



**KUTATELADZE
INSTITUTE OF
THERMOPHYSICS**



NHRF
Theoretical & Physical
Chemistry Institute

INTERNATIONAL WORKSHOP

28-29 June 2024, Novosibirsk — Athens

BOOK OF ABSTRACTS

**THE FIRST SIBERIAN-ATTICA
INTERNATIONAL WORKSHOP
ON LASER PROCESSING
FOR THERMOPHYSICAL
APPLICATIONS**



KUTATELADZE INSTITUTE OF THERMOPHYSICS SB RAS
(NOVOSIBIRSK, RUSSIA)
THE NATIONAL HELLENIC RESEARCH FOUNDATION
(ATHENS, GREECE)

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Novosibirsk
2024

УДК 621.373.826:536.2(063)

ББК Ж618Я431

F56

Organizations

Kutateladze Institute of Thermophysics SB RAS (Novosibirsk, Russia)

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F56 The First Siberian-Attica International Workshop on Laser Processing for Thermophysical Applications. 28–29 June 2024, Novosibirsk — Athens : Book of Abstracts. Novosibirsk: NSU Publishing House, 2024. — 22 p.

ISBN 978-5-4437-1667-1

This book contains abstracts of reports presented at International Workshop on laser processing in thermophysics application (28–29 June 2024, on-line). The goal of the workshop is to exchange experience in the rather labor-intensive field of science and technology, which simultaneously involves many areas of physics, chemistry, and materials science.

The workshop was supported by the Ministry of Science and Higher Education of the Russian Federation (grant 075-15-2024-620).

УДК621.373.826:536.2(063)
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ISBN 978-5-4437-1667-1

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Advanced functional nanomaterials by laser technologies

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Surface nanotexturing is a crucial technological procedure allowing to improve basic characteristics of the practically important materials and interfaces or even endow them with new functionalities appearing when the feature sizes shrinks down to nanometer scales. Bright examples of such material improvement can be readily found in almost all aspects of modern engineering and device fabrication, where appropriate patterning allows to control electrical, optical, biological, tribological and wetting properties of the processed surfaces. Material processing using short and ultrashort laser provides a set of non-lithographic techniques showing continuously growing popularity for both scientific and industrial communities. Such interest is largely boosted by the development of the laser market offering cheaper, high-speed and stable pulsed sources, more precise beam shaping and scanning systems as well as the growth of fundamental understanding of laser-matter interaction via development of computer-aided simulation tools and analysis techniques based on machine learning.

In this report, we summarize our recent results on fabrication of advanced nanomaterials in the form of nanoparticle suspensions and nano-textured interfaces via several straightforward laser-assisted fabrication strategies: direct laser patterning, laser-induced period surface structuring (LIPSS) and laser ablation in liquids (LAL). In particular, we focus on advanced opportunities of the mentioned laser technologies for achieving deep-subwavelength feature sizes and hybrid multi-elemental compositions such as metal-semiconductor ones. Diverse applications areas of the produced nanomaterials for resonant light manipulation, light-to-heat conversion, security labeling and sensing are also discussed.

The work was supported by Russian Science Foundation (project No. 24-19-00541)

Micro/nano-structured functional surfaces and devices by laser processing

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The need for advanced materials and systems with new functionalities has motivated the development of micro/nanostructures on solid surfaces, which are necessary for the fabrication of functional devices for novel applications. In this talk, we will discuss the development of functional micro/nanostructures, based on laser-processed surfaces. Laser micro/nanofabrication presents distinct advantages, such as low cost, simplicity (tabletop apparatuses, maskless processes), large-scale potential, high spatial resolution (localized modifications, of the order of the optical wavelength).

Coating micro/nanostructures with thin metallic films results in plasmonic substrates with enhanced electromagnetic response across the entire visible range, which are used for plasmonic optical trapping [1] and surface-enhanced Raman spectroscopy (SERS) [2]. “Smart” surfaces of controllable extreme wetting states are obtained by combining thermoresponsive polymers or photoresponsive metal oxides with micro/nanostructured substrates [3]. Also, surfaces with controlled topography, either at the micro- or at the nano-scale, for targeted cell cultures for biomedical applications [4]. We will present recent advances in these fields and discuss future applications.

