

# Fiber Optics in the Multiphoton Ionization Regime

Mario Ferraro, Fabio Mangini, Yifan Sun, Mario Zitelli, Maria Caterina Crocco, Vincenzo Formoso, Raffaele Giuseppe Agostino, Riccardo Cristoforo Barberi, Antonio de Luca, Alessandro Tonello, et al.

# ▶ To cite this version:

Mario Ferraro, Fabio Mangini, Yifan Sun, Mario Zitelli, Maria Caterina Crocco, et al.. Fiber Optics in the Multiphoton Ionization Regime. 2022 Italian Conference on Optics and Photonics (ICOP 2022), Jun 2022, Trente, Italy. 10.1109/ICOP56156.2022.10044375. hal-04670712

# HAL Id: hal-04670712 https://hal.science/hal-04670712v1

Submitted on 12 Aug 2024

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

# Fiber Optics in the Multiphoton Ionization Regime

1st Mario Ferraro

Department of Information Engineering, Department of Information Engineering, Department of Information Engineering, Electronics and Telecommunications Sapienza University of Rome Via Eudossiana 18, Rome, Italy mario.ferraro@uniromal.it

## 4th Mario Zitelli

Department of Information Engineering, Electronics and Telecommunications Sapienza University of Rome Via Eudossiana 18, Rome, Italy mario.zitelli@uniroma1.it

7th Raffaele Giuseppe Agostino STAR Research Infrastructure and Physics Department University of Calabria Via Pietro Bucci, Rende, Italy raffaele.agostino@fis.unical.it

10th Alessandro Tonello XLIM Research Institute University of Limoges, UMR CNRS 123 Avenue Albert Thomas, 87000 Limoges, France alessandro.tonello@unilim.fr

2<sup>nd</sup> Fabio Mangini

Electronics and Telecommunications Sapienza University of Rome Via Eudossiana 18, Rome, Italy fabio.mangini@uniroma1.it

> 5th Maria Caterina Crocco STAR Research Infrastructure and Physics Department University of Calabria Via Pietro Bucci, Rende, Italy mariacaterina.crocco@unical.it

8th Riccardo Cristoforo Barberi STAR Research Infrastructure and Physics Department University of Calabria Via Pietro Bucci, Rende, Italy riccardo.barberi@fis.unical.it

11th Vincent Couderc XLIM Research Institute University of Limoges, UMR CNRS 123 Avenue Albert Thomas, 87000 Limoges, France vincent.couderc@xlim.fr

3rd Yifan Sun

Electronics and Telecommunications Sapienza University of Rome Via Eudossiana 18, Rome, Italy vifan.sun@uniromal.it

> 6th Vincenzo Formoso STAR Research Infrastructure and Physics Department University of Calabria Via Pietro Bucci, Rende, Italy vincenzo.formoso@unical.it

> 9th Antonio De Luca Physics Department University of Calabria Via Pietro Bucci, Rende, Italy antonio.deluca@unical.it

12th Stefan Wabnitz Department of Information Engineering, Electronics and Telecommunications Sapienza University of Rome Via Eudossiana 18, Rome, Italy stefan.wabnitz@uniroma1.it

Abstract—We investigate an extreme regime of nonlinear propagation in standard multimode optical fibers, where intense laser pulses trigger multiphoton ionization effects. These influence the dynamics of spatiotemporal solitons which are formed from the fission of input laser pulses. In particular, we observed a nonmonotonic Raman wavelength shift over a time scale of several hours. Moreover, the material ionization generates irreversible damages, which are visualized by means of optical microscopy and X-ray computed microtomography.

