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Review of the PhD Thesis:
Widely-Tunable All-Fiber Laser Source for Coherent Raman Scattering Microscopy
by M.Sc. Cassia Corso Silva, M.Sc

The doctoral dissertation of Ms. Cassia Corso Silva, M.Sc., titled *"Widely-Tunable All-Fiber Laser Source for Coherent Raman Scattering Microscopy,"* was prepared as part of the Warsaw-4-PhD doctoral program at the Institute of Physical Chemistry of the Polish Academy of Sciences, under the supervision of Prof. dr hab. Yuriy Stepanenko and Dr. eng. Katarzyna Krupa.

The dissertation is experimental in nature and represents a mature, interdisciplinary approach to topics at the intersection of photonics, fiber laser engineering, nonlinear fiber optics, and advanced biomedical imaging. It presents the design, development, and validation of three novel, widely-tunable all-fiber laser sources capable of operating across both the fingerprint and C-H stretch regions of the Raman spectra.

Particularly noteworthy is the implementation of innovative spectral broadening techniques based on Self-Phase Modulation (SPM) and Degenerate Four-Wave Mixing (D-FWM) in chirped pulse regimes—entirely in fiber-based configurations. These approaches demonstrate not only high performance but also practical advantages such as robustness, compactness, and environmental stability, making them suitable for real-world biomedical applications.

A significant highlight of the work is the successful integration of these laser systems into Stimulated Raman Scattering (SRS) and Coherent Anti-Stokes Raman Scattering (CARS) microscopes, achieving high-resolution, label-free imaging of biological samples such as leukemic cells and polystyrene beads. This directly supports the potential of these sources in clinical diagnostics and advanced biomedical imaging.

The first developed laser source is a tunable dual-wavelength system for SRS imaging, based on a Yb-doped fiber oscillator mode-locked using a Nonlinear Optical Loop Mirror (NOLM). Tunable pump and Stokes beams were generated through spectral filtering—of a supercontinuum (SC) produced in photonic crystal fiber (PCF) for the pump, and of the oscillator output for the Stokes. The Stokes beam's tunability was further extended using a novel all-fiber approach, employing self-phase modulation (SPM) of chirped pulses. Both pump and Stokes spectra were filtered and finally amplified using appropriate fiber amplifiers. The laser system covers the fingerprint region, i.e., the spectral range from 950 cm^{-1} to 1600 cm^{-1}.

The second all-fiber laser source developed is also based on a NOLM-mode-locked Yb-doped fiber oscillator. However, it offers even broader tunability by utilizing the Degenerate Four-Wave Mixing (D-FWM) effect, which is generated and enhanced within a fiber optical parametric oscillator (FOPO) ring cavity. This tunability is achieved by adjusting the central wavelength of the FOPO pump. By combining the tunable D-FWM beam with the oscillator beam fixed at 1030 nm, Raman measurements could be performed across an exceptionally wide spectral range—from 929 cm^{-1} to 3990 cm^{-1}.

The third developed light source, which is the simplest in terms of design, is based on a commercial ultrafast fiber laser operating at a fixed wavelength of 1030 nm. As the laser pulses propagate through a polarization-maintaining fiber (PMF) followed by a microstructured (photonic) fiber (PCF), the D-FWM phenomenon occurs as a result of self-phase modulation (SPM). The wavelength shift of the frequency sidebands in the D-FWM spectrum is controlled by adjusting the chirped pulse duration of the pump beam. As a result, when combined with the pump beam, this laser source enables Raman measurements in the range of 873 cm^{-1} to 3624 cm^{-1} or from 1512 cm^{-1} to 3738 cm^{-1}, using the D-FWM signal or idler beam, respectively.

The doctoral dissertation of Ms. Silva, M.Sc., was written in English and comprises lists of publications, figures, tables, and acronyms, a total of 125 pages. It contains 74 figures and 3 tables. The thesis begins with an Introduction, followed by the main part consisting of 6 chapters. The dissertation concludes with a bibliography and 2 appendices.

