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Abstract: We show that high efficiency stimulated Raman scattering can be obtained using hollow core photonic crystal fiber with the core filled with a low refractive index nonlinear liquid. This new architecture opens new perspectives in the development of nonlinear functions as any kind of nonlinear liquid media can now be used to implement them, with original properties not accessible with silica core fibers.

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OCIS codes: 060.4370 Nonlinear optics, fibers, 060.2320 Fiber optics amplifiers and oscillators, 190.5650 Raman effect

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Although discovered long time ago [1] and heavily studied in the past [2], applications of Stimulated Raman Scattering (SRS) show presently a renewed interest as well for compensation of inline losses for optical fibers transmissions [3], as for the development of new tunable laser sources [4] or for low noise amplification of optically carried radiofrequency signals [5]. For all these applications, the optimization of devices is based on a compromise between the value of three parameters: the Raman nonlinearity of the chosen medium, the interaction length and the pump beam intensity. To increase the effective interaction length, the Raman medium can be inserted in doubly resonant cavity [6], but in such a case the intensity stays small outside the focal zone. Conventional optical fibers are another example of a possible compromise [7]. They allow a good confinement of light (that can be pushed towards its maximum by the use of microstructured optical fibers [8]) and a very long interaction length (due to their very low absorption) but they use silica, which presents a very low Raman efficiency, as an active medium.

The ideal solution combining the performance of fiber guidance with a Raman medium of high efficiency has soon been proposed, with the use of liquid filled capillaries [9-12]. Unfortunately, as the guidance efficiency is linked to the total internal reflection phenomenon, the refractive index of the nonlinear medium must be greater than that of silica that is usually used for the capillary (some other cladding media with lower index were proposed [13] but without further developments). Moreover, when used with high index liquids these capillaries very easily become multimode (unless using very small diameter capillary), what leads to low quality spatial characteristics of the generated beam. All these conditions greatly limit the possibilities among the possible Raman liquids, to those having a refractive index slightly above the one of silica and completely prevents the use of gases with indices close to unity.

The recent apparition of Hollow-Core Photonic Crystal Fibers (HC-PCF) (or more generally Hollow-core Microstructured Optical Fibers) [14, 15], consisting of a hollow core surrounded by large air holes running down the length of the fiber, gives new opportunities in the realization of liquid- or gas-core fibers. For instance, stimulated Raman scattering has been demonstrated in gas cells fabricated with HC-PCFs filled with hydrogen or nitrogen [16-18]. Moreover, the presence of air-holes in the cladding allows to tailor the effective index of the cladding. It is thus possible to decrease the cladding index below that of low-index liquids (such as ethanol with n around 1.36 at visible wavelengths [19]) to ensure a waveguidance based on total internal reflection at the liquid-filled core/cladding interface. We present in this paper a first experiment of stimulated Raman scattering realized in an ethanol filled core microstructured optical fiber.

The air-silica fiber used in our experiments has been fabricated at IRCOM by the stack and draw technique. Scanning Electron Microscope (SEM) photographs of the transverse section are shown in Figure 1(a). The hole-to-hole spacing in the cladding is around 4µm and the air-filling fraction is 84%. The central hole has a diameter of 11µm (corresponding to a surface of the fiber core $S=9.5 \times 10^{-7} cm^{-2}$). In order to have a good versatility of the designed fiber, we have designed a fiber able to ensure optical guidance with most liquids. To do so, the air-

filling fraction has been calculated to reach an effective index of the cladding around 1.25, i.e. smaller than the refractive index of most existing liquids giving a large choice of Raman liquids. However this advantage is obtained at the expense of a multimode propagation for most liquids that could be used. For instance, assuming the ethanol index to be 1.36 in the visible region, the numerical aperture of the filled fibre is around 0.6 indicating that the fibre is highly multimode for the 11µm-core diameter (the V-parameter is equal to 39 at 532 nm). This last point will not be essential for preliminary tests such as the one described here, and the obtained results will be used to design microstructured optical fibers optimized to ensure single mode operation for a given liquid.

The holes constituting the cladding were closed at both extremities of the fiber using a fiber fusion-splicer. By a proper choice of the fusion parameters the central hole expands whereas the peripheral holes collapse, allowing in such a fiber to fill only the central hole with the low index liquid (Fig. 1(b)). The core is then surrounded by air filled holes of the cladding, ensuring a lower effective index of the cladding than the liquid core.