Index Terms-Optical fibers, Optical breakdown, Multiphoton ionization, Multimode fibers

#### I. INTRODUCTION

Nonlinear optical effects are generally associated with the reversible variation of the refractive properties of a material, as a result of the propagation of intense laser beams [1]. However, nonlinear optics may also involve absorption properties of materials, which lead to a class of phenomena which are

We acknowledge the supports of funding from European Research Council (740355), Sapienza University of Rome Grant (RG12117A84DA7437, AR22117A8AFEF609, AR22117A7B01A2EB), Italian Ministry of Education and Research (MIUR - R18SPB8227, PIR01-00008), and French National Agency for Research (ANR - 18-CE080016-01, 10-LABX-0074-01).

known as multiphoton absorption (MPA) effects. MPA is often neglected in glass materials, such as optical fibers, at variance with semiconductors. However, because of the need of scalingup the power of optical devices, such as fiber lasers, the contribution of MPA may eventually become significant at sufficiently high pulse peak powers. Although many studies have been reported about the MPA propagation regime in bulk and nanoparticles [2]-[4], first results on the impact of MPA on the propagation of ultrashort pulses in multimode optical fibers (MMFs) have been only recently reported [5].

Generally speaking, the appearance of MPA-related phenomena in glasses requires powers which may lead to damage the material. These phenomena may be due to different causes, and may give rise to material modifications. In particular, when the damages are caused by thermal effects, glasses become fully opaque, thus preventing any light propagation. Clearly, in this condition no investigation of beam propagation dynamics is possible. This happens when the duration of the laser pulses is relatively long, i.e., when operating with picosecond or quasi-continuous wave sources [6], [7]. Conversely, when using femtosecond laser pulses, thermal effects can be avoided

[8], thus facilitating the observation of MPA-induced beam dynamics. Material damages which are not caused by thermal effects are, instead, due to material ionization. The ionization phenomena driven by MPA are referred to as multiphoton ionization (MPI) effects. These lead to irreversible material modifications, which do not totally hamper its transparency to light. Although the MPI propagation regime has been widely studied in bulk glasses, studies of the ionization regime in optical fibers have been mainly limited to the case of photonic crystal fibers [9], where supercontinuum generation results from the ionization of the gas that fills the photonic crystal structure.

Here we study the propagation dynamics of femtosecond laser beams in the MPI regime of standard silica MMFs. The ionization of silica glass produces irreversible material damages. These are formed over a time scale of a few hours, thus giving rise to an unique propagation regime. Specifically, the optical transmission of the MMF is quenched, as long as the damaging process is on-going, and, only after a few hours, the light-fiber system reaches an equilibrium condition, whereby the fiber transmission no longer varies. The slow transmission drop affects the dynamics of spatiotemporal solitons, whose Raman-induced self-frequency shift (SSFS) progressively reduces, as the damaging process builds up. Experimental observations are in good qualitative agreement with numerical simulations based on the generalized multimode nonlinear Schrödinger equation model. Finally, the local modifications of the refractive index induced by MPI are analysed by optical microscopy and X-ray computed microtomography (μCT). In terms of applications, these results are of interest for the development of MMF lasers and MMF-based highpower devices, as well as for the study of new techniques for geometrically non-trivial glass micromachining.

## II. BEAM DYNAMICS IN THE MPI REGIME

#### A. Experimental observation

In order to trigger MPI, we use a femtosecond Yb-based laser (Lightconversion PHAROS-SP-HP), generating pulses of 174 fs at 100 kHz repetition rate and 1.03  $\mu$ m wavelength, with Gaussian beam shape. By means of an optical parametric amplifier (Lightconversion ORPHEUS-F), the laser wavelength and the pulse duration are converted to 1.55  $\mu$ m and 67 fs, respectively. The beam at the fiber output is characterized by means of a broadband thermopile power meter (Gentec XLP12-3S-VP-INT-D0) and a real-time multiple octave spectrum analyzer (Fastlite Mozza). In our experiments, we use a 30 cm span of standard 50/125 GRIN MMF (Thorlabs GIF50E).

