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REVIEW
of Michal Kamrádek doctoral dissertation entitled
"Preparation and characterization of rare-earth-doped optical fibers for fiber lasers operating around 2 micrometers"

The review concerns Michal Kamrádek doctoral dissertation entitled "Preparation and characterization of rare-earth-doped optical fibers for fiber lasers operating around 2 micrometers", prepared under the supervision of assoc. prof. Ing. Pavel Peterka, Ph.D. (Institute of Photonics and Electronics Czech Academy of Sciences).
Supervision Specialists: prof. Ing. Václav Kubeček, DrSc. – Guarantor (Czech Technical University in Prague, Faculty of Nuclear Sciences and Physical Engineering, Department of Physical Electronics) and Dr. Ing. Ivan Kašík – Consultant (Institute of Photonics and Electronics, Czech Academy of Sciences).
The review was prepared at the request of doc. Ing. Vaclav Cuba, Ph.D. the Dean of the Faculty of Nuclear Sciences and Physical Engineering (the letter Ref. No.: 7006-V1/22/14922/Zab).

1. Introduction

Silica optical fibers definitely revolutionized our world enabling the control of light propagation. This applies to broadband telecommunications and laser radiation sources – both awarded with the Nobel Prize. This recognition is due to the important features of the fiber, such as: laser generation in the continuous and pulsed operation regime, scalability to the level of hundreds of kW and fs pulses with >100 mJ energy, excellent quality of the laser beam M^2=1.1 and broadband emission sources ability. In results the unique optical fiber design allows for numerous applications not only in the field of telecommunications, but above all in industry, metrology and medicine. In particular, the last two are related to the doctoral dissertation because they concern the area of radiation above 1.5 μm, defined as the eye-safe range.

2. Layout

Numbering 125 pages, the work is formulated in a classical way i.e. containing theoretical (chapters: 1. Introduction and 2. State of the art) and experimental (chapters: 4. Methods, 5. Results and 6. Conclusions) parts. Besides the dissertation also includes: the objectives of the thesis (chapter 3), list of abbreviations, references (chapter 7) and list of author's publications (both related and outside of the thesis).
In the theoretical part, which is less than 25% of work, after a clear introduction to the general scope which is improving 2 μm emission in optical fibers doped with Tm and Ho ions, the second chapter presents the last achievements in optical fiber technology. Author pointed out the requirements for the modification of silica glass in the MCVD method and indicated other methods such as crucible or sol-gel, including also the latest REPUSIL (reactive powder

sintering) one. He described RE doping problems in the MCVD method, paying attention to the advantages and disadvantages of the Solution Doping (SD) and Chelate Doping (CD) methods, taking into account the latest technique of embedding nanocrystals with lanthanides (2.2-2.3). The second essential section describes the conditions for obtaining a laser action in the 2 μm range using Tm and Ho ions. Author cites here the latest achievements concern to power and efficiency pointing out energy level diagrams of holmium and thulium discussing in detail the energy transitions processes taking place in thulium (2.4-2.6).

Author stated that despite the present achievements in the optimization of silica fibers doped with thulium and holmium, these issues still require research, especially in the field of 1) searching for new RE doping methods (like Nanoparticles Doping - ND) for silica fibers, 2) optimization of glass composition and obtaining higher quantum efficiency within Ho ions and 3) studying the energy transfer (ET) coefficients in optical fibers doped with Tm.

The theoretical background is presented clearly and is strictly related to the subject of the conducted research. The analysis of the state of the art was made on properly selected, up-to-date literature covering 129 positions (138 in total). The substantive basis of the work is the series of seven publications, presented in full version as the research results in the following chapters:

5.1. Nanoparticle and solution doping for efficient holmium fiber lasers

[1] M. Kamrádek, I. Kašík, J. Aubrecht, J. Mrázek, O. Podrazký, J. Cajzl, P. Vařák, V. Kubeček, P. Peterka, P. Honzátko, "Nanoparticle and solution doping for efficient holmium fiber lasers," IEEE Photonics Journal 11(5), 1-10 (2019). doi:10.1109/JPHOT.2019.2940747

