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# Optimization of liquid filled hollow core photonic fibers for nonlinear applications

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## 1. Liquid filled hollow core photonic bandgap fibers

In 1991, the discovery of photonic bandgap fibers opened a novel area for conceiving new optical fiber components [1]. The design of the photonic structure allows both to position the transmission band and to precisely manage the dispersion. This dispersion management is crucial in most nonlinear optical experiments such as supercontinuum generation.

The optical nonlinearity is most often restricted to the nonlinearities of the silica from which fibers are drawn. Silica can be doped, for instance to realize laser amplifiers. Alternatively, inserting a nonlinear material during the fabrication of the silica fiber has been demonstrated. Photonic crystal fibers made from other materials, such as chalcogenides for working in the mid-infrared, were also demonstrated. Nevertheless, in the visible and near-infrared range a greater versatility is obtained by using hollow core photonic crystal fibers. The high quality of the silica structure is preserved and the hollow core can be filled with a desired nonlinear material: a gas or a liquid for instance [2, 3]. The mesh silica structure of these hollow core fibers can be made very thin, so that the influence of the silica structure on the nonlinearities becomes negligible compared to the nonlinearity of the material filling the holes of the structure. Using large fiber lengths eventually compensates for the small nonlinearities of the filling material. Because of the bandgap structure, the guiding properties are kept even if the refractive index of the filling material is lower than the refractive index of silica. These features open this technique to a very large choice of filling materials and thus broaden the field of applications.

We have been working with these liquid filled hollow core photonic crystal fibers for more than 10 years. In the following we are going to describe the optimization that can be made by a correct choice of the liquid (or mixture of liquids) taking two recent devices we developed as examples: an efficient wavelength converter based on stimulated Raman scattering; a parametric generator of high purity pairs of photons.

## 2. Optimization of liquid filled fibers for Raman wavelength conversion

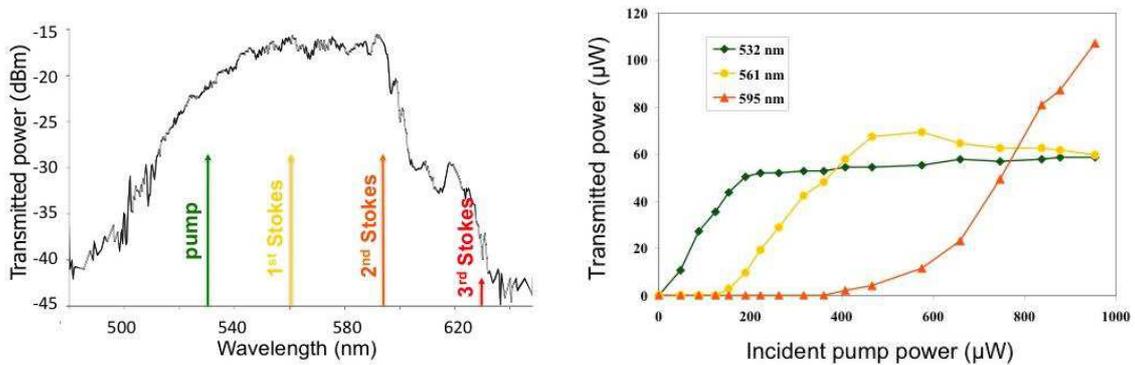
Compared to parametric generation, stimulated Raman scattering is easy to handle. Raman scattering indeed does not require to fulfill a phase matching condition so that the design of the converters does not need to take into account dispersion. Stimulated Raman scattering is thus a robust technique to convert any wavelength into a larger one.

During their propagation inside the liquid filling the fiber, some of the pump photons are first scattered by spontaneous Raman scattering into Stokes photons at a longer wavelength; part of them propagates inside the mode of the fiber. These latter are further amplified at the expense of the pump beam by stimulated Raman scattering. The frequency shift between the pump and Stokes wavelengths depends on the selected Raman liquid and can be chosen among a very large panel up to a few thousands of  $\text{cm}^{-1}$ , for instance about  $3400 \text{ cm}^{-1}$  for the main Raman line of water. Raman cascades (e.g. further scattering of the Stokes photons) allow reaching larger shifts. Although the Raman spectra of most liquids are rather complex and present many Stokes lines, usually only the largest one significantly contributes to the stimulated Raman scattering in fibers. Indeed, because the stimulated Raman scattering is an exponential process, the line whose Raman gain is the largest first depletes the pump beam prior to the emergence of other Stokes lines.

Nevertheless, as explained above the converted Stokes photons can themselves serve as a pump and can be scattered again into photons with a doubled frequency shift. This cascading process can be repeated many times provided that the new converted photons remain in the transmission band of the fiber. Therefore, in designing a liquid core photonic fiber for efficient Raman conversion, one has thus just to make sure that the Raman cascade is stopped at the desired Stokes order, say the  $n^{\text{th}}$  order. In other words, both the pump wavelength and the desired  $n^{\text{th}}$  Stokes order wavelength must lie inside the transmission

band of the liquid filled fiber. The next order, the  $n+1^{th}$ , should be set outside the transmission band. In such a case all the pump energy accumulates on the desired  $n^{th}$  order, and large efficiencies are obtained. We already built several such converters by stopping the cascade either on the first or on the second order. By carefully selecting the liquids and adjusting the transmission band, starting from microlasers delivering 532 nm sub-nanosecond pulses of about 1  $\mu$ J, we already demonstrated the conversion towards the following wavelengths: 556 nm; 561 nm; 582 nm; 595 nm; 612 nm; 630 nm; 667 nm; 772 nm.

