BULGARIAN ACADEMY OF SCIENCES

GEORGI NADJAKOV

INSTITUTE OF SOLID STATE PHYSICS

ANNUAL RESEARCH REPORT 2022

Edited by K. Buchkov

Institute of Solid State Physics - BAS
72, Tzarigradsko Chaussee Blvd.
1784 Sofia, BULGARIA
tel: (+359 2) 875 80 61
e-mail: director@issp.bas.bg http://www.issp.bas.bg

Contents

Foreword

Georgi Nadjakov Institute of Solid State Physics of the Bulgarian Academy of Sciences (ISSP-BAS) is an institution with half-century research traditions in fundamental and applied sciences dedicated to important social areas as science, education, healthcare, historical and cultural heritage.

Since its first days ISSP-BAS have been developing in modern fields of condensed matter physics, atomic, plasma and laser physics; physics of liquid crystals and of soft matter, in the theoretical description of diverse complex solid state phenomena; in the synthesis of novel materials with unique properties; quantum, bio- and nano-technologies; acoustoelectronics, nanoelectronics and sensing.

The past year marked ISSP 50th anniversary. To commemorate this event, we have honoured our scientific heritage and shared new unexplored scientific horizon's with inspiration for higher research levels and socially significant contributions, not only to Bulgarian, but also to European and world science.

The scientific projects, throughout 2022, were worked out in a close collaboration with various academic institutions and under the frames of different programs. It is worth to note the participation of ISSP in the joint initiatives "ELI-EIC-BG" together with the Institute of Electronics-BAS and the Faculty of Physics of Sofia University within the framework of the "National Roadmap for Research Infrastructure (2020-2027)" and the National Centre of Mechatronics and Clean Technologies (Grant No BG05M2OP001-1.001-0008) funded by the Operational Programme "Science and Education for Smart Growth" 2014-2020 and the European Regional Development Fund. In addition, there are also several national and international scientific projects financed by the Bulgarian National Science Fund.

Scientific infrastructure

In 2022, the Gas Sensor test system Kenosistec KGAS4S was installed as part of the implementation of research equipment within the National Center for excellence "Mechatronics and clean technologies". This platform provides various options for detailed electrical characterization of nanostructured gas-sensing devices depending on broad set of parameters as concentration, pressure, humidity, temperature. It is particularly designed for the detection of toxic gases (CO_2, NO_2 and NH_3) and to simulate real atmospheric conditions.

ISSP maintains:

- Scientific facilities for thin films and nanostructures synthesis - clean room laboratory (10 000) with two high class deposition systems - Beneq TFS 200 atomic layer deposition of oxide thin films and Oxford Nanofab Plasmalab 100 for plasma stimulated chemical deposition (PE CVD) for synthesis of graphene and carbon nanotubes (low dimensional materials).
- Technological platform for routine microelectronic operations and preparation of structures, sensors and devices based on silicon thin film technology together with a contemporary equipment for analysis of electrical and sensing characteristics.
- Multifunctional laboratory filtration system (MaxiMem, Prozesstechnik GmbH) for substance extraction for pharmaceuticals, membrane filtration, purification and recycling.
- Technological line and measurement equipment for research/testing of mass sensitive quartz resonators for a wide range of sensor applications (e.g. gas and thermometric).

The most recent development of this methodology (in 2022) is in context of reproductive medicine.

- Various laser systems and technologies with broad spectrum of applications for monitoring ultrafast processes and dynamic measurements, study of living structures, nanoscale material modifications, archeological artefacts analysis. Femtosecond laser systems and metal vapor based lasers.
- Equipment for optical characterization of thin films, such as the spectroscopic ellipsometer M2000D, spectrophotometer Perkin Elmer-Lambda 1050 equipped with a Fourier transform infrared spectrophotometer Vertex 70 and module "150 mm integrating sphere".
- CellASIC™ ONIX automated microfluidic system, handheld cytometer Scepter 2.0 for experiments on the soft matter/nanostructures interactions
- Combined system of galvanostat and potentiostat SP-200
- Scanning probe microscope (VEECO, Multimode, USA) for precise surface characterization at the nanoscale.

The traditional XXII SCHOOL OF CONDENSED MATTER PHYSICS (State of the Art in Functional Materials & Technologies) was held in the period August 29 - September 2, 2022 in Varna. The biannual scientific forum is organized since 1980, and in 2022 was dedicated also to the 50th anniversary of ISSP-BAS with lecturers and participants from the USA, France, Germany, Poland, Slovenia, Serbia, Hungary, Poland, Romania, Italy, Belgium and Bulgaria.

Publishing activity

In 2022, were published in total 112 papers and overall 93 in peer-reviewed journals indexed in the international scientific databases Web of Science and/or SCOPUS.

Scientific articles for 2022	**112**
Articles indexed in WoS и SCOPUS with corresponding quartiles	**93**
Q1	**30**
Q2	**23**
Q3	**13**
Q4	**8**
Book chapters	**2**
WoS & Scopus citations	**1644**

Patents

ISSP currently holds intellectual property rights on 25 BG patents and two recent applications filed during 2022 - I. K. Kostadinov, D. N. Astadjov, K. A. Temelkov, G. P. Yankov: Gas-discharge laser, № 67473 / 15.11.2022 and K. D. Esmeryan and Y. I. Fedchenko, A method for the fabrication of ultra-durable extremely water-repellent carbon soot coatings №113502/15.03.2022.

GEORGI NADJAKOV INSTITUTE OF SOLID STATE PHYSICS
Bulgarian Academy of Sciences

72, Tzarigradsko chaussee Blvd., 1784 Sofia, BULGARIA
Tel: (+359 2) 875 80 61, ***http:// www.issp.bas.bg***

Director:
Professor
Hassan Chamati, Ph.D., D.Sc.
tel.: 02 979 5758
director@issp.bas.bg

Deputy Director:
Professor
Albena Paskaleva, Ph.D., D.Sc.
tel.: 02 979 5742
paskaleva@issp.bas.bg

Scientific Secretary:
Associate Professor
Krastyo Buchkov, Ph.D.,
tel.: 02 979 5706
buchkov@issp.bas.bg

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tel.: 02 979 5717
eiordanova@issp.bas.bg

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Daniel Iliev,
tel.: 974 60 52, 979 5700
diliev@issp.bas.bg

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Ljubomila Dedinska, Dipl. Eng.
tel.: 875 80 61, 979 5792
buba@issp.bas.bg

Accountant Office:
Eleonora Popova
tel.: 875 50 59, 979 5744
nora@issp.bas.bg

Administration Office:
Irina Velkova, Dipl. Eng.
tel.: 877 34 92, 979 5780
velkova@issp.bas.bg

Switch board:
02 979xxxx

Mission: The Institute is committed to generating, disseminating and preserving fundamental knowledge in the fields of condensed matter physics, optics, spectroscopy and laser physics, and application of this knowledge for the creation of novel materials, devices and analytical methods for micro- and nano-technologies, and to explore new scientific endeavors and to expand the knowledge to bear on Bulgaria's great challenges.

The main research topics investigated at ISSP-BAS are:

- Synthesis of advanced multifunctional (semiconductor, dielectric, magnetic and superconducting) materials and detailed study of their complex physical behavior with the aid of a broad set of electrical, optical, magnetic, structural, microscopic and others methods.

- Physics and Technology of Thin and Nanoscale Inorganic Dielectrics and semiconductor layers, carbon nanostructures and low-dimensional (1D/2D) systems, biomaterials and nanostructured liquid crystals and their applications in nanoelectronics, opto and acoustoelectronics, sensing devices.

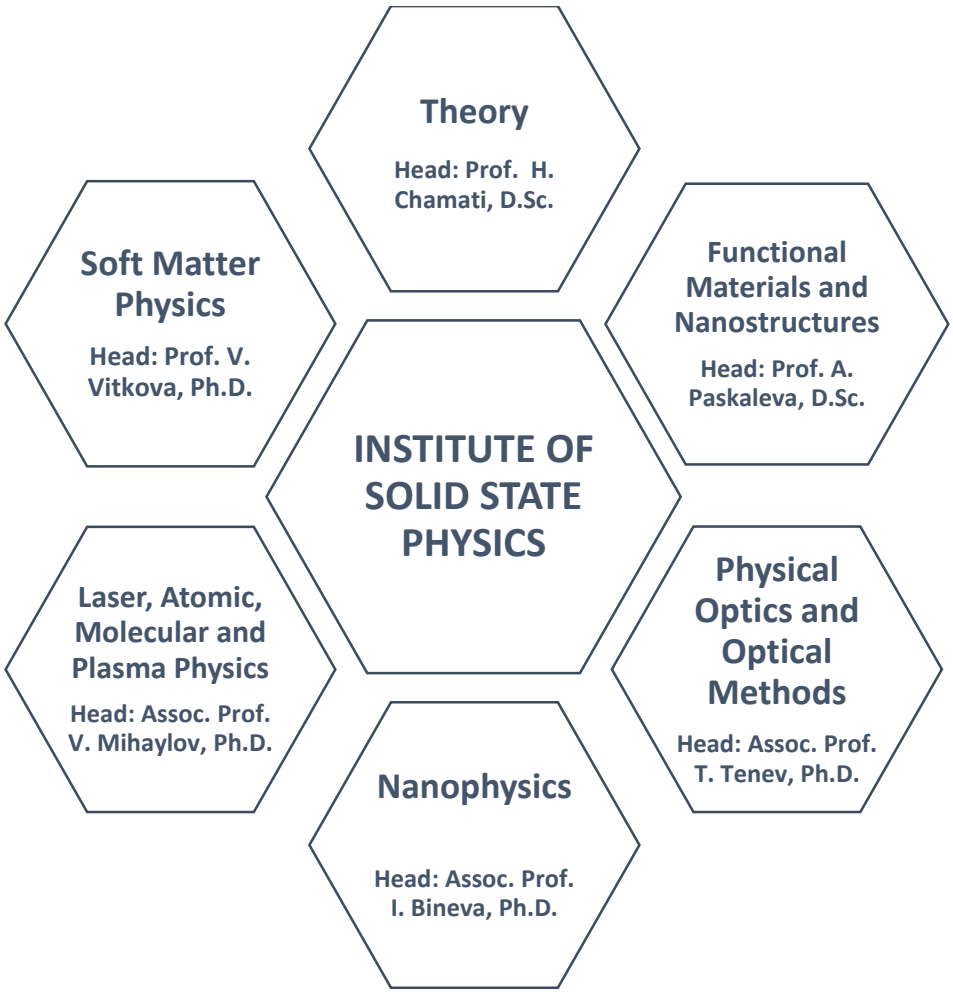
- Theoretical modeling of physico-chemical processes, properties, phases and phase transitions in condensed matter, dynamics of non-linear systems.

- Lasers, atoms and plasma, applications and technologies (nanotechnologies, laser technologies, archaeometry, medicine, ecology).

- Photonics, optics and spectroscopy of nonlinear and optically anisotropic media.

HISTORICAL REFERENCE: ISSP at BAS was established by the Council of Ministers of the Republic of Bulgaria by Decree No 362 of October 16, 1972. This Government Decree divides the existing Institute of Physics with Atomic Scientific Experimental Centre at BAS, founded by Academician G. Nadjakov in 1946, into ISSP and INRNE (Institute for Nuclear Research and Nuclear Energy), starting from January 1, 1973. Since February 16, 1982, the Institute of Solid State Physics was named after Academician Georgi Nadjakov. Since its establishment, ISSP was managed by the directors Acad. Milko Borissov, Prof. Nikolay Kirov and Acad. Alexander G. Petrov, as well Prof. Nikolay Tonchev and Prof. Kiril Blagoev, who acted as interim directors

ORGANIZATION OF THE INSTITUTE OF SOLID STATE PHISICS



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THEORY

LABORATORY

THEORY GROUP

HEAD: **Prof. Hassan Chamati, D.Sc.**
tel: 979 5778; e-mail: chamati@issp.bas.bg

TOTAL STAFF: **15**
RESEARCH SCIENTISTS: **15**
ASSOC. MEMBERS: **2**

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Associated members: Prof. N.S. Tonchev, D.Sc.; Assoc. Prof. M. Primatarowa, Ph.D.

RESEARCH ACTIVITIES:

Mononuclear molecular magnets that possess an energy barrier to the reversal of magnetization are among the most attractive nanomagnetic systems for both theorists and experimentalists working in this research field. Known also as single-ion magnets, these small units of matter stand as promising candidates for the design and assembly of the future quantum technologies. By virtue of their unique anisotropy behavior and hence distinct fine structure, they have continued to captivate the researcher attention for more than two decades.

Focused on single-ion magnets that exhibit eminent magnetization-reversal barrier, slow magnetic relaxation and high blocking temperature, during the present year we review the progress made in the synthesis of 3d and 4f single-ion magnets in the last two decades and discuss the observed magnetostructural properties underlying the anisotropy behavior and the corresponding remanence. Moreover, we emphasize on the fundamental theoretical aspects elucidating the complex behavior of these nanosized magnetic entities. That includes analyses of key notions, such as the zero-field splitting, anisotropy energy and quantum tunneling of the magnetization. The interdependence of these features is closely studied within the cases of $3d_2$ and $3d_8$ single-ion magnets possessing different crystal field symmetry and with V_3^+ and Ni_2^+ metal centers, respectively. Of particular interest for the performed studies was the problem related to the occurrence of huge zero-field splitting and giant magnetic anisotropy in Ni_2^+ complexes, especially these of trigonal bipyramidal geometry.

The magnetic properties of the considered single-ion magnets were computed with the aid of the exact diagonalization method. The overall theoretical approach results in a multi-configurational self-consistent scheme for evaluating the contribution of the spin exchange interactions, crystal field and spin-orbit interactions on the fine structure of the corresponding energy spectrum. The used method allows one to trace the direct correspondence between the obtained fine structure and the probability to observe each one of the electronic initial quantum basis states, hence ensuring the correct determination of the zero-field splitting and magnetic anisotropy.

A thermodynamic model of free energy expansion up to eighth order of magnetisation to describe the complex magnetic phase transitions in ferromagnetic superconductor UGe_2 was

proposed. The model successfully describes transitions between ordered phases, which take place without changing of magnetic structure but only of magnetization, which is the case of UGe_2 where two magnetic phases with the same structure, but with different magnitude occur with decreasing of temperature. The influence of pressure on the magnetic transitions was considered in the simplest form by including it only in pressure dependence of the Curie temperature for the transition between the paramagnetic and the magnetic phase with the lower magnetic moment FM1. The results show that for pressures lower than some limiting pressure the transition between low- and high-magnetisation phase remains of crossover type. Above this limiting pressure this transition changes to real thermodynamic transition of first order.

Several types of materials have been numerically studied in order to obtain a qualitative picture on their structure after irradiation with heavy ions, neutrons or helium (within the experimental conditions available at JINR). The focus mainly was on getting parameters relevant for the positron annihilation studies, which can shed light on the structural changes caused by irradiation. Using the ABINIT and MIKA software packages, numerical calculations of defects applying the positron lifetime method were performed in Si, WC, WC+n%Co materials after hydrogen/helium implantation. Other materials were also considered, such as graphene on metal substrates, tungsten and boron carbides (WC+6wt%Co and B_4C), anatase (one of the TiO_2 phases), W_2B obtained in the sintering of a certain W, B_4C, TiC, C powder mixture, and in Au thin films. In addition, some semiconducting properties of ZrO_2 with Y as dopant where considered, in connection with development of applications.

High-temperature superconductivity in optimally doped cuprates was studied. Using the Shubin-Kondo-Zener s-d exchange interaction with the Linear Combination of Atomic Orbitals (LCAO) Hamiltonian for the CuO_2 plane, the anisotropy of the electron scattering rate and lifetime observed by Angle Resolved Photo-Emission Spectroscopy (ARPES) were evaluated. The performed qualitative analysis reveals that “cold spots” correspond to nodal regions of the superconducting phase where the superconducting gap is zero, since the exchange interaction vanishes. On the other hand, “hot spots” and intensive scattering in the normal state correspond to regions with maximal gap in the superconducting phase. It is obtained that a separable kernel postulated in the Fermi liquid approach to the normal phase is the same kernel which is exactly calculated in the framework of the s-d approach in the LCAO approximation for the CuO_2 plane. In this sense, at least on the qualitative level, the superconducting cuprates are described by one and the same Hamiltonian applied to their superconducting and normal properties and contributions are: (1) The well-known Pavarini et al. [Phys. Rev. Lett. 87, 047003 (2001)] correlation between the critical temperature $T_{c,max}$ and the shape of the Fermi contour of the optimally hole-doped cuprates is explained. (2) The linear temperature dependence of the Ohmic resistance is caused by wave scattering by thermal density fluctuations in a layered system. (3) Propagation of zero sound in layered transition metal perovskites is predicted in framework of s-d exchange interaction with ferromagnetic interaction. This propagation is possible only close to cold spots directions of the normal charge carriers along the Fermi contour.

