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# Raman Probe Based on Optically-Poled Double-Core Fiber

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**Abstract:** A novel Raman probe based on an optically-poled double-core fiber is reported. Efficient in-fiber second-harmonic generation allows for Raman spectroscopy at 532 nm when illuminating the fiber with 1064 nm laser light. The concentric cores of the fiber provide independent paths for the delivery of the excitation light and the collection of Raman scattering. Spectra of a sample of dimethyl sulfoxide acquired with this device are presented.

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## 1. Introduction

Due to its ability to provide unambiguous and detailed molecular information on nearly any sample in a fast and non-destructive way, Raman spectroscopy represents an invaluable diagnostic and monitoring technique. In particular, when combined with fiber optics Raman spectroscopy can benefit from their flexibility and reduced size, therefore becoming the perfect candidate for remote *in vivo* analysis. Efforts to integrate in fiber the functions required for Raman analysis are thus motivated. The main challenges related to fiber-based Raman spectroscopy are the generation of the excitation light in the fiber and the efficient collection and filtering of the scattered light. In the case of single-fiber geometries the path separation between excitation and collection is also a major issue. While in-fiber Rayleigh-rejection filtering has previously been demonstrated [1, 2], monolithic fiber lasers in the visible range are not so trivial to realize. Nevertheless, by optically poling the core of the fiber responsible for the delivery of the excitation light it is possible to achieve efficient second-harmonic generation (SHG) [3], thus being able to perform Raman spectroscopy at visible wavelengths despite having injected infrared (IR) light into the fiber. In-fiber SHG of up to 236 mW has been reported [4]. As for single-fiber geometries, coaxial double-core fibers provide an intrinsic separation between the excitation and collection paths (the inner and outer core, respectively), also allowing for a reduction of the fiber spectral background. Propagating the pump light in a small core allows for the illumination of the sample with high intensity, as required for nonlinear processes such as Raman scattering. Probes based on double-core fibers and double-core fiber couplers for fluorescence and optical coherence tomography systems have previously been reported [5].

A complete all-in-fiber Raman spectroscopy system may be constructed by integrating a fiber laser, an optically-poled double-core fiber, a double-core fiber coupler and an in-fiber Rayleigh-rejection filter. As an intermediate step towards this final goal, this work focuses on demonstrating that by optically poling the inner core of a double-core fiber it is possible to generate enough monochromatic visible light to perform Raman spectroscopy, and furthermore that the same waveguide is able to deliver the generated excitation light to a given sample through the inner core, and at the same time to collect the Raman scattering in its outer core.

## 2. Materials and Method

The double-core fiber used in this work, shown in Fig. 2.1 (a), is a step-index structure characterized by a central 7.5  $\mu\text{m}$  diameter germanium-doped inner core surrounded by a 260  $\mu\text{m}$  pure silica outer core and by a fluorine-doped cladding of lower refractive index, for a total fiber diameter of 300  $\mu\text{m}$ . For similar N.A. values, from simple geometrical considerations the collection in the outer core is 1200 times more efficient than in the inner core of the fiber.

### 2.1. Optical Poling

Prior to its employment in the Raman setup schematically illustrated in Fig. 2.1 (c), the double-core fiber was prepared for SHG by self-organization of a quasi-phase matched grating. The growth of the second harmonic (SH) light is known

to be parametrically dependent on the amount of SH already present in the core [3], therefore it can be accelerated by briefly *seeding* the fiber with SH light, together with the fundamental IR [6]. For this reason, at first the 1064 nm light from a Q-switched mode-locked Nd:YAG laser (Quantronix) was frequency-doubled by means of a KTP crystal and coupled into the inner core of the fiber by means of a dichroic mirror designed to operate at 1064 nm and a 10x focusing lens (Nikon). After a few seconds of illumination with both green and IR light the nonlinear crystal was removed and the growth of the SH continued as a self-sustained process. Saturation occurred after a few hours, with approx 0.83 mW average power of green light generated along the inner core of the fiber, out of the average 650 mW of IR light initially injected into it. It is worth noting that SHG happens in the first few centimeters of the waveguide, therefore in case the distal fiber tip gets damaged it is possible to re-cleave it without any change in the generated green light.

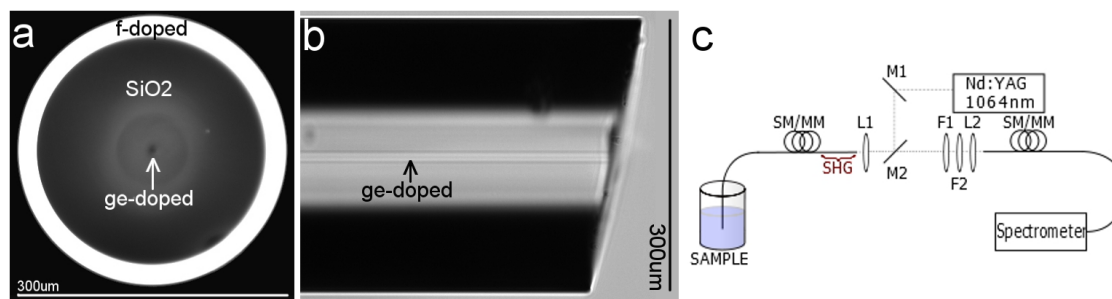


Fig. 1. (a) Cross-section of the dual-core fiber; (b) Angle-cleaved tip (about  $11^\circ$ ) on the sample side; (c) Schematic illustration of the setup: M1,2 are mirrors (M2 is dichroic), L1,2 are 10x focusing lenses, F1 is a dual notch filter, F2 is a KG5 glass filter. The double-core fiber is indicated as SM/MM. The spectrometer is connected to a computer (not shown).