5.2. Holmium-doped optical fibers for efficient fiber lasers

[2] M. Kamrádek, I. Kašík, J. Aubrecht, J. Mrázek, O. Podrazký, J. Cajzl, P. Vařák, V. Kubeček, P. Peterka, P. Honzátko, "Holmium-doped optical fibers for efficient fiber lasers," in Proceedings of SPIE 11355, Micro-Structured and Specialty Optical Fibres VI, 113550C (2020). doi: 10.1117/12.2559176

5.3. Holmium-doped fibers for efficient fiber lasers at 2100 nm

[3] M. Kamrádek, J. Aubrecht, M. Jelínek, M. Frank, P. Peterka, P. Honzátko, J. Mrázek, P. Vařák, O. Podrazký, F. Todorov, V. Kubeček, and I. Kašík, "Holmium-doped fibers for efficient fiber lasers at 2100 nm," in OSA Highbrightness Sources and Light-driven Interactions Congress 2020, OSA Technical Digest, paper MTh3C.5 (2020). doi: 10.1364/MICS.2020.MTh3C.5

5.4. Spectral properties of thulium doped optical fibers for fiber lasers around 2 micrometers

[4] M. Kamrádek, J. Aubrecht, P. Peterka, O. Podrazký, P. Honzátko, J. Cajzl, J. Mrázek, V. Kubeček and I. Kašík, "Spectral properties of thulium doped optical fibers for fiber lasers around 2 micrometers," in Proceeding of SPIE 10232, Micro-structured and Specialty Optical Fibres V, 1023205 (2017). doi:10.1117/12.2265697

5.5. Energy transfer coefficients in thulium-doped silica fibers

[5] M. Kamrádek, J. Aubrecht, P. Vařák, J. Cajzl, V. Kubeček, P. Honzátko, I. Kašík, P. Peterka, "Energy transfer coefficients in thulium-doped silica fibers," Optical Materials Express 11(6), 1805-1814 (2021). doi: 10.1364/OME.427456

5.6. Broadband thulium-doped fiber ASE source

[6] J. Aubrecht, P. Peterka, P. Honzátko, O. Moravec, M. Kamrádek, I. Kašík, "Broadband thulium-doped fiber ASE source," Optics Letters 45(8), 2164-2167 (2020). doi: 10.1364/OL.389397

5.7. All fiber mode-locked thulium-doped fiber laser using a novel femtosecondlaser-inscribed 45°-plane-by-plane-tilted fiber grating

[7] K. Nithyanandan, A. Theodosiou, J. Aubrecht, P. Peterka, M. Kamrádek, K. Kalli, I. Kašík, P. Honzátko, "All fiber mode-locked thulium-doped fiber laser using a novel femtosecond-laser-inscribed 45°-plane-by-plane-tilted fiber grating," Laser Physics Letters 16(9), 095104 (2019). doi: 10.1088/1612-202X/ab39db

In above publications [1-5], the Michal Kamrádek is the first and corresponding author. The work for each publication contains detailed information on his role in the conception, execution and analysis of the research carried out. The articles were divided thematically, discussing holmium [1-3] and thulium [4-7] doped optical fibers, maintaining the logical continuity of the following steps from the optimization of the preform process, through their characterization and drawing of the optical fiber and application works on laser action and broadband source. It should be emphasized that the research scope presented in the publications is in line with the attached description, emphasizing the author's contribution.

The contribution of the student's own research and coherence of the published articles and the research topic of the dissertation

Author's contribution in the presented publications has been identified before each of them. His contribution relay on: preparation and characterisation of the preforms, their preparation for drawing optical fibers, participating in characterization of optical fibers, including measurement of the fluorescence decay curves and modelling. The author contributed to writing of the publications. All pointed out contributions are reflected to the proposed the goal of the thesis and results described in the conclusions. He was a key person who optimised preform compositions according to the settled goals and proposed their modifications as a feedback to the obtained results (it is also clearly seen in perspectives).