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Highly-Uniform Thermochemical Laser-Induced Periodic Structures Formed by Femtosecond Laser Pulses

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Laser-induced periodic surface structures (LIPSS) is a remarkable phenomenon observed on surfaces of almost any materials under an impact of laser radiation with high intensity. LIPSS represent a periodic modulation of relief height and are formed due to the interference effects between incident light and scattered waves, which create an intensity pattern on the surface with periodicity and orientation defined by the parameters of irradiation, e. g. wavelength, polarization, angle of incidence, as well as properties of material and ambient environment [1]. At laser fluence under an ablation threshold, the regimes of thermochemical LIPSS based on a thermally stimulated reaction of oxidation were observed on surfaces of thin metals and amorphous semiconductors films with outstanding regularity and optical properties opening pathway for various applications including optical sensors, biophotonics, and solar light harvesting [2]. In this work we review our recent results on thermochemical LIPSS formation under impact of femtosecond laser pulses on Ti, Cr, Hf and semiconductors amorphous Si and Ge thin films. The sub-wavelength structures with morphology depending on the processing parameters and strategy of laser micromachining were revealed. The different physical mechanisms responsible for thermochemical LIPSS formation along with practical applications of obtained structures are discussed.

The work was supported by the Russian Science Foundation grant (No. 21-72-20162).

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Electromagnetic phenomena in laser processing of materials

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The nanoscale patterning of materials and surfaces is extremely important for a broad range of practical applications. Nanopatterning techniques that are fast, efficient, and do not require a mask are particularly attractive. Here, I will discuss two such examples. The first concerns the formation of Laser Induced Periodic Surface Structures (LIPPS) upon irradiation with high-energy fs pulses [1, 2]. I will focus on metals and I will discuss the case of thin films [3]. In such cases, both symmetric and antisymmetric surface plasmon waves can be excited. Their interference shapes the distribution of energy deposition (absorption) and can thus largely dictate the resulting surface topology. I will show that it can be tailored through the film thickness and the sub-/super-strate properties. The second concerns the utilization of nanoparticles to produce locally-enhanced fields (photonic nanojets) [4]. This field enhancement can improve the structuring capabilities of a laser beam, reducing the power requirements and enhancing the resolution. By varying the characteristics of the particle (diameter, material), one can tailor the properties of the formed nanojet, namely, maximum achieved field enhancement, “focal distance”, and spot size.

In both cases, we numerically calculate the electromagnetic fields by rigorously solving Maxwell’s equations with the Finite Element Method (FEM) [5]. Following energy deposition (absorption) on the surface, a complex physical process initiates that involves heat transfer and mass transfer (material melting and resolidification) or removal (ablation) and ultimately leads to the formation of the resulting surface topology. Tracing the connection between the distribution of the energy deposition and the resulting surface topology is a standing open issue [6].

The author acknowledges collaborators Dr. G. Perrakis, Dr. G. Tsibidis, Dr. E. Stratakis, Dr. P. Lingos and Dr. M. Kandyla, Dr. S. Starinskiy, N. Chouchoumi.

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An Investigation of Laser Annealed and Metal-Induced Crystallized Polycrystalline Silicon Thin Film

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Today semiconductor electronics is used in many fields. For example, thin-film polycrystalline semiconductor materials are promising for micro-, nano-, optoelectronics [1] and photovoltaics [2]. One of the key needs for industrial production is the cheapening of such materials, in particular, the use of glass and plastic as substrates. The main method for producing thin films of polycrystalline silicon is thermal annealing of amorphous silicon films. However, thermal annealing is demanding to external conditions: high temperature of the process (more than 600 °C), the need to maintain a high vacuum, duration (more than 10 hours). There are alternative methods of crystallization, for example: laser-induced crystallization (LIC)[3] and metal-induced crystallization (MIC)[4]. It was aimed to combine MIC and LIC to obtain a fast and undemanding crystallization method.

