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## Stimulated Raman scattering in hollow core photonic crystal fibres

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### Abstract

We present an experimental demonstration of stimulated Raman scattering in a hollow core photonic crystal fibre filled with ethanol. By combining the original transmission properties of these fibres with a highly nonlinear liquid we have realized a very efficient Raman generator. This technique can be applied to other nonlinear mechanisms and opens the way towards the realisation of new fibered components for optical systems.

### 1 INTRODUCTION

Invented one decade ago photonic crystal fibres [1, 2] present original properties of light propagation that make them particularly attractive for applications in nonlinear optics, such as the generation of supercontinuum for spectroscopy and biophotonics or correlated photons pairs for quantum optics. However, for these applications, the performances of these fibres are limited by the weak nonlinearity of the core material, which is silica. A first idea was to use hollow core capillaries and to fill the core with a highly nonlinear liquid. This solution is limited to the use of liquids with refractive index slightly higher than the index of silica (1.45) in order to have a singlemode guidance by total internal reflection. Moreover, in this case, the use of gases whose refractive index is near 1 is not possible. A solution is to use hollow core photonic crystal fibres (HCPCF) and to fill them with a highly nonlinear medium such a gas [3] or a liquid [4, 5]. The use of HCPCF filled with a material with a strong nonlinear susceptibility such as a liquid or a gas should enable to improve the performances of nonlinear fibered components and offers new perspectives thanks to the wide choice of gas and liquids that can fill the fibre. In this work we have studied HCPCF filled with liquids. In the following

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we present the two types of guidance that are possible in such fibres. Then we present an experiment of stimulated Raman scattering in a liquid filled HCPCF and the results that we have obtained.

## 2 MECHANISMS OF GUIDANCE IN HCPCF FILLED WITH LIQUIDS

In HCPCF the central hole of the fibre is surrounded by a cladding containing air holes in a periodic structure. In HCPCF filled with a liquid whose refractive index is smaller than the one of silica, two types of guidance are possible.

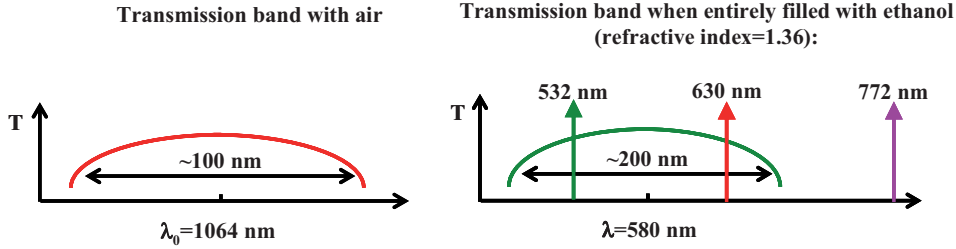
On the one hand, when only the central hole is filled with the liquid, the effective index of the cladding can be lowered below the index of the liquid thanks to the presence of the air holes in the cladding. In this case, guidance by total internal reflection can be achieved. However, in order to have a singlemode guidance, the index of the liquid has to be slightly higher than the effective index of the cladding. For a given fibre the choice of the liquid is then limited. Moreover this technique requires closing the holes of the cladding at both extremities, for example with a fibre fusion-splicer, in order to only fill the core.

On the other hand, an original property of HCPCF is the presence of transmission bands in which the propagation of light is independent from the indices of the core and of the cladding. In this case, the guidance of light can be achieved in air by photonic bandgap effect. When the fibre is entirely filled with a low-index liquid, the transmission band of the fibre is down-shifted according to the following formula [6]:

$$\lambda = \lambda_0 \sqrt{\frac{n_{\text{si}}^2 - n_{\text{liquid}}^2}{n_{\text{si}}^2 - n_{\text{air}}^2}}.$$

$\lambda_0$  is the central wavelength of the transmission band in air,  $\lambda$  is the central wavelength of the transmission band when filled with the liquid.  $n_{\text{si}}$ ,  $n_{\text{air}}$  and  $n_{\text{liquid}}$  are respectively the refractive indexes of silica, air and of the liquid.