In the 3-page Introduction (**Chapter 1**), the author presents the motivation and objective of the work, specifically: the development of widely tunable, all-fiber laser radiation sources for use in Raman microscopy systems—specifically SRS and CARS. The Introduction also outlines the structure of the entire dissertation. **Chapter 2**, spanning 10 pages, provides a concise overview of the principles of light propagation in optical fibers. It also discusses various types of optical fibers and the key parameters that characterize them. **Chapter 3**, which spans 11 pages, presents—at very general level—the fundamental nonlinear optical effects that can occur in optical fibers, such as self-phase modulation, four-wave mixing, optical wave breaking and supercontinuum generation. **Chapter 4**, comprising 11 pages, covers the fundamentals of Raman optical microscopy. It outlines the different types of Raman scattering and explores the various detection methods utilized in this technique. The next three chapters (**Chapter 5 to 7**) form the core, original part of this doctoral dissertation. These chapters describe the design, operation, and performance parameters of the three developed laser sources. Finally, the 3-page **Chapter 8** provides a summary of the dissertation. It highlights the main achievements of the work and discusses potential improvements to the developed light sources, as well as possible directions for their future development.

The strongest aspects of the dissertation include:

• **Innovative approach:** The author developed three different concepts for tunable laser sources in a fiber-optic configuration, based on nonlinear optical effects such as self-phase modulation (SPM) and degenerate four-wave mixing (D-FWM). Each of the presented systems was thoroughly described theoretically, experimentally built, commissioned and tested in a practical context.

• **Practical application:** The systems were successfully integrated with SRS and CARS microscopes, enabling imaging of both biological and synthetic material, exemplified by leukemia cells and polystyrene beads, respectively. The obtained images and Raman spectra demonstrate the high effectiveness of the sources in biological applications.

• **Attention to experimental validation:** The dissertation contains a rich set of experimental results characterizing the developed light sources, some of which are supported by numerical simulations. This reflects well on the author's competence in both experimental work and data analysis.

• **Publication record:** The author can boast three scientific publications (including in recognized journal *Optics Letters*), numerous presentations at international conferences, and co-authorship of patent applications – all of which confirming both the originality of the research and its implementation potential.

• **Editorial diligence:** The text of the dissertation is logically structured, written in scientific yet accessible language, with attention to clarity of argument and terminological accuracy. The graphical elements (schematics, charts, images and photos) are mostly well-prepared and support the understanding of the content.

Although the substantive level of the dissertation is very high and the presented results are original and convincingly documented, it is worth pointing out several aspects that may be considered potential limitations of the work or areas that require further exploration in future research:

1. **Simplified model in numerical simulations:** In the theoretical part concerning nonlinear simulations of pulse propagation phenomena in optical fibers, a scalar form of the generalized nonlinear Schrödinger equation (GNLSE) was used. Although this approach is commonly employed to simplify computations, the omission of polarization may be significant when performing precise analysis of nonlinear effects, especially in optical fibers with strong anisotropic properties, such as the LMA-PM-5 photonic fibers used in this work. Extending the model to include a full vectorial analysis could for instance improve the accuracy of spectral sideband predictions and better reflect the influence of birefringence.
2. **Lack of full quantitative validation of simulation models:** The work includes valuable qualitative comparisons between experimental results and simulations; however, the author did not present a complete quantitative validation of the numerical models by directly comparing simulation results with experimental outcomes (e.g., in terms of the spectral locations of sidebands or the intensities of D-FWM signals). Including thorough quantitative validation of the models would enhance their credibility.
3. **Complexity of source synchronization and experimental repeatability:** The synchronization of pump pulses and the generation of the appropriate D-FWM spectrum required very precise adjustment of fiber lengths and timing synchronization (e.g., in FOPO or CARS setups). The thesis does not present an analysis of the repeatability of these measurements or information about system tolerances, which could be important when planning practical implementations.
4. **Lack of in-depth comparative analysis with existing solutions:** Although the introduction outlines the general advantages of fiber-based systems over systems based on nonlinear crystals, it lacks a more detailed comparative analysis of

parameters such as power, tuning efficiency, noise levels, and long term reliability relative to currently available commercial or laboratory systems. Such an analysis could further emphasize the advantages of the proposed solutions.

