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# Kagome hollow-core photonic crystal fiber probe for Raman spectroscopy

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We demonstrate the use of a large-pitch Kagome-lattice hollow-core photonic crystal fiber probe for Raman spectroscopy. The large transmission bandwidth of the fiber enables both the excitation and Raman beams to be transmitted through the same fiber. As the excitation beam is mainly transmitted through air inside the hollow core, the silica luminescence background is reduced by over 2 orders of magnitude as compared to standard silica fiber probes, removing the need for fiber background subtraction. 

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There is a strong demand to combine the chemical selectivity of Raman spectroscopy with the practical ease of use offered by optical fibers [1,2]. The applications are numerous, and cover all fields where conventional light microscopy cannot be used, from material science to medical diagnosis and forensics [2–4]. Hence, a wide range of fiber-based Raman spectrometers have been commercially developed over recent years. However, the use of conventional silica fibers is severely limited by the high luminescence background generated in the silica cores of the excitation and collection fibers [5]. Generally, this background has to be subtracted from the measured spectra, or specific designs of fiber optic probes including optical filters have to be used [1,6], which complicates the signal processing and/or the probe implementation. An alternative approach is to use hollow capillary type excitation fibers [7], but such fibers require large (500 μm) inner diameters to avoid otherwise high transmission losses. Such large mode diameters may be impractical or inefficient for some applications, specially those related to biomedical investigations [3,4].

Hollow-core photonic crystal fibers (HC-PCFs) that confine the light in an air core side-step the limitations of silica-core or hollow capillary fibers [8,9]. Since only a small fraction of the light propagates in the glass, the effect of material nonlinearities is significantly reduced, making HC-PCFs attractive for high peak power laser pulse delivery [10], stimulated Raman scattering [11], four-wave mixing [12], and coherent anti-Stokes Raman scattering [13]. Hence, the high transmission and low background in HC-PCF probes hold great promise for portable Raman spectrometers [14].

HC-PCFs designed for Raman spectroscopy have to meet two technical challenges. First, to enable back-coupling the Raman signal into the fiber probe, the transmission window should exceed 70 nm to ensure observing Raman shifts up to 1500 $cm^{-1}$ corresponding to the Raman fingerprint region. Second, the efficiency of back-coupling the Raman signal into the HC-PCF has to be maximized. To meet these criteria, current strategies use separate solid-core fused silica fibers, or double-cad HC-PCFs to collect the Raman signal from the sample [13,14]. However, these approaches complicate the fiber probe design and/or its fabrication. A more desirable configuration would be for both excitation and Raman scattered light to be counterpropagating in the same fiber.

In this Letter, we demonstrate the suitability of a large-pitch Kagome-lattice HC-PCF for Raman spectroscopy and endoscopy. Thanks to a large transmission window, both the excitation and Raman beams can be transmitted through the same HC-PCF. Of particular interest are (i) the simple optical configuration, (ii) the 2 orders of magnitude reduction in silica background noise, and (iii) the large bandwidth for Raman shifts.

We fabricate the Kagome-lattice fiber using the conventional stack-and-draw technique [9]. Figure 1(a) shows a scanning electron micrograph of the end-face of the fiber. This fiber has a one-cell core defect. Its transmission spectrum, measured by cutback measurement is shown on Fig. 1(b). The main transmission band has a

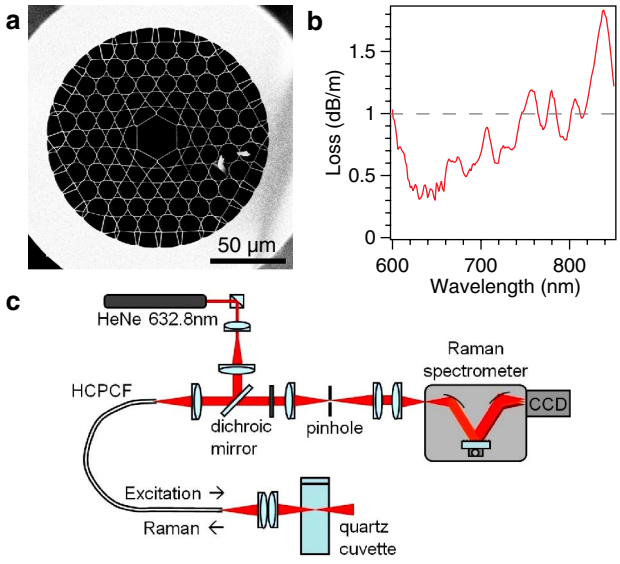


Fig. 1. (Color online) (a) Scanning electron micrograph of the Kagome-lattice HC-PCF. (b) Transmission losses. (c) Experimental setup.

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loss below 1 dB/m spanning over a 150 nm wide range, with a minimum loss of 0.31 dB/m at 630 nm.