This paper presents the results of an experimental study of amorphous silicon crystallization using nanosecond pulsed laser annealing in combination with the metal induced crystallization method. The sample is a layered structure of a 30 nm thick gold film and a 130 nm thick amorphous silicon film. Two wavelengths, 532 and 1064 nm, were used for laser annealing. The modification thresholds for the wavelengths used were determined. IR radiation modifies the material and has a large operating energy range. When exposed to visible radiation, tearing and delamination of the composite film was observed. Similar experiments were carried out in vacuum. RAMAN spectroscopy of the material showed that silicon crystallizes under the influence of IR radiation. The morphology of the film after laser treatment was studied by electron scanning microscopy

For the first time gold was chosen as a donor metal for such a process, since it forms with silicon the eutectic point with the lowest temperature and does not oxidize. The possibility to crystallize amorphous silicon by the proposed method, both in air and in vacuum, was shown. In the future, other possible modes of op-

eration will be searched for and the dependence of the process on the laser pulse duration will be investigated.

The work was performed under the state contract with the Institute of Thermophysics SB RAS (No. 121031800214-7).

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Development of n^+ -ZnO/ n -Si photodetector with wavelength-selective operation

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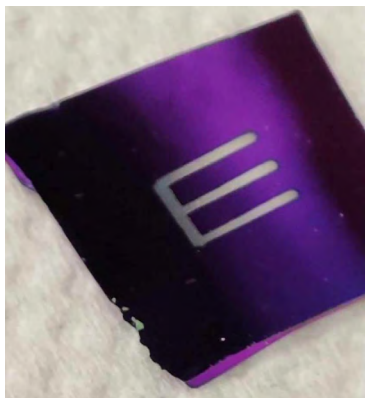


Figure 1: n^+ -ZnO/ n -Si photodiode

In this presentation we will discuss the development and operation of broadband n^+ -ZnO/ n -Si photodetectors. Three ZnO thin films were developed by Atomic Layer Deposition (ALD) on n -Si at three different temperatures (100 °C, 150 °C, and 200 °C), forming three different photodetecting devices.

Current-voltage measurements exhibit broadband photoresponse of the devices, across the UV-visible-NIR. The photodetectors are able to distinguish between UV-vis and NIR illumination by producing opposite photocurrents, demonstrating wavelength-selective operation. By utilizing pulsed light, the response times of the

photodetectors were investigated for different pulse frequencies and wavelengths. The devices show response times as low as 40 μ s at a pulse frequency of 400 Hz. The ZnO film developed at 150 °C exhibited the most promising results with a responsivity of 0.242 A/W and a detectivity of $42 \cdot 10^{12}$ Jones.

Dynamic of water droplets impacting on laser textured surfaces

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In the study of surfaces with various properties, the process of droplet spreading and splashing upon collision is a key phenomenon that plays a significant role in various natural and practical applications [1]. Examples of such applications include inkjet printing, coating spraying, pesticide spraying, spray cooling, and anti-icing measures [2]. The interaction of droplets with solid surfaces, depending on wetting properties, involves a range of diverse processes, including spreading, rebounding, and splashing. These phenomena are influenced by surface properties such as roughness and wettability, as well as the physicochemical properties of liquids, including density, viscosity, surface tension, and droplet velocity. Additionally, the interaction of droplets with heated surfaces is a crucial phenomenon in spray cooling, capable of dissipating significant heat flux and holding substantial potential for cooling electronic devices [3]. The dynamics of droplet interaction with heated surfaces is a complex process dependent on numerous parameters, including environmental conditions, droplet characteristics, and surface attributes such as roughness, micro- and nanostructure, as well as wettability properties [4].

In this paper, we review our recent results on the study of the dynamics of interaction between a droplet and a textured surface. Different interaction modes are found depending on the type of microstructure and surface temperature. Experimental and numerical results are compared.

