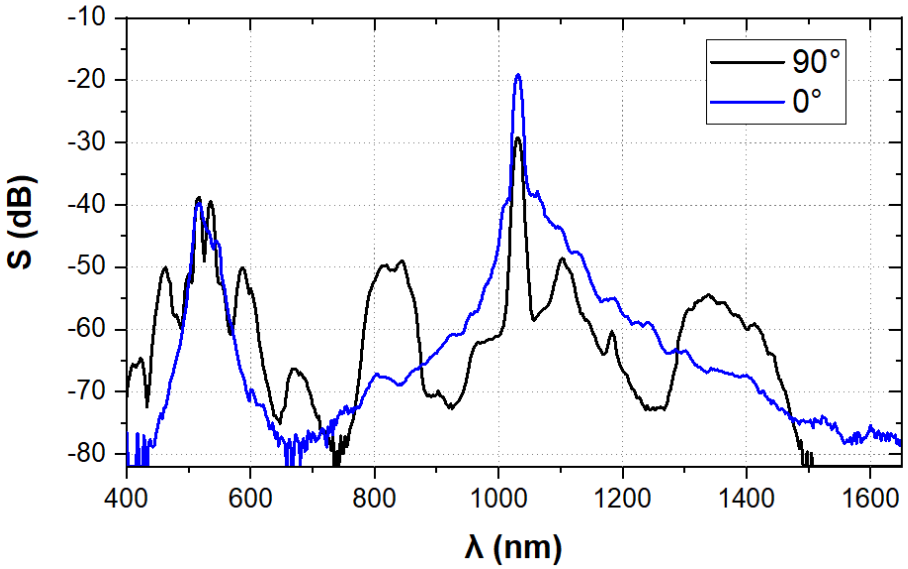

Figure 2: SC spectra generated in a PPLN crystal for two orientations of the input polarization (0°- 90°), pulse duration: 3 ps.

3.2 Impact of the pulse duration and the crystal temperature

We investigated the impact of the pulse duration and the temperature on the generation of the supercontinuum, which can be used as the pump and the Stokes waves in M-CARS measurements. We pumped the PPLN crystal with optical pulses ranging from 270 fs and up to 100 ps with a constant pulse energy. The temperature of the PPLN crystal was fixed at 200°C which corresponds to a second harmonic process obtained with a large phase mismatch conditions (Phase matching temperature: 41 °c). Thus, the spectrum ranges from 450 nm to 1600 nm with a minimum at 700 nm. The upper part (800 nm-1600 nm) of the SC exhibits a triangular shape for short pulse duration i. e. between 250 fs and 3ps. Indeed, in that configuration, the group velocity mismatch becomes larger and no longer superimposition between the fundamental and the second harmonic is obtained. Then, the SC is mainly due to the Kerr effect by means of self-phase modulation. By increasing the pulse duration, the shape of the SC is changed and nonlinear conversions between 1200 nm and 1400 nm are clearly visible. Therefore, for very long pulse (100 ps), the peak power drastically falls, and no spectral broadening is observed (fig. 3(a)).

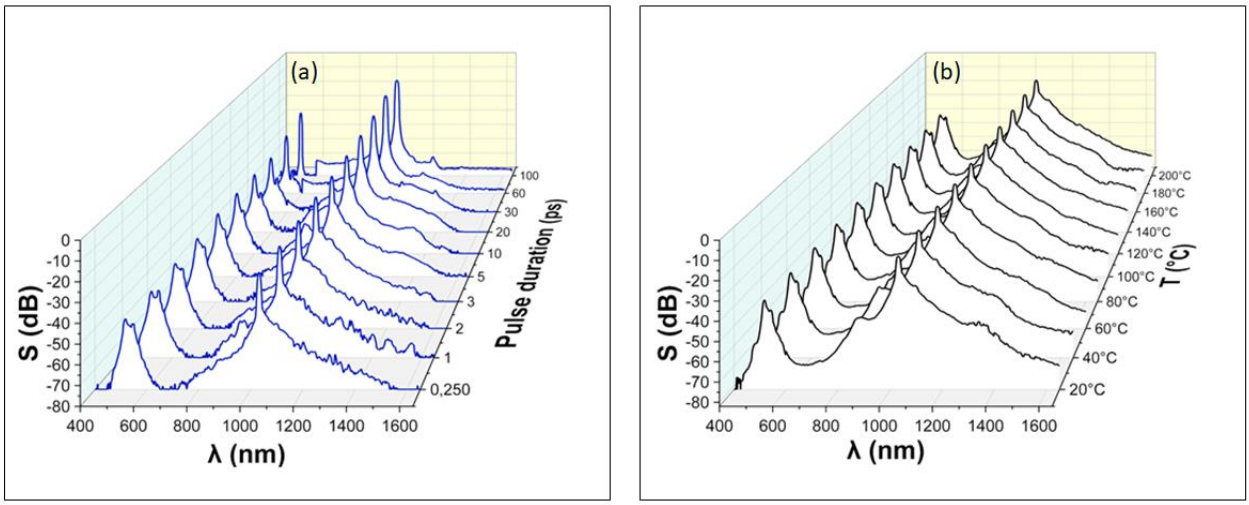

Figure 3: (a) Evolution of SC generated in a PPLN crystal vs. input pulse duration for a temperature of 200°C and a laser repetition rate of 30 kHz, (b) vs. temperature for an optical pulse duration of 3 ps.

Next, we studied the effects of the variation of the PPLN temperature on SC generation. For an optical pulse duration of 3 ps, we varied the PPLN temperature from 20°C to 200°C. The evolution of the SC spectrum is shown in Figure 3(b). As observed the spectral shape of the SC does not evolve so much versus the temperature which demonstrates that the main spectral broadening is driven by the Kerr effect. However, lateral shoulders are visible for temperature close to the phase matching configuration (41°), but progressively disappear for higher phase mismatch configurations. Thus, the SC maintains its spectral extension ranging from 450 nm and up to 1650 nm.

