Frontiers in Applied Optics: Materials, Methods and Sensors

Complete Abstracts of the Workshop

Session 2

Dependence of CuMnAs quench-switching on excitation laser pulse parameters

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Antiferromagnetic (AF) materials are the focus of a new area of spintronic research [1,2]. Currently, AFs are less utilized in commercial spintronic devices than their ferromagnetic (FM) counterparts but promise some major advantages for applications in future devices. The absence of net magnetization and stray fields of AFs allows for higher integration density and AFs also exhibit orders of magnitude faster dynamics compared to FMs. Some functionalities of AFs can be directly derived from FMs, like readout using anisotropic magnetoresistance or magnetic axis reorientation using current-induced spin-orbit torque [3]. Specific effects are unique to AFs, like quench-switching first observed in CuMnAs [4]. Quench-switching is a heat-based effect that allows using both electrical and optical excitation stimuli. In its core, quench-switching is based on changes in the magnetic domain structure at temperatures close to the material's Neél temperature, followed by rapid cooling. This shattering of the large domains into nano-scale domains is detectable in the resistivity change, reaching tens of percent at room temperature. The quench-switching signals exhibit a temperature-dependent relaxation with characteristic times in the range of seconds at room temperature [4].
The quench-switching of CuMnAs was first reported using electrical and ultrashort optical stimuli [4]. This contribution explores further laser pulse parameters and their effect on switching performance. It was also previously demonstrated that the quench-switching behaves universally across various differently grown CuMnAs films. Due to this universal behavior, this analysis investigates dependencies on the laser pulse parameters only for 20 and 50 nm thick stoichiometric CuMnAs films. With increasing fluence of the laser pulses, we encountered two distinct thresholds – quench-switching and damage thresholds. While the devices quench-switch even above the damage threshold, the central area of the Gaussian laser spot exhibits permanent changes in the film that are detectable also in its resistivity. This analysis focuses on fluences between the two thresholds and explores dependencies above the damage threshold.

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Anomalous Magneto-Optical Kerr Effect in Unconventional Antiferromagnetic MnTe

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In recent years, antiferromagnets (AFMs) received a lot of attention due to their application potential in the field of spintronics, such as materials for memory media. It is a requirement for such media, that one can manipulate the magnetic moments. In the case of AFMs, the magnetic moments can be manipulated in several ways, for example by spin-orbit torques, strain control, field-cooling or by strong magnetic field. Recently, a new group of unconventional AFMs was reported [1], exhibiting alternating spin directions in both, real and momentum space. This leads to the spin splitting of the bands, allowing time reversal symmetry breaking phenomena, such as anomalous Hall effect (AHE),
or magneto-optical Kerr effect (MOKE).

[1] Šmejkal, L. et al., Phys. Rev. X 12, (2022).

Optically-induced magnetic dynamics in altermagnetic candidate MnTe

J. Střihavková, J. Kimák, Z. Sadeghi, D. Kriegner, H. Reichlová, G. Springholz, E. Schmoranzerová,

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Spintronics is a part of electronics that utilizes spin of electrons for information storage and processing, which can improve speed and memory capacity of electronic devices. [1] Very promising materials for spintronic applications are altermagnets, recently described materials with antiparallel ordering of magnetic moments and a spin-split electronic band structure, enabling observation of phenomena otherwise typical for ferromagnets, such as giant magnetoresistance (GMR) and spin-transfer torque (STT). [2] Despite the promising potential, altermagnetic properties have not yet been studied using all-optical methods. We have investigated magnetic dynamics of thin film antiferromagnetic MnTe, an altermagnetic candidate, using time-resolved magnetooptics.

We have observed laser-induced dynamics of magnetic origin on two regions on the sample, displaying the same magnetoopticaleffect, but with different symmetry. This can indicate different orientations of the Néel vector in these regions.

[1] Němec, P. et al., NaturePhys. 14, 229-241 (2018)
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Optical and magneto-optical spectroscopies of half- and full-Heusler Rh-Mn-Sb thin films