## 2.2. Raman Spectroscopy

The generated 532 nm light was used to perform Raman spectroscopy on a sample of Dimethyl Sulfoxide (DMSO, Merck), according to the setup shown in Fig. 2.1 (c). In particular, the sample was illuminated by the light exiting the inner core of the fiber, while the Raman-scattered light was collected by the outer core of the same waveguide. The fiber was inserted vertically into the sample, and a Raman spectrum was recorded. Subsequently, the fiber tip was extracted from the sample and held at a distance of about 10 cm from its surface, and a second spectrum was acquired to account for the interfering Raman signal generated in the fiber. Both spectra were acquired over a 15 s time, repeated twice to avoid artefacts. The fiber tip on the sample side had previously been cleaved at an angle of about  $11^\circ$  by means of Vytran LDC400 fiber cleaver (see Fig. 2.1 (b)) to minimize reflections.

After propagating through the outer core of the fiber, the collected Raman signal was transmitted through the same dichroic mirror used to couple the IR laser light into the fiber, and was filtered by a dual 532 and 1064 nm notch filter (Edmund Optics), to eliminate the Rayleigh-scattered light and the IR light reflected by the other optical surfaces. A KG5 glass provided an additional filtering of the 1064 nm light. The collected Raman signal was then coupled into another piece of the same double-core fiber by a second 10x focusing lens (Nikon), and conveyed to a CCD-based spectrometer (QE65000, Ocean Optics), thermo-electrically cooled down to  $-20^\circ\text{C}$  to reduce dark current noise. The aperture slit of the spectrometer is 1 mm high and  $50\mu\text{m}$  wide, therefore limiting the collection efficiency of the setup.

## 3. Results and Discussion

The generated 0.83 mW of 532 nm light proved to be intense enough to perform Raman spectroscopy on a sample of DMSO. Fig. 3 shows the recorded Raman spectrum, normalized to the spectrum acquired when the fiber tip was held outside the sample. Raman peaks at  $1046$ ,  $1424$ ,  $2919$  and  $3005\text{ cm}^{-1}$ , together with the double peak at  $671$  and  $699\text{ cm}^{-1}$ , are clearly identifiable. All the detected peaks match well the values reported in the literature [7].

The spectral normalization proved to be an easy and efficient way to extract the peaks located at low wavenumbers from the intense fiber spectral background, which would otherwise mask them, as shown in the inset of Fig. 3. This interfering signal is at least one order of magnitude more intense than the signal from DMSO, up to the point where it saturates the detector for wavenumbers smaller than  $500\text{ cm}^{-1}$ , even with acquisition times as short as 1 s. For this reason, the spectral difference is only significant for wavenumbers larger than  $500\text{ cm}^{-1}$ . It is reasonable to think that

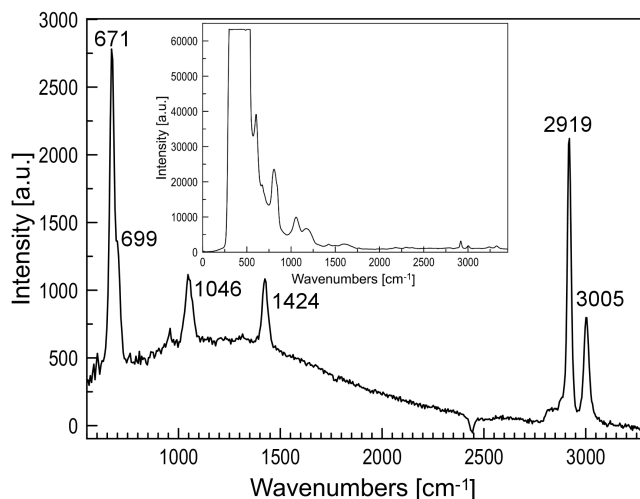


Fig. 2. Raman spectrum of DMSO acquired with the presented double-core fiber-based probe, normalized to the spectrum of the fiber. Inset: spectrum of DMSO as it appears before the normalization.

the strong fiber spectral background is related to that portion of the Raman background signal which is generated by the excitation light in the inner core but cannot be confined in it, due to its scattering angle, and therefore ends up being guided in the outer core. This was partially proved by the significant reduction of the Raman signal from the fiber achieved by angle-cleaving its tip. The notch appearing at about  $2450\text{ cm}^{-1}$  is due to the spectral normalization.

#### 4. Conclusions

Efficient SHG in a double-core fiber was demonstrated. The amount of generated green light proved to be enough to excite Raman scattering in sample of DMSO. The double-core structure allowed for the delivery of the generated excitation light to the sample through the inner core and the efficient collection of the scattered light in the outer core. The work described here represents a significant step towards an all-in-fiber Raman system in the visible range.

#### 5. Acknowledgments

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