The possibility of performing coherent control over the state of a qubit by the propagation of a magnetic soliton in an anisotropic Heisenberg chain is investigated. The physical problem is described by a Hamiltonian involving the interaction between the soliton and the qubit. Under certain conditions the generic Hamiltonian may be mapped on that of a qubit two-level system with matrix elements depending on the soliton parameters. The action of a bright and a dark soliton depending on the driving nonlinear wave function is considered. It is shown, that the qubit can be flipped and/or returned in its initial state, and an equal superposition of qubit “up” and “down” states can be generated. In the case of a local interaction of the qubit with its closest spin on the chain, the off-resonance and effective resonance regimes are studied. To achieve an effective resonance regime for a bright soliton, a fine tuning of the z component of the qubit coupling is required, while in the case of dark

soliton this is not needed due to the smallness small magnitude of the coupling around the time origin.

A way to simplify the circuit of quantum random walk search algorithm, when the traversing coin is constructed by both generalized Householder reflection and an additional phase multiplier is examined. If an appropriate relation between the corresponding parameters is realized, the algorithm becomes more robust to deviations in the phases. In this modification the marking coin is not needed, and all advantages from the above-mentioned optimization to the stability, are preserved. It is shown explicitly how to construct such walk coin in order to obtain more robust quantum algorithm.

The expressive power of the quantum restricted Boltzmann machine Ansatz in order to represent faithfully the operation process of a quantum spin-glass system undergoing an adiabatic quantum computation process is discussed. The representability and the trainability problems associated to the restricted Boltzmann machine Ansatz are addressed when we try to describe the ground state of a quantum Ising spin-glass in a transverse field. In our approach, we obtain the ground state of the system using exact diagonalization and find the optimal restricted Boltzman machine. The preliminary results show that due to the complex spin-glass landscape, for some topologies of the restricted Bolztmann machine, the parameters of the representation are hard to be found.

Half of the world population resides in cities and urban segregation is becoming a global issue. One of the best known attempts to understand it is the Schelling model, which considers two types of agents that relocate whenever a transfer rule depending on the neighbor distribution is verified. The main aim of the study is to broaden our understanding of segregated neighborhoods in the city, i.e. ghettos, extending the Schelling model to consider economic aspects and their spatial distribution. To this end, we have considered a monetary gap between the two social groups and five types of urban structures, defined by the house pricing city map. The results show that ghetto sizes tend to follow a power law distribution in all the considered cases. For each city framework the interplay between economical aspects and the geometrical features determines the location where ghettos reach their maximum size. The system first steps shape greatly the city's final appearance. Moreover, the segregated population ratios depend largely on the monetary gap and not on the city type, implying that ghettos are able to adapt to different urban frameworks.

Segregation affects millions of urban dwellers. The main expression of this reality is the creation of ghettos which are city parts characterized by a combination of features: low income, poor cultural level. Segregation models have been usually defined over regular lattices. However, in recent years, the focus has shifted from these unrealistic frameworks to other environments defined via geographic information systems (GIS) or networks. Nevertheless, each one of them has its drawbacks: GIS demands high-resolution data, that are not always available, and networks tend to have limited real-world applications. Our work tries to fill the gap between them. First, we use some basic GIS information to define the network, and then, run an extended Schelling model on it. As a result, we obtain the location of ghettos. After that, we analyze which parts of the city are segregated, via spatial analysis and machine learning and compare our results. For the case study of Washington D.C., we obtain an 80% accuracy.

The evolution analysis of networks whose links are either positive or negative, representing opposite relationships such as friendship and enmity, has been revealed to be particularly useful in sociological contexts. Using a large relational dataset containing the last two centuries of state-wise geopolitical information (the correlates of war–alliance conflicts), a machine learning approach is presented to predict network dynamics. The combination of geometric, as well as information–theoretic measures to characterize the resulting discrete time series together with the power of deep learning machines is used to generate a model whose predictions are even accurate on the few days in two centuries of international relations when

the typical value (i.e., Alliance or Neutral) changed to a war or a conflict. In other words, the model can predict the next state of the network with a probability of error close to zero

The symplectic and scalable algorithm for spin lattice dynamics embedded in LAMMPS was used to model the coupled relaxation processes of the spin and lattice subsystems to investigate the phonon dispersion of bcc Fe at T = 300 K. The atomic interactions were modelled via three semi-empirical many-body potentials within the embedded atom method, while the distance-dependent spin coupling relied on the Heisenberg-type Hamiltonian. In the state of mutual equilibrium of the spin and atom ensembles, the dynamical matrix and the phonon spectra in bcc iron have been calculated. It has been found that for small to moderate absolute values of the wave vector, the phonon dispersion curves agree well with experimental results obtained from inelastic neutron scattering, while discrepancies between theory and experiment are observed for larger values of the wave vector, especially near the zone boundaries. Moreover, the impact of magnons on the phonon spectra is pronounced for all employed potentials.

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ONGOING RESEARCH PROJECTS:

- Quantum effects in low-dimensional and nanostructured magnetic systems
- Phases and excited states of highly frustrated magnetic systems – BNSF
- Liquid crystal approach for model lipid membrane functions optimization by nanoparticles insertion – BNSF
- Synthesis and theoretical studies of graphene nanostructures – Bulgaria & JINR Dubna
- Magnetic quantum effects in low-dimensional and nanostructured spin systems – BNSF
- Dynamics of low dimensional spin systems – Post doc
- Exchange interactions in nanomagnetic systems

INTERNATIONAL COLLABORATION:

- University of Bielefeld, Germany
- JINR Dubna, Russia

TEACHING ACTIVITIES:

Latex Basics
Computer modeling of complex systems

DEPARTMENT FUNCTIONAL MATERIALS AND NANOSTRUCTURES

LABORATORY

PHYSICS OF MATERIALS AND LOW TEMPERATURES

HEAD: **Prof. Peter Rafailov, PhD**
tel: 979 5718; e-mail: rafailov@issp.bas.bg

TOTAL STAFF: **17**
RESEARCH SCIENTISTS: **12**
ASSOC. MEMBERS: **2**

Prof. D.Z. Dimitrov, PhD; Assoc. Prof. B.S. Blagoev, PhD; Assoc. Prof. K.M. Buchkov, PhD; Assist. Prof. L.K. Yankova; Assist. Prof. V.T. Tomov, PhD; Assist. Prof. K. Pavlov, PhD; S. Boyadjiev, PhD; V. B. Mehandzhiev, MSc;
B. Georgieva, MSc; E. Milanov, MSc; M. Valkovski, MSc.
Technicians: L. Nikova, S. Simeonov, P. Zashev.
Associated member: Prof. M. Gospodinov, DSc, Assoc. Prof. E. Nazarova, DSc

RESEARCH ACTIVITIES:

Growth and characterization of crystals and thin layers with optical, X-ray, electron-microscopic and other methods

The ability to control the orientation of charge carrier spins in various directions is a major goal in spintronics. The phenomenon of charge-spin interconversion (CSI) depends strongly on symmetry. Systems with reduced crystal symmetry allow an anisotropic CSI effect with non-conventional components where the charge and spin currents and spin polarization are not mutually perpendicular to each other. Here we demonstrate experimentally that CSI in the graphene-WTe_2 system generates spins with components in all three spatial directions. Through multi-terminal nonlocal spin precession experiments and with specific magnetic field orientations, we show that it is possible to distinguish CSI from the spin-Hall effect and the inverse spin-galvanic effects.

Continuous layers and individual clusters of tungsten diselenide (WSe_2) were synthesized on quartz substrates by the «thermally assisted conversion» (TAC) method. With optical microscopy and atomic force microscopy mapping, isolated flakes of different shapes are found, mainly concentrated near the edges of the substrate, which tend to form clusters and further overlap into continuous layers, towards the central part of the substrates. Raman spectroscopy and photoluminescence measurements confirmed the existence of atomically thin flakes and continuous layers of 2H-WSe_2. The measured current-voltage characteristics show ohmic behavior, suggesting unlimited prospects for the integration of WSe_2 into various heterostructures.

AlN thin films were grown in a Beneq TFS-200 atomic layer deposition (ALD) reactor with 550 ALD cycles at 330 °C substrate temperature. TMA (trimethylaluminum) and NH_3 were used as precursors. TMA and NH_3 dosing was applied in the form of 180 ms and 90 ms pulses followed by 2s and 9s nitrogen purging, respectively. To study the morphological evolution of AlN thin films, substrates providing different surface kinetics were used: Si-face and C-face at 4° on-axis and along-axis 4H-SiC, and graphene grown on 4H-SiC by sublimation. As shown by atomic force microscopy (AFM), the lowest RMS surface roughness of about 0.8 nm was found for the AlN film deposited on the Si face along the 4H-SiC axis due to its higher surface energy, which provides better nucleation. These results are promising

for further investigation of the properties of AlN thin films for application in surface acoustic wave (SAW) devices.

The object of the following study is a biogenic material obtained from neutrophilic iron-forming bacteria of the genus Leptothrix, cultivated in an artificially created environment. In their life cycle, as a waste product, these bacteria form hollow tubes with a cross-section of hundreds of nanometers and a length of the order of microns. Optical microscopy and scanning electron microscopy were used for characterization. The X-ray diffraction study of a control sample (without inoculated bacteria) demonstrated the important role of the bacteria in Fe^{2+} transformation. In contrast to the biogenic product, where lepidocrocite nanoparticles were present, no traces of stable single-phase iron oxide or oxyhydroxide were detected in the bacteria-free control sample. Biotically produced nanoscale single-phase lepidocrocite was investigated for the presence of a magnetostrictive effect at 4.2 K and 100 K. A positive magnetostrictive effect of the order of 10^{-5} was found in the material, the effect being stronger at lower temperature. Understanding the structural and magnetic properties of biologically produced non-toxic iron oxyhydroxide nanoparticles is important for fundamental research, but may also offer the development of practical applications in electronics and biotechnology.

Carbon-nanostructure research

A theoretical and experimental investigation of the terahertz (THz) conductivity of graphene on metal-based substrates is presented. Analytical equations are derived for the general problem of oblique incidence of the THz beam in time-domain-tuned spectroscopy (TDS) operating in reflection mode. The recorded time-domain signals are then processed to determine the thickness of the substrate, its dielectric frequency dispersion, and the complex graphene conductivity frequency dispersion, which is described by a generalized Drude-Smith model. The method was tested on two samples of graphene deposited by chemical vapor deposition, transferred onto polyethylene terephthalate and cycloolefin polymer substrates with sub-millimeter thickness and characterized by Raman spectroscopy. By working only with the amplitude spectra, the proposed method avoids the problems arising from the phase uncertainties that usually affect reflection-mode TDS measurements. Rapid, non-destructive characterization of graphene sheets is possible directly integrated into the production flow of graphene-based passive or active components using metal-based resonant cavities, such as THz absorbers, metasurface lenses or waveguide antennas.

Samples of predominantly single-layer graphene grown on Cu foil were investigated with respect to the strength of the Cu-graphene bond and the oxidation of Cu to Cu_2O as a way of its relaxation. Correlating results were obtained from Raman and XPS characterization and ellipsometry. We find strong Cu-graphene bonding on the unoxidized Cu grains and a gradual release of this bonding along the “strain” line without significant doping effects. Our ellipsometric results provide an estimate of the typical thickness of the oxide layer, which ranges from fractions of a nanometer to 6-7 nm. We confirm that in the case of a polycrystalline film, the oxidation of copper to Cu_2O under the graphene layer is selective to the grain orientation, with (001)-oriented grains being the most resistant and (011)-oriented being the most susceptible to oxidation. From visualization of graphene grains by liquid crystal coating, it is found that graphene grown on electropolished Cu foil exhibits larger grains with lower defect density.

Alumina nanofilms (ALO) were grown on Si, Cu and graphene/Cu substrates by atomic layer deposition (ALD). Trimethylaluminum (TMA) and deionized (DI) water were used as precursors for the ALD process. To obtain an ALD temperature window for the aforementioned precursors, ALO nanofilms were deposited on Si substrates at temperatures ranging from 50 °C to 300 °C. Based on ellipsometric measurements of the film thickness, the growth rate of the resulting ALO films was between 1.15 Å/cycle and 1.25 Å/cycle.

Ellipsometric and XPS analyzes show that the optimal growth temperature for ALO films is between 150 °C and 200 °C. A graphene layer grown on a Cu foil by chemical vapor deposition (CVD) was covered by the ALD method with an ALO nanofilm. Raman spectroscopy was used to characterize the effect of the deposition of ALO films on the graphene layer. The ALO films in the ALO/graphene/Cu samples are uniform and smooth as seen by spectroscopic ellipsometry, and in addition, an increase in Raman intensity is observed for all graphene spectral bands. The graphene in the ALO/graphene/Cu samples retains its good quality; thus, the obtained heterostructure can be considered as a promising candidate for graphene transfer template.

High-temperature superconductors

The stability of the electro-transport characteristics of a Fe-based superconductor before and after a seven-year period of storage under natural conditions was investigated. The stability of superconducting materials over time is essential from the point of view of high-current practical applications. The comparative analysis of changes in the resistive transition of FeSe (melt) sample in the temperature range 4.2-300K shows a degradation of the parameters and mechanisms of superconductivity associated with the formation of defects in the material.

PUBLICATIONS:

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ONGOING RESEARCH PROJECTS:

- National Scientific Research Fund: Projects КР-06-ПН58/12, КР-06-Н38/10 and КР-06-Н28/8;
- M-ERA 2D-SPIN-MEM project КР-06-DO 02/3; COST Action project КР-06-COST/13;
- BG05M20P001-1.001 National Centre for Mechatronics and Clean Technologies.

INTERNATIONAL COLLABORATIONS:

- National Ciao Tung University, Department of Electrophysics, Hsinchu, Taiwan;
- CNR-Istituto per la Microelettronica e Microsistemi (IMM), Rome, Italy
- Department of Physics ‘E.R. Caianiello’, CNR-SPIN, University of Salerno, Salerno, Italy.

DEPARTMENT FUNCTIONAL MATERIALS AND NANOSTRUCTURES

LABORATORY

PHYSICAL PROBLEMS OF MICROELECTRONICS

HEAD: Assoc. Prof. D. Spassov, Ph.D.
tel: 029795731; e-mail: d.spassov@issp.bas.bg

TOTAL STAFF: **15**
RESEARCH SCIENTISTS: 7

Prof. D.Sc. A. Paskaleva, Assoc. Prof. Ph.D. D. Spassov, Assoc. Prof. Ph.D. Ts. Ivanov, assist. T. Stanchev, physicist E. Gajdarzhieva, technologist S. Tsvetanov, technologist M. Stoicheva, technologist Ch. Petkanov, technologist M. Atananasov

Electromagnetic sensor group: Assist. Prof. L. Mihailov, assist. P. Todorov, mechanical eng. N. Mihaylov, technician V. Altynova, technician N. Ilieva

RESEARCH ACTIVITIES:

The research activities of the Laboratory in the recent years have been focused on nanoelectronics and cover the investigations of thin and ultra-thin nanolaminated and doped high-*k* dielectrics and semiconducting oxides for advanced non-volatile memories. The dedicated Electromagnetic sensor group is mainly engaged in experimental investigations of the Electromagnetic Echo Effect (EMEE) and development of sensors for rapid and contactless analysis of solids, liquids and gases. The research activities during 2021 can be summarized as follows:

The effects of charge trapping in capacitor structures designed for application non-volatile charge-trapping memories have been studied. The investigated memory cells are based on HfO_2/Al_2O_3 nanolaminate multilayer dielectrics obtained by atomic layer deposition (ALD). Two types of memory capacitors in respect to the thickness of the tunnel SiO_2 layer (2.4 and 3.5 nm) were studied. It is shown that the use of 3.5 nm tunnel SiO_2 provides a higher density of trapped positive charge in the HfO_2/Al_2O_3 stack. The record operation in the studied cells (corresponding to trapping of electrons in them) strongly depends on the concentration of electrons in the surface layer of p-Si substrate. At a high concentration of electrons (provided by illumination of the sample in visible light range), a voltage pulse of 1ms duration provides a trapped negative charge density of about 60% of the value obtained with a 10 s pulse at which saturation of the charge trapping occurs due to traps filling. Without illumination, the amount of electrons that can be injected into the dielectric layer is determined by the rate of thermal generation/recombination processes in the p-Si surface region. Since these processes are slow at room temperature, long time pulses are required to fill the traps of the charge trapping dielectric. It was found that the positive charge trapping is slower than electron trapping and does not depend on the illumination of the sample during the application of the corresponding voltage pulse, since the holes injected in this case are main carriers in p-Si. The observed changes in the shape of the capacitance-voltage (C-V) characteristics obtained after positive charge trapping were analyzed and it was demonstrated that they can be related to the release of a part of the trapped holes during the characteristic measurement.