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The Heusler compounds have gained significant amount of attention in the past years owing to high application potential. Because of the highly tunable electronic structure and extraordinary magneto-optical (MO), magnetoelectric, and magnetocaloric properties, Heusler compounds have found use in a vast number of research fields, including superconductivity [1], magnetic shape memory [2] and, most recently, topological insulators. They have also proven to be more than suitable for utilization in spintronics because of the high spin polarization some of them exhibit [3]. Compounds containing heavier atoms can benefit from large spin-orbit coupling and related phenomena, such as magneto-optical effects or anomalous transport.
Here we present a systematic study of the field dependent magneto-optical spectroscopy together with spectroscopic ellipsometry on full-Heusler Rh_2MnSb and half-Heusler RhMnSb thin films prepared by magnetron sputtering. The magneto-optical spectra were taken in polar Kerr geometry in the spectral range from 1.2 to 5 eV at nearly normal incidence and in external magnetic fields up to 1T. The spectroscopic ellipsometry was taken by Mueller matrix ellipsometer Woollam RC 2 in the spectral range from 0.7 to 6 eV.
The samples exhibited large magneto-optical response with respect to their thickness and its field dependence suggested an in-plane magnetic anisotropy with negligible remanent signal for out-of-plane external magnetic field. Comparing the spectral dependencies between half- and full-Heusler samples, the main differences were observed between 2 and 4 eV. In this energy region the manganese d states are responsible for optical transitions and therefore the differences in magneto-optical response most likely origin from modification of manganese d orbitals when changing the Heusler structure. This was confirmed by the deduction of spectral dependence full permittivity tensor of these materials.

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[2] I. Aaltio, A. Sozinov, Giant Magnetorestrictive Materials, Elsevier, 1st edition, 2016.
[3] M. Jourdan, Jet al., Nat. Comm. 5, 3974 (2014)

Large magneto-optical response of $Eu_{0,5}Bi_{2,5}Fe_5O_{12}$ thin garnet films prepared by metal-organic decomposition

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Ferrimagnetic iron garnets ($R_3Fe_5O_{12}$, where R is a rare earth element) are promising materials for various applications, such as magneto-optical isolators, spintronic and spin wave devices. They crystallize in a cubic lattice which consists of three sites defined by the oxygen positions: iron ions are situated in tetrahedral and octahedral sites while the rare-earth sits at the dodecahedral site. Their magnetic and optical properties can be tailored by a variety of elemental substitutions.
Here we report on large magneto-optical response of $Eu_{0,5}Bi_{2,5}Fe_5O_{12}$ thin films in terms of Faraday rotation. The samples were prepared by metal-organic decomposition using the solutions made of Eu, Bi, and Fe carboxylates [1]. The final metal-organic liquids were prepared by mixing each solution to obtain desired chemical compositions. The solutions were spin-coated on GGG (100) and (111) substrates and subsequently dried using a hot-plate. To decompose organic materials and to obtain amorphous oxide films, the samples were pre-annealed at 450 °C. This procedure was repeated, from spin coating to pre-annealing, several times to obtain appropriate thickness. Finally, the samples were annealed for crystallization in a furnace at 700 °C for several hours.
The magneto-optical characterization was done using a Faraday magneto-optical spectrometer based on rotating analyzer technique [2] in the spectral range from 1.2 to 5 eV and external magnetic fields up to 0.7 T. All experiments have been done at room temperature.
The samples exhibited large amplitude of magneto-optical Faraday rotation with respect to their thickness of approx. 50 nm, exceeding 7.5 degree at 2.9 eV. The spectral dependence of Faraday rotation resembles the one reported for Bi doped $Y_3Fe_5O_{12}$ [3] with a broad absorption band between 2.5 and 3.5 eV and with enhanced tail of Faraday rotation towards the lower energy side. The energy slices of the field dependent spectra revealed almost square shape hysteresis loops, suggesting strong out-of-plane magnetic anisotropy. This might be due to the Eu doping or via the strain induced by the presence of the substrate.

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Session 3

Ultrafast spectroscopy of single light-harvesting complexes

May Myat Noe Shein, Pavel Malý

Ultrafast optical spectroscopy has greatly enhanced our understanding of energy transfer in complex organic systems. However, traditional techniques average over vast molecular ensembles, often masking the intricacies of dynamic molecular behaviors. This averaging effect limits our ability to uncover the fundamental mechanisms underlying processes in systems with inherent disorder and heterogeneity.
To address this challenge, we combine ultrafast spectroscopy with single-molecule fluorescence detection, establishing a multi-excitation ultrafast optical spectroscopy platform. This novel technique is capable of tracking excitation dynamics within individual molecules as they interact within complex systems such as photosynthetic light-harvesting complexes. We will construct a dedicated time-resolved fluorescence microscope, which will allow us to examine the transient spectra of individual complexes, quantify the relative signal magnitude, and determine which spectral features can be distinguished within specific measurement times. In this contribution, we will describe the setup, present measurements that characterize its performance, and show the
first results on molecular systems. Additionally, our setup will enable us to observe how the spectroscopy reflects changes when complexes switch between states, spectral characteristics, and kinetic behaviors throughout the measurement process.