For example, to reach 595 nm we selected toluene as the Raman medium. A cascade up to the 2<sup>nd</sup> Stokes order of the main Raman line (1004  $\text{cm}^{-1}$ ) allows reaching 595 nm from a pump beam at 532 nm [4]. We started from a commercial fiber (HC 1550-02 from NKT) whose transmission band is centered at 1550 nm. To shift the transmission band over the pump and the Stokes wavelengths, we needed a liquid whose refractive index is 1.42 [4]. We therefore mixed the Raman liquid (toluene whose refractive index is 1.49) with methanol (refractive index 1.33) to obtain the targeted refractive index. As seen in Fig 1 left, the measured transmission band of the filled fiber covers the pump, the first and the second Stokes orders. In Fig. 1 right, we have plotted the mean output powers at the pump, first and second Stokes wavelengths versus the mean power of the incident pump (not corrected from the injection losses). The microlaser delivers 560 ps pulses at a 6 kHz repetition rate. This figure demonstrates that the energy is successively transferred from the pump to the first Stokes wavelength and then accumulates on the second Stokes wavelength. In this specific experiment, correcting from the injection losses, the conversion efficiency from the pump to the second order is 42 %.



**Figure 1** Left, transmission band of a filled hollow core fiber with the positions of the pump and Stokes orders; right, output powers at the wavelength of the pump, 1<sup>st</sup> and 2<sup>nd</sup> Stokes orders versus the incident mean pump power.

As seen in this Fig. 1 right, the power of the second Stokes order does not saturate and goes on rising with the pump beam power. The energy accumulates on this order without significant depletion by the 3<sup>rd</sup> order set outside the transmission band.

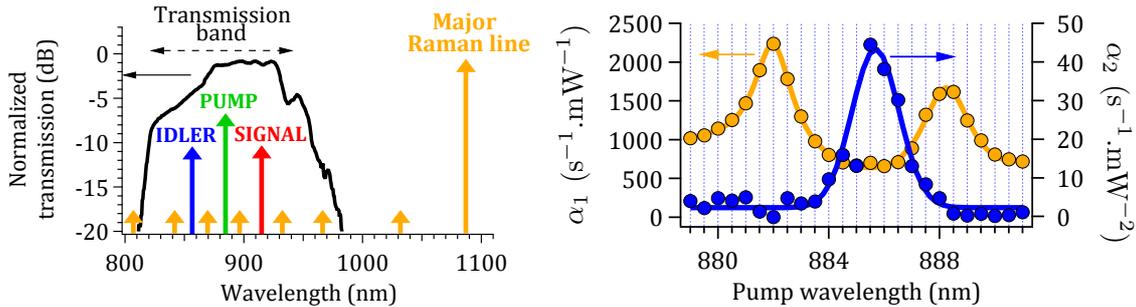
### 3. Optimization of liquid filled fibers for parametric generation

Four-wave mixing is an elastic process in which two pump photons are transformed into a signal photon, of higher wavelength, and an idler photon of lower wavelength. It requires fulfilling the phase matching condition that is achieved around the zero dispersion wavelength of the fiber. Using solid core photonic crystal fiber is convenient as the nonlinear coefficient is provided by the small silica core while the zero dispersion wavelength is tuned by the design of the photonic structure [1]. Such solid core fibers are used to produce correlated signal and idler photon pairs. However the quality of the photon pairs is corrupted by the unavoidable presence of spontaneous Raman scattering in silica. The Raman band of silica is indeed quite large and always overlaps the signal wavelength: efficient filtering of this noise is impossible.

In order to circumvent this problem, we recently proposed the use of hollow core fiber, HC-1550-PM-01 from NKT, filled with deuterated acetone [5]. As depicted in Fig. 3, the transmission band and thus the zero dispersion wavelength of the filled fiber are shifted around 900 nm, close to the pump wavelength. Conversely to a silica core fiber, the Raman spectrum of liquids is made of lines rather than large bands. This feature allows us to put the major Raman line of deuterated acetone outside the transmission band of the fiber (Fig 2 left). Because of the complexity of the Raman spectra, other numerous, but less intense, secondary Raman lines also lie under the transmission band. Nevertheless, we demonstrated that the

signal and idler photons can be emitted between these lines. To do so, we detected the output photons transmitted by a filter set at a wavelength of 857 nm. Keeping this detection frequency fixed, we varied the pump wavelength from 879 nm to 891 nm. At each of these pump wavelengths we varied the pump power in order to identify, on the one hand, the idler photons from the parametric process and, on the other hand, the Raman photons. This identification relies on the fact that the spontaneous Raman scattering is proportional to the pump power while the idler dependence is quadratic. Therefore, the detection rates,  $\alpha_1$  and  $\alpha_2$ , for the Raman and idler photons are respectively expressed in  $\text{s}^{-1}\text{mW}^{-1}$  and  $\text{s}^{-1}\text{mW}^{-2}$ . We plotted these detection rates in Fig. 2 right. This figure evidences that the Raman noise can effectively be minimized (at 886 nm in this example).

Using such a fiber, we recently demonstrated the production of high quality correlated photon pairs [5].



**Figure 2** Left, transmission band of the filled hollow core fiber with the position of the main wavelengths relevant in the parametric process; right, evolution of the detection rates for the idler and Raman photons versus the pump wavelength.

#### 4. Conclusions

Photonic crystal fibers are already versatile components. However, liquid filling hollow core photonic fibers adds to this versatility by providing additional and independent linear and nonlinear optical parameters. The silica photonic crystal structure provides a low transmission in a well-defined band while the liquid is selected for its nonlinear properties. As exemplified above, the combination of the liquid and photonic structure can greatly improve the properties of many devices. Similar optimizations can be advantageously conducted by immersing tapered silica nanofibers in liquids [6,7].

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