The work was performed under the state contract with the Institute of Thermophysics SB RAS (No. 121031800214-7).

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Utilizing Optical Tweezers for Measuring the Elasticity and ζ -potential of Red Blood Cells

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Optical trapping, achieved using optical tweezers, is an advanced and highly sensitive technique capable of measuring forces in the femto-Newton range. This sensitivity allows it to detect minute variations in biological properties [1]. With this method, microscopic objects such as living cells, bacteria, and viruses can be captured at the focus of a laser beam, without physical contact, enabling the study of their fundamental physical properties, such as elasticity and membrane viscosity [2].

Using optical tweezers developed at the National Hellenic Research Foundation, we measured the elasticity of red blood cells (RBCs) in plasma, to simulate human body conditions. Additionally, the ζ -potential of RBCs was determined using the electrophoresis method in a custom-built chamber. These measurements provide a foundation for further investigation into the physical properties of RBCs and how they are influenced by medication or diseases, such as Mediterranean anemia (thalassemia).

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Investigation of optical properties of catalytic particles synthesized by pulsed laser ablation in liquid

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Colloidal solutions, due to their properties such as stability, gel formation, and the ability to transport and distribute various substances in liquids, find application in many industries [1]. In this work, a colloidal solution of titanium dioxide was used because it occupies an important niche in photocatalysis, for example, in the photocatalytic decomposition of harmful organic compounds. The most commonly used modification of titanium dioxide as photocatalysts is anatase, which is associated with its high photocatalytic activity.

For the synthesis of the colloidal solution in this work, Pulsed Laser Ablation in Liquid (PLAL) was used due to several advantages, including the flexibility of parameter selection, localized action, and compatibility with other technologies. Despite large amount of data on controlling the crystalline phase described in the literature, obtaining samples using pulsed laser ablation remains a challenging task, as rutile is predominantly the dominant oxide phase in colloidal solutions synthesized by this method. This is explained by the fact that the phase transition from anatase to rutile occurs at high pressure and temperature and is irreversible.

It was hypothesized that there is an energy window in which the use of picosecond pulses can produce stable colloids with anatase as the dominant oxide phase [2].

The experimental setup consisting of a controlled coordinate mechanism and a laser system was used for the synthesis of the colloidal solution. Fluence was varied in the range of 10–14 J/cm² for nanosecond pulse duration and was 2.2 J/cm² for picosecond pulse duration.

The optical properties of the obtained colloidal system were analyzed using an SF-2000 spectrometer. It is assumed that the modification of titanium dioxide obtained is anatase. It was demonstrated that with nanosecond pulses, rutile is the predominant oxide phase, while with picosecond pulses, anatase is the most prevalent form at an fluence of 2.2 J/cm².

The work was performed under the state contract with the Institute of Thermophysics SB RAS (No. 121031800214-7).

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Femtosecond direct laser writing and micromachining

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We explore the capabilities of femtosecond lasers in direct laser writing (DLW) applications, emphasizing their advantages and the impact of various parameters on the micromachining process. Femtosecond lasers, with their ultrashort pulse durations and high peak powers, offer high precision and minimal heat-affected zones eliminating thermal effects during micromachining. These attributes make them ideal for intricate micromachining tasks and the generation of a variety of structures at the micro- and nanoscale. Our investigation focuses on the significance of process parameters such as pulse energy, focusing depth, and repetition rate, all of which influence the feature size, shape, and quality of the laser-modified materials. In this work, we present micromachining results in a range of various material platforms such as Silica, Silicon, and polymers materials. Additionally, we examine various distinct applications such as silica on silicon optical circuits for quantum applications [1, 2], laser-patterned silicon substrates explored for cancer cell culture [3], surface modifications in polymer Intraocular Lenses (IOLs) for personalized vision correction, polymer films for aiding cytological screening, and also PMMA thin films and polymer optical fibers, for information storage or sensing applications. Through these experiments, we aim to confirm surface modifications and identify the optimal irradiation parameters for precise laser ablation by eliminating thermal effects accumulation during laser processing.