The propagation in GRIN MMFs of high-energy femtosecond pulses of  $1.55 \mu m$  wavelength leads to the generation of multiple spatiotemporal solitons via the fission of the input pulse [10]. These solitons represent peculiar solutions of the coupled nonlinear Schrödinger equations, which arise from the compensation of fiber dispersion by nonlinearity. It has been recently shown that, owing to their intrinsic multimode nature, these solitons have unique features when carrying power close

to the value which leads to triggering MPI. For example, unlike singlemode solitons, the oputut soliton duration barely depends on their energy [10]. Because of the Raman effect, the solitons undergo SSFS, which increases as their energy grown larger [11]. However, the soliton wavelength cannot keep increasing indefinitely. Above a certain input pulse energy threshold, MPA becomes significant, and clamps the output soliton energy as well as its maximum wavelength, say  $\lambda_M$ . As long as the input pulse power is insufficient for triggering MPI, the material is not damaged, and the energy lost by the beam via MPA is dissipated by material defects [5]. The other way around, whenever the input power overcomes the MPI threshold, the material is irreversibly damaged, and the power transmission of the fiber slowly drops in time [12].

MPI is responsible for local variations of the local refractive index, that affect the beam dynamics. This is shown in Fig. 1a, where we report the measured output spectrum vs. time. In order to highlight the energy lost because of MPI, we used a second horizontal axis (placed on the top of the figure) that accounts for the output energy. We monitored the output power and spectrum for about 2 hours. As it can be seen by Fig. 1a, the beam dynamics exhibits a slow and nontrivial evolution, which is characterized by two different time scales. Indeed, a fast spectral variation occurs in the first 500 s, where  $\lambda_M$  decreases from 2.4  $\mu$ m down to 2.2  $\mu$ m; whereas slow variations take place over a time scale of the order of one hour, during which  $\lambda_M$  oscillates around 2.2 μm. Note that, although the most red-shifted solitons settles its wavelength after 2000 s, other spectral peaks at lower wavelengths keep varying their energy for much longer times. This indicates that, despite weaker, the MPI-induced damaging process is still on-going. Eventually, after about 2500 s, the output power remains stable; correspondingly, an equilibrium spectral distribution is reached.

#### B. Numerical simulations

Our experimental observations can be qualitatively reproduced by numerical simulations based on the generalized multimode nonlinear Schrödinger equation [13]. We took into account the self-steepening effect, Kerr and Raman nonlinearities, as well as chromatic dispersion (up to the fourthorder). Since SSFS pushes the soliton wavelength above 2 μm, where multiphonon absorption is relevant, we also include wavelength-dependent linear losses [12]. Indeed, the fiber loss coefficient increases by four orders of magnitude when  $\lambda$  grows from 1.55  $\mu$ m up to 2.1  $\mu$ m [14]. On the other hand, we did not include in simulations any nonlinear term for describing the presence of MPI, since its contribution to spatiotemporal beam dynamics acts over a time-scale which is much shorter than that associated with damages formation. For this reason, we mimicked the process of damage generation by just decreasing the energy of the input laser pulses.

The beam dynamics is strongly influenced by the fiber multimodality. Indeed, if in our input condition we consider only one mode, the Raman-induced SSFS produces a monotonic red-shift of  $\lambda_M$ . This can be seen in Fig. 1b, where we report the output spectrum vs. the laser pulse input energy. Conversely, whenever we take into account multiple modes, the Raman-induced SSFS does not monotonically increase as the input pulse energy grows larger. This is shown in Fig. 1c, where we report the cases of input conditions such that only the three radially symmetric lowest-order modes (expressed in the Laguerre-Gauss base) are populated. Indeed, the wavelength positions of the output spectral lobes move back and forth, as the input pulse energy grows larger, as in the experiments shown in Fig. 1a. Therefore, we may associate the input pulse energy with the experimental long time-scale of evolution of the output energy.