The electric breakdown of memory capacitors with HfO_2/Al_2O_3 nanolaminate layers were studied in dependence on the thickness of the SiO_2 tunnel layer and the post deposition oxygen annealing technological step. Two methods were used to characterize the electrical breakdown: ramp voltage stress and constant voltage stress. It is shown that under the employed parameters

of the layers building up the memory capacitor, the average values of the breakdown voltage do not depend on the thickness of the tunnel oxide and are determined by the HfO_2/Al_2O_3 nanolaminate and the blocking Al_2O_3 layer. The oxygen annealing step results mainly in a certain increase of the low-field breakdowns. The O_2 treatment, however, leads to an increase in the amount of electric charge passed through the structure until the breakdown event, and this effect is more pronounced for capacitors with 3.5 nm SiO_2. The electrical breakdown in the studied structures is accompanied by the accumulation of positive charges and the generation of fast interface states at the SiO_2/Si interface.

Ellipsometric studies of multilayer dielectric structures consisting of 3 different types of dielectrics: Al_2O_3, HfO_2/Al_2O_3 nanolaminate and SiO_2 were conducted. The data obtained from the spectral ellipsometry were analyzed with a special algorithm in order to obtain information about the individual thicknesses of the HfO_2 and Al_2O_3 sublayers building the nanolaminate and their deviation from the nominal one, determined by the deposition rates. It was shown that the method allows a clear distinction of the interfaces between the sublayers. The interface regions were identified as a mixture of HfO_2 and Al_2O_3 oxides and the interface region thickness and HfO_2/Al_2O_3 ratio varies along the stack thickness.

The structural, magneto-optical and dielectric properties of the ZnO layers obtained by atomic layer deposition and doped with Co, Fe and Ni were investigated, as a function of the technological conditions of deposition and the type of the dopant. The doped layers show a pronounced magneto-optical Kerr effect with a strong magnetic response and very high values of the polarization angle, especially in the case of ZnO/Fe. The investigations of the electrical characteristics indicated existence of a ferroelectric behavior in the Fe-doped zinc oxide layers. The rich range of physical phenomena observed in these nanostructured materials opens new perspectives for their application in modern electronic, spintronic and optical devices.

The field effect in samples of polymethylmethacrylate (PMMA) implanted with Si+ ions was investigated. The possibility of forming a conducting channel in the "ion-implanted buried" planar layer with a thickness of about 100 nm, made of nanoclustered amorphous carbon, has been demonstrated. The values of the mobility of the charge carriers in the channel, the contact resistance and the leakage current through the gate in the field effect transistor constructed with organic materials (PMMA) have been determined.

The cleaning properties of mists for various types of pollutants in the air was investigated and optimized.

The effects of equation of state quantization for electron motion on the equilibrium composition of the magnetar crust are investigated. The composition of the crust and the limit beyond which atomic nuclei decay by neutron emission are numerically determined for different magnetic field strengths. Experimental atomic mass measurements are used, supplemented with theoretical masses calculated from Hartree-Fock-Bogolyubov nuclear models. According to numerical simulations, the magnetic fields inside neutron stars could be so strong that even the structure may be changed.

The ground state electron energies, mass correction and mass polarization of low and multiply charged helium-like ions are calculated analytically and numerically. The two-electron Schrödinger equation is solved with a perturbation approach based on explicitly correlated wave functions. The corrections due to extra dimensions are investigated and different approaches to the minimization procedure are considered. The results obtained correspond to all physical and mathematical principles and measurements.

The developed algorithms “Optimal Distribution of Locomotives" and "Dynamic Traction Integrator" have been implemented in the railways with great efficiency and became the basis of the working systems for planning and controlling the movement of trains. With assistance from the laboratory, the algorithms are included in scientific and applied systems of national importance with a large number of workplaces. The "National Railway Infrastructure Company" and almost all other domestic railway carriers as well as the "Technical University"

- Sofia, "Higher Transport University "T. Kableshkov"" and the Kazakh Academy of Transport and Communications have employ the applications. The products are also used in The Corporate Information System for Strategic Analyzes in the BDZ Holding and the systems for up-to-date information on the movement of trains, for online ticketing and feeding of time-table boards in the railway stations.

PUBLICATIONS:

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Veljkovic, S., Mitrovic, N., Davidovic, V., Golubovic, S., Djoric-Veljkovic, S., Paskaleva, A., Spassov, D., Stankovic, S., Andjelkovic, M., Prijic, Z., Manic, I., Prijic, A., Ristic, G., Dankovic, D.. Response of Commercial p-Channel Power VDMOS Transistors to Irradiation and Bias Temperature Stress. Journal Of Circuits, Systems, And Computers, 31, (2022), 2240003. JCR-IF: 1.278

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Todorov P., Peshev Z., Ilieva N., Pérez-Díaz J.L., Ivanov O., Contactless Determination of the Number and Diameter of Fog Droplets Using Gravitational Separation and Measurement of Electrical Signals, Machines. Technologies. Materials, 2022, Vol. 16, Issue 8, 283-285

Ivanov O., Todorov P., Pérez-Díaz J. L., Tiankov T., Principle of Operation and Advantages of a Sensor for Fog Contamination Detection, Security & Future, 6, 2, 2022, 84-86

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Electrical Signals, Proceedings of the XIX International Congress "Machines. Technologies. Materials", 07.09 – 10.09.2022, Varna, Bulgaria, 324-326, IV Technologies

Ivanov O., Todorov P., Pérez-Díaz J. L., Principle of Operation and Advantages of a Sensor for Fog Contamination Detection, Proceedings of VI International Scientific Conference CONFSEC 2022, 8, 1, 2022, 2603-2953

AWARDS:
The project COUNTERFOG carried out by Electromagnetic sensors group received the best executed and most innovative project funded by the EC award.

ONGOING RESEARCH PROJECTS:

Funded by Bulgarian Science foundation:

- Resistive switching and magnetorsistanse effects in ZnO layers doped with transition metals (Co, Ni, Fe) for multifunctional applications
- Multilayered high-*k* dielectric structures for application in non-volatile flash memories
- Investigation of crystallization mechanism of graphene and carbon nanotubes on catalytic surfaces.

INTERNATIONAL COLLABORATION:

- Fraunhofer Inst. of Integrated Systems and Device Technology, Erlangen, Germany.
- Institute of Electronic Engineering, Slovak Academy of Sci., Bratislava, Slovakia.
- Insitute of Physics, St.St. Cyril and Methodius Universty, Skopie, Macedonia.
- University of Nish, Serbia.
- Institute of Physics, Polish Academy of Science. Warsaw, Poland.
- Universidad Autonoma de Baja California, Mexicale, Mexico.

DEPARTMENT FUNCTIONAL MATERIALS AND NANOSTRUCTURES

LABORATORY

ACOUSTOELECTRONICS

HEAD: **Assoc. Prof. Karekin Esmeryan**
tel: +359 02979 58 11; e-mail: karekin_esmerian@abv.bg

TOTAL STAFF: **6**
RESEARCH SCIENTISTS: 2
ASSOC. MEMBERS: 3

Assoc. Prof. Karekin Esmeryan, PhD.; Assist. Prof. Yuliyan Lazarov Ph.D., chemist Yulian Fedchenko, MsC.; chemist Teodor Grakov, BsC.; eng. Lazar Vergov, Stefan Staykov

Associated members: Cor. Mem. Lozan Spassov, DsC.; Assoc. Prof. V. Georgieva; Assoc. Prof. Ekaterina Radeva

RESEARCH ACTIVITIES:

Resonant Structures Using Rayleigh Surface Acoustic Waves (RSAW), Bulk Acoustic Waves (BAW) and Surface Transverse Waves (STW). Applications to Sensors.

Piezoresonance sensors based on Rayleigh Surface Acoustic Waves (RSAW) have been developed, whose active surface is functionalized with single and multilayer Langmuir-Blodgett (LB) films, optimized by thickness and density for the detection of volatile organic compounds. Monolayers of phospholipid (SLP) dissolved in hexane arachidic acid (HDAA) and chloroform arachidic acid (CDAA) were used for this purpose. Several layers of these compounds are deposited on top of each other on the sensing surface of 434 MHz dual-input RPAV resonators in an LB trough to fabricate a highly sensitive quartz surface microbalance (QSM) sensing various organic vapors. The frequency shift as a function of vapor concentration is detected with a vector network analyzer (VNA). Saturated vapors of hexane, chloroform, methanol, acetone, ethanol, and water were investigated after each deposited layer to study the behavior of QSM input attenuation, loaded Q-factor and vapor sensitivity to propose an optimal compromise between these parameters with the aim for maximum sensor performance in real-world conditions. With 2200 ppm and 3700 ppm chloroform sensitivity, QSM devices with HDAA and CDAA coatings reach optimal performance at 15 and 11–15 monolayers, respectively. Surface pressure-optimized single monolayers of phospholipid LB films were found to provide up to 530 ppm chloroform vapor sensitivity with a negligible increase in insertion loss and a decrease in loaded Q-factor. This vapor sensitivity is higher than the mass of the sensing layer itself, making SLP films an excellent choice for QSM functionalization.

The sensitivity of 16-MHz quartz microbalances (QCMs) coated with immobilized electrocross-linked amorphous titanium oxide fibers towards different concentrations of NO_2 was investigated. The morphology, phase, and chemical composition of the sensitized material were determined by high-resolution scanning and transmission electron microscopy (SEM, HRTEM), transmission high-energy electron diffraction (THEED), and X-ray photoelectron spectroscopy (XPS). Shifts of the resonant frequency of the QCM-TiO_2 system over a wide range of NO_2 concentrations. Experimental evidence of reversible gas sorption was found at the lowest tested concentration of 50 ppm. An additional response of the sensor to gradually increasing concentrations of NO_2 up to 5000 ppm was observed. Electrocrosslinked

amorphous titanium oxide fibers have been shown to be promising sensor materials for the detection of harmful gases.

Superhydrophobic Soot Coatings

The possibility of fabricating superhydrophobic carbon soot coatings that support bouncing of supercooled water droplets under dynamic collision, even when the solid surface is covered with frost, has been experimentally confirmed (thereby preventing and/or minimizing the formation of glazed ice under conditions of sub-zero temperatures and high air humidity). By analyzing the impact dynamics of water droplets on two groups of frosted soot coatings, it is shown for the first time that the impalement of liquid meniscus to the surface can be avoided when the soot is mainly composed of macropores and oxygen functional groups below 7 at. %. Such a surface configuration ensures minimal energy losses (qualitatively determined) immediately after the dynamic collision due to the negligible amount of "ice bridges" formed between the individual condensates, which helps to suppress the heat transfer at the solid-liquid interface. The results of this research represent a useful platform for the development of universal icephobic surfaces functioning under severe operating conditions.

A new method for the development of economically expedient and wear-resistant ultra-non-wettable carbon soot coatings is proposed. The deposition of soot from a burning rapeseed oil flame on various synthetic fabrics and their subsequent functionalization with cyanoacrylate glue and fluorocarbon compounds endows the fabrics with exceptional durability to mechanical and temperature interventions (for example, sandpaper abrasion, blade scraping, immersion in liquid nitrogen ($T \sim -196$ °C), twisting, high-velocity water jetting), as well as improved fire resistance, while retaining their water repellency and oleophobicity. The as prepared soot fabrics can adhered to a solid surface and facilitate the recovery of ~60% of the initial motility of human spermatozoa after cryopreservation procedures or used as stand-alone non-wettable membranes. This invention can be considered as the first fundamental stage of safely (without any health concerns) integration of carbon soot in reproductive medicine and development of a new generation of cryogenic and antibacterial medical devices.

Cryopreservation of Human Gametes

Sperm cryopreservation is vital in the battle against human infertility and the use of nanotechnology to deposit hydrophobic carbon soot nanoparticles on the inner walls of cryovials outlines exciting new directions in the development of cryobiology. Doubts related to possible toxicity of this material question its practical applicability, but the analysis of the cytotoxicity of three types of rapeseed oil soot, differing by morphology, chemical composition and surface charge, shows that this category of carbon nanoparticles is non-cytotoxic or at worst slightly toxic to the male gametes. Moreover, increased progressive sperm motility was observed in ~50-60% of patients whose ejaculate was treated with carbon soot due to the occurrence of electrostatic repulsion and biochemical changes in the seminal plasma. These fascinating results open new horizons for the use of rapeseed oil soot as functional activator of human spermatozoa prior to in vitro fertilization.

Review activity

The laboratory is intensively engaged with extended review activity for notable journals (Surface Topography: Metrology & Properties, Surface and Coatings Technology, Applied Surface Science, Surfaces and Interfaces, Applied Thermal Engineering, PLOS One, Nanomaterials, Colloids & Surfaces A) from Academical Publishers as: Elsevier, Institute of

Physics, Wiley, MDPI, American Chemical Society, Royal Society of Chemistry, World Scientific and for institutions as Bulgarian National Science Fund as well.

PUBLICATIONS:

Esmeryan K., Vargas S., Gyoshev S., Castano C., Water droplet bouncing on pre-frosted superhydrophobic carbon soot – a step forward in designing passive icephobic surfaces, Diamond & Related Materials 123 (2022) 108850. JCR-IF: 3.806 Q1

Esmeryan K., Rangelov I., Chaushev T., Hydrophobic soot nanoparticles as a non-cytotoxic motility activator of human spermatozoa, Nanoscale Advances 4 (2022) 2806-2815. JCR-IF: 5.598 Q1

Esmeryan K., Fedchenko Y., Gyoshev S., Lazarov Y., Chaushev T., Grakov T., On the development of ultradurable extremely water-repellent and oleophobic soot-based fabrics with direct relevance to sperm cryopreservation, ACS Applied Bio Materials 5 (2022) 3519-3529.

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Starbova K., Georgieva V., Starbov N., Stefanov P., Georgieva B., Lazarov J., Quartz crystal microbalance sensor for NO_2 detection based on electrospun amorphous titanium oxide fibers, *J. Phys. Conf. Series* 2240 (2022) 012017. SJR: 0.21

Avramov I., Ivanov G., Layer by layer optimization of Langmuir-Blodgett films for surface acoustic wave (SAW) based sensors for volatile organic compounds (VOC) detection, Coatings 12 (2022) 669. JCR-IF: 3.236 Q2

PATENTS:

K. D. Esmeryan and T. A. Chaushev, Method for cryopreservation of human cells and tissues using superhydrophobic carbon soot coatings, *ref. №67427 B1*, April 2022, Sofia, Bulgaria.

K. D. Esmeryan and T. A. Chaushev, A system for complex biochemical analysis of human urine based on a superhydrophobic quartz crystal microbalance, *ref. №67464 B1*, October 2022, Sofia, Bulgaria.

PATENT APPLICATIONS:

K. D. Esmeryan and Y. I. Fedchenko, A method for the fabrication of ultradurable extremely water-repellent carbon soot coatings, filled on 15.03.2022г, Sofia Bulgaria, №113502.

ONGOING RESEARCH PROJECTS:

- Studying the impact of physicochemical characteristics of super-nonwettable carbon soot coatings on their icephobic properties, KP-06-H37/7/06.12.2019.
- An innovative engineering approach for cryopreservation of human gametes, KP-06-H57/1/15.11.2021.