The journey towards weighing biomolecules with light at atomistic resolution

Evangelos Ofreas Efraimidis, Barbora Špačková, Tereza Roesel, Niklas Hansen, Vladimíra Petráková

The ability to measure the mass of single biomolecules in their native state is critical for advancing our understanding of molecular functions and interactions. Conventional mass spectrometry, while powerful, requires ionization and fragmentation of biological samples and typically operates in the gas phase. These requirements limit its ability to analyze biomolecules in their native, solution-state environment, hindering the study of delicate or dynamic systems and prohibiting real-time exploration of functional phenomena.
Nanofluidic Scattering Microscopy (NSM) addresses these challenges by enabling real-time imaging of biomolecules diffusing freely in solution. Analysis of the Brownian motion of diffusing molecules allows for the determination of their hydrodynamic radius. Furthermore, by leveraging the relationship between integrated optical contrast and molecular polarizability, NSM provides accurate molecular weight (MW) determination across a range from tens of kDa to several MDa. This approach enables optical mass spectrometry, allowing MW characterization of individual

molecules directly from solution in their native state. Additionally, NSM supports simultaneous MW measurement of different molecules, facilitating the profiling of heterogeneous samples.
The resolution of MW measurements in NSM improve with longer observation times, as the uncertainty is inversely proportional to the square root of the molecule's observed trajectory length. Extended observation times can be achieved through single-molecule manipulation techniques, such as controlled flowrates and electrostatic trapping. Advances in rigorous data processing also hold great promise for enhancing mass resolution.
Using DNA origami as a model system, this work explores the potential of achieving atomic-level MW resolution by integrating single-molecule manipulation with advanced data processing. These developments could greatly enhance the study of protein aggregation, phase transitions like liquid-liquid phase separation, providing deeper insights into biomolecular behavior.

High-sensitivity optical tomography of gas jet for laser plasma accelerator

Vojtěch Janota, U. Chaulagain, M. Lamač, M. Raclavsky, J. Nejdl

Optical probing is essential for exploring nature yet analyzing transparent materials with a refractive index near unity, like low-density gas jets, challenges its sensitivity limits. We present an enhanced optical probing technique using multiple passes through the sample [1-3], boosting phase sensitivity. This method, tested on supersonic gas jets, increased sensitivity and enabled detailed visualization of complex structures like shocks. Using wavefront sensor we capture intensity and phase projection of propagated wave for tomography reconstruction of 3D gas density distribution, important for applications like laser wakefield acceleration and compact X-ray sources[4-5].

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Bayesian optimization for ultrafast X-ray generation from dual-stage laser-plasma accelerators

Dominik Čáp, Orsolya Morvai, Uddhab Chaulagain, Jaroslav Nejdl, Marcel Lamač

Laser wakefield acceleration (LWFA) [1] has proven highly effective in producing high-energy electron beams [2, 3], which then generate bright femtosecond X-ray pulses through betatron oscillations [4, 5]. This method offers a compact and nearly spatially coherent X-ray source suitable [6] for many applications such as phase contrast imaging and X-ray time-resolved spectroscopy [7] but is limited by its low conversion efficiency and relatively low photon flux. One of the promising approaches to increase the intensity of X-ray radiation is so-called passive plasma lensing [8]. In

this dual-stage scheme, electrons are injected and accelerated in the first gas jet – the accelerator – and the transverse momentum of the oscillations of the electron beam are then amplified in the second gas jet with higher density – the radiator. It has been shown that in the radiator, the electron beam is focused, and the magnitude of the betatron radiation is increased [9]. Using Bayesian optimisation and multidimensional particle-in-cell simulations, we demonstrate how the process of passive plasma lensing can be optimised to maximise X-ray production and even reduce the number of computationally intensive simulations needed for a thorough parametric scan. Parameters are chosen to correspond to the capabilities of the Gammatron beamline at ELI Beamlines for possible future experiments.

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Resonant betatron X-ray generation using chirped laser pulses

Orsolya Morvai, Dominik Čáp, Jaroslav Nejdl, Uddhab Chaulagain, S. V. Bulanov, Marcel Lamač