5. **Scope of practical application:** Although the application in Raman microscopy was convincingly demonstrated, the thesis lacks a broader discussion of potential implementation barriers, such as production costs, miniaturization, or integration with existing clinical platforms.
6. **Clear representation of the ongoing processes and the resulting spectral changes:** Due to the highly complex optical systems of the developed light sources and engaged numerous nonlinear optical effects—I believe it would be helpful for readers if simplified diagrams were included for each source. These diagrams should illustrate the spectral changes at each key stage, particularly after optical elements that significantly alter the spectral characteristics of propagating pulses.

In fulfilling my duties as a reviewer, I feel obliged to address some errors and inaccuracies identified during the review of the thesis, which I have detailed below.

page 31, equation (2.1):	Instead of the refractive index n_1, it should be n, which refers to the medium surrounding the optical fiber.
page 36, Fig. 2.5(c):	The shading in the drawing does not correspond to the distribution of the refractive index in the fiber core.
page 40, line 3 from the bottom:	Instead of "of index h", it should say "of the n-th order"
page 41, line 1:	Instead of "We will simplify an approach by considering such tensors as simple coefficients…", it should say "We will simplify the approach by considering $\chi^{(n)}$ as scalar quantities…"
page 43, line 3:	Instead of "input pulse power", it should say "input pulse intensity"
page 44, line 9, page 45, line 10:	"mode index" should be replaced with "mode refractive index"
page 47, line 8:	The author writes: "*Considering a CW beam propagating in a lossless optical fiber, where the NLS equation governs the behavior of the pulse, given by [26]:*". In fact, the equation does not refer to a continuous (CW) laser beam, but rather to sufficiently long pulses, longer than many optical cycles – let's say around 100 fs.
page 65, Figure 5.3(d):	Looking at the diagram, I wonder what is the short- and long-term stability of the power and wavelength (expressed, for example, by the standard deviation) of the presented pulses?
page 66, lines 15-16:	The author writes "*As a result, a Stokes beam tunable from 1023 nm to 1036 nm was achieved, covering the entire spectral range of the initial fiber oscillator. The beam had a spectral bandwidth of 2.0 nm and an average power of up to 100 mW at each wavelength, as shown in panel (a) of Figure 5.5. The pulse duration was measured to be approximately 12.2 ps (see Figure 5.5b), which resulted from adding a 200 m long segment of fiber (Coherent, PM980-XP) before the first amplifier. This modification was made to minimize undesired spectral broadening.*"

	I wonder whether the length of this fiber segment was somehow optimized and the effect verified?
page 82, lines 5:	Looking at Figure 6.2, the long-wavelength limit of the idler beam is about 1140 nm, not 1089 nm as stated.
page 86, Figure 6.7(a-b)	The presented graphs would be much clearer if they were shown in the form of a color map.
page 86, Figure 68(a-b)	I believe that due to the large differences in power values, the data should be presented on a logarithmic scale.
page 89, Figure 6.13:	It seems to me that the legend should include the resonance cavity lengths of the OPO, not the power of the generated beam.
page 90, line 4:	I would consider the statement of excellent agreement between the simulation and the experiment to be too strong; the agreement is simply good.
page 94, line 3 from the bottom:	Tuning the chirped pulse duration from 4 ps to 30 ps, λ_{SPM} varies from 1041 nm to 1035 nm, not the other way around.
page 99, paragraph 7.3.2	Why were the numerical simulations performed for a pump beam power of 31 kW, instead of the 10 kW used in the experiment?
Page 103, equation (7.5):	It is not the definition of the pulse spectrum, but rather of the frequency Fourier transform of its amplitude.

Despite the above remarks, which are constructive in nature and do not undermine the scientific value of the work, the dissertation by Ms. Cássia Corso Silva is distinguished by a high level of academic and research quality. The author has demonstrated scientific independence, great proficiency in conducting experiments, and an excellent understanding of complex optical phenomena. Her work significantly expands the capabilities of Raman microscopy, steering it toward more compact, cost-effective, and potentially implementable light sources for clinical practice.

I hereby certify that the doctoral dissertation of Ms. Cássia Corso Silva, meets the requirements for doctoral dissertations and request that the Author be admitted to further stages of the PhD process.

Kraków, 28.04.2025