DEPARTMENT NANOPHYSICS

LABORATORY

PHOTOELECTRICAL AND OPTICAL PHENOMENA IN WIDE BAND GAP SEMICONDUCTORS

HEAD: Assoc.Prof. Irina Bineva, PhD
tel: 979 5743; e-mail: irka@issp.bas.bg

TOTAL STAFF: **8**
RESEARCH SCIENTISTS: **5**
ASSOC. MEMBERS: **5**
HONORARY MEMBER: 2

Assoc. Prof. Z. Levi, PhD; Assoc. Prof. P. Terziyska, PhD; Assoc. Prof. T. Hristova-Vasileva, PhD; Prof. D. Nesheva, D.Sc.; Eng. R. Dzhurkova; Eng. V. Dzhurkov; Technologist: E. Zaharincheva;

Associated members: Assoc. Prof. Z. Ivanova, PhD; Assoc. Prof. A. Szekeres, PhD; Prof. S. Alexandrova, D.Sc.; Assoc. Prof. S. Simeonov, PhD; Assoc. Prof. N. Peev, Ph.D;
Honorary member: Prof. E. Vateva, D.Sc, Prof. D. Nesheva, D.Sc.

RESEARCH ACTIVITIES:

ZnSe films with thickness of 50, 70 and 100 nm were prepared on Corning 7059 glass substrates at room temperature by applying continuous or periodically interrupted physical vapour deposition at very low deposition rates (Vd = 0.2 and 0.5 nm/s) and low substrate temperature (~20^{o}C). Part of the as-deposited films was annealed at 200^{o}C for 60 minutes in an inert atmosphere. Spectroscopic ellipsometry results have shown that the porosity of the periodically deposited (PD) films (22-50%) is significantly higher than that of the continuously deposited (CD) ones (<10 %). It has also been found that independently of the film thickness the porosity of the as-deposited PD films obtained at Vd = 0.2 nm/s is very high (~50 %). These observations have been connected with the number of atoms reaching the surface per unit time as well as with the atom diffusion and cluster formation on the substrate surface. The comparison of the experimental optical transmission spectra with simulated spectra of compact ZnSe layer has shown that the CD films transmittance is very similar to that of a compact layer, while the transmittance of the as-deposited PD films depends on their porosity. It has been inferred that the pores in the films prepared at Vd = 0.2 nm/s are of various sizes, some of them being comparable with the incident light wavelength λ, while in the films deposited at Vd = 0.5 nm/s, which have lower porosity, the pore size is significantly smaller than λ. The obtained high porosity is essential for enhancing the gas sensitivity of the films and increases their potential for application as chemical sensors.

Comparative studies of the properties of two groups of ZnO films prepared using a modified sol-gel method and spin-coating technique have been carried out. Monoethanolamine (MEA) was used as a complexing agent for the preparation of the **first group** of layers, and diethanolamine (DEA) for the **second group**. Heating with a hot air flow was applied immediately after the deposition of the precursor solution on the Si/SiO2 substrate, followed by a conventional furnace treatment at 140°C. A part of the layers were additionally annealed at 400°C or 600°C. X-ray diffraction studies have revealed that the presence of MEA in the solution led to crystallization of ZnO (wurtzite) after the furnace treatment at 140°C, while after the same treatment the layers obtained using DEA are amorphous. The size of the ZnO

film crystallites is larger in the films annealed at higher temperature, and under the same annealing conditions, the samples from the DEA group have a smaller crystallite size. The optical band gap Eg of the ZnO films was calculated from their optical transmission spectra in the visible and ultraviolet (UV) regions. It has been found that for both groups of samples, the increase of the annealing temperature leads to Eg decrease (from 3.4 eV at 140°C to 2.27 eV at 600°C).

The photocatalytic activity of one series of ZnO films from each group was measured in terms of malachite green dye degradation under irradiation with UV or visible light. It has been found that all samples demonstrate good photocatalytic activity, successfully degrading the dye up to six consecutive cycles. The layers of the second group have shown higher degradation rates under both types of illumination. These studies were carried out jointly with colleagues from the Institute of General and Inorganic Chemistry and the Institute of Mineralogy and Crystallography at the Bulgarian Academy of Sciences.

Ellipsometric investigation of thin metal oxide films of (ZnO, AZO) layers deposited on Si substrates by Atomic Layer Deposition (ALD), was performed using Woollam M2000D (193-1000 nm) spectroscopic ellipsometer in order to determine thicknesses and optical constants of the layers. Spectroscopic ellipsometry was also used for optimization and control of the FeO deposition (by ALD) on Si substrates. AlN thin films were also investigated (in collaboration with the Institute of Electronics) in order to optimize the deposition process of AlN on SiC substrates.

ZnO/ZnO and ZnO/Ag-nanoparticles/ZnO nanolaminates deposited by RF sputtering were also investigated by Spectroscopic ellipsometry. It was shown that the experimental ellipsometry data for Ψ and Δ dispersion can be used as a criterion for the presence of silver particles. (This work is performed in collaboration with Central Laboratory of Solar Energy and New Energy Sources, Bulgarian Academy of Sciences).

PUBLICATIONS:

Nesheva, D., Grujić-Brojčin, M., Šćepanović, M.J., Levi, Z., Dzhurkov, V., HristovaVasileva, T., Vasić, B., The effects of deposition manner and rate on structure and morphology of porous ZnSe nanolayers: Modification of Phonon Confinement Model for resonant Raman conditions. Journal of Alloys and Compounds, 927, 166942, 2022. Elsevier, SJR: 1.027, JCR-IF: 6.371 Q1

Paskaleva, A., Buchkov, K., Galluzzi, A., Spassov, D., Blagoev, B., Ivanov, Tz., Mehandzhiev, V., Avramova, I., Terzyiska, P., Kovacheva, D., Polichetti, M., Magneto - Optical and Muliferroic Properties of Transition-Metal (Fe, Co, or Ni)-Doped ZnO Layers Deposited by ALD. ACS Omega, 7, 47, 43306-43315, 2022. ACS Publications, SJR: 0.708, JCR-IF: 4.132 Q1

Rafailov, P.M., Sveshtarov, P.K., Mehandzhiev, V.B., Avramova, I., Terziyska, P., Petrov, M., Katranchev, B., Naradikian, H., Boyadjiev, S., Cserháti, C., Erdélyi, Z., Szilágyi, I.M., Growth and Characterization of Graphene Layers on Different Kinds of Copper Surfaces. Molecules, 27, 6, 1789, 2022. JCR-IF: 4.927 Q1

Simeonov, S., Szekeres, A., Spassov, D., Anastasescu, M., Stanculescu, I., Nicolescu, M., Aperathitis, E., Modreanu, M., Gartner, M., Investigation of the Effects of Rapid Thermal Annealing on the Electron Transport Mechanism in Nitrogen-Doped ZnO Thin Films Grown by RF Magnetron Sputtering. Nanomaterials, 12, 1, 19, 2022. JCR-IF: 4.358 Q1

Todorov, R., Hristova-Vasileva, T., Katrova, V., Atanasova, A., Milushev, G., Electronic structure and plasmonic activity in co-evaporated Ag-In bimetallic alloys. Journal of Alloys and Compounds, 897, 163253, 2022. Elsevier, 163253, SJR: 1.027, JCR-IF: 6.371 Q1

Butcher, K.S.A., Georgiev, V., Georgieva, D., Gergova, R., Terziyska, P., Binsted, P.W., Downstream Electric Field Effects during Film Deposition with a Radio Frequency Plasma and Observations of Carbon Reduction. Coatings, 12, 1581, 2022. SJR: 0.482, JCR-IF: 3.236 Q2

Todorov, R., Hristova-Vasileva, T., Atanasova, A., Katrova, V., Thin Ag/Bi coatings as epsilon-near-zero material with low optical losses. Optical Materials, 124, 112040, 2022. Elsevier, SJR: 0.583, JCR-IF: 3.754 Q2

Vitanov, P., Ivanova, T., Dikov, H., Terziyska, P., Ganchev, M., Petkov, N., Georgiev, Y., Asenov, A., Effect of a Discontinuous Ag Layer on Optical and Electrical Properties of ZnO/Ag/ZnO Structures. Coatings, 12, 9, 1324, 2022. SJR: 0.482, JCR-IF: 3.236 Q2

Milenov, T., Terziyska, P., Avdeev, G., Karashanova, D., Georgieva, B., Avramova, I., Genkov, K., Valcheva, E., Structure and Phase Composition Study of Heavy Doped with Carbon Thin Films of TiO2 : C Deposited by RF Magnetron Sputtering. Russian Journal of Inorganic Chemistry, 67, 10, 1509-1520, 2022. SJR: 0.292, JCR-IF: 1.63 Q3

Blagoev, B.S., Delibatov, D.A., Mehandzhiev, V.B., Sveshtarov, P., Terziyska, P., Avramova, I., Rafailov, P.M., Optimization of atomic layer deposition of Al2O3 films as possible template for graphene transfer. Journal of Physics: Conference Series, 2240, 012002, 2022. IOP, SJR: 0.21

Beshkova, M., Blagoev, B.S., Mehandzhiev, V., Yakimova, R., Georgieva, B., Avramova, I., Terziyska, P., Strijkova, V., Morphological evolution of thin AlN films grown by atomic layer deposition. Journal of Physics: Conference Series, 2240, 012005, 2022. IOP, SJR: 0.21

Gergova, R., Sendova-Vassileva, M., Popkirov, G., Dikov, Hr., Grancharov, G., Terziyska, P., Organic bulk heterojunction solar cells spin-coated in ambient environment on flexible and glass substrates. Journal of Physics: Conference Series, 2240, 012030, 2022. IOP, SJR: 0.21

Katrova, V., Hristova-Vasileva, T., Atanasova, A., Strijkova, V., Todorov, R., Optical properties of nanostructured bimetallic films from the Ag-In and Ag-Sb systems and their surface-enhanced fluorescence application. Journal of Physics: Conference Series, 2240, 012007, 2022. IOP, SJR: 0.21

Milanova, M., Donchev, V., Georgiev, S., Kirilov, K., Terziyska, P., Effect of growth temperature on nitrogen incorporation into GaAsN during liquid-phase epitaxy. Journal of Physics: Conference Series, 2240, 012047, 2022. IOP, SJR: 0.21

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Rabadzhiyska, S., Ormanova, M., Valkov, S., Dechev, D., Terziyska, P., Petrov, P., Study of the structure, roughness and optical properties of HfO2 coatings deposited on microscopic glass substrates. Journal of Physics: Conference Series, 2240, 012011, 2022. IOP, SJR: 0.21

ONGOING RESEARCH PROJECTS:

Financed by the Bulgarian Academy of Sciences:

- "Preparation and investigation of nanostructured thin films suitable for environmental protection"

Financed by the Bulgarian Ministry of Education and Science:

- "Development and application of spectrophotometric methods for determination of thin films optical constants" (NSF - No КП-06 ПН57/5) Project coordinator: Assoc. Prof. Tihomir Tenev. Assoc. Prof. I. Bineva, Assoc. Prof. P. Terziyska, Eng. V. Dzhurkov, Eng. R. Dzhurkova-team members.

- "Effects of resistive switching and magnetoresistance in transition metal (Co, Ni, Fe) doped ZnO layers for multifunctional applications" (NSF - КП-06-Н28/9). Project coordinator: Prof. Albena Paskaleva. Assoc. Prof. P. Terziyska - team member.

- "Controlled hybrid ALD/CVD synthesis of 2D transition metal dichalcogenides" (NSF - КП- 06 ПН58/13). Project coordinator: Prof. Dimitre Dimitrov. Assoc. Prof. P. Terziyska-team member.

- "Buffer layer- single crystalline Si substrate" (NSF - КП-06-Н58/2). Project coordinator: Assoc. Prof. Teodore Milenov, Institute of Electronics, BAS. Coordinator of ISSP team: Assoc. Prof. Tihomir Tenev. Assoc. Prof. P. Terziyska-team member

Funded by the Operational Programme Science and Education for Smart Growth, cofinanced by the European Union through the European Regional Development Fund

- Project BG05M2OP001-1.001-0008 "National Center of Mechatronics and Clean Technologies", Operational Program: Science and Education for Smart Growth, Creation and Development of Centers of Excellence. Team members: Prof. D. Nesheva, Assoc. Prof. I. Bineva, Eng. V. Dzhurkov.

INTERNATIONAL COLLABORATION:

Center for Solid State Physics and New Materials, Institute of Physics, SASA, Belgrade, Serbia.

Institute of Engineering, Autonomous University of Baja California, Mexicali, Mexico.

DEPARTMENT SOFT MATTER PHYSICS

LABORATORY

LIQUID CRYSTALS AND BIOMOLECULAR LAYERS

HEAD: Prof. Victoria Vitkova, Ph.D.
tel: +3592 979 5825; e-mail: victoria@issp.bas.bg

TOTAL STAFF: **13**
RESEARCH SCIENTISTS: **7**
HONORARY MEMBERS: **1**
ASSOC. MEMBER: **1**

Prof. Yordan Marinov, D.Sc.; Prof. Julia Genova, Ph.D.; Assoc. Prof. Angelina Stoyanova-Ivanova, Ph.D.; Assist. Prof. Lidia Popova, Ph.D.; Assist. Prof. Zdravka Slavkova, Ph.D.; Assist. Prof. Todor Vlakhov; Chem. Eng. Maria Dencheva-Zarkova; Chem. Eng. Peter Lilov; Biologist Velizar Georgiev; Physicist Ognyan Petkov, M.Sc (part-time); Techn. Assist. Nevena Yotova (part-time); Dr. Poornima Santosh, Postdoctoral Fellow
Honorary members: Academician Alexander G. Petrov, D.Sc. Fellow of the Bulgarian Academy of Sciences; Prof. Isak Bivas, D.Sc.
Associated member: Assoc. Prof. Antonia Zheliaskova, Ph. D.

RESEARCH ACTIVITIES:

In view of chemical biosensor applications, the electrical impedance response of a phospholipid DPPE LB film to methanol vapors was studied at room temperature by applying electrochemical impedance spectroscopy (EIS). DPPE LB film deposited on interdigitated microelectrodes was examined at various concentrations of methanol vapors. The results show that the changes in the complex electrical impedance of DPPE LB film due to adsorption of methanol gas molecules are well detectable. In particular, the reduction of the surface resistivity of the DPPE LB film deposited on interdigitated microelectrodes can be used for detection of methanol vapors. Besides the electrical impedance characteristics of the DPPE LB film, the adsorbed methanol gas molecules lead to frequency-dependent variations in its dielectric properties. Such changes of the impedimetric response can be also used to detect methanol vapors. The results obtained from this study are certainly promising and the methodology will be further investigated. Combining EIS and phospholipid LB films one can develop electrochemical biosensor micro-devices. Further, EIS measurements can provide information on the electrical and dielectric properties of phospholipid LB films in the in-plane direction, as well as on their change upon vapors of volatile organic compounds, such as methanol.

Preparation and investigation of polymer nanocomposite electrolyte membranes by incorporating various nano-fillers as well as sodium and magnesium ions into the host matrix obtained from PEO/PVP mixed polymers was carried out. Attention was paid to the experimental investigation and analysis of polymer nanocomposite membranes (PNMs) obtained by means of the layer-by-layer (LBL) deposition technique. This technique increases the number of “polymer/nanofillers” interfaces, which significantly enhance the room temperature ionic conductivity; mechanical stability; robustness and cycling capability. Fabrication of advanced PNMs opens a new window for the possibility of developing a new

class of smart materials with exceptional properties. Such fabrication procedures of PNMs are economically affordable. They do not require any kind of vacuum technologies, high level of experimental set-up etc.

We studied the influence of a phytoalexin with pharmacological potential on the mechanical and dielectric properties of lipid membrane structures. The structure, mechanical and electrical properties of phosphatidylcholine membranes in the presence of resveratrol were investigated in connection with its use in the prevention of oxidative stress damage in the human body. The degree of hydration, the bending modulus of the membrane and its specific electrical capacity were determined. An increase in the order parameter of the lipid molecules was found at all investigated concentrations of biphenol, which supports the hypothesis of its transmembrane conformation parallel to the lipid molecules and its role as a 'sealer' of the lipid bilayer. The effect of resveratrol on membrane bending elasticity was investigated by analyzing the thermal shape fluctuations of quasispherical lipid vesicles. An increase in the mechanical constant was found with increasing the resveratrol concentration. This result is in agreement with the data from fluorescence spectroscopic measurements indicating the denser packing of the phosphatidylcholine molecules in the presence of the biphenol.

