# Study of composite optical nanofibers for 2<sup>nd</sup> and 3<sup>rd</sup> order nonlinearities

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**Abstract:** We present the design of composite optical nanofibers coated with different nonlinear materials (PMMA, PMMA/DR1 and TiO<sub>2</sub>) for the realization of new all-solid Raman wavelength converters and sources of correlated photon pairs having an efficiency enhanced by a factor of 1000 compared to bare nanofibers. Two coating processes have been successfully developed, inducing only relatively low losses comprised between 0.5 dB and 1.76 dB.© 2024 The Author(s)

## 1. Introduction

Optical nanofiber (ONF – figure 1), i.e., the homogeneous section of a stretched and tapered silica optical fiber (sub micrometer or micrometer diameter on length of up to more than 10 cm) between two tapered transitions, has been presented a widespread use in science and engineering applications since more than thirty years as an elementary optical component easily integrated by its nature in an all-fibered network. The expanding use of ONF is due to its physical properties. The optical modes guided by the ONF have large intensities due to their strong transverse confinement, present very low losses (below 0.005 dB/cm, far beyond other micro/nano waveguides) and exhibit an evanescent part outside the ONF and therefore in interaction with the external medium.

Due to these intrinsic properties, ONF-based technologies have addressed a large versatility of domains from fundamental to applications such as quantum information devices, nonlinear optics and remote sensor devices for the most active ones. We have already performed several experimental demonstrations in nonlinear optics using ONF. Among them we realized highly efficient wavelength converters based on Stimulated Raman Scattering in the evanescent field of ONF immersed in liquids with conversion efficiencies from the pump source emitting at 532 nm to the first Stokes order of ethanol at 630 nm as high as 60% [1]. We also experimentally demonstrated the emission of photon pairs by four wave-mixing in a bare ONF with high Coincidence to Accidental Ratio [2].



Figure -1. Principle of a composite ONF (left). Mode propagating in an ONF having a radius of 500 nm, functionalized by a 200 nm thick layer of PMMA and encapsulated in silicone (right).

In this work, we investigate the possibility to add a new degree of freedom to silica ONF by its functionalization with a nonlinear material coating, such as a polymer or other material of interest such as  $TiO_2$ . We show that optical nonlinearities can be obtained and even enhanced in such composite ONF by studying the design of new all-solid Raman converters and sources of photon pair by Spontaneous Parametric Down Conversion (SPDC) [3]. The encapsulation of ONF, bare or functionalized, is also studied, as this is a critical point for the protection and manipulation of an ONF-based component (see fig. 1).

## 2. 3rd order nonlinearity using a composite ONF: all-solid Raman wavelength converters

In these converters, the Stokes photons are generated in the functionalizing material of thickness e and refractive index  $n_g$  by the Raman scattering of the pump photons present in the evanescent field. These Stokes photons then couple to a guided mode of the composite ONF.

We have studied two functionalizing materials, PMMA, whose strong adhesion to silica surfaces makes it an attractive coating material for silica ONF, and TiO<sub>2</sub>, which presents a high Raman gain. In the first configuration the ONF with PMMA coating is encapsulated in silicone for protection ( $n_{silicone} = 1.39$ ). We performed simulations on the propagating modes using a three-layer modelling (see fig. 1 (right) for a calculated spatial mode example). The parameters used in our simulations are summarized in Table 1. The chosen ONF radius is 0.5 µm. Based on our previous results on ethanol Raman ONF converters [1] we believe that the modal Raman gains in both configurations should enable to reach Raman threshold by using a pulsed pump in the ns second regime.

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	$\lambda_p (\mu m)$	$\Delta \sigma_R (cm^{-1})$ Raman shift	$\lambda_s (\mu m)$ Stokes	g <sub>R</sub> (cm/W) Raman coefficient gain	e (nm)	ng	g (m <sup>-1</sup> W <sup>-1</sup> ) Raman modal gain
PMMA + silicon	1	2957	1.42	3.7 10-10	200	1.49	0.3
$TiO_2 + air$	1.30	140	1.32	8.3 10-10	60	2.238	0.4
110 <sub>2</sub> + un	1.50	110	1.52	0.5 10	00	2.200	0.1

Tableau 1. Parameters used for the calculation of the modal Raman gains in ONF coated with PMMA and TiO2.

## 3. 2<sup>nd</sup> order nonlinearity: photon pair generation by Spontaneous Parametric Down Conversion

We have also studied PMMA/DR1 coated silica ONF for enhancing the second-order nonlinear susceptibility of the composite material and propose a novel source of correlated photon pairs based on SPDC [3]. We estimated photon pair generation spectral densities for two configurations: bare silica ONF and PMMA/DR1-coated silica ONF. We studied a modal phase matching scheme utilizing a  $TM_{01}$  pump mode at 775nm to generate photon pairs in the  $HE_{11}$  mode around 1550nm. Our simulations have shown a 1000-fold enhancement in photon pair generation efficiency with the coating compared to bare silica ONF.

## 4. Fabrication of the composite nanofibers

ONF were fabricated following the classical "pull and brush" technique to create the tapers and the uniform part [1]. We have developped a process to coat the ONF with PMMA (and PMMA/DR1) based on multilayer dip-coating to obtain thicknesses of a few hundreds of nm (see fig. 2a). We also performed the encapsulation of a PMMA coated ONF with silicone. We measured relatively low additional losses of 1.76 dB after the 2 processes, decreasing from 57% to 38%. For the coating with  $TiO_2$ , we have used Atomic Layer Deposition technique (see fig. 2b). The additional losses are only 0.5 dB on 2 cm.



Figure -2. (Left) MEB cross-section in a taper of an ONF coated with PMMA (e = 850 nm). (Right) MEB cross-section of an ONF coated with TiO<sub>2</sub> (e = 48 nm). TiO2 thickness can be accurately measured thanks to the partial delamination of the thin film during fiber cleaving.

## **5.** Conclusions and perspectives

The preliminary results presented in this study open the way to a new family of composite ONF for different applications in nonlinear optics. We have studied as examples the realization of all-solid Raman converters and new sources of pairs of photons for quantum communications. Many other experiments can be imagined thanks to the wide range of possible materials for coatings.

#### 6. References

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