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Report on PhD thesis of Ing. Juraj SLÁDEK

This report is related to the PhD thesis of Mr. Ing. Juraj SLÁDEK, entitled *"Modification of bandgap materials by ultra-short laser pulses"*, submitted to the *Czech Technical University in Prague* within the Degree Programme "Applications of Natural Sciences" for obtaining the doctorate degree in the field of Physical Engineering in the Academic Year 2022/2023.

The work was carried out by Mr. Ing. Juraj SLÁDEK at the *HiLASE Centre*, under the supervision of Dr. Yoann LEVY (Czech Academy of Sciences, Institute of Physics, HiLASE Centre). Some results were obtained by Juraj SLÁDEK during an internship at the *Center for Physical Sciences and Technology* (FTMC) in Vilnius, Lithuania, at the beginning of this PhD project.

The thesis manuscript is written in English, contains 130 pages, and is logically arranged in six *Chapters* along with an *Introduction*, a brief summary of the *Aims of the dissertation*, an *Appendices* section, a bibliography part citing 187 *References,* and a *List of the author's publications*. The work is complemented by an *Abstract*, a *Bibliographic entry*, and an *Acknowledgements* section. All Chapters start with a brief introduction. If relevant, support from colleagues and collaborating institutions are clearly indicated in the manuscript text and also explicitly stated in the Acknowledgements section.

In general, in this PhD thesis manuscript, the processing of technical materials (semiconductors, dielectrics, polymers, metals) upon irradiation with ultrashort laser pulses through different experimental strategies (different laser systems, various spatial beam profiles, spot- and scanning-approaches, irradiation with single-pulse or double-pulse bi-color sequences, choice of scanning vs. polarization directions, presence of an overlayer, etc.) is investigated. Special attention is paid to the precise determination of *laser-induced surface damage thresholds*, the generation of so-called *laser-induced periodic surface structures* (LIPSS), a systematic comparison of the damage and surface morphology behavior among the different bandgap materials, and the practical/technical difficulties to obtain precise/correct results and to optimize them. For that different *ex-situ* material characterization techniques (several variants of optical microscopy, confocal laser scanning microscopy, scanning electron microscopy, atomic force microscopy, Raman spectroscopy, etc.) or *in-situ* investigations (e.g. transmission experiments) were employed. In very few cases the results are complemented with detailed theoretical modelling.

More specifically, the thesis manuscript starts in Chapter 1 (14 pages) with a summary of the theoretical background of ultrafast laser-matter interactions and discusses the current state-of-the-art in the field of LIPSS. Chapter 2 (29 pages) then gives an overview of laser surface processing as well as the post-irradiation characterization techniques used in the thesis work. Here, the experimental methods for the evaluation of the laser-induced surface damage, spatial and temporal beam shaping, laser beam scanning and synchronization issues are detailed. The following Chapter 3 (24 pages) reports results on several specific effects that are affecting the materials' damage threshold fluence and the resulting material-specific surface modifications - a prerequisite for the fine tuning of laser-processing parameters (peak fluence, spot overlap, etc.) necessary for processing regular LIPSS on large surface areas. Here, a focus is set on the comparison of the processing of LIPSS on the semiconductor silicon and the dielectric fused silica upon scanning with Gaussian-shaped laser beam profiles. In Chapter 4 (12 pages) additionally techniques of spatial beam shaping are explored, including doughnut-shaped beam profiles, variable sized Gaussian beams, flat-top profiles, and line-shaped foci generated through cylindrical lenses. Chapter 5 (4 pages) summarizes attempts of modifying silicon with ultrashort laser pulses in the mid-IR spectral range and exploring the associated limiting nonlinear effects. Finally, Chapter 6 (2 pages) provides some conclusions of the thesis work and an outlook, followed by six Appendices (in total 12 pages).

The topic of the thesis is up-to-date. This work is timely conducted at a moment, where the knowledge already gained from microscopic spot-processing is being transferred into large-area surface processing with high-power lasers in order to meet the current industrial demands for enabling technical applications of laser-textured surfaces. For that, important aspects were studied in detail, such as the questions of optimum laser beam shapes, processing strategies, as well as electronic synchronization aspects of Galvo-scanner-coupled laser systems, the proper selection of suitable sample characterization and precise damage evaluation methods, and finally the morphological/structural optimization of the large-area laser processing results.

The thesis manuscript considers all relevant developments in its field. It properly discusses the current state-of-the-art and goes beyond that, particularly through the new insight describing the morphological transitions and its scaling behaviour close to the damage thresholds in laser scan-processing approaches for silicon and fused silica, specifically upon irradiation with fs- or ps-pulse durations (Chapter 3). The same applies for the new insights reported for the LIPSS regularity with respect to the laser beam spot size (Chapter 4). The quality of the results presented in the core Chapters 3-5 of the thesis (resulting from challenging and complex experiments) is excellent. The overall scientific quality of the data, their visualization (figures) and the thesis manuscript text in terms of clarity and their presentation is very good too. Some sections are rather short, but this is acceptable and appears constrained to avoid an overlength thesis manuscript.

The main goals of the dissertation are briefly summarized at the beginning of the thesis manuscript in four topics (p. 8). All these goals are suitably addressed in the thesis manuscript and were successfully achieved, although – due to time constraints and prioritization to other experiments - rather few results on the attempts to perform volumetric material modifications in semiconductors in the mid-IR spectral range with the Perla-C laser at the HiLASE Center were presented in Chapter 5. The thesis work is mainly experimentally oriented (at an excellent level) and - from a (personal) physicist's point of view - it could have benefited from some more detailed theoretical modelling, supporting some of the presented experimental results. However, in Chapter 2 describing the experimental methods, in the core Chapters 3-5, and in the Appendices section, one can see an excellent in-depth-level of understanding of the complex technologies and all measurement techniques, further supporting the very strong commitment of Mr. Ing. Juraj SLÁDEK to experimental and physical engineering work.

The scientific value of this work lies in its new results made available to the laser processing community. This also applies for many experimental details and their technological relevance, which are usually neither properly controlled, nor sufficiently documented - in contrast to the thesis presented here. The high scientific quality, relevance, and novelty contained in this PhD thesis manuscript is also reflected by six peer-reviewed scientific publications published between 2019 and 2023, mostly in high-impact factor journals with Juraj SLÁDEK being the first author of three of these papers and co-author of the three others. Moreover, he is a co-author of two contributions presented at the important international scientific conference *2020 Conference of Lasers and Electro-Optics* (USA).

There are some minor technical issues in the thesis text, which can be clarified during the thesis defense.

In conclusion, given the high scientific and technical quality of the results and their good presentation in this PhD thesis manuscript, I am recommending to admit Mr. Ing. Juraj SLÁDEK to the presentation & defense of this thesis at the *Czech Technical University in Prague* for obtaining the doctoral degree.

Dr. Jörn Bonse, Berlin, July 30th, 2023