The shift of the transmission band of a HCPCF is illustrated in Figure 1. In this example the transmission band of the HCPCF in air is centred at 1064 nm with a bandwidth of about 100 nm. When entirely filled with a liquid with a refractive index of 1.36 such as ethanol the central wavelength of the transmission band is shifted towards 580 nm with a bandwidth of about 200 nm. We see that the shift of the transmission band can be controlled thanks to the refractive index of the liquid that fills the holes of the fibre. Then the transmission bands can be used to favour or to prevent a given non linear effect. For example, in Figure 1, the pump line is at 532 nm. The line at 630 nm corresponds to the first Raman Stokes order of ethanol. Both lines are inside the transmission band and then will see few losses. On the contrary the second Stokes order at 772 nm lies outside the band and will see too much losses to be generated. The Raman cascade can be stopped at the first order thanks to the transmission band of the fibre.

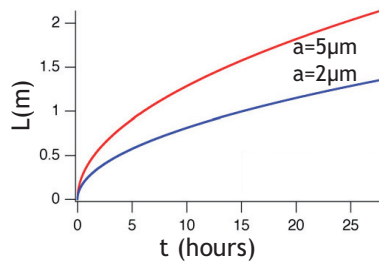


**Figure 1.** (Color online) Shift of the transmission band of a HCPCF entirely filled with a liquid with a refractive index of 1.36.

### 3 EXPERIMENTAL SETUP

The nonlinear liquid that we have used in our experiments to fill the fibres is ethanol. Its refractive index is 1.36, well below the index of silica (1.45).

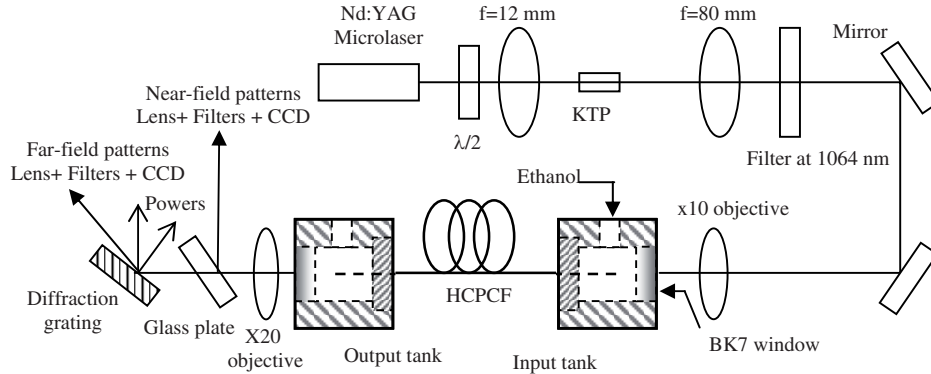
Both extremities of the fibre are fixed in tanks filled with ethanol. The fibre is filled from one extremity using capillary forces. When the liquid reaches the other end of the fibre the second tank is filled with the liquid. This ensures a proper filling of the fibre without any air bubbles. To estimate the time that is necessary to fill our fibres we have used the theory of capillaries filling that gives the filled length  $L$  versus time  $t$  for a given radius  $a$  of the capillary [7]. In Figure 2 we have plotted  $L$  versus  $t$  for different values of  $a$  and for ethanol. At the beginning the filling evolves rapidly. When the time  $t$  tends to infinity, the theoretical length  $L$  evolves as the square root of  $t$ , so there is no theoretical upper limit for  $L$ . In practise the time that is necessary to fill a 1-m-length 2- $\mu\text{m}$ -radius capillary is about 20 h. As we can follow the progression of the interface between the liquid and air inside the fibre by coupling a visible laser into the fibre, we have observed that this calculated time is in good agreement with the experimental filling. Note that the filling of the fibre is possible with any liquid that wets silica.



**Figure 2.** (Color online) Length filled with ethanol for different capillary radius versus time.

The experimental setup is shown in Figure 3. The pump source is a Nd:YAG microlaser that is frequency doubled inside a KTP crystal to provide the light at 532 nm. The pulse duration is 560 ps and the frequency rate repetition is 6 kHz. The maximum energy is 1.26  $\mu\text{J}$ .

The pump source is focused into the fibre with a microscope objective and we can observe at the output of the fibre the near-field and far-field patterns of the