The study was performed at NHRF, supported by the Hellenic Foundation for Research and Innovation (H.F.R.I.) under the “First Call for H.F.R.I. Research Projects to support Faculty members and Researchers and the procurement of high-cost research equipment” (Project Number: HFRI-FM17-640).

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Laser induced chemical deposition of tungsten nanostructures from the gas phase

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Laser chemical vapor deposition (LCVD) is a method for selective deposition of solid materials via localized chemical reaction driven by a focused laser beam. This method holds great potential for the production of small and complex metal and ceramic parts [1], and capable of producing true-3D microstructures with micron or submicron-sized features [2]. LCVD printing is mostly limited by the laser and its optical system which define minimum size of structures. We focused our attention on pyrolytic LCVD where the energy of the focused laser beam is absorbed by reagent gases, leading to the decomposition of gas molecules and the formation of a thin solid film on the substrate. In this work, we review our recent results of tungsten deposition on silicon substrate by pulsed solid-state Nd:YAG laser at atmospheric pressure. The number of grown spots is presented and analyzed; composition and deposition rate of the structures are discussed.

The study was performed with the support of state task of the Kutateladze Institute of Thermophysics (IT SB RAS), number of State Registration 121031800214-7.

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Investigation of water droplet geometry evolution under uniform electrical field on rose petal and surface modified by pulse laser ablation

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The study of droplet evaporation from various surfaces is an active problem that is widely investigated and has many technical applications (intensification of heat transfer, deposition of colloidal solutions). Biomimetic surfaces, which repeat the properties of some natural surfaces, are particularly interesting. In this work biphilic surfaces — superhydrophobic surfaces with superhydrophilic areas, on which the droplets were placed, which were made by pulse laser ablation. Such surfaces have great potential in various applications, as they can be used for controlled movement of liquids, controlled deposition and heat transfer intensification. Also, of great interest are the studies of droplet geometry changes in the electric field. In the present work, the evolution of the geometry of water droplets on a biphilic surface and a rose petal in a constant electric field of different strength. The data were obtained on the dynamics of droplet geometry as a function of field strength for the surfaces used. It is found that the droplet height grows nonlinearly with increasing field strength. The results of this work will allow us to expand our understanding of the kinetics of evaporation, the dynamics of droplet geometry and the field strength for the surfaces used.

The study was performed at the Kutateladze Institute of Thermophysics (IT SB RAS) with the support of № 121031800214-7.

Deposition of functional layers by pulsed laser deposition

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Nanoscale thin films are widely used in various technological and scientific domains, forming the basis for numerous advancements that have propelled human progress, thanks to their highly tunable functional properties based on chemical composition. Pulsed laser deposition is one of several physical vapor deposition methods employed to fabricate thin films, utilizing laser energy to eject material from a target as plasma. A substrate, often a single-crystal oxide, is positioned in the path of the plasma plume and acts as a template for the incoming species from the target to coalesce and self-assemble into a thin film. This technique is incredibly useful for producing crystalline films due to the broad range of atmospheric conditions and the potential chemical complexity of the target. However, this flexibility introduces significant complexity, often necessitating meticulous optimization of growth parameters to achieve high-quality crystalline films with the desired composition. In this work, we present our results on the synthesis of Au [1], SiO_x [2] and Al₂O₃ [3] films by pulsed laser deposition in vacuum or background gas.

The dynamics of heating and vaporization of gold in vacuum under the action of laser pulses of low intensity, when the main ablation products are neutral particles, is investigated by mass spectrometry and numerical modeling. The laser-ablation plume is found to consist of gold atoms and dimers with kinetic energies considerably higher than their thermal energy upon evaporation. Nanostructured gold films are synthesized by depositing the ablation products onto a substrate.