The synthesis of new hemorphin analogues by substitution of amino acid residues in the chain of the endogenous heptapeptide VV-hemorphin-5 (valorphin) allows for tailoring the biological activity and morphinomimetic properties of valorphin. The interactions of synthetic morphinomimetics with biomembrane models were investigated. The influence of valorphin analogues on the physicochemical properties of lipid bilayers was investigated to unravel the membrane-related interaction mechanisms of opioid peptides with cells and subcellular structures. The membrane bending modulus was found to decrease and increased specific electrical capacitance was measured in the presence of modified valorphins with nociceptive activity. The partition coefficient of the studied peptides between the bulk phase and the lipid bilayer was assessed by isothermal titration calorimetry. The obtained results revealed that the inclusion of non-proteinogenic amino acids with different numbers of methylene groups changes the valorphin affinity of valorphin to the membrane. The highest binding coefficient to the lipid bilayer was found for a hemorphine, which in previous studies has been shown to exhibit a variable dose- and time-dependent effect on the visceral nociception in mice. Amongst all valorphins studied the analogue with the highed antinociceptive activity *in vivo* showed the most pronounced changes in the membrane dipole and electrokinetic potentials.

The mechanical characteristics and physicochemical properties of phospholipid membranes were investigated in the presence of amphiphilic peptides exhibiting selective toxicity and antimicrobial and antiproliferative effect as shown in a previous study. The membrane bending modulus in the presence of the studied peptide was determined by thermal shape fluctuation analysis of quasispherical lipid vesicles. Reduced bending rigidity is reported at a certain volume concentration of the peptide testifying to peripheral orientation of the peptide at the lipid bilayer. An estimate of the partition coefficient of the peptide between the aqueous phase and the membrane is presented. The obtained results may be useful in the development of new peptide-based pharmacological substances with improved pharmacokinetic properties and enzymatic stability.

In recent years, special attention has been paid to the preparation and characterization by various structural methods of multifunctional materials, namely ceramics and metal alloys, with regards to their practical application.

Bulk samples of the systems: Y-Ba-Cu-O (with nominal composition: 123; 13-20-33) and Dy-Ba-Cu-O (with nominal composition 123) were obtained by solid-phase synthesis. They were doped with Ag_2O and/or Fe_3O_4 nanopowder, which are introduced after the second stage of their synthesis. According to the literature, silver is believed to reduce porosity and lead to the greater formation of a conductive phase with an orthorhombic structure, while iron improves the magnetic properties of the composite. Various methods such as scanning electron

microscopy (SEM), X-ray diffraction (XRD), energy dispersive analysis (EDX) and magnetic measurements (AC/DC) were used to investigate them. Crystal lattice volumes of pure and doped Dy-123 samples were calculated by XRD. The addition of Fe_3O_4 does not change the lattice volume. EDX and SEM measurements reveal that doping with 2 wt% Fe_3O_4 does not hinder the formation of the superconducting Dy-123 phase as well as the $BaCuO_2$ and CuO phases. On EDX, small amounts of Fe were observed around the crystals of Dy-123, $BaCuO_2$ and CuO, indicating that the dopant does not react with the other elements, does not form its own phase and does not enter the crystal lattices of the available phases. The difference in the XRD-calculated oxygen content results of the two samples was not statistically significant. Results show that doping with 2 wt% Fe_3O_4 does not affect the superconducting properties of the bulk composite ceramic material.

The obtained bulk samples from the Y-Ba-Cu-O system with nominal composition: 123 and 13-20-33 doped with 0.20 wt.% Ag_2O and 0.20 wt.% nano Fe_3O_4 are non-monophasic. The presence of the superconducting Y123 and the non-conducting Y211 phases with an orthorhombic structure, as well as the presence of Y_2O_3, were recorded. The iron additive reacts with the ceramic, forming the magnetic phases $BeFe_2O_4$ and YFe_2O_4, which have a positive effect on the properties of the sample. Silver is uniformly distributed over the surface of the sample with Ag_2O addition. The $BaCuO_2$ phase is observed for both samples. Regardless of the presence of other phases in them, they are superconducting and the transition is very narrow ($\Delta \sim 6$ K) for the iron-doped Y-Ba-Cu-O system, and the presence of new phases with magnetic properties can expand their fields of application.

The obtained results of the conducted research show that the additions of Ag_2O and/or Fe_3O_4 nanopowder with low concentrations to the Re(Y; Dy)-Ba-Cu-O systems improve the superconducting properties by increasing the critical temperature and critical current density of the obtained polycrystalline bulk ceramics with a heterogeneous composition. A homogeneous distribution of iron and silver is observed on their surface, respectively.

For the purpose of electrochemical applications (such as Ni-Zn batteries), we have experimentally studied the electrical impedance of electrochemical systems (ECSs) in which the anodes are obtained from a nano-sized ZnO material doped with a high-temperature superconducting ceramic $Bi_{1.7}Pb_{0.3}Sr_2Ca_2Cu_3Oy$ (Bi(Pb)SCCO 2212) at a concentration of 5, 7 or 10 wt%. The electrically conductive properties of such alkaline electrolyte ECSs were characterized at ambient temperature by electrochemical impedance spectroscopy measurements in the frequency range 0.1 Hz – 100 kHz. The effect of ceramic additives on the complex electrical impedance and electrical conductivity of the studied ECSs was evaluated. It was demonstrated that the incorporation of B(Pb)SCCO 2212 HTSC ceramics at a concentration of 7 wt% into the ZnO anode material of ECSs leads to an increase in their static (DC) electrical conductivity. In addition, the AC conductivity of the ECSs is also improved and approaches the values corresponding to ECSs with undoped ZnO anode through identical ECSs geometrical configurations. These positive effects of the B(Pb)SCCO 2212 ceramics observed by the tested electrochemical systems, combined with the significantly improved properties and other advantages of such additions to the Zn active mass of the electrodes make this ceramic a promising doping material for the development of new electrochemical systems as well as for use in different devices.

Two types of multi-strength orthodontic archwires (Tritanium®, American orthodontics; Bioactive®, GC) measuring 0.016×0.022 inches were studied. They are made of a nickel-titanium alloy, and apply biologically tolerable forces along their length, progressively increasing from front to back. The frontal arch segment distributes the weakest force, the premolar - more and the molar - the largest. The influence of their clinical use on morphology and mechanical properties was determined for periods up to and over 8 weeks. For this purpose, scanning electron microscopy (SEM) and a three-point bending test conducted at a temperature of 36°C, which is close to that of the oral cavity, were used. The obtained data

were analyzed statistically. It was found that for the studied periods, the arcs retain the properties of shape memory and superelasticity. A differential application of forces was also observed along the length of the examined arcs with an increase in the distal direction (from front to back). The released forces in the three areas (frontal, premolar and molar) increase with increasing arch deformation, i.e. at 2 mm deformation, the greatest forces are recorded, which suggests their use in oral breathing patients. No statistically significant differences were observed. The obtained results are extremely important in treatment with fixed orthodontic technique.

Melatonin, a hormone secreted by the pineal gland in the brain, plays a key role in the regulation of sleep-wake cycles. Due to its antioxidant and free radical scavenging properties, it has shown promising protective effects against cancer, and neurodegenerative disorders. Studies pertaining to the use of melatonin and its biophysical effects on cell membranes are important for their biomedical implications. The main goal of our study is to investigate the effect of melatonin on the thermal and structural properties of 1-stearoyl-2-oleoyl-sn-glycero-3-phosphocholine (SOPC) phospholipid membranes using sensitive techniques such as Fourier transform infrared (FTIR) spectroscopy and differential scanning calorimetry (DSC). For this purpose, SOPC lipid bilayer membranes were prepared with different melatonin concentrations (10–50 mol%). DSC results have indicated that melatonin strongly interacts with the membrane lipids in a concentration-dependent fashion to form domains and alters the phase behavior by reducing the main phase transition temperature, lipid order, and increasing membrane fluidity. FTIR data have shown that increasing the melatonin concentration caused a reduction in the wavenumber of C=O and PO_2 antisymmetric stretching vibrations, indicating the possibility of strong hydrogen bonding between melatonin and the SOPC lipid with the nearby water molecules in the environment, suggesting its preferential location near lipid head groups at the interfacial region. Using differential scanning calorimetry, Raman spectroscopy, and molecular dynamics simulations, the joint effect of cholesterol and melatonin on the structural properties and phase behavior of a synthetic lipid membrane was investigated. The results show the presence of a competing effect of the two substances. The effect of cholesterol on increasing the ratio of trans/gauche conformations, accompanied by an increase in bilayer thickness, indicates that cholesterol is embedded and interacts primarily in the region of the hydrocarbon chains. In addition, these interactions have been shown to be modulated by the presence of melatonin, which influences the dynamics of the hydrocarbon chains. Understanding the organization of melatonin in membranes and its influence on membrane properties is important to gain thorough knowledge and to develop safe melatonin-based formulations for therapeutic applications.

Due to their unique physicochemical properties, gold nanoparticles (AuNPs) have gained significant interest in a broad range of biomedical applications such as sensors, diagnosis, and therapy. AuNPs are generally synthesized via different conventional physical and chemical methods, which often use harmful chemicals that induce health hazards and pollute the environment. To overcome these issues, green synthesis techniques have evolved as alternative and eco-friendly approaches to the synthesis of environmentally safe and less-expensive nanoparticles using naturally available metabolites from plants and microorganisms such as bacteria, fungi, and algae. A thematic overview has been prepared, which examines the progress and advances in the synthesis of AuNPs using different biological resources with examples, and their profound applications in biomedicine. A special focus on the biosynthesis of AuNPs using different medicinal plants and their multifunctional applications in antibacterial, anti-inflammatory, and immune responses are featured. Additionally, the applications of AuNPs in cancer theranostics, including contrast imaging, drug delivery, hyperthermia, and cancer therapeutics, are comprehensively discussed.

A series of red wine (Mavrud) nanofiltration experiments were carried during the year. Nanofiltration runs were carried out at a constant-pressure cross-flow on MaxiMem,

Prozesstechnik GmbH filtration system, equipped with a nanomembrane Alfa Laval NF99HF (thin film composite polyester, MWCO 300 Da). Cooling temperature regime using the thermostatic cooling bath LAUDA Alpha RA 8 was applied. Effect of transmembrane pressure and cross-flow rate on the permeate flux were studied in view of membrane fouling. Transmembrane pressure and cross-flow rate were varied in the range of 10 to 50 bar and 1-3 l/min respectively. The observed average permeate flux was found to increase linearly by increasing the operating pressure in the range of 20-40 bar. The observed decrease in the permeate flux at 50 bar can be explained by membrane compression and fouling (including concentration polarization and cake layer resistance). At the same time, the commented effects should be considered reversible, since before each subsequent experiment the solvent flux was completely restored by water filtration. The increase in cross-flow velocity improved the hydrodynamic conditions such as shear stress field close to the membrane. Forced convection imposed by the cross-flow velocity has been expected to reduce concentration polarization and to increase permeate flux. Factors such as solute-membrane interactions as well as membrane cell construction might be also considered. A minor positive influence of the cross-flow rate on permeate flux has been observed for Alfa Laval NF99HF membrane, due to the narrower range of cross-flow rates, within which the measured differences in the permeate fluxes were comparable to the experimental error.

Permeate flux can be significantly improved by applying higher pressure and cross-flow velocity, the former having stronger effect. The obtained results can be explained in terms of membrane fouling. Higher operating pressures enhance the permeate flux but may also lead to increased membrane fouling. Calculations based on the total membrane fouling index have been reported to be in the range of 0.02 to 0.04. In general, the obtained results have confirmed low and reversible membrane fouling. The latter has been estimated by the water flux before and after filtration (and washing). In has been shown that washing the system with water restores the flux through the membrane as the fouling index remains in the limits of 0-2% for all nanofiltration runs.

The reasons for the observed results might be attributed to the membranes characteristics, such as hydrophobicity and molecular weight cut-off (MWCO). Another explanation could be partial blocking of the pores by larger solute molecules. These effects have been usually accompanied by a more pronounced deviation from the mass balance in relation to the permeated solutes, as well as the fact that time duration between washing and filtration affects the nanofiltration kinetics, possibly due to relaxation change of the membrane, or a release of dissolved solute by the membrane polymer.

The available in the literature research on the application of polymer membranes for concentration and purification of bioactive compounds from natural extracts has been summarized, paying special attention to the most commonly used polymer materials. In order to obtain increased efficiency and higher permeability, various polymer membranes have been considered. For these special membrane types through additional cross-linking and modeling of the hydrophobic-hydrophilic behavior of the membrane surface, optimal filtration conditions have been determined for various investigated substances. Chemically stable membranes, with potential application in aggressive organic environments, have been also discussed and commented. Novel membrane systems that successfully overcome the limitations of conventional membrane filtration technologies have been reviewed and discussed to gain higher membrane selectivity in certain specific separation cases.

AWARDS:

Best poster award by European Physical Journal (EPJ), BPU11 Congress, 28 August – 1 September 2022, Belgrade, Serbia, Ognyan Krasimirov Petkov for the poster entitled „Effect

of Melittin on Human Erythrocytes and Rat Liver Mitochondrial Membranes“ by V. Doltchinkova, R. Nikolov, M. Shkodrova, A. Soyanova-Ivanova, O. Petkov and V. Vitkova.

Second Prize for the presentation “Nano-thin phospholipid molecular monolayers: response to cadmium ions studied by recording electrochemical impedance spectra” by Todor E. Vlakhov at 19th National Youth Scientific and Practical Conference, held on November 17 and 18, 2022

PUBLICATIONS:

Vitkova, V., Staneva, G., Hazarosova, R., Georgieva, St., Valkova, I., Antonova, K., Todorov, P.. Interaction of new VV-hemorphin-5 analogues with cell membrane models. Colloids and Surfaces B: Biointerfaces, 220, Elsevier, 2022, 112896. JCR-IF: 5.999 Q1

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Stoyanova-Ivanova, A.K., Lefterova, E.D., Ivanova, G.D., Marinov, Y.G., Hadjichristov, G.B., Stoyanova, A.E.. Impedance spectroscopy study of Ni-Zn electrochemical alkaline systems with anode of nano-sized ZnO doped with conducting ceramics $Bi_{1.7}Pb_{0.3}Sr_2CaCu_2O_y$. Compt. Rend. Acad. Bulg. Sci., 75, 3, 2022, 358-366. SJR: 0.19, JCR-IF: 0.329 Q3

Vlakhov, T. E., Hadjichristov, G.B., Marinov, Y. G., Scaramuzza, N.. Ion Conductivity of Nanocomposite Solid Polymer Electrolyte PEO-PVP-NaIO4 with Added TiO2 Nanoparticles. Comptes rendus de l'Academie bulgare des Sciences, 75, 3, 2022, 349-357. SJR: 0.19, JCR-IF: 0.329 Q3

Vlakhov, T. E., Marinov, Y. G., Hadjichristov, G. B., Scaramuzza, N.. Electrical Conductivity Properties of Solid Polymer Electrolytes PEO-PVP-NaIO4 Filled with TiO2 Nanoparticles.