Undoubtedly, publications presented as the substantive scope of the work are coherence in line with the doctoral thesis.

In conclusion the structure of PhD dissertation makes it possible to evaluate Michal Kamrádek's achievements.

3. Evaluation

The choice of the research topic and the quality of the formulation, research target and methodological approach

In general the choice of a PhD topic concerns the work on silica optical fibers sources emitting in the range of 2μm. The major scope relays on optimisation of thulium and holmium doping using MCVD method, monitoring the spectroscopic properties of final optical fibers. From the scientific and utilitarian point of view, lasing and broadband emission in the 2 μm range is constantly developed due to metrological and medical applications (lasers, OCT). However the achievements in this area are very limited comparing with 1 μm range. It is caused by complex energy scheme of Tm ions (Cross-Relaxation - CR, Up-conversion phenomenon - UP) and difficulties in power scaling of holmium-doped lasers.

In case of the RE doping in MCVD – someone can say that it is well known method, but notice that it is incompatible with the number of research centres capable to produce active preforms with high quality spectroscopic properties leading to the development of new optical fiber sources. Were it not for the fact that the PhD student works in one of the most experienced research teams (UFE, ACR) in the field of active optical fibers production using the MVCD method, the implementation of the research goals would certainly not be possible.

Belong to them the following topics:

„ - Investigation and development of solution-doping and nanoparticle-doping methods for preparation of preforms for active optical fibers doped separately with thulium and holmium.
- Comparison of both doping methods from the point of view of spectroscopic parameters and laser characteristics of the prepared optical fibers.
- Finding key material parameters of holmium-doped fibers for highly efficient lasers.
- Determination of energy transfer coefficients between thulium ions in silica fibers eligible for numerical models of fiber lasers."

The above tasks respond to the challenges and shortcomings related to the achievements in the range of 2 μm fiber lasers. In particular, the new aspects include:

- optimization of the Nanoparticle Doping - ND and comparison with the currently used Solution Doping - SD method,
- analysis of wide range of Al/RE ratio and RE concentration on the example of holmium ion,
- critical analysis of key technological parameters (SD, ND) in relation to spectroscopic properties (lifetimes, efficiency, laser threshold) of optical fibres,
- optimisation of holmium optical fiber versus high quantum efficiency and low threshold of lasing,
- determination of energy transfer (ET) coefficients as a result of the analysis of the lifetime of optical fibers with different concentrations of thulium ions,
- applications of optimised Tm doped optical fibers for lasing and broadband sources.

In order to implement the above assumptions, Author used properly selected technological and metrological methods in the field of laser spectroscopy. He obtained active preforms by SD and ND methods in a wide range of RE and Al_2O_3 concentrations. Electron probe microanalysis (EPMA) and Photon Kinetics A2600 refractive index analyser were used to characterize the preforms. Silica optical fibers were produced on typical 7-meter-high drawing tower and characterised using the following measurements:

- absorption background in the range from 400 nm - 1600 nm (10–30 $dB \cdot km^{-1}$) and OH^- content (2–5 molar ppm) - cut-back method,
- rare-earth concentration determined due to measurement of Ho and Tm absorption bands - Nicolet 8700 FT-IR spectrometer,
- refractive index profile of fibers - IFA-100 (Interfiber Analysis) analyzer,
- lifetime measurements of Tm and Ho doped fibers – developed setup with side detection from 1mm length of fiber,
- laser parameters - developed setup with Fabry-Pérot resonator arrangement.

Technological and metrological methods were selected correctly and they belongs to the advance techniques in the field of optical fiber fabrication, characterisation and application.
Summing up the aim of the work was directed to answer new and unsolved issues concerned with technology and application of Tm and Ho silica optical fibers for 2 μm emission range.
In my opinion the assumptions, aims and methodology of the work have been formulated correctly.