Still, when comparing Fig. 1a with Fig. 1c, one may notice that the latter shows some macroscopic features which are missing in the former, e.g., the occurrence of discontinuities of the soliton wavelength when increasing the input energy. This difference vanishes when considering non-radially symmetric modes. In Fig. 1d, in fact, it can be seen that the soliton SSFS does not present any discontinuity, when considering the six lowest-order modes only. In this regard, it is worth to underline that the non-monotonic trend of the SSFS can lead to the collision of spatiotemporal solitons [15], which is observed in the short time scale of evolution of the output spectrum, e.g., at 250 s in Fig. 1a. However, these are not captured by our numerical model, which provides an almost flat spectrum at input energies above 75 nJ (cfr. Fig. 1d). In order to describe soliton collision at such high values of input energy, it would be necessary to take into account much more modes, thus increasing the dynamics complexity and the computation time, which is beyond the scope of this work.

Finally, one may notice that the input energy range in Fig. 1b differs from that of Fig. 1c and 1d. This is because, when considering a single-mode excitation, the input energy is confined in a quite small portion of the fiber core (the fundamental mode waist is  $8.79~\mu m$ ). Therefore, the local intensity reaches values so high, that our numerical code diverges.

#### III. IMAGING OF THE REFRACTIVE INDEX MODIFICATION

In order to visualize the refractive index modification induced by MPI, we relied on the  $\mu$ CT technique, which has been recently proposed as a tool for retrieving the refractive index profile of MMFs [16]. For this purpose, we used a microfocus source (Hamamatsu L12161-07), which emits a conical polychromatic X-ray beam of 10 W average power and a maximum energy of 60 keV. Whereas, the intensity profile of the transmitted X-ray beam was measured with a flat panel detector (Hamamatsu C7942SK-05).

A reconstructed slice of the fiber section is shown in Fig. 2a. This was obtained by means of the software Fiji and Avizo [17]. In order to highlight the presence of damages, the  $\mu$ CT image has been post-processed. Specifically, we applied three different filters to the image intensity (see Fig. 2b). In that figure, each color accounts for different intensity intervals, i.e., to different zones inside the fiber volume. In particular, the yellow zone is enclosed inside the fiber core, and it contains

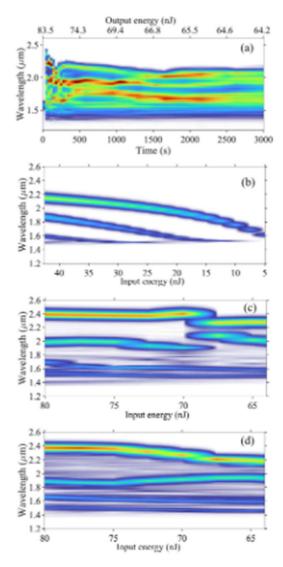


Fig. 1. (a) Experimental output spectrum as a function of time. The horizontal axis on the top of the figure represent the measured output energy. The laser pulse input energy is 103 nJ. (b-d) Numerically calculated output spectra for different input conditions. Specifically, in (b) only the fundamental mode is populated; in (c) the mode 1, 6, and 15, which are radially symmetric, have a population of 0.5570, 0.2878, and 0.1552, respectively; in (d) the population of the six lowest order modes is 0.3847, 0.1740, 0.0106, 0.0220, 0.0000, 0.4086, respectively.

the fiber axis; the red zone still belong to the core but it does not include the fiber axis; the blue zone is fully located inside the cladding.

Some features, indicating the presence of damages, could be detected from Fig 2c, which is a bi-dimensional reconstruction of the fiber longitudinal profile. Indeed, it can be appreciated that the red and green zones have rather flat edges. Whereas the most inner zone experiences local shrinkages, which are

associated with the presence of MPI-induced damages. This is further highlighted in Fig. 2d, where we plot the trend of the diameter variation of the three zones. Visibly, the yellow curve shows more pronounced spikes with respect to the other curves. This is ascribable to the fact that the red and green zones are far from the fiber axis, where the damages are induced.