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Vlakhov, T. E., Marinov, Y.G, Hadjichristov, G. B., Scaramuzza, N. ELECTRICAL CONDUCTIVITY OF COMPOSITES FROM POLYMER POLY(ETHYLENE OXIDE) AND NEMATIC LIQUID CRYSTALS E8. Comptes rendus de l'Acad´emie bulgare des Sciences, 75, 8, 2022, 1115-1122. SJR: 0.19, JCR-IF: 0.329 Q3

Vitkova, V., Staneva, G., Hazarosova, R., Georgieva, St., Valkova, I., Antonova, K., Todorov, P.. Valorphins alter physicochemical characteristics of phosphatidylcholine membranes: Datasets on lipid packing, bending rigidity, specific electrical capacitance, dipole potential, vesicle size. Data in Brief, 45C, Elsevier, 2022, 108716. SJR: 0.13 Q4

Vlakhov, T. E., Hadjichristov, G. B., Marinov, Y.G.. Impedimetric response of phospholipid Langmuir-Blodgett films to methanol vapors. Bulgarian Chemical Communications, 54, B2, 2022, 88-93. SJR: 0.17, JCR-IF: 0.398 Q4

Vitkova, V., Hazarosova, R., Antonova, K., Mitkova, D., Yordanova, V., Momchilova, A., Staneva, G.. Resveratrol stiffens 1-palmitoyl-2-oleoyl-sn-glycero-3-phosphocholine bilayers. Lecture Notes in Networks and Systems: Contemporary Methods in Bioinformatics and Biomedicine and Their Applications, Springer Verlag, 2022, SJR: 0.19 Q4

Dencheva-Zarkova, M., Yankov, D., Genova, J., Tsibranska, I.. Flux and separation efficiency in nanofiltration with mixed solvents. Bulgarian Chemical Communications, 54, 2, 2022, DOI:10.34049/bcc.54.2.5461, SJR: 0.17, JCR-IF: 0.4 Q4

Stoyanova-Ivanova A., Kolev S., Petrova V., Petkov O., Tran L. M., Babij M., Zaleski A., Mikli V., Kovacheva D.. Physicochemical study of bulk Dy-123 doped with nano-Fe_3O_4. Bulgarian Chemical Communications, 54, 2022, 71-76. SJR: 0.17 Q4

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Slavkova, Z, Genova, J, Chamati, H, Boev, V, Yancheva, D. Silver nanoparticles synthesis and their effect on the SOPC lipid structure. Journal of Physics: Conference Series, 2240, 2022, SJR: 0.2

Budime Santhosh, P., Genova, J., Chamati, H.. Green Synthesis of Gold Nanoparticles: An Eco-Friendly Approach. 4, 2, Chemistry, 2022, 345-369

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Vlakhov T., Hadjichristov G. Electrochemical impedance spectroscopy for the study of Na+-ion conducting PEO/PVP solid polymer electrolytes doped with TiO_2 nanoparticles, 2022, 94-100

Vlakhov T., Marinov Y. Nano-thin phospholipidic molecular monolayers: response to Cadmium ions, as studied by electrochemical impedance spectra, 2022, ISSN:1314- 8931, 88-93

Todorov, Stefan T., Popova, Lidia T. On characteristics of some natural waters. 2022. 112-117

ONGOING RESEARCH PROJECTS:

- Research Project “Liquid crystal nanocomposites for applications in photonics, sensorics and biomedicine”, National Science Fund (NSF) of Bulgaria contract № KP-06-N58/6 from 19.11.2021 granted under "Competition for financing fundamental scientific research - 2021". ISSP-coordinator Prof. DSc Y. Marinov

- Research Project “New effects in nano-thin ordered organic films (Langmuir and Langmuir-Blodgett) and their use for conceptual development of a new generation of biosensors for working in a fluid environment at ambient conditions and real-time monitoring of hard-to-find water pollutants (anti-terrorism) or early diagnosis by tumor markers (acronym NanoBioSensors)” (contract № KP-06-OPR 03/9) by the Ministry of Education and Science, through the National Science Fund of Bulgaria (2019 – 2021), ISSP-coordinator Prof. Y. Marinov.

- Research Project “Advanced Nanocomposite Polymer Membranes for Na-, Mg-ions conductive electrolytes, Proton-Exchange, and Chromogenic Applications”, under National Scientific Program “Petar Beron i NIE” (P. Beron), BNSF project ANAPOM (2020-2022), contract № KP-06-DB-1/16.12.2019, Dr. Koduru, ISSP-coordinator Prof. Y. Marinov.

- Research Project “Obtaining and study of nanostructured materials by optical methods and impedance spectroscopy (2018-2021), Bulgarian Ministry of Education and Science under the National, Research Programme “Young scientists and postdoctoral students” approved by DCM #577 / 17.08.2018, coordinator Assist. Prof. T. Vlakhov

- Research Project “Model membrane systems in the presence of biologically active macromolecules: physical and physicochemical parameters in norm and pathology” National Science Fund, Bulgaria – Grant KP-06-N38/14/2019), Coordinator, principal investigator: Assoc. Prof. V. Doltchinkova, Sofia University “St. Kliment Ohridski”; Coordinator from ISSP-BAS /partner organization/: Prof. V. Vitkova

- Bilateral Research Project /ISSP-BAS and Tallinn University of Technology, Estonian Academy of Science (Estonian projects TAR16016 and IUT-T4 / Preparation of composite materials with multifunctional properties: structural studies and application) Coordinator: Assoc. Prof. Dr Angelina Stoyanova-Ivanova

- Research Project “Influence of hydrophobic gold nanoparticles on bending elasticity, stability, phase transition and fluidity of SOPC lipid model systems”, under National Scientific Program “Petar Beron i NIE” (P. Beron), BNSF project AULIP (2020-2022), Grant № KP-06-DB-1/01.12.2020, Dr. Santhosh, ISSP-BAS coordinator Prof. J. Genova

- Research Project “Valorization of valuable bioactive substances and ethanol from grape products through innovative nano- and ultrafiltration membrane processes” National Science Fund, Bulgaria – Grant № KP-06-N47/1/26.11.2020), Coordinator, principal investigator: Prof. D. Christova, Institute of polymers, BAS”; Coordinator from ISSP-BAS /partner organization/: Prof. J. Genova

INTERNATIONAL COLLABORATION:

- BELGIUM: Dr. Christophe Minetti, Faculté des Sciences Appliquées, Université libre de Bruxelles
- ESTONIA: DSc Valdek Mikli, Senior Scientist, Tallinn University of Technology, Estonian Academy of Science
- FRANCE: Prof. Miglena Angelova, Université Paris VI: Pierre and Marie Curie and Paris VII: Diderot; Dr Thomas PODGORSKI, Laboratoire Interdisciplinaire de Physique, UMR 5588 (CNRS – Université Grenoble-Alpes)
- GERMANY: Dr. habil. PD Rumiana Dimova, Department of Theory and Bio-Systems, Max Planck Institute of Colloids and Interfaces
- ITALY: Prof. Nicola Scaramuzza, Dr. Marco Castriota, University of Calabria, Department of Physics and Department of Chemistry and Chemical Technologies
- INDIA: Prof. Krishna Prasad, Centre for Nano and Soft Matter Sciences (CeNS)
- POLAND: Prof. Andrzej Zaleski, Institute of Low Temperature and Structure Research, Polish Academy of Sciences, Wroclaw
- RUSSIA: Prof. Evgeny V. Yurtov, Prof. Marina Koroleva, Dmitry Mendeleev University of Chemical Technology of Russia
  Prof. Grygory Arzumanyan, Joint Institute for Nuclear Research (JINR), Dubna, Russian Federation
  Prof. Yury Ermakov, Dr Oleg Batishchev, Frumkin Institute of Physical Chemistry and Electrochemistry, Russian Academy of Sciences
- SLOVENIA: Prof. Ales Iglic, Dr. Samo Penic, University of Ljubljana

TEACHING ACTIVITIES:

PhD Student Todor Vlakhov (ISSP-BAS); Supervisors: Prof. DSc. Yordan G. Marinov and Prof. Georgi Hadjichristov
PhD Student Eng. Peter Lilov (ISSP-BAS); Supervisor: Assoc. Prof. Dr Angelina Stoyanova-Ivanova

DEPARTMENT PHYSICAL OPTICS AND OPTICAL METHODS

LABORATORY

OPTICS AND SPECTROSCOPY

HEAD: **Assoc. Prof. T. Tenev, Ph.D.**
tel: 979 5752; e-mail: tenev@issp.bas.bg

TOTAL STAFF: **11**
RESEARCH SCIENTISTS: **7**
ASSOC. MEMBERS: ***9***

Prof. K. Panayotov D.Sc.; Prof. G. Hadjihristov, Ph.D.; Assoc. Prof. M. Kuneva, Ph.D; Assoc. Prof. B. Katranchev, Ph.D.; Assist. Prof. I. Milushev, Ph.D.; Assist. Prof. M. Goranova; Physicist E. Stoyanova; Physicist M. Molerova; Engineer O. Avramov; Technician Y. Sarafov.

Associated members: Prof. M. Petrov, D.Sc.; Prof. S. Rashev, D.Sc.; Assoc. Prof. T. Tsvetkova, Ph.D.; Assoc. Prof. R. Peeva, Ph.D.; Assoc. Prof. A. Andreev, Ph.D, Assoc Prof. K. Antonova, Ph.D.; Assoc. Prof. H. Naradikian Ph.D., Assoc. Prof. L. Tsonev, Ph.D.; Assoc. Prof. S. Tonchev

RESEARCH ACTIVITIES:

Spectroscopy of solid ion-conducting polymer nanocomposite electrolytes with added nanoparticles

The role of TiO_2 nanoparticles (10 nm in size) as nanoadditives to improve the ionic conductivity of a complex system composed of the polymers poly(ethylene oxide) (PEO) and poly(vinylpyrrolidone) (PVP) and also the salt $NaIO_4$ as an ion donor. The ion-polymer nanocomposite system PEO/PVP/$NaIO_4$/TiO_2 is novel and specific, and of practical interest. Based on this electrolyte conducting sodium ions, multifunctional materials can be developed, e.g. for efficient Na+-ion batteries as well as applications in organic electronics, mechatronics and sensors. The ionic conductivity, alternating current conductivity, as well as the electric modulus and dielectric relaxation of the studied Na^+-ion electrolyte were determined, at concentrations of TiO_2 nanoparticles from 1 to 5 wt%. The effect of TiO_2 nanoparticles is investigated and explained in terms of dipole reorganization in the ion-conducting nanocomposite complex (and solid polymer electrolyte) PEO/PVP/$NaIO_4$/TiO_2 when an external alternating electric field is applied to this dielectric material. The results of this study are relevant for the preparation of nanocomposite polymer electrolytes with improved ionic conductivity.

Spectroscopy of liquid crystal-based composites and nanocomposites

By means of complex electrical impedance spectroscopy and dielectric spectroscopy in the range from 1 Hz to 1 MHz, samples of ion-conducting composites synthesized from poly(ethylene oxide) (PEO) and nematic liquid crystal E8 were characterized. The obtained experimental data are processed and analyzed, the results are explained. The ionic conductivity, as well as the frequency dependence of the complex electric modulus and the dielectric permittivity of flexible thin films (150 μm thick) of PEO/E8 composites (in composition ratio PEO:E8 = 70:30 wt%) were determined. These films were investigated as a function of temperature in the range 25 – 50 °C, i.e., below the glass transition temperature of

the PEO/E8 composite. Information on the dielectric relaxation processes associated with electrically-controlled applications of the polymer/liquid crystal composite PEO/E8 as a soft-matter dielectric is obtained. Conductivity relaxation was found to have an important role for ion transport influenced by the contribution of polymer segmental relaxation in the PEO/E8 composite. The determined dielectric properties and key characteristics, as well as their temperature dependence, are of practical interest for the application of the soft-hard PEO/E8 composites. The obtained results show that the investigated ion-conducting dielectric material is promising for practical applications in molecular electronics and flexible organic electronics, as well as for dielectric devices, using the unique properties of nematic liquid crystals under electromagnetic field effects. By introducing flexible conductive electrodes, PEO/E8 flexible thin films can be integrated into a variety of electrical/optical/electro-optical/optoelectronic flexible devices widely used at present.

Impedance spectroscopy of phospholipid Langmuir-Blodgett molecular monolayers

In view to the practical application as biosensors, and more specifically detectors for health-hazardous volatile organic compounds, the effect of methanol vapor exposure on previously prepared nano-thin (3–6 nm) Langmuir-Blodgett (LB) phospholipids was investigated. Such films containing the phospholipid Dipalmytoyl Phosphatidyl Ethanolamine (DPPE) deposited on interdigital microelectrodes deposited on quartz substrates were characterized by complex electrical impedance spectroscopy. Impedance spectra of DPPE LB films were measured for different concentrations of the investigated vapors, in the range from 80 to 420 mg/dm^3, at room temperature. The obtained electrical characteristics of the films in the frequency range from 0.1 Hz to 1 MHz were analyzed and interpreted. The effect of the vapor sorption of the studied compounds on the real and imaginary parts, the modulus and the phase of the complex electrical impedance, as well as on the electrical conductivity of the DPPE LB films, for different vapor concentrations was established. The possibility of detecting the investigated vapors, as well as determining their concentration with nano-thin DPPE LB films and with the applied registration technique, was estimated. The detection limit for the specific application case of impedance spectroscopy and interdigital microelectrodes is determined.

Optical spectroscopy of photochromic fluorescent spirooxazines

The fluorescence properties and photo-switching behavior of newly synthesized spirooxazine derivatives were investigated in dilute solutions (10^{-4}–10^{-5} M), under photoexcitation with low-intensity continuous light (1–10 mW/cm^2) in the UV spectral region, as well as by irradiation with light in the red region of the spectrum. The photo-induced fluorescence emission from the investigated solutions is in the blue-green spectral region. The most favorable wavelength of the irradiating UV light was determined to achieve optimal fluorescence photoswitching. It was established that the interaction of spirooxazines with Mg^{2+}, Ca^{2+}, Cd^{2+}, Zn^{2+}+ or Pb^{2+} ions leads to the formation of metal-organic coordination complexes of merocyanine form. As a result, the fluorescence spectra of these photochromic compounds in solution are significantly changed. The obtained photo-physical characteristics are useful for elucidating the photo-chemical mechanism of the photo-transformations of the involved molecular conformers, which allows various their photo-controllable applications. In addition to the optical characterization of the studied spirooxazine derivatives and their complexes with metal ions, quantum-chemical calculations were performed to optimize the electronic and stereochemical structure of the spirooxazine compounds, and accordingly – to optimize their synthesis. For this purpose, experimental FT-IR spectral data were compared with results for vibrational infrared absorption spectra obtained by quantum chemical

calculations for the molecular conformers. The results show that by specific structural modification of the studied spirooxazine derivatives, their fluorescence and photo-switching efficiency can be improved.

Nanoelectronics with ion-implanted polymers

The nano-structure formed in the near-surface region of the polymer polymethyl methacrylate (PMMA) implanted with silicon (Si^+) ions at low energy (50 keV) and an irradiation dose of 10^{16} ions/cm^2 was investigated. With controlled local chemical modification at a depth of 150 – 200 nm, a nano-thin bilayer configuration consisting of an ion-modified layer and an ion-implanted carbon-nanostructured layer was created in PMMA. By electrical measurements, the trans-conductivity of such a complex nano-layered configuration of the organic material in the Si^+-implanted PMMA was determined. The formed ion-implanted planar layer (ca. 100 nm-thick channel) of nano-clustered amorphous carbon as an organic semiconductor gives an electrical field effect in the ion-implanted PMMA. For such an organic field-effect transistor configuration, the values of the key parameters such as carrier mobility, contact resistance, and leakage current, which are of scientific and applied interest, were determined.

Graphene films were grown by chemical vapour deposition (CVD) on Cu foil. We investigated single-layer graphene-coated Cu foils with respect to the Cu-graphene coupling and Cu oxidation to Cu_2O as a way for its relaxation. Correlating results were obtained from Raman and XPS characterization, ellipsometry and the EBSD technique. We find strong Cu-graphene coupling on the nonoxidized Cu grains and gradual release of this coupling along the "strain" line without significant doping effects. Our ellipsometric results provide an estimate for the typical oxide layer thickness beneath the graphene after 6 months of sample aging, which varies from fractional parts of a nanometer to 6–7 nm. We confirm that in the case of polycrystalline foil oxidation to Cu_2O of graphene-coated copper is grain-selective, with (001)-oriented grains being most resilient and (011)-oriented ones most susceptible to oxidation. Correspondingly, the strong Cu-graphene coupling and the strain in the graphene lattice is released very nonuniformly and over different time scales. From a visualization of graphene grains by means of coating by liquid crystal, it is found that graphene grown on electropolished Cu foil exhibits larger grains with lower defect density. It is found that the Cu-graphene coupling alters the frequency and the intensity ratio of the G and 2D band, and a determination of the number of graphene layers from Raman spectral parameters should preferably be made after transfer on isolating substrate.

While surface plasmon excitation assisted by nematic liquid crystal layers has been comprehensively studied in different aspects, application of cholesteric structures for surface plasmon excitation remains an unexplored area. Moreover, structures including cholesteric layers and metal grating for surface plasmon excitation have never been considered, to the best of our knowledge. We studied theoretically and experimentally such structures focusing on different regimes of propagation of normal modes in the cholesteric liquid crystal layer. The application of such structures for sensing is accordingly discussed.