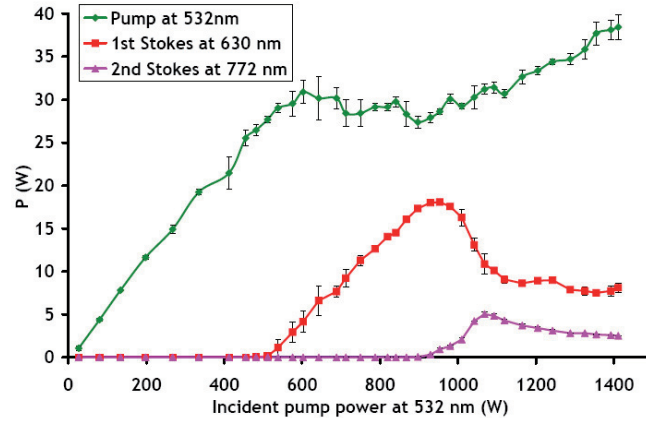


**Figure 3.** Experimental setup.

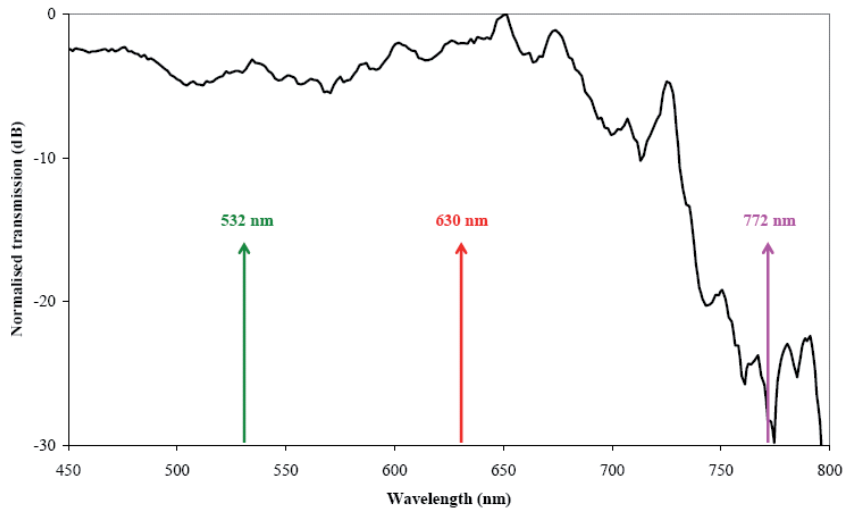
different lines through coloured filters. We can also measure at the same time the different output powers and the output spectrum. The Raman shift of ethanol being  $2928\text{ cm}^{-1}$ , the first Stokes line is at 630 nm and the second Stokes line is at 772 nm.

We have studied two types of fibre with this experimental setup. The first one was fabricated in XLIM (Limoges, France). The results are more detailed in reference [2]. In this fibre the holes of the cladding at both extremities were closed using a fibre fusion-splicer. This technique enables to only fill the central core of the fibre [8]. We have generated the two first Stokes of ethanol at 630 nm and 772 nm with this fibre. These first results present two limitations. Firstly the generated beams have a multimodal structure which is a problem if used as a Raman source. Secondly, as illustrated in Figure 4, where we have plotted the transmitted powers of the different lines *versus* the incident peak pump power, the generation of the second Stokes line at high incident pump powers depletes the first Stokes line. The conversion efficiency from the pump to the first Stokes line is then limited. However, despite these limitations, these results were the first experimental demonstration of stimulated Raman scattering in a HCPCF filled with a liquid and are in good agreement with the simple theoretical model of multi-Stokes order generation that we have developed [4].

The second fibre that we have used is a commercial fibre from Crystal Fiber (reference HC 1060-02) with guidance by photonic bandgap effect. The length of the fibre is 1.14 m and the diameter of the central hole is  $10\text{ }\mu\text{m}$ . When used with air the transmission band of the fibre is centred on  $1\text{ }\mu\text{m}$  with a 3 dB bandwidth of about 100 nm. The experimental transmission band of the fibre filled with ethanol is shown in Figure 5. This spectrum was obtained by coupling a supercontinuum laser source inside the fibre. Once entirely filled with ethanol (central hole + cladding holes) the transmission band of the fibre is shifted towards 580 nm with a 3 dB bandwidth of about 200 nm. One important point is that the pump line at 532 nm and the first Stokes line of ethanol at 630 nm lie inside the transmission band, whereas the second Stokes line at 772 nm is outside the transmission band and then will not be generated.



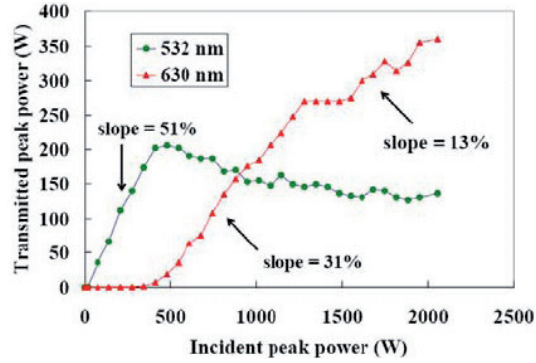
**Figure 4.** (Color online) Transmitted powers of the pump, first Stokes and second Stokes lines *versus* the incident pump power (Xlim fibre, only the central hole is filled with ethanol).