The relationship between the variation of the size of the  $\mu$ CT zones and the damages can be understood by comparing Fig. 2d and Fig. 2e. The latter is an image of the fiber tip during the damaging process which was captured with a digital microscope (Dinolite-AM3113T). Indeed, one can see bright light spots (white and violet) at the self-focusing point and its self-image, respectively. The latter is generated because of the index-grading of the GRIN MMF [18]. The edges of the damages, whose position is determined by that of the luminescent spots in Fig. 2e, corresponds to the spikes in Fig. 2d. Indeed, since  $\mu$ CT is sensitive to refractive index mismatches, non-negligible oscillations of the yellow curve in Fig. 2d only appear at the edges of the damages.

As a final note, it is important to underline that the results in Fig. 2d and 2e do not refer to the same fiber. Indeed, the former was obtained when damaging the fiber with the laser source at 1.03  $\mu$ m of wavelength, whereas the latter was obtained by using the laser pulses at 1.55  $\mu$ m, i.e., those that provide the dynamics described in the previous section. Still, since the distance between the self-focusing point and its replica does not depend on the beam wavelength [19], the comparison between Fig. 2d and Fig. 2e is legitimate.

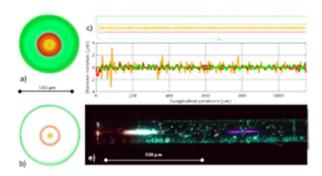


Fig. 2. Imaging of the MPI-induced damages. (a) μCT image of the GRIN MMF fiber transverse section. (b) Same as (a) after filtering post-process. (c) μCT image of the GRIN MMF fiber longitudinal section. (d) Trend of the diameter variation of the three zones (yellow, red, and green) in (c). (e) Digital microscope image of the tip of a GRIN MMF during the MPI damaging process.

### IV. CONCLUSION

We reported the experimental investigation of the nonlinear evolution of ultrashort pulses as determined by combining MPI with the complex dynamics of glass MMFs. We found a nontrivial evolution of the spectrum at the MMF output, which is accompanied with an irreversible loss of the fiber transmission. The experimental results found good qualitative agreement with numerical simulations The fiber damages produced by the MPI were imaged by means of optical microscopy and  $\mu$ CT. In perspective, our results will find applications to MMFbased high-power devices, such as MMF lasers, and to the development of novel methods for glass micromachining, and microstructured glass characterization at X-ray frequencies.