A nanocomposite built of the hydrogen-bonded dimer liquid crystal 4-heptyloxybenzoic acid (7OBA) and Graphene oxide (GO) exhibiting a cascade of phase transitions and phases not typical for pristine 7OBA, was investigated by optical polarization microtexture analysis and Raman spectroscopy. We use the 7OBA/GO nanocomposite beacause of the effective functionalizing properties of GO to study the generation and development of the ferroelectric smectic CG phase in its bi-tilted configuration. For the appearance of the CG phase with its substructures we propose an explanation based on the HB formation between 7OBA dimers in closed and open conformations and the GO sheets, aided by the π–π interaction with graphene hexagons which are free from functional groups. We

propose molecular and macroscopic models for the substructures CG_{cl} and CG_{ln} and indicate their triclinic C_1 symmetry, characterized by a polar vector directed oblique to the layers. This confirms that the bi-tilted CG and its substructures are ferroelectric in the bulk similarly to solid ferroelectrics, thus showing potential as material for controllable photonic functional devices.

The work on determining optimal modes of the technological process for the realization of films with the necessary characteristics (refractive index, density, strength, adhesion, etc.) with new optical materials with the Symphony 9 vacuum deposition system of the Tecport Optics company, purchased at Operational program "Development of the competitiveness of the Bulgarian economy".

Multilayer coatings for the visible and near-UV region - anti-reflective and specular - are being developed using resistant oxides - ZrO_2, TiO_2, Al_2O_3 and SiO_2. Spectrophotometric methods are being developed to determine the optical constants of thin layers. The sensitivity of the multilayer coatings to the accuracy of the determined optical constants and technological parameters is investigated.

A large number of spectrophotometric studies have been carried out in various spectral regions by colleagues from the Institute, from institutes in the BAS and external users. The institute has signed framework agreements for cooperation and joint activity with companies from the industry involved in the development or use of optical coatings - Optics AD, Kimcoop Holding Ltd., Milkotronic Ltd., Opteko and Partners Ltd., Technooptics Ltd..

We continue our research on stable dissipative light bullets in Kerr cavities. These three-dimensional (3D) localized structures consist of either an isolated light bullet, bound together, or could occur in clusters forming well-defined 3D patterns. They can be seen as stationary states in the reference frame moving with the group velocity of light within the cavity. We demonstrate the existence of breathing dissipative light bullets in a birefringent optical resonator filled with Kerr media. The propagation of light inside the cavity for each polarized component, which is coupled by cross-phase modulation, is described by the coupled Lugiato-Lefever equations. The space-time dynamics of breathing light bullets are described using Stokes parameters and frequency spectra.

We consider coupled weakly birefringent cavities filled-in with nonlinear Kerr material and subject to linearly polarized optical injection. Light propagation in such a system is described by a system of discrete Lugiato–Lefever-type equations for each linear polarization component of the electric field into each cavity, coupled by the cross-phase modulation terms and the neighboring waveguides field overlap integrals. We demonstrate that this system supports stable three-dimensional vector localized structures often called discrete vector light bullets. We consider both anomalous and normal dispersion and show that it results in the generation of, respectively, bright and dark discrete vector light bullets. Due to the polarization multistability of the system, we demonstrated coexisting light bullets with polarization at the light bullets peaks as different as predominantly linear to predominantly circular. We have shown that chaotic spatiotemporal dynamics can be realized even for such an injection strength for which the light bullets distribution in the system is stationary by increasing the coupling strength between the cavities.

We investigate the formation of vector solitons in weakly birefringent high-Q resonators. The presence of nonlinear polarization mode coupling in optical resonators subject to a coherent optical injection allows stabilizing up to two families of bright or dark vector dissipative solitons, depending on the dispersion properties of the system: anomalous or normal dispersion regimes. In these regimes, the input–output characteristics possess either a bistable or a tristable homogeneous response. The stabilization mechanism of these localized states is attributed to a subcritical modulational instability in the case of anomalous dispersion and to a front-locking mechanism in the normal dispersion regime. Their bifurcation diagrams

exhibit either a homoclinic or a heteroclinic snaking type of instability, depending on the dispersion properties.

We experimentally demonstrate a new approach of broadband comb generation based on the polarization mode competition in single-mode VCSELs. More specifically, we analyze nonlinear dynamics and polarization properties in VCSELs when subject of optical injection from a frequency comb. When varying injection parameters (injection strength and detuning frequency) and comb properties (comb spacing), we unveil several bifurcation sequences enabling the excitation of free running depressed polarization mode. Interestingly, for some injection parameters, the polarization mode competition induces a single or a two-polarization comb with controllable properties (repetition rate and power per line). We explain our experimental findings by utilizing the spin-flip VCSEL model (SFM) supplemented with terms for parallel optical injection of frequency comb. We provide a comparison between parallel and orthogonal optical injection in the VCSEL when varying injection parameters and SFM parameters.

We investigate experimentally the polarization dynamics of a vertical external-cavity surface-emitting laser with a saturable absorber mirror in the cavity. We demonstrate that the normalized Stokes parameters and degree of polarization are functions of time reaching extreme values around the pulse peaks. Our experiments show that light is elliptically polarized, being able to have a circular right-handed or left-handed component, depending on the orientation of the saturable absorber mirror

A complex study of the impact of substrate pre-annealing on the formation of proton-exchanged planar optical waveguides in X-cut lithium niobate has been performed using structural (XRD and IR absorption spectroscopy) and optical (optical microscopy and mode spectroscopy) methods. It has been established that homogenizing thermal pre-treatment reduces the strains and dislocation density in proton-exchange waveguides. It is shown that structural phase transformations during post-annealing occur more rapidly in homogenized by pre-annealing samples. The results obtained contribute to better understanding of processes which take place during the proton exchange and the post-exchange annealing and their dependence on the state of crystal surface. These results are of importance for producing lithium niobate based phase and amplitude modulators with stable parameters used in navigation systems, telecommunications and sensors.

This year marked the 50th anniversary of ISSP–BAS and of one of its largest laboratories – the Laboratory of optics and spectroscopy. The review on the work of the scientists of the Laboratory in three parts is dedicated to this anniversary. The first part comprises some historical notes and a short overview of the scientific and applied achievements in two of the nine areas of the Laboratory's work – thin-layer and multilayer structures and integrated optics.

PUBLICATIONS:

Hadjichristov, G.B., Ivanov, Tz.E. Near-Surface Nanostructuring of Polymethylmetha-crylate by Silicon Ion Implantation. Journal of Nano Research, 72, 95-112, 2022. JCR-IF: 1.78

Panajotov, K, Tlidi, M, Song, Y, Zhang, H. Discrete vector light bullets in coupled chi3 nonlinear cavities. Chaos, Solitons and Fractals, 163, 112532 Elsevier, 2022. JCR-IF: 9.9

Rafailov, P. M., Sveshtarov, P. K., Mehandzhiev, V.B., Avramova, I., Terziyska, P., Petrov, M., Katranchev, B., Naradikian, H., Boyadjiev, S., Cserháti, C., Erdélyi, Z., Szilágyi, I. M. Growth and Characterization of Graphene Layers on Different Kinds of Copper Surfaces. Molecules, 27, 6, 1789, MDPI, 2022. JCR-IF: 4.927

Stoyanova-Ivanova, A.K., Lefterova, E.D., Ivanova, G.D., Marinov, Y.G., Hadjichristov, G.B., Stoyanova, A.E. Impedance spectroscopy study of Ni-Zn electrochemical alkaline systems with anode of nano-sized ZnO doped with conducting ceramics $Bi_{1.7}Pb_{0.3}Sr_2CaCu_2O_y$. Compt.Rend.Acad.Bulg.Sci., 75, 3, 358-366

Vlakhov, T. E., Hadjichristov, G. B., Marinov, Y. G. Impedimetric response of phospholipid Langmuir-Blodgett films to methanol vapors. Bulgarian Chemical Communications, 54, B2, 88-93, 2022

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Vlakhov, T. E., Marinov, Y. G., Hadjichristov, G. B., Scaramuzza, N. Electrical Conductivity Properties of Solid Polymer Electrolytes PEO-PVP-NaIO4 Filled with TiO_2 Nanoparticles. 75, 6, Comptes rendus de l'Académie bulgare des Sciences, 804-811, 2022

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Wu, L, Fan, T, Wei, S, Xu, Y, Zhang, Y, Ma, D, Shu, Y, Xiang, Y, Liu, J, Li, J, Panajotov, K, Qin, Y, Zhang, H. All-optical logic devices based on black arsenic–phosphorus with strong nonlinear optical response and high stability. Opto-Electronic Advances, 5, 200046, 2022. JCR-IF: 8.933

Котликов Е.Н., Лавровская Н.П., Тенев Т.К., Милушев И.К. Исследование и применение оптических плёнок оксида алюминия Al_2O_3 в ультрафиолетовом диапазоне спектра. Оптический журнал, 89, 12, 82-89, 2022

Сосунов А. В., Петухов И. В., Журавлев А. А., Пономарев Р. С., Мололкин А. А., Кунева М. К. Влияние предварительного отжига пластин ниобата лития на характеристики протоннообменных волноводов. Кристаллография, 67, 6, 982-989, 2022

ONGOING RESEARCH PROJECTS:

Financed by the Bulgarian Academy of Sciences (budget subsidy)**:**

- " Optics, electro-optics and spectroscopy of new materials and systems", 2021/2023, budjet subsidy from the Bulgarian Academy of Sciences (BAS)

Financed by the Bulgarian Ministry of Education and Science:

- Research project No КП-06 ПН57/5 (from 19.11.2021):
  "Development and application of spectrophotometric methods for determination of thin films optical constants" funded by the Ministry of Education and Science of Bulgaria, through the National Science Fund of Bulgaria

- Research project No КП-06-Н58/2 (from 19.11.2021):
  "Study of the possibilities for deposition of two- or multi- layered structures of type: graphene bufer layer- single crystalline Si substrate" funded by the Ministry of Education and Science of Bulgaria, through the National Science Fund of Bulgaria

- Research project No. КП-06-N58/6/2021 (from 19.11.2021):
  "Liquid crystal nanocomposites for applications in photonics, sensorics and biomedicine",
  funded by the Ministry of Education and Science of Bulgaria, through the National Science Fund of Bulgaria

- Research project No. КП-06-Russia/8 (from 11.12.2020) in the frame of Bilateral Cooperation Program 'Bulgaria – Russia':
  "Investigation of the deposition of Langmuir-Blodget nano biofilms and their interaction with various types of acoustic waves in piezoelectric structures",
  funded by the Ministry of Education and Science – the National Science Fund of Bulgaria

- Research project No. КП-06-OPR 03/9 (from 5 Sep 2018):
  "New effects in nano-thin ordered organic films (Langmuir and Langmuir-Blodgett) and their use for conceptual development of a new generation of biosensors for working in a fluid environment at ambient conditions and real-time monitoring of hard-to-find water pollutants (anti-terrorism) or early diagnosis by tumor markers (acronym NanoBioSensors)", funded by the Ministry of Education and Science, through the National Science Fund of Bulgaria (current) (with extension until the end of 2022)

- "Improvement of ion exchange methods for obtaining optical waveguides for highly stable integrated optical systems", Contract with Ministry of Education and Sciences of Perm region - № C-26/848 and with Russian Federation research Funf and Perm region, Project № 20-42-596001. Term: 2021-2023

INTERNATIONAL COLLABORATION:

- Department of Applied Physics and Photonics (TW-TONA), Vrije Universiteit Brussel, B-1050 Brussels, Belgium
- Faculté des Sciences, Université Libre de Bruxelles, Campus Plaine, C.P. 231, Brussels B-1050, Belgium
- Shenzhen Engineering Laboratory of Phosphorene and Optoelectronics, Collaborative Innovation Center for Optoelectronic Science and Technology, Institute of Microscale Optoelectronics, Shenzhen University, Shenzhen 518060, China
- Chaire Photonique, LMOPS, CentraleSupélec, 2 Rue Edouard Belin 57070 Metz, France
- Łukasiewicz Research Network–Institute of Microelectronics and Photonics, Al. Lotników 32/46, 02-668, Warsaw, Poland
- Laboratoire de Physique des Lasers, Atomes et Molécules, CNRS UMR 8523, Université Lille 1 - 59655 Villeneuve d'Ascq Cedex, France

TEACHING ACTIVITIES:

Assoc. Prof. M. Kuneva, Ph.D

- 120 hours Introductory course of physics for students on English, University of Architecture, Civil Engineering and Geodesy, Sofia, 2022
- 60 hours laboratory exercises in physics, bachelor program, Technical University, Sofia, 2022
- A series of 10 invited lectures for the Perm State Research University on the topic: "Optical waveguide layers obtained by proton exchange in lithium niobate and lithium tantalum: technology and research methods", 2022

Department LASER, ATOMIC, MOLECULAR AND PLASMA PHYSICS

LABORATORY

ATOMIC SPECTROSCOPY

HEAD: **Assoc. Prof. Valentin Mihailov,PhD**
tel: 979 5740; e-mail: valentin@issp.bas.bg

TOTAL STAFF: **6**
RESEARCH SCIENTISTS: **6**

Assoc. Prof. Boian Torosov, Ph.D.; Assoc. Prof. Galina Malcheva, Ph.D.;
Assist. Prof. Hristina Hristova Ph.D.; Assist. Prof. Vani Tankova Ph.D
Assist. Prof Vasilka Steflekova

RESEARCH ACTIVITIES:

The laboratory "Atomic spectroscopy" carries out scientific research in the field of quantum optics and analytical atomic spectroscopy.

In the field of quantum optics, comprehensive experimental tests of various composite pulse sequences were performed using one of IBM's open-access quantum processors based on superconducting (transmon) qubits. Experiments were performed controlling the pulses interacting with the qubits, using the low-level access to the backend provided by IBM Quantum. A wide variety of broad-spectrum, narrow-spectrum, and rectangular excitation profiles were obtained, producing any preselected target probabilities ranging from zero to one. Universal composite pulses that compensate for errors in each experimental parameter were tested. Excellent agreement between the theoretical and experimental excitation profiles was observed in all experiments. This proves both the effectiveness of composite pulses as a very efficient and flexible quantum control tool and the high quality of IBM's quantum processor. As an extreme example, a distinct narrowband excitation profile was tested and observed for a composite sequence of 1001 pulses.

A new method of quantum control using "polychromatic pulse trains" consisting of pulses with different frequency with respect to the qubit transition frequency was also developed and experimentally tested. Multiple polychromatic pulses are extracted that generate broad-spectrum, narrow-spectrum, and rectangular excitation profiles for different target transition probabilities. This makes it possible to create high-accuracy excitation profiles that are either (i) robust to variations in experimental parameters, which is attractive for quantum computing, or (ii) more sensitive to such variations, which is attractive for noise elimination from cross-excitation as well as for quantum sensors. The method has been demonstrated experimentally using one of IBM's superconducting quantum processors, in very good agreement between theory and experiment. These results demonstrate the accuracy, robustness and flexibility of the proposed quantum control technique. They also show that the frequency difference is a control parameter that is as effective as the pulse phase commonly used in composite pulses. Therefore, the method opens up various perspectives for quantum control in areas where phase manipulation is difficult or imprecise.

The work in the field of analytical atomic spectroscopy was related to archeometric study of 22 ceramic fragments of vessels from the Neolithic settlement near the village of Galabnik, located on the territory of today's Western Bulgaria, Pernik region. The aim was to determine the elemental composition of red, brown and black ornaments on the examined ceramics and on this basis to classify them into separate technological groups.

Analyzes of the samples were performed in both the decoction and the agoba using a portable LIBS system. The results of the study showed that all the ceramic samples contained main elements Fe, Al, Si, Ca, Mn and Mg and minor elements Ti, Sr, Ba, Na, K and Li, as well as traces of Cu. These elements are found in both decorations and engobes, but with different ratios of the intensities of the emitted spectral lines and therefore with different concentrations. The focus of the research was on the semi-quantification of manganese and iron in the decorations, since the content of manganese and iron oxides in the pigment determines its color. The 475.4 nm atomic spectral line for manganese and 302.06 nm for iron were used to distinguish samples with increased amount of Mn from those with higher amount of Fe. In seven of the ceramics, an increased manganese content was found in the decoration compared to the engobe.