3.3 Multiplex CARS measurement

In our configuration, the pump and the SC Stokes waves are directly obtained from the initial beam coupled into the PPLN crystal. Thus, and because of the low impact of the dispersion along the propagation in the nonlinear crystal, all the waves remain temporally synchronized. The SC spectrum was re-shaped by using different filters, to remove the visible part of the SC (useless and disturbing for the M-CARS signals), and to narrow the pump bandwidth (thus enhancing the spectral resolution of CARS). The polychromatic beam is sent into the microscope and is focused onto the sample with a 60X objective. The resulting forward M-CARS signals are collected through a 60X objective and then sent into a spectrometer. The measurement of a polystyrene bead demonstrates the capabilities of our approach to obtain a M-CARS process. A vibrational signature of a polystyrene sample was obtained in the C-H zone at -3055 cm^{-1} and simultaneously in the fingerprint region (Figure 4a). Figure 4b shows the image of the polystyrene bead around -3055 cm^{-1} peak, with its corresponding bright-field image in Figure 4c.

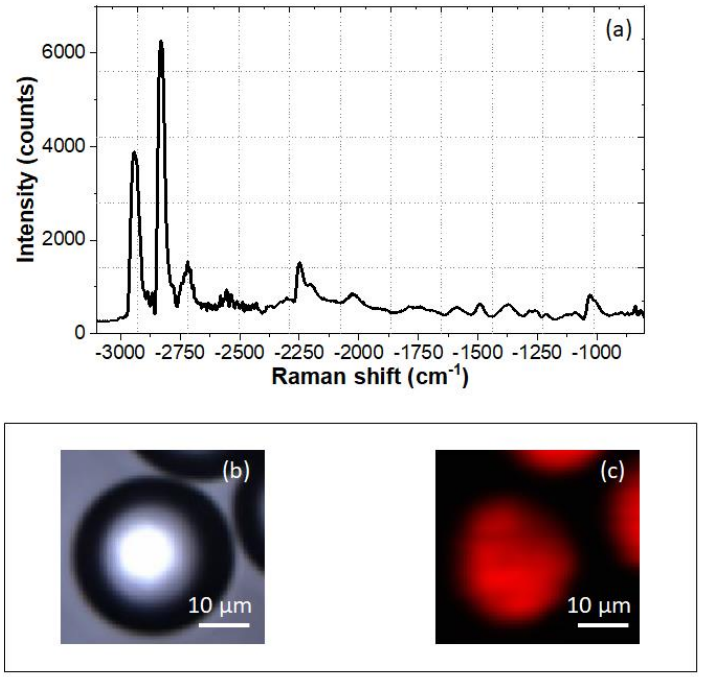

Figure 4: (a) M-CARS spectrum of methanol, (b) M-CARS image of a polystyrene bead at -3055 cm^{-1} extracted from M-CARS data, (c) corresponding bright-field image of the polystyrene bead.

A second series of experiments were carried out on cellulose by using two modes of imaging i.e. M-CARS process and second harmonic generation, introduced by the remaining part of the pump wave. These results are displayed on the Figure 5.

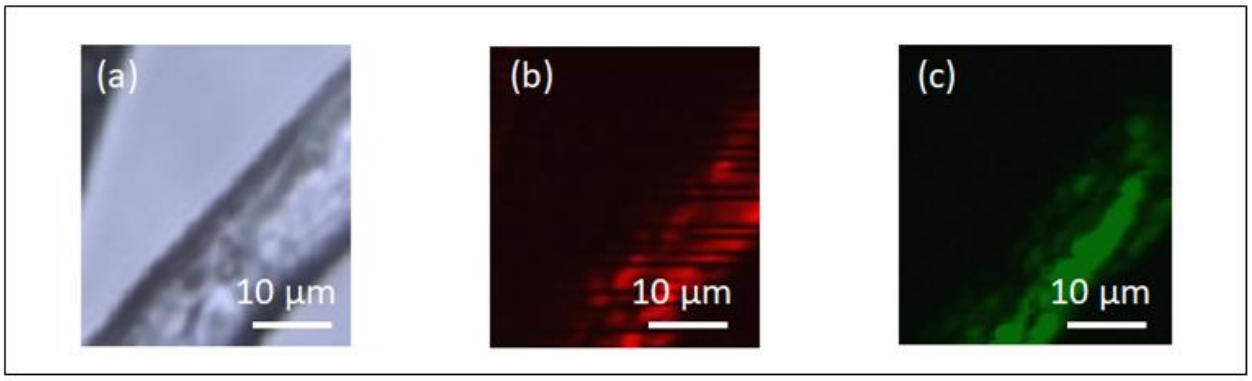

Figure 5. (a) Bright field image of cellulose fibers; (b) M- CARS image at -2896 cm^{-1};(c) Second harmonic image.

4. CONCLUSION

We reported on SC generation in a PPLN crystal by mixing second and third-order nonlinearities. We demonstrated the possibility to control the shape of the SC spectrum by varying the input pulse duration and the optical beam polarization. The short length of the PPLN crystal allows for the generation of an ultra large spectrum, comprising directly synchronized pump and Stokes waves (used for M-CARS experiment), with no spatial degradation of the beams. Thus,

we developed a M-CARS setup without any optical delay line. We, then, simultaneously measured the spectrum of a polystyrene bead in the C-H and in the fingerprint regions and achieved a 2D image of the polystyrene bead at -3055 cm^{-1}. Multimode imaging process has been demonstrated by adding an additional image at the second harmonic obtained on cellulose fibers.

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