The nanosecond laser ablation technique was used to synthesize thin silicon oxide films of various stoichiometry in vacuum and in a background gas. The local oxidation degree of specimens was evaluated using three different characterization methods. It was found that, on increasing the distance to the laser-plume axis, there occurred a monotonic increase in the oxygen content of the films due to their oxidation inhomogeneity. A profound decrease in ablated mass, related to an increased reverse flow of substance to the target, was found to occur when the pressure of the ambient mixture was increased from 20 to 60 Pa. A comparison was made of the oxidation efficiencies of the films heated at the stage of their

synthesis and at the stage of annealing of already formed films. It is shown that the composition of the films could be controlled by varying the inert-gas pressure at the constant pressure of the chemically active component in ambient mixture.

Transparent aluminum oxide nanostructures with extreme wetting properties were synthesized by nanosecond laser deposition in background oxygen. The transparency and morphology of the samples were analyzed. Non-monotonic behavior of the transmittance coefficient was observed with varying background oxygen pressure in the range of 20 to 140 Pa, attributed to differences in the kinetics of ablation product dispersion. The evolution of the contact angle was studied, ranging from ~ 5 to $\sim 120^\circ$, during the storage of coatings in air under normal conditions.

The work was performed under the state contract with the Institute of Thermophysics SB RAS (No. 121031800214-7).

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Microtexturing of materials by nano- and femtosecond laser pulses

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In recent years, laser processing of materials has been actively used to change their physical and chemical properties. Nanosecond and femtosecond pulses are most often used. Nanosecond lasers are characterized by thermal effects on materials. The main applications of nanosecond lasers include micro-processing, medical procedures such as laser surgery and skin treatment, and chip manufacturing in the semiconductor industry. Their advantages are high pulse energy and relatively low cost of equipment, but significant thermal effects remain the main disadvantage. Femtosecond lasers minimize the thermal effect on the material, which localizes its heating and ensures more accurate and clean processing. This makes them ideal for precision micro-machining, including high-precision cutting and drilling, as well as for microelectromechanical systems (MEMS). Femtosecond lasers are also widely used in medical applications such as laser vision correction (LASIK) and microsurgery.

This work presents the results of processing materials with nanosecond and picosecond laser pulses. As a result of exposure to nanosecond pulses, it was possible to achieve a developed morphology of the surface and its composition contributing to the achievement of superhydrophilic wetting properties. Femtosecond radiation has been used to create microchannels in transparent materials.

The work was performed under the state contract with the Institute of Thermophysics SB RAS (No. 121031800214-7).

Photonic nanojet for laser treatment thin metal films

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Laser processing of materials at the submicron level is increasingly attracting attention due to the potential application of optical technologies in a wide range of sensing tasks. These include medicine, biology, microelectronics, and others. Focusing radiation at the scale of the diffraction limit is a rather non-trivial task. In this work, we report on how optical tweezer technologies and photonic nanostructures can be combined to achieve more precise focusing of radiation. Developed algorithms, together with modeling results of optical processes during light transmission through micron-sized objects captured by optical tweezers, as well as thermal effects of laser radiation on metallic coatings, have demonstrated the effectiveness of this technology. Unlike earlier studies, the methodology has been applied to cases involving highly absorbing object, namely gold palladium thin film, yet optimal processing parameters have enabled the localization of the treatment area.

This work has received funding from the European Union's Horizon Europe research and innovation programme under the Marie Skłodowska-Curie Postdoctoral Fellowship №101067553 (LANAFUSEHA).

Scientific edition

**THE FIRST SIBERIAN-ATTICA INTERNATIONAL WORKSHOP
ON LASER PROCESSING FOR THERMOPHYSICAL APPLICATIONS**

28–29 June 2024, Novosibirsk — Athens

BOOK OF ABSTRACTS

Signed to publish on 24.08.2024.

Format 60 × 84 1/16. Pub. sheet-copy 1.3. Print. sheet 1.27.

Order No. 90.

NSU Publishing House

2 Pirogov str., 630090, Novosibirsk

ISBN 978-5-4437-1667-1