**Figure 5.** (Color online) Experimental transmission band of the HCPCF filled with ethanol.

## 4 RESULTS

We have obtained efficient stimulated Raman scattering on the single first Stokes line of ethanol at 630 nm. We have plotted the transmitted powers of the pump line and of the first Stokes line *versus* the incident pump power in Figure 6. Raman threshold is obtained for an incident pump power of approximately 450 W. Below the threshold we observe a linear transmission of the pump power with a slope efficiency of 51%. Then the transmitted pump power saturates and the transmitted

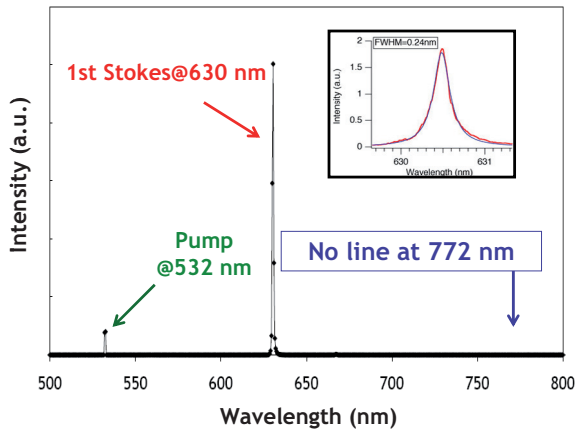


**Figure 6.** (Color online) Transmitted powers of the pump line (circle) and the first Stokes line (triangle) versus incident peak pump power.

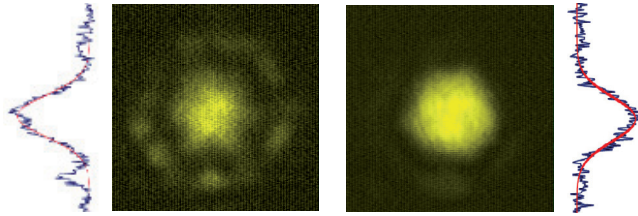
power of the first Stokes presents a double slope behaviour. Below 1200 W we are in the regime of saturation of the pump and we measure a slope efficiency of 31%. Above 1200 W we are in the conversion regime and we measure a slope efficiency of 13%. The important point to note is that there is no saturation of the conversion even at the maximum incident pump power we were able to reach, which corresponds to 5 times the Raman threshold. This result is to be compared with our first results, where the second Stokes appeared at two times the first order Raman threshold in agreement with the theory of multi Stokes order generation (Fig. 4).

To confirm this result we have observed the spectrum of the output beam at maximum incident pump power (Fig. 7). As expected there is no apparition of the second Stokes line at 772 nm which was prevented to appear thanks to the transmission band of the fibre. In the insert we have also measured a first Stokes linewidth of  $6 \text{ cm}^{-1}$  which is well below the spontaneous Raman linewidth of ethanol ( $17 \text{ cm}^{-1}$ ), as expected for a stimulated generation.

We have also observed the near-field patterns of the two lines at 532 nm and 630 nm (Fig. 8). The pump beam is slightly multimode, which may be due to a mismatch between the pump mode and the mode of the fibre at 532 nm. The first



**Figure 7.** (Color online) Output spectrum at maximum incident pump power.



**Figure 8.** (Color online) Near-field patterns of the pump (left) and of the first Stokes line (right).

Stokes beam is almost perfectly Gaussian, which is due to the nonlinear origin of the first Stokes line. In both cases we measured of full width at half maximum of  $4 \mu\text{m}$  on the Gaussian fit.

This Raman generator at  $630 \text{ nm}$  is therefore compatible with standard fibered components.

## 5 CONCLUSION

To conclude we have presented an experimental demonstration of efficient Raman scattering in hollow core photonic crystal fibres filled with ethanol. We have managed to stop the Raman cascade to the first order thanks to the transmission band of the fibre in a guidance by photonic bandgap effect. This has enabled to create an efficient Raman generator on a single line, in a singlemode propagation. This work opens the way towards the study of other nonlinear effects in optics, such as parametric generation by four-wave-mixing. This technique should enable the realisation of new optical nonlinear devices for applications in quantum optics or signal processing.

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