The significance, originality and novelty of the research conducted by the student and the international quality of the results within the research field

The main achievement of Michał Kamrádek's is optimisation of Tm and Ho doped preforms and then optical fibers with simultaneous modification of RE concentration and

Al_2O_3/RE ratio. First SD method was used to develop 7 preforms with content of Al_2O_3 (1.26-4.30 mol%) and Ho ions (501-3184 ppm) obtaining Al/Ho concentration ratio in the wide range of 10-113 (Table 1 [1-3]). In the case of the ND method, 9 preforms were created with the following parameters: content of Al_2O_3 (2.35-5.92 mol%) and Ho ions (433-4687 ppm), Al/Ho concentration ratio 8.6-118 (Table 1 [1-3]). Justifying the results Author obtained intended features of optical fibers like: refractive index profiles (e.g. Fig.1 [1], Fig.28) and parameters of attenuation (10-30 dB km^{-1}) and water content (0.1-0.3 dBm^{-1}). They confirm a very good quality of the optical fibers even though preformed glass modifications were significant. More to notice that also assumed concentrations of Ho and Al_2O_3 in form of $AlCl_3$ and γ-Al_2O_3 nanocrystals (50nm) were embedded successfully. A clear confirmation of the correct preform optimisation and the high quality of optical fibers can be derived from the measured values of lifetimes for Ho (1348μs, $^5I_7$, [1] – world's record) and Tm (585μs, $^3F_4$) levels. These values are among the highest reported in the world literature. It definitely confirm that assumed technological goal was achieved.

Investigation of the second goal so optimisation of the Al/RE ratio was carried for both SD and ND methods. In result, Author found out that the key parameter, which condition the achievement of high quantum efficiency values (> 80%) and the laser threshold (155mW), is independent of the method (Fig. 4, [2]) and consists in obtaining a molar ratio in the case of Al/Ho> 60 (Figs. 4-6, [1]). It was proofed by analysing collective compilations of lifetime dependencies, slope efficiency and laser threshold for both methods. An achievement that should be emphasized as an evident is the comparison of two Ho doped fibers: i) 2000 ppm Ho, 6 mol.% Al_2O_3, Al/Ho ratio = 55 and ii) 600 ppm Ho, 3.5 mol.% Al_2O_3, Al/Ho = 55 where for both constructions the same spectroscopic parameters like: fluorescence lifetimes above 1.2 ms, laser threshold under 200 mW and slope efficiencies greater than 80% have been achieved. In both cases, the output power at 2100 nm was almost 6W (table 1, [3]). They are valuable scientific results as they clearly indicate the technological requirements that must be met. It must be admitted that so comprehensive approach is a rarity in studies on MCVD technology.

Moving on to the Tm doped system, the most important issue was to determine the absorption coefficients and analyze the impact of the cross-relaxation (CR) effect on the spectroscopic properties of optical fibers. For this purpose, Author initially made two series of optical fibers characterising with i) increasing concentration of Tm (500, 2000 and 4000 ppm), constant content of Al_2O_3 (~2 mol.%) and GeO_2 (~1 mol.%) and ii) two extremely different concentrations of Tm (40, 11200 ppm) and high Al_2O_3 content (~7 and 9 mol%). In the first series, he found out a linear decrease in the lifetime of $^3F_4$ level from 487 μs (500 μs is typical value for Tm doped silica fibers) to 378 μs, explaining this by the ET and clustering among Tm ions (fig.5a., [4]). Author characterized optical fibers giving the absorption values for the Tm levels (1180 nm, 1640 nm), as well as background losses (~14-30 dB/km), confirming the possibility of laser generation on the example of the fiber doped with Tm content below 1000 ppm [4]. The obtained values of slope efficiency (44.5%) and laser threshold (391mW) are typical for silica fibers. In the second series, energy transfer coefficients in thulium-doped fibers were determined by rate equation modelling [5]. The main achievement was determination of the absorption coefficients k_{3011}, k_{1130} and k_{1120}, where Author accomplished this task by measuring lifetimes at two wavelengths, 792 nm and 1620 nm, while fluorescence decay curves were monitored at 800 nm and 2 μm (fig. 1 and fig. 4, [5]). Additionally using various pump power showed significant lifetimes decrease of $^3F_4$ and $^3H_4$ Tm levels (fig. 5, [5]). Indeed very beneficial results concern measured and modelled fluorescence decays in SG1598 fiber as different phenomena are seen like high tendency to up-conversion processes (fig. 5d). I agree with the Author that so comprehensive investigations on Tm doped optical fibers have not been done yet. The scientific goal were achieved - however still many interesting optimalisation issues appeared - in majority mentioned in the future perspectives section 6.2. The final works dealt with the applications where i) 150 nm - broadband ASE source at 1.85 μm (90 mW output power) and ii) femtosecond laser (862 fs) at 1870 nm with energy close to 1 nJ were proposed. In both constructions optimized optical fibers were developed.