(a)   (b)
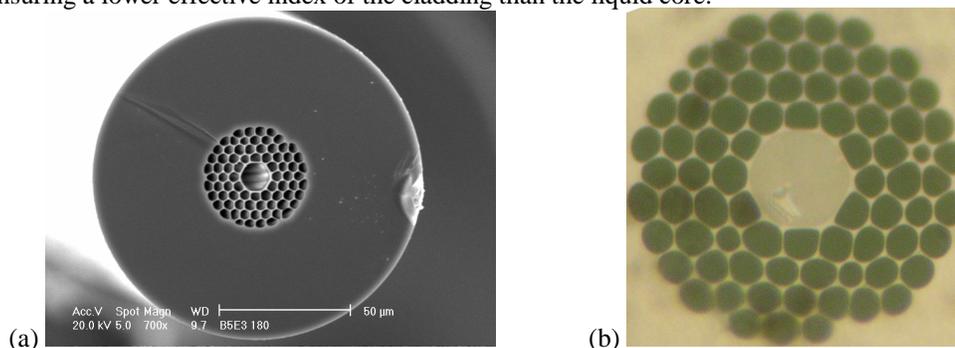
Fig. 1. (a) SEM image of used fiber, (b) Optical microscope image of the end of the fiber filled with water and illuminated from above. The peripherical holes were closed on the side where liquid was inserted, and the fiber is observed on the cleaved other side. Only the central hole is filled with the liquid.

Both extremities of the fiber are fixed in tanks filled with the liquid. The fiber is filled from one extremity using capillary forces (filling is accelerated using an overpressure in the tank). When the liquid reaches the end of the fiber the second tank is filled with the liquid. The process ensures a proper filling of the fiber without any air bubbles in the fiber. The fiber ending is brought close to the tank window to assure good coupling of light with small working distance microscope objectives. The length of the fiber is 2.8m.

The nonlinear experiments conducted at LCFIO used the set-up shown in Figure 2. The laser source is a Nd:YAG microlaser delivering 560ps duration pulses at 1064nm with energy 7.8µJ and a repetition rate of 6kHz. The pulses are frequency doubled in a KTP crystal with an efficiency around 20 to 40%. The Raman shift of ethanol is 2928cm^{-1} with a Raman gain g=5cm.GW^{-1} [20]. It induces a first Stokes line at 630nm and a second Stokes line at 772nm for the used 532nm pump beam. The third Stokes line at 997nm was never seen probably due to the strong absorption of ethanol at this wavelength. The wavelength width of the first Stokes line is 0.237nm (Fig. 3). From this measurement we deduce a Stokes linewidth of 6cm^{-1} for ethanol. As expected for stimulated amplification, this value is smaller than the spontaneous linewidth of the 2928cm^{-1} line (17.4cm^{-1}) found in the literature [21].

We have also measured the transmitted intensities of the different lines (pump and first and second Stokes) as a function of the incident power (Fig. 4). The pump beam intensity saturates when the first Stokes appears; it itself saturates when the second Stokes appears. The inset shows the strong increase of the Stokes line intensities around the threshold (500W and 900W for the first and second Stokes lines respectively) due to the exponential amplification of the Raman signal.
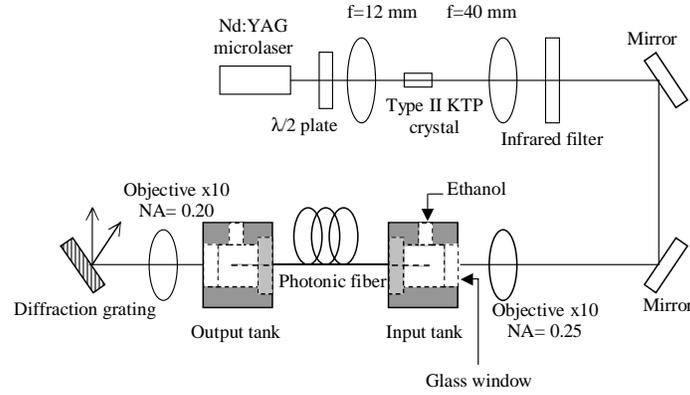
Fig. 2. Experimental set-up for the measurement of Stimulated Raman Scattering in liquid core photonic crystal fiber.
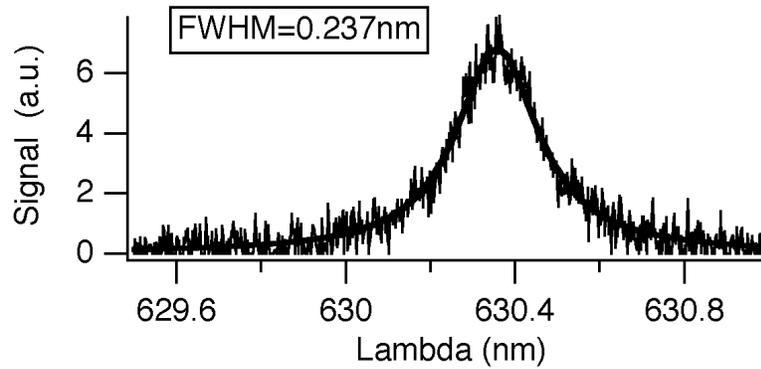
Fig. 3. Spectrum of first Stokes lines at 630nm. The bold line represents an adjustment with a Lorentzian profile, with the given width.