Laser wakefield acceleration (LWFA) [1] is a state-of-the-art technique that uses intense laser pulses in a plasma medium for compact electron acceleration and X-ray generation. The X-rays generated by this method hold significant promise for applications requiring femtosecond-scale X-ray pulses. However, the lack of sufficient photon flux leaves room for exploration of new approaches to enhance their efficiency. Recent studies have introduced various approaches to optimize electron properties, thereby improving the quality of X-ray sources. Some of these methods include utilizing a capillary discharged waveguide [2], adjusting plasma density [3], modifying laser properties by adding chirp to the driver pulse [4-5], or employing resonant effects through the use of additional pulses behind the driver [6]. In this study, we investigate the effect of

chirped laser pulses on betatron X-ray generation, focusing on how varying the chirp parameter affects the electrons' transverse momentum and the energy of emitted X-rays Introducing a chirp to the laser pulse can alter the wakefield's evolution without changing the pulse energy, potentially increasing the charge and energy of the accelerated electrons. Additionally, adjusting the chirp parameter influences the laser pulse dynamics through group velocity dispersion, which may strengthen the resonant interaction between the electrons and the probe pulse, thereby enhancing the electrons' transverse momentum. This effect can significantly impact both the flux and critical energy of the emitted X-ray radiation. These insights can contribute to future betatron X-ray generation and LWFA experiments, aiming to achieve higher control over the acceleration mechanism and radiation generation.

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Session 4

Plasmon-enhanced fluorescence imaging biosensor for single molecule detection

Katharina Schmidt, Naoto Asai, Gizem Aktug, Jakub Dostalek

We report on an ultrasensitive biosensor advanced for protein biomarker detection in real-world samples. [1] The Rolling circle amplification (RCA) serves as an enzymatic based enhancement technique. [2] It is based on a circular probe binding to a primer sequence for prolongation, leading to long single-stranded DNA. It serves as a scaffold to either subsequently bind fluorophore-tagged labelling strands or provide the possibility to incorporate fluorophores during the amplification process. The real-time labelling is monitored by a combined surface plasmon resonance and plasmon-enhanced fluorescence (PEF) set-up. Additionally, a home-built PEF imaging device was developed for single-molecule resolution.

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Facile fabrication of large-area biofunctionalized metallic nanostructures for high-performance affinity plasmonic biosensors

Dario Cattozzo Mor, Adam Benjamin Plšek, Prasanth Asokan, Van Truc Vu, Gizem Aktug, Ladislav Fekete, Zdeněk Hubička, Chun Jen Huang, Jakub Dostalek

Surface plasmons are associated with optical resonances originating from collective oscillations of charge density at metal/dielectric interfaces. These resonances produce a tightly confined electromagnetic field and are widely used in bioanalytical methods, including surface plasmon resonance (SPR) biosensors, surface enhanced Raman spectroscopy (SERS) or plasmon-enhanced fluorescence (PEF).
The enhancement of the electromagnetic field intensity and the quality factor of plasmonic resonances depend significantly on the materials used. For gold nanostructures on glass, an adhesion layer, typically chromium or titanium, is required, but can introduce dampening to the surface plasmon field.
We present a preparation approach for periodic arrays of gold nanoparticles with controlled plasmonic properties in combination with facile adhesion-promoting coating based on non-damping molecular linkers.
The nanofabrication is based on UV-laser interference lithography combined with monitoring of resist mask preparation step and novel Argon milling dry etching method to improve sample-to-sample reproducibility. Furthermore, we exploit a "chemical toolbox" composed of thiol and silatrane molecules to function as adhesion-promoting layers, antifouling coatings, and biofunctional moieties for the gold nanoparticle arrays. Silatranes, with their silane-based tricyclic caged structures, offer high homogeneity and allow for organized coating of glass surfaces. Each component is engineered to selectively bind to specific materials, ensuring an orthogonal and targeted coating process.

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FAST optical fluorescence telescopes: analysis of miniarray sensitivity

Zuzana Svozilíková

The Fluorescence-detector Array of Single-pixel Telescopes (FAST) is a proposed next-generation cosmic-ray observatory based on an array of simplified fluorescence telescopes each equipped with four photo-multiplier tubes with a diameter of 200 mm positioned at the focal plane of a segmented mirror with a diameter of 1.6 m. The future array is aimed at observing ultra-high cosmic rays with energies above 10^19 eV with unprecedented statistics. In order to make the future FAST array feasible on a grid of tens of thousands of square kilometers, it is necessary to ensure that the individual telescopes are easily and reasonably replicable, fully autonomous and require only low maintenance. To this end, the FAST collaboration is testing the first prototypes at two observatories, the Telescope Array (USA) and the Pierre Auger Observatory (Argentina). It will be at the Pierre Auger Observatory that stereo detection is used for the first time, where two telescopes already placed in one place will be supplemented by two other telescopes at a distance of about 10 km and after some period also by two more telescopes at third position to create a simple triangular array. The contribution will present the current status of the project, preliminary results from the data analysis, as well a recent development towards a mini-array of FAST telescopes. Expected sensitivity of this mini-array to cosmic ray showers will be discussed.