In conclusion, I confirm that Michał Kamrádek achieved the intended goals of his doctoral dissertation, and the obtained results clearly contribute to the technology and optimization of the spectroscopic properties of Tm and Ho doped silica fibers. Taking into account the achievements indicated by the Author, listed in section 6.1, I confirm that they represent a significant contribution to science and they were verified on the basis of the latest papers.

The goals of Michał Kamrádek's doctoral dissertation were achieved and proved.

Remarks and questions

Assessing doctoral dissertation I found some issues for discussion:

1. Have you noticed any differences in background losses between fibers developed both SD and ND methods?
2. On the fig. 4 [2] cross-section of the preforms (SD, ND) with clearly visible cores are shown – have you checked the existence (survival) of Al_2O_3 nanocrystals? It is a curiosity if the consolidation and collapse heat treatments are a final stage of crystal dissolution.
3. In the paper [4] three optical fibers were fabricated with different Al/Tm ratios (~80, 20, 10). Following the conclusions with the holmium fibers can we justify Tm lifetime decrease (fig.5, [4]) mainly by clustering effect? The answer for this question is probably in the next paper [5] where even in low 40ppm doped fiber the $^3H_4$ level lifetime decreases (where Al/Tm ratio is huge).
4. My curiosity is connected with the fact that Al/Tm ratio for SG1598 fiber is low (~12) so do we expect the correct value of the k_{1120} coefficient?
5. Can you comment the possible influence of the phonon energy comparing published k_{1120} value $3.5x10^{-23}$ m^3s^{-1} [5] with $0.15x10^{-23}$ m^3s^{-1} in YAG crystal?

Overall evaluation

Michal Kamrádek verified own assumptions and results with the latest knowledge at each level of the thesis. What is important in such process as MCVD he also mentioned about older works which were significant in the field. It was a good starting point to perform experimental part and propose alumina nanocrystals using different RE concentrations. He showed clearly that the chosen thesis aims were selected in consequence of lack or not sufficient results in the literature. Finally he achieved significant results in: i) nanoparticle doping as a method suitable for preparation of highly-doped optical fibers for efficient lasers, ii) comprehensive study of wide content of holmium and Al/Ho molar ratios, iii) determination of energy transfer coefficients in thulium-doped fibers, iv) developing and characterisation of high quality optical fibers proofed by record of lasing in Ho fiber laser, broadband emission in optimised Tm doped fiber and generation of femtosecond pulses at 1.87 mm.
Moreover Michal Kamrádek is very active young scientist which is proved by numerous publications and conference presentations. His achievements in numbers includes reported in

the thesis 17 publications related to the doctoral thesis and 8 publications outside. In total he has 30 papers, 117 citations and *h*-index 7 (Scopus).
I am convinced that above achievements proves his scientific quality.

4. Conclusion

Summing up, Ing. Michal Kamrádek solved the scientific and research problem which required both a wide range of theoretical knowledge, as well as advanced skills in the field of technology and metrology of photonics materials. The achievements presented in the thesis allow me to declare that it meets the requirements set for doctoral dissertations and recommend it for presentation and defence. Moreover taking into account his achievements I propose to distinct his thesis.

Krakow, 29.04.2022 r.