#### REFERENCES

- [1] Robert W. Boyd, Nonlinear optics. Academic press, 2020
- [2] Guang S. He, Loon-Seng Tan, Qingdong Zheng, and Paras N Prasad, "Multiphoton absorbing materials: molecular designs, characterizations, and applications," Chem. Rev., 108(4), 1245-1330 (2008).
- [3] Vaidya Nathan, Arthur H. Guenther, and Shashanka S. Mitra, "Review of multiphoton absorption in crystalline solids," JOSA B 2.2, 294-316 (1985).
- [4] Richard A. Farrer, Francis L. Butterfield, Vincent W. Chen, and John T. Fourkas, "Highly efficient multiphoton absorption induced luminescence from gold nanoparticles," Nano letters 5.6, 1139-1142 (2006).
- [5] M. Ferraro, F. Mangini, M. Zitelli, A. Tonello, A. De Luca, V. Couderc, and S. Wabnitz, "Femtosecond nonlinear losses in multimode optical fibers," Photon. Res. 9, 2443-2453 (2021)
- [6] B. C. Stuart, M. D. Feit, A. M. Rubenchik, B. W. Shore, and M. D. Perry, "Laser-induced damage in dielectrics with nanosecond to subpicosecond pulses," Phys. Rev. Lett., 74(12), 2248 (1995).
- [7] D. Du, X. Liu, G. Korn, J. Squier, and G. Mourou, "Laser-induced breakdown by impact ionization in SiO2 with pulse widths from 7 ns to 150 fs," Appl. Phys. Lett., 64(23), 3071-3073 (1994)
- [8] M. D. Perry, B. C. Stuart, P. S. Banks, M. D. Feit, V. Yanovsky, and A. M. Rubenchik, "Ultrashort-pulse laser machining of dielectric materials," J. Appl. Phys., 85(9), 6803-6810 (1999).
- [9] P. Hölzer, W. Chang, J.C. Travers, A. Nazarkin, J. Nold, N.Y. Joly, M. F. Salch, F. Biancalans, and P. St.J. Russell, "Femtosecond nonlinear fiber optics in the ionization regime," Phys. Rev. Lett. 107.20, 203901 (2011).
- [10] M. Zitelli, F. Mangini, M. Ferraro, A. Niang, D. Kharenko, and S. Wabnitz, "High-energy soliton fission dynamics in multimode GRIN fiber," Opt. Express 28, 20473-20488 (2020)
- [11] J. Santhanam, G. P. Agrawal, "Raman-induced spectral shifts in optical fibers: general theory based on the moment method," Optics Communications, 222, 413-420 (2003)
- [12] M. Ferraro, F. Mangini, Y. Sun, M. Zitelli, A. Niang, M. C. Crocco, V. Formoso, R. G. Agostino, R. Barberi, A. De Luca, A. Tonello, V. Couderc, S. A. Babin, and S. Wabnitz, "Multiphoton ionization of standard optical fibers," Photon. Res. 10, 1394-1400 (2022)
- [13] L. G. Wright, Z. M. Ziegler, P. M. Lushnikov, Z. Zhu, M. A. Eftekhar, D. N. Christodoulides, and F. W. Wise, "Multimode Nonlinear Fiber Optics: Massively Parallel Numerical Solver, Tutorial, and Outlook," IEEE Journal of Selected Topics in Quantum Electronics, vol. 24, no. 3, pp. 1-16, May June 2018
- [14] G. P. Agrawal, "Nonlinear fiber optics," Nonlinear Science at the Dawn of the 21st Century, Springer, Berlin, Heidelberg, 195 211 (2000)
  [15] Y. Sun, M. Zitelli, M. Ferraro, F. Mangini, P. Parra-Rivas, and S.
- [15] Y. Sun, M. Zitelli, M. Ferraro, F. Mangini, P. Parra-Rivas, and S. Wabnitz, "Multimode soliton collisions in graded-index optical fibers," Opt. Express 30, 21710-21724 (2022)
- [16] M. Ferraro, M. C. Crocco, F. Mangini, M. Jonard, F. Sangiovanni, M. Zitelli, R. Filosa, J. J. Beltrano, A. De Luca, R. C. Barberi, R. G. Agostino, V. Coudere, S. Wabnitz, V. Formoso, "X-ray computed μ-tomography for the characterization of optical fibers," Opt. Mat. Exp., in press.
- [17] J. Schindelin, I. Arganda-Carreras, E. Frise, V. Kaynig, M. Longair, T. Pietzsch, S. Preibisch, C. Rueden, S. Saalfeld, B. Schmid, J.Y. Tinevez, D.J. White, V. Hartenstein, K. Eliceiri, P. Tomancak, and A. Cardona, "Fiji: an open-source platform for biological-image analysis," Nat. Meth., 9(7), 676-682 (2012).
- [18] F. Mangini, M. Ferraro , M. Zitelli, A. Niang, A. Tonello, V. Coudere, and S. Wabnitz, "Multiphoton-Absorption-Excited Up-Conversion Luminescence in Optical Fibers," Phys. Rev. Applied 14, 054063 (2020).
- [19] T. Hansson, A. Tonello, T. Mansuryan, F. Mangini, M. Zitelli, M. Ferraro, A. Niang, R. Crescenzi, S. Wabnitz, and V. Couderc, "Nonlinear beam self-imaging and self-focusing dynamics in a GRIN multimode optical fiber: theory and experiments," Opt. Express 28, 24005-24021 (2020)