Using the statistics program - SPSS Statisticsc, a component analysis (Principal Components Analysis (PCA)) of the obtained experimental data was performed. In addition to the data for Mn and Fe concentration, the ratios in the concentration of all other elements in the decorations and engobe of the studied ceramics are also included. The results showed that increased Fe concentration was observed in samples with red decoration. In part of the samples with brown and black decoration, high concentrations of Mn were found, and in another part - a high concentration of Fe. From the samples with increased Mn content (7 pieces), five have a brown decoration color, and two have a black color. Based on the obtained experimental results, the studied objects are divided into separate groups according to the probable technologies used for their production.

In ancient times, one of the ways to obtain brown and black coloring was the use of manganese and iron oxides. A dark brown to black color can be achieved by two different methods. First, by firing an iron-based pigment in reducing conditions, and the second technique is by applying a manganese-based pigment, which gives a dark brown color regardless of the firing conditions. From the results obtained by us, it can be concluded that the dark brown color in 5 of the fragments and the black color in 2 of them were probably made by applying mineral pigments with a high content of manganese oxides, while in the remaining fragments with brown decoration red pigments were used as iron reduction technique was applied. Determination of the mineral composition of the ornaments by FTIR is pending.

PUBLICATIONS

Kisov, H., Blagoev, K., Tankova, V., Georgieva, B., Strijkova, V., Petrova, P., Dyankov G. Organic random laser generation by stimulated cascaded four-wave mixing, Optics and Laser Technology, 148, Elsevier Ltd., 2022, 1-7. JCR-IF: 4.94

Yankov G., Karatodorov S., Mihailov V., Tankova V., Iordanova E., Nedyalkov N., Damage threshold in ablation regime induced by femtosecond laser irradiation on transparent media, Comptes Rendus de l'Académie Bulgare des Sciences, 322, 2022. JCR-IF: 0.326

Torosov B. and Vitanov N., Experimental Demonstration of Composite Pulses on IBM's Quantum Computer, Physical Review Applied 18, 034062 (2022). JCR-IF: 4.931

Ivanov S., Torosov B., and Vitanov N, High-Fidelity Quantum Control by Polychromatic Pulse Trains, Physical Review Letters 129, 240505 (2022). JCR-IF: 9.185

ONGOING RESEARCH PROJECTS:

- Applications of laser ablation for the study of traditional materials in cultural heritage (under the Academy's bilateral agreements with National Institute for the

Development of Optoelectronics - INOE 2000, Bucharest, Romania).

- Atomic and Plasma Physics (funded by the budget subsidy of BAS),
- Contract КР-06-ПМ-68/1 dated 30.11.2022 "Spectroscopic analysis of pigments used to decorate Neolithic and Chalcolithic ceramics by laser-induced plasma spectroscopy (LIBS) and Fourier transform infrared spectroscopy (FTIR)"

INTERNATIONAL COLLABORATION:

- National Institute for Research and Development of Optoelectronics (INOE 2000), Magurele, Romania,
- French National Research Center CNRS, Institute of Physics,
- Faculty of Physics, Jagiellonian University, Krakow, Poland,
- Faculty of Physics, University of Belgrade, Serbia.

DEPARTMENT – LASER, ATOMIC, MOLECULAR AND PLASMA PHYSICS

LABORATORY

METAL VAPOUR LASERS

HEAD: **Professor Krassimir Temelkov, PhD**
tel: 979 5708; e-mail: temelkov@issp.bas.bg

TOTAL STAFF: **20**
RESEARCH SCIENTISTS: **15**
HONORARY MEMBERS: **1**
ASSOC. MEMBERS: **1**

Assoc. Prof. Dimo Astadjov, PhD; Assoc. Prof. Ekaterina Iordanova, PhD; Assoc. Prof. Peter Zahariev, PhD; Assoc. Prof. Lyubomir Stoychev, PhD; Assoc. Prof. Todor Petrov, PhD; Assist. Prof. Stefka Slaveeva, PhD; Assist. Prof. Georgi Yankov, PhD; Assist. Prof. Stefan Karatodorov, PhD; Assist. Prof. Danka Yordanova, PhD; Assist. Prof. Ognian Sabotinov, PhD; Physicist Viktoria Atanassova, PhD; Physicist Kaloyan Zlatanov, PhD; Assist. Ivan Kostadinov; Assist. Krassimir Dimitrov; Physicist Blagovela Blagoeva; Physicist Mariam-Rima Halil Shehadi, B. Sc.; Sc. Eq. Worker Lyubomir Kandov

Associated member: Margarita Grozeva, PhD;
Honorary members: Academician Nikola Sabotinov, DSc, PhD, member of BAS

RESEARCH ACTIVITIES:

A unique master oscillator – power amplifier (MO–PA) strontium vapor laser system delivering diffraction-limited laser radiation (M^2 = 1) in the middle infrared (MIR) spectral range with a record-high average laser power of 14 W was developed and studied. The lifetime of the two sealed-off laser tubes exceeded 500 hours, which is typical lifetime for the sealed-off laser tubes produced by the manufacturing companies. Using a parabolic short-focus mirror for the laser beam focusing, spherical aberrations were overcome and a crater diameter and a channel width of the order of the laser wavelength (6.45 μm) were made for the first time through drilling and cutting. Precise microprocessing (microdrilling, microcutting, microwelding, etc.) of various materials, such as optical grade fused quartz, sapphire, stainless steel (SS), hard biological tissues and polymers, was also accomplished. Microcutting of 2-mm optical grade silica and 0.6-mm sapphire was realized via multi-contour trepanning. Microcraters with high aspect ratio, which is defined as the ratio of the crater depth to the crater diameter, were also drilled in optical grade fused quartz with wave-guide-assisted microdrilling. Diffraction-limited laser radiation (M^2 = 1) was obtained for the first time on self-terminating metal transitions, using a small-bore discharge tube placed in flat-flat stable cavity instead of the unstable cavity from the negative or positive branch with certain magnification *M*.

A unique MO–PA copper bromide vapor laser system yielding diffraction-limited laser radiation (M^2 = 1) in the visible spectral region at the atomic copper lines 510.6 nm and 578.2 nm was developed and investigated. The highest beam quality (M^2 = 1.02 ± 0.02) achieved so far through laser system oscillating in the visible spectral range on the self-terminating metal transitions was confirmed with detailed study on the laser beam divergence, applying two independent methods. For the single-transverse-mode (TEM_{00}) solid-state laser system made by leading enterprises the beam propagation factor M^2 is well in the range 1.05–1.3. When focusing, a laser power flux exceeding 1 $TW.cm^{-2}$ was obtained that significantly expanded the application possibilities. Precise microdrilling, microcutting and microscribing of different

materials, namely optical grade silica, various ceramics, silicon (Si) and SS, were accomplished. Though the aspect ratio of the crater (300) drilled in optical grade fused quartz through the wave-guide-assisted method was three-times higher than the one obtained so far (100), a new method was also developed and utilized to limitlessly increase the aspect ratio for transparent materials.

Multilevel system, whose intermediate state is continuum, was studied at Raman-type excitation. Based on the results obtained a new method for chiral resolution was developed, at which for specific laser field settings one of the two enantiomer was ionized considerably more than the other. It allowed two enantiomers in a mixture to be spatially separated and to determine the percentage content of the two enantiomers in the sample. The Morris – Shore transformation was investigated and expanded to include various experimental cases with time-dependent laser field. Such a transformation permitted the use of already-known adiabatic techniques, namely RAP, STIRAP, etc., in the multilevel problems.

A study on possibilities for modification, functionalization and structuring of different types of synthesized biopolymer materials (2D thin layers and 3D matrices) was carried out with femtosecond laser radiation, as the laser parameters (laser pulse energy, pulse repetition frequency, number of the applied pulses, scan speed, etc.) were optimized for each material and the type of processing. The laser ablation and destruction thresholds were also experimentally determined for transparent media (doped or pure borosilicate glasses with gold nanoparticles with concentration of 1%) via femtosecond laser radiation at the fundamental wavelength of 800 nm.

Using an advanced z–scan system equipped with the new femtosecond laser system Pharos PH2-SP-HP (LIGHT CONVERSION Inc.), nonlinear refractive n_2 and absorption β coefficients were determined for broadband semiconductors GaN and AlN. Applying femto- and subpicosecond laser pulses, the laser destruction threshold was also experimentally determined for both materials, as the structural changes after the laser-matter interaction and the two values of the laser destruction threshold were determined – the first, at which initial changes were observed in the sample structure without any optical effect, and the second, at which there were visible residual changes. Developing a special holder for the samples studied and using advanced thermal insulators made by Aspen – CrioAerogel, analogous measurements of the same semiconductors were made for temperature in a wide range.

Precise determination of the losses along the optical path for the laser beam utilized for cooling of rubidium atoms in magneto-optical trap.

Elemental and mineralogical composition of 22 pigments used for decoration of Neolithic ceramic dishes was determined on the territory of Pernik. Spectral investigation were carried out through the laser breakdown plasma spectroscopy (LIPS), as the focus of research was in the ratio measurement of composition of Mn and Fe, which were determinative of the fragments' colour. For determination of the mineralogical composition FTIR method was used in the infrared spectral range (4000–400 cm^{-1}). The presence of hematite was identified in most samples, as only in some samples traces of magnetite, manganite and maghemite were detected. Gypsum, calcite, quartz, silicate and feldspar, which are natural ingredient of the pigments, were also found. Statistical analysis (Principal Components Analysis – PCA) of the experimental data obtained with LIPS and FTIR methods was also carried out. The PCA results were applied for the classification of the investigated ceramics in separate groups and conclusions concerning the technologies for their crafting were made on this basis.

AWARDS:

Academician Nikola Vassilev Sabotinov, DSc was awarded by the President of Republic of Bulgaria with the order necklace "Sts Cyril and Methodius" on the 5th of

May 2022 (Decree № 260 on the 4th of October 2021 published in the State Gazette № 84 on the 08th of October 2021) for particularly significant merits in the field of science.

The scientific applied development "New patented bipolar electrical scheme for excitation of gas discharge lasers with copper bromide and strontium vapour delivering laser radiation in the visible and middle infrared spectral ranges with record-high average laser power and beam quality" by a scientific team led by **Prof. Dr. K. A. Temelkov** and including **I. K. Kostadinov**, **K. A. Temelkov**, **D. N. Astadjov**, L. T. Popova, **S. I. Karatodorov**, **S. I. Slaveeva**, **G. P. Yankov** was awarded by the Scientific Council of the Georgi Nadjakov Institute of Solid State Physics, Bulgarian Academy of Sciences for the Best Scientific Applied Achievement for 2022.

PUBLICATIONS:

Kostadinov I., Temelkov K., Popova L., Ivanov B., Slaveeva S., "Compact 10-W Sr vapor laser oscillating in middle infrared spectral range on Sr atomic self-terminating transitions", *Optical and Quantum Electronics*, 54, 718, 2022, JCR-IF: 2.794 Q2

Iordanova E., Yankov G., Karatodorov S., Kovachev L., Diffraction-free femtosecond optics", *Optik*, 267, 16968, 2022, JCR-IF: 2.840 Q2

Zlatanov K. and Vitanov N., "Chiral resolution based on laser-induced continuum structure", *Optics Communications*, 520, 128514, 2022, JCR-IF: 2.335 Q2

Zlatanov K., Rangelov A., Vitanov N. "Extension of the Morris-Shore transformation to arbitrary time-dependent driving fields", *Journal of Physics B: Atomic, Molecular and Optical Physics*, 55, 204001, 2022 JCR-IF: 1.655 Q2

Iordanova E., Yankov G., Stankova N., Nedyalkov N., Modification and activation of the surface of medical-grade PDMS after irradiation by ultrashort laser pulses", *Journal of Physics: Conference Series*, 2240(1), 012051, 2022 SJR: 0.210

Karatodorov S., Shehadi M., Stoychev L., Yankov G., Tsankov D., Shivachev B., Petrov T. Nonlinear refractive index and multiphoton absorption measurements of wide bandgap semiconductors materials by femtosecond z-scan method, Optica Advanced Photonics Congress, JW3A. 40, 2022

PATENTS:

Maintained patents:

1. Gas-discharge laser, № 67473 from 15.11.2022	**I. K. Kostadinov**, **D. N. Astadjov**, **K. A. Temelkov**, **G. P. Yankov**
2. Method for determination of laser ablation threshold of solid state materials, No. 67278 from 11.03.2021	O. Ivanov, V. Mihailov, **S. Karatodorov**, José Luis Pérez-Díaz

Patents in procedure:

1. Method and system for trapping, cooling and compression of neutral atoms, molecules and particles with laser pulses, per. № 113628 от 14.12.2022	L. M. Kovachev, **E. I. Iordanova**, **G. P. Yankov**

ONGOING RESEARCH PROGECTS:

- Research and development of an innovative solution for the assessment of microplastic contamination in the aquatic environment funded by Bulgarian National Innovative Fund (BNIF) of the Executive Agency for the Promotion of Small and Medium Enterprises (EAPSME) of the under grant № 13if-02-6 07.12.2022.
- Spectral analysis of the pigments used for decoration of ceramics from the Neolithic and Chalcolithic era via Laser Induced Breakdown Plasma Spectroscopy (LIPS) and Fourier Transform Infrared Spectroscopy (FTIR) funded by BNSF under grant KP-06-PM68/1 30.11.2022.
- Optical cooling and acceleration of neutral particles with femtosecond laser pulses funded by COST under grant KP-06.
- ELI ERIC BG for research infrastructure funded by contract D01-401 17.12.2021
- Dynamics and formation of plasma induced by infrared femtosecond laser pulses in transparent medium funded by BNSF under grant KP-06-PH58/11 2021.
- Basic research and development of high-beam-quality high-power laser system oscillating in visible spectral range funded by BNSF under grant KP-06-H37/2 06.12.2019.
- Functionalization of 3D printed fibrous matrixes by femtosecond laser modelling funded by BNSF under grant KP-06-PH-38/4 06.12.2019.
- Basic research and development of high-beam-quality high-power laser system oscillating in middle infrared spectral range funded by BNSF under grant KP-06-H27/5 08.12.2018.
- Experimental and theoretical investigation on ultrafast dynamics of processes, induced by subpicosecond laser nanomachining of wide gap semiconductors funded by BNSF under grant DN 18/07 2017.
- Laboratory “Laser Technologies” at Centre of Excellence “Mechatronics and Clean Technologies”.
- Lasers, laser technologies and applications funded by the budget subsidy of BAS.
- Femtosecond laser applications in material microprocessing under the Academy’s bilateral agreements with IFFM, Gdansk, PAS, Poland.
- Material processing and analysis by ultrashort laser pulses under the Academy’s bilateral agreements with National Institute for Lasers, Plasma and Radiation Physics, RAS, Romania.

INTERNATIONAL COLABORATION:

The **METAL VAPOUR LASERS,** has international collaborations with:

- Institute of Fluid-flow Machinery - Polish Academy of Sciences, Gdansk, Poland;
- Institute for Laser Science – UEC, Chofu-shi, Tokyo, Japan;
- TU/e, Eindhoven, The Netherlands
- Tomsk State and Tomsk Polytechnic Universities, Tomsk, Russian Federation
- National Institute for Lasers, Plasma and Radiation Physics, RAS, Romania

TEACHING ACTIVITIES:

Assoc. Prof. Todor Petrov is a full-time lecturer in the Technical University – Sofia. Prof. Dr. K. Temelkov organized the traditional 25th Winter Seminar of PhD Students and Young Scientists in Physics (Webinar), 06–07 December, 2022. Prof. Dr. K. Temelkov and Assist. I. Kostadinov gave an invited lecture "HIGH-POWER DIFFRACTION-LIMITED SEALED-OFF LASER SYSTEMS OSCILLATING IN MIDDLE INFRARED AND VISIBLE SPECTRAL RANGES ON STRONTIUM AND COPPER ATOMIC SELF- 49 TERMINATING TRANSITIONS" at the 15th Spring Seminar of PhD Students and Young Scientists in Chemistry (Webinar), 22–23 June, 2022.

Assoc. Prof. E. Iordanova and Assist. Prof. G. Yankov gave an invited lecture "The future is vast: Regenerative medicine perspectives and technology trends in advanced implantable biomaterial strategies" at the 22nd International School on Condensed Matter Physics "State of the art in Functional Materials & Technologies", 29 August – 2 September, Varna, Bulgaria, 2022.

Assoc. Prof. E. Iordanova gave an invited lecture "Regenerative medicine – development in technologies for implantable biomaterials" at the 25th Winter Seminar of PhD Students and Young Scientists in Physics (Webinar), 06–07 December, 2022.