dr hab. inż. Karol Tarnowski
Department of Optics and Photonics
Faculty of Fundamental Problems of Technology
Wroclaw University of Science and Technology
Wybrzeże Wyspiańskiego 27
50-370 Wrocław
e-mail: karol.tarnowski@pwr.edu.pl
phone: +48 71 320 41 17

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Report on the doctoral thesis „*Widely-Tunable All-Fiber Laser Source for Coherent Raman Scattering Microscopy*” submitted by MSc Cássia Corso Silva

The thesis submitted by Ms. Cássia Corso Silva reports the development and characterization of three laser sources designed for application in coherent Raman scattering (CRS) microscopy. This imaging technique enables the acquisition of high-resolution images and the label-free identification of chemical compounds. At the same time, it requires dual-wavelength operation of the laser source. The cornerstone of the thesis is the exploitation of nonlinear optical phenomena taking place in optical fibers to achieve broad spectral tunability of generated bands needed for CRS microscopy. The work was carried out under the supervision of prof. dr. hab. Yuriy Stepanenko at Institute of Physical Chemistry, Polish Academy of Sciences. Dr. eng. Katarzyna Krupa acted as the auxiliary supervisor.

The aim of this work was to build widely tunable sources for CRS microscopy, as clearly presented in the introductory chapter (Chapter 1). The PhD candidate formulated three research questions:

> *“1. Is it possible to build a tunable fiber light source that enables measurements in the fingerprint Raman region based on supercontinuum generation in the normal dispersion regime?*
>
> *2. Is it possible to build an optical parametric oscillator in an all-fiber configuration that enables a broad tunability range through the four-wave mixing nonlinear effect when pumped with a tunable laser system based on a fixed wavelength oscillator?*
>
> *3. Is it possible to achieve broad tuning of four-wave mixing sidebands when pumped with laser systems at a fixed wavelength?”*

In the following three chapters, Ms. Cássia Corso Silva presents the theoretical background for optical fibers (Chapter 2), nonlinear optical phenomena (Chapter 3), and coherent Raman spectroscopy (Chapter 4). Next – in Chapters 5 to 7 –the development of the tunable light sources and their application in the CRS imaging systems are reported. The thesis is concluded with a summary presented in Chapter 8.

Chapter 2 explains the principle of operation of optical fibers, describes their key properties: attenuation and chromatic dispersion; and presents different types of optical fibers. Finally, it introduces the two photonic crystal fibers used in the studies. In my view, this description is over simplified, which has led to the following inaccuracies.

(i) The definitions of incidence angle, refracted angle and critical angle in the text are not consistent with the symbols used in Figure 2.1.

(ii) The attenuation constant α in equation 2.6 is not expressed in dB/km as stated in the text. The relation between α and α_{dB} is missing.

(iii) The definition of M used in equation 2.15 is missing.

(iv) The shading used for graded index fiber in Figure 2.c is incorrect.

Chapter 3 provides the concise description of nonlinear phenomena in optical fibers. It starts by introducing Kerr nonlinearity, goes through self-phase modulation, optical wave breaking, four-wave mixing, stimulated Raman scattering and reaches supercontinuum generation. This description focuses on the nonlinear effects which are important in the context of presented research and is sufficient to follow the further parts of the thesis. In this part, I noticed the following flaws.

(i) The expressions for instantaneous optical frequency given in equations 3.17 and 3.18 are not consistent with the data presented in Figure 3.1 due to sign.

(ii) The sentence preceding equation 3.29 is incorrect. In both types of four-wave mixing (degenerated and non-degenerated) the energy of two photons is transferred to two other photons.

Chapter 4 presents the principles of spectroscopic measurements using spontaneous Raman scattering, stimulated Raman scattering and coherent anti-Stokes Raman scattering. This chapter is valuable because it gives an important application context for the results presented in further chapters that report the development of the laser sources.

The description of the first laser source is presented in Chapter 5. The developed source is based on mode-locked Yb-doped fiber oscillator operating around 1030 nm. In the pump arm, the supercontinuum is generated in all-normal dispersion fiber and subsequently filtered and amplified, resulting in the pump band tunable from 913 nm to 930 nm. The Stokes arm uses efficient spectral broadening of chirped pulses by self-phase modulation to achieve tunability range from 1020 nm to 1070 nm. I greatly appreciate the implementation of this novel all-fiber approach, which was critical for obtaining broad tunability of Stokes band.

The development of the second laser source is reported in Chapter 6. The source uses degenerated four-wave mixing in fiber optical parametric oscillator. The developed source generates pulses tunable in range from 730 nm to 940 nm enabling broadband spectroscopic measurements.

Finally, the last laser source is described in Chapter 7. In this source degenerated four-wave mixing is also used and the tunability of generated bands is achieved by chirping the pump pulses. With the fixed spectral position of pump (1030 nm) the tunability of four-wave mixing bands was achieved in range 750 nm – 945 nm (for signal band) and 1220 nm – 1675 nm (for idler band). I highly appreciate the application of the proposed innovative approach to achieve such broad tunability. Chapters 5 and 7 present also the implementation of developed fiber sources in coherent anti-Stokes Raman scattering microscopes.

The thesis is closed with conclusions and perspectives presented in Chapter 8. In my view, it lacks explicit reference to the research questions stated in the introduction. This is particularly striking, as answers to all questions are positive as was proven on the course of the thesis.

In my opinion, Ms. Cássia Corso Silva defined in the thesis the current research problem – the design and development of tunable fiber laser sources – and presented an original solution of this problem. A distinguishing feature of this thesis is its comprehensive scope, spanning from the observation of fundamental phenomena – documented in the published scientific paper [Optics Letters 48(21): 5531, (2023)] – through the practical implementation of a functioning device to patent applications [P.444762, EP24174041]. Ms. Cássia Corso Silva is the first author of the manuscript and patent applications what proves her leading role in the presented research.

Moreover, the presented practical implementations make the connection to the chemical sciences as a discipline of science of the thesis. However, the presented research would perfectly fit the discipline of physical sciences.

Finally, I have a few minor remarks.

(i) No information is provided about the mechanisms inducing birefringence in the used polarization-maintaining fibers. Was birefringence considered in the mode analysis presented in Chapter 6?

(ii) Paragraph indentation is inconsistent throughout the thesis, and indentation after equations is incorrect.

(iii) Bibliography formatting should be unified: author names appear inconsistently, and abbreviations are set in lowercase.

Summarizing, it was a challenge for Ms. Cássia Corso Silva to present her research – comprehensive in scope – in concise dissertation. I would like to acknowledge her efforts to keep the thesis easy to follow. Still, the dissertation proves the expertise of Ms. Cássia Corso Silva in the chemical sciences (particularly, in the field of Raman spectroscopic imaging) and confirms the ability to conduct independent scholarly research. The results presented in the thesis constitute an original solution to a scientific problem. In my opinion, the presented thesis meets all the requirements specified by the Act on Academic Titles and Degrees (article 187) and may serve as a basis for applying for the doctoral degree in the discipline of chemical sciences. Consequently, I evaluate the dissertation positively and recommend that Ms. Cássia Corso Silva be admitted to the subsequent stages of the doctoral proceedings.

Karol Tarnowski