To model these curves a simple Raman generation model can be used, with the following equation for the amplitude A_i of the different beams (i = p, s1 and s2, for the pump, first and second Stokes respectively) (derived from [22]) :

$$\frac{dA_p}{dz} = -\frac{\alpha_p}{2} A_p - \frac{G_0}{2} A_p \left| A_{s1} + N_{s1} A_p \right|^2 \tag{1}$$

$$\frac{dA_{s1}}{dz} = -\frac{\alpha_{s1}}{2} A_{s1} + \frac{G_0}{2} \frac{n_p \lambda_p}{n_{s1} \lambda_{s1}} \left(\left| A_p \right|^2 (A_{s1} + N_{s1} A_p) - A_{s1} \left| A_{s2} + N_{s2} A_{s1} \right|^2 \right) \tag{2}$$

$$\frac{dA_{s2}}{dz} = -\frac{\alpha_{s2}}{2} A_{s2} + \frac{G_0}{2} \frac{n_p \lambda_p}{n_{s2} \lambda_{s2}} \left| A_{s1} \right|^2 (A_{s2} + N_{s2} A_{s1}) \tag{3}$$

where α_i is the absorption coefficient and n_i the refractive index of ethanol at the wavelengths λ_i (Table 1) and the power P_i in Watt being given by $P_i = \left| A_i \right|^2$. $G_0 = g/S_{eff}$ where g is the Raman gain coefficient and S_{eff} is the effective area of the mode (supposed here to be identical for all lines and equal to the surface S of the central hole). N_{s1} and N_{s2} deal with the amplitude of the spontaneous Raman noise emitted in the fiber mode, for the two Stokes lines. These

parameters are related by $N_{s1}\lambda_{s2}^2 = N_{s2}\lambda_{s1}^2$ and N_{s1} is thus the only adjustable parameter that allows to control the position of the threshold of the two Stokes lines.
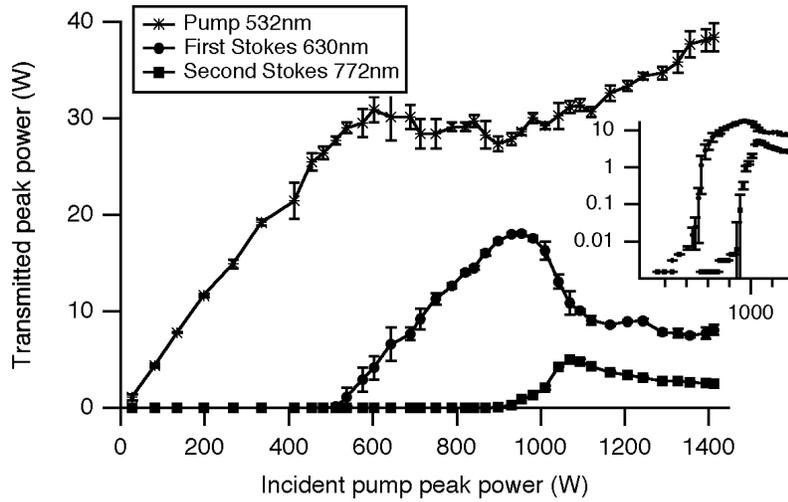
Fig. 4. Intensity dependence of the transmitted pump, and Stokes lines as a function of the incident pump beam intensity. The insert shows the Stokes intensities on a log scale allowing a precise determination of the threshold.