The excitation source for Raman spectroscopy is a linearly polarized CW He–Ne laser operating at 632.8 nm. The laser beam is coupled into the 1.5 m long Kagome HC-PCF by a 150 mm lens, matching the 0.02 NA of the HC-PCF. The total transmission of the HC-PCF, including coupling and transmission losses, is about 60%. The laser beam exiting the fiber is focused into a standard 1 cm quartz cuvette for spectroscopy by a stack of two achromatic doublets with 25 mm focal length. The same lenses are used for back-coupling the Raman scattered light into the fiber, see Fig. 1(c). To separate the Raman scattered light from the reflected and elastically scattered laser light, we use a dichroic mirror (Omega Filters 650DRLP), a long-pass filter (Omega Filters 640AELP) and a notch filter (Kaiser Optical HNF-632.8-1.0). A 20 μm pinhole is conjugated to the fiber output with a 1∶1 magnification to spatially filter the HC-PCF mode carrying the Raman scattered light. The Raman signal is finally detected by a spectrometer (Horiba Jobin Yvon iHR320) with a Peltier-cooled CCD camera. Each Raman spectrum is the average of four acquisitions with 10 s integration time.

Figure 2(a) shows the Raman spectrum of chlorobenzene measured with the Kagome-lattice HC-PCF. The main Raman bands at 415, 705, 1007, 1027 and 1088 $cm^{-1}$, and also the weakest bands at 610, 1126, 1162, 1178, and 1589 $cm^{-1}$ are all clearly visible above the background, which is remarkably flat except for two small peaks at 1563 and 1657 $cm^{-1}$. For comparison, we also display in Figs. 2(b) and 2(c) the Raman spectra of chlorobenzene acquired respectively with a standard silica fiber probe (Ocean Optics QP200-2-UV-VIS) and with a confocal microscope equipped with a $63 \times 1.2$ NA objective. For these latter two cases, the pinhole diameter is set to 50 μm so as to optimize the signal-to-noise ratio. Remarkably, the Raman spectrum acquired with the HC-PCF [Fig. 2(a)] compares very positively with the confocal Raman spectrum, both in Raman peak positions and intensities. This is in strong contrast to the Raman spectrum acquired with the silica fiber probe [Fig. 2(b)], where the Raman peaks can hardly be distinguished on the raw spectrum, and the background subtraction procedure has to be used. We also display in Fig. 3 the Raman spectra acquired with the HC-PCF for toluene and ethanol (all solvents are HPLC grade with purities >99.9%). In the experiments, the maximum range available to measure Raman shifts is about 2000 $cm^{-1}$, and is limited by transmission losses in the HC-PCF and by nonperfect correction for chromatic aberrations in the collection optics.

A key parameter for Raman fiber probes is the spectrally broad background noise that is generated inside the fiber. Importantly for the Kagome HC-PCF, the spatial distribution of this background noise is very different from the HC-PCF mode, and thus can be spatially filtered by a pinhole conjugated to the fiber output tip. Figure 4(a) is an image of the fiber output (without any Raman sample) showing the spatial distribution of the luminescence background, while Fig. 4(b) displays the HC-PCF mode carrying the Raman light scattered by the sample. The six-fold symmetry of the Kagome lattice can be clearly distinguished in the background

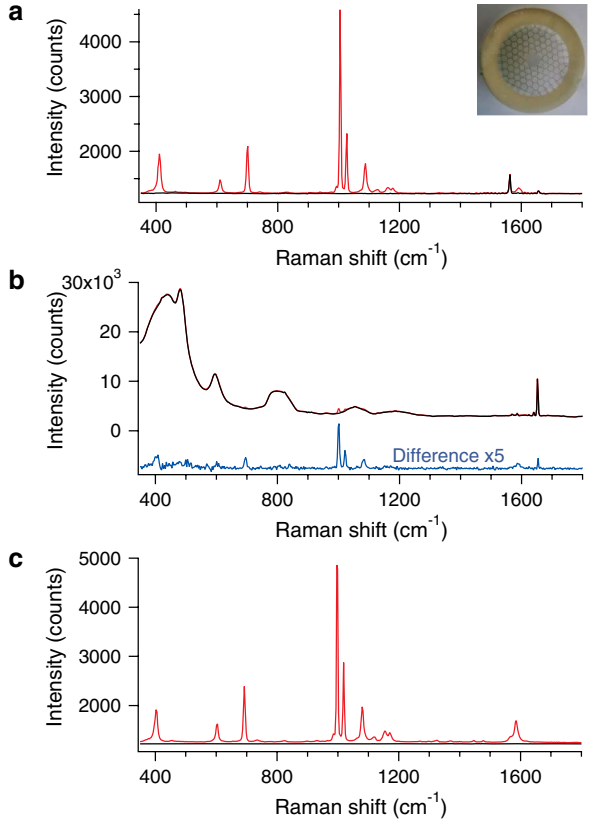


Fig. 2. (Color online) Raman spectra of chlorobenzene (red lines) measured (a) with the Kagome-lattice HC-PCF, (b) with a standard commercial silica fiber, and (c) with a confocal microscope. The black lines correspond to the background in the absence of sample. For all cases, the excitation power incident on the sample is 6 mW, the integration time is 10 s. In (b), the blue line displays the Raman spectrum after background subtraction, and is shifted vertically for clarity.