Table 1. Numerical value of the absorption and refractive index of ethanol used in numerical calculations. The absorption coefficients were measured with a 2 cm tank using a spectrophotometer. The refractive indices were calculated from the Cauchy dispersion formula with the coefficients given in [19]

Wavelength (nm)	Absorption (cm^{-1})	Refractive index
532	1.5×10^{-3}	1.3637
630	3×10^{-3}	1.3605
772	15×10^{-3}	1.3578

The equations are numerically solved (Fig. 5) and a reasonable accordance for the position of the threshold is obtained for a value of the spontaneous noise equal to $N_{s1} = 10^{-3}$. In order to take into account the poor coupling efficiency that we have experimentally obtained and attributed to insertion losses in the fiber, we have considered a coupling coefficient T of the incident pump power in the fiber. T can be estimated from the slope ($T e^{-\alpha_p l} = 5.4\%$) of the low intensity part of the experimental curve (see figure 4) where l is the fiber length. We thus find T = 8.3%. The apparition of the second Stokes line indicates that we are in a strongly depleted pump regime and the transmitted pump intensity should normally go to zero [23]. To explain for the linear increase of the transmitted pump intensity at high incident power we need to suppose that part of the incident light is coupled to propagation modes that are always below Raman threshold and thus propagate linearly. The proportion of light coupled into those two modes (the "nonlinear" and the "linear" modes) can be simply evaluated from the ratio of slopes of the transmitted pump beam intensity at low (giving $T=T_L+T_{NL}$) and high power (giving T_L). We have $T_{NL} = 5.1\%$ and $T_L = 3.2\%$. The bold curve in Fig. 5 gives the calculated transmitted pump power using these parameters. Note by the way that using these transmission values the internal intensity thresholds are only 25 and 45W for the first and second Stokes lines respectively. This strongly indicates that continuous wave Raman lasers could be easily obtained using optimized HC-PCF.
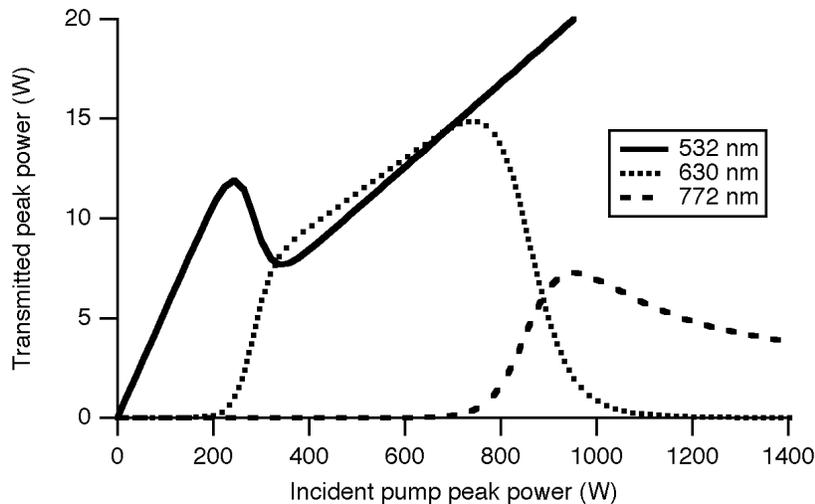
Fig. 5. Theoretical dependence of the pump and Stokes lines intensities.

Considering the simplicity of the model the accordance between the theoretical and experimental curves is rather good, taking into account that only one parameter is used in the adjustment. More sophisticated models including Stokes-Antistokes coupling between the second Stokes line and the pump beam may be developed, with better accordance but at the expense of additional adjustment parameter. Further experiments with for example alternative fiber length will be needed and performed in the near future for comparison with these improved models. This will give way to an optimization of the fiber structure that should allow to observe single mode Raman generation with CW pump lasers.

The use of photonic crystal fibers gives new possibilities for the elaboration of nonlinear fibers in which the silica core can be replaced by more efficient nonlinear liquids or gases. The microstructured cladding can be optimized to allow good guiding properties with any kind of liquid with refractive index lower than the one of silica, greatly enlarging the range of usable liquids. The first experimental results presented here show how this can be used efficiently for Raman generation but the presented results can be extended without any problem to other nonlinear mechanisms such as stimulated Brillouin scattering, parametric conversion or Kerr effect giving rise to new implementation of nonlinear functions for optical devices.