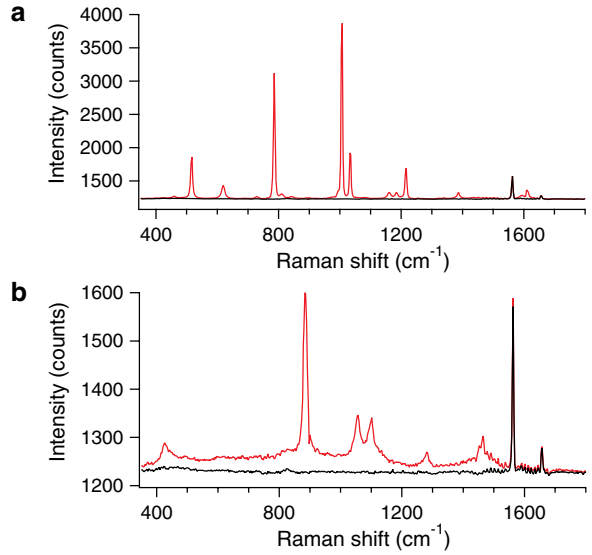


Fig. 3. (Color online) Raman spectra of (a) toluene and (b) ethanol measured with the Kagome-lattice HC-PCF [same conditions as Fig. 2(a)].

spatial distribution, confirming that the background luminescence stems from the tiny silica bridges between the hollow channels. Figure 4(c) explores the influence of

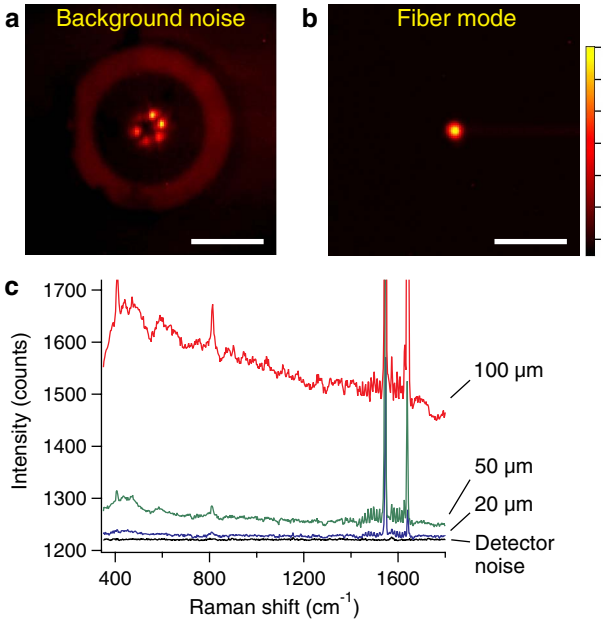


Fig. 4. (Color online) Image of the fiber output (a) without and (b) with the Raman sample. The scale bar is 100 µm. (c) Luminescence background spectra for increasing pinhole diameters [same conditions as Fig. 2(a)].

the pinhole diameter used to filter the background noise. A 20 µm diameter appears the best compromise for efficient collection of the HC-PCF mode and rejection of the background luminescence. As the pinhole diameter is increased, more and more luminescence from the silica bridges is collected by the spectrometer, while the useful Raman signal intensity does not significantly increase. Nevertheless, we point out that the noise in the most disadvantageous case of 100 µm diameter remains over 1 order of magnitude below the noise of a standard silica fiber probe.

The results demonstrate the suitability of a large-pitch Kagome-lattice HC-PCF for Raman spectroscopy and endoscopy, where both the excitation and Raman beams are transmitted through the same fiber. Remarkably, the background noise is reduced by over 2 orders of magnitude compared to standard silica fiber probes. Compared to commercially available HC-PCFs, our Kagome design has a larger core and thinner bridges between the hollow channels. These features enable a better reduction of the luminescence background. Nevertheless, the similar transmission bandwidth found in some commercially available HC-PCFs should enable a straightforward transposition of our results toward commercial applications. We also point out that the fiber tip could be fused shut and used for direct measurement of Raman or fluorescence spectra in immersion into the liquid without losing much of the signal as compared to the current results. We believe such probes offer crucial advantages for spectroscopy in remote and/or highly scattering media.

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