O 7: Plasmonics and Nanooptics I

Time: Monday 10:30–13:00 Location: MA 043

O 7.1 Mon 10:30 MA 043

Plasmonic Twistronics: Discovery of Plasmonic Skyrmion Bags — •Julian Schwab[1], Alexander Neuhaus[2], Pascal Dreher[2], Shai Tsesses[3], Anant Mantha[1], Florian Mangold[1], Bettina Frank[1], Guy Bartal[3], Frank-J. Meyer zu Heringdorf[2], Timothy J. Davis[1,2,4], and Harald Giessen[1] — [1]4th Physics Institute, Research Center SCoPE, and Integrated Quantum Science and Technology Center, University of Stuttgart, Germany — [2]Faculty of Physics and Center for Nanointegration, University of Duisburg-Essen, Germany — [3]Andrew and Erna Viterbi Department of Electrical Engineering, Technion-Israel Institute of Technology, Israel — [4]School of Physics, University of Melbourne, Australia

We explore the application of twistronics in plasmonic systems by superimposing two plasmonic skyrmion lattices to create a moiré skyrmion superlattice. We combine this concept with plasmonic topological excitations and demonstrate that the topology of moiré skyrmion lattices contains skyrmion bags as complex topological quasiparticles that so far have been demonstrated only in liquid crystals and chiral ferromagnets. The size of plasmonic skyrmion bags can be controlled by the twist angle and its center of rotation. The resulting electric field distribution of a skyrmion bag is derived numerically and verified experimentally using time-resolved two-photon photoemission electron vector microscopy (PEEM).

O 7.2 Mon 10:45 MA 043

Photoelectron imaging of topological edge states in one-dimensional plasmonic Su–Schrieffer–Heeger chains — •Luisa Brenneis[1], Benedikt Schurr[2,3], Matthias Hensen[1], Philipp Kessler[1], Bert Hecht[2,3], and Tobias Brixner[1] — [1]Institut für Physikalische und Theoretische Chemie — [2]NanoOptics Biophotonics Group, Experimental Physics 5 — [3]Würzburg-Dresden Cluster of Excellence ct.qmat, [1-3]: Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

Plasmonic nanostructures exhibiting different topological phases are proposed to offer unique properties like energy flow along the phase separating interfaces [1] or localized edge states [2]. Here, we use photoemission electron microscopy (PEEM) to directly image the mode pattern of one-dimensional nanochains with trivial and nontrivial topology, representing a plasmonic analogue of a Su–Schrieffer–Heeger chain [3]. To ensure sufficient electron delivery for the photoemission process we exploit Babinet's principle by investigating nanoslit chains written in monocrystalline gold flakes. The precision of the helium ion beam milling used for this purpose enables us to produce the individual chain elements with a distance down to 8 nm. Therefore, we can precisely control the dipole–dipole coupling between the individual chain elements. By comparing the experimental results with finite-difference time-domain simulations we identify the presence of edge states.

[1] M. Proctor, Appl. Phys. Lett. 118, 091105 (2021).
[2] S. R. Pocock, ACS Photonics 5, 22712279 (2018).
[3] W. P. Su, Phys. Rev. B 22, 2099 (1980).

O 7.3 Mon 11:00 MA 043

Attosecond electron microscopy by free-electron homodyne detection — John H. Gaida[1,2], Hugo Lourenço-Martins[1,2], •Murat Sivis[1,2], Thomas Rittmann[1,2], Armin Feist[1,2], F. Javier García de Abajo[3,4], and Claus Ropers[1,2] — [1]MPI for Multidisciplinary Sciences, Göttingen, Germany — [2]4th Physical Institute, University of Göttingen, Germany — [3]ICFO-Institut de Ciencies Fotoniques, Castelldefels (Barcelona), Spain — [4]ICREA-Institució Catalana de Recerca i Estudis Avançats, Barcelona, Spain

Photon-induced near-field electron microscopy (PINEM) enables the imaging of near-field intensities [1,2]. However, access to the evolution of nanoscale fields and structures within the light cycle requires a sensitivity to the optical phase [3,4].

Here, we introduce Free-Electron Homodyne Detection (FREHD) for phase-resolved electron microscopy of optical responses [5], which we demonstrate for the plasmonic fields of a gold nanoprism with few-nanometer spatial and sub-cycle temporal resolutions.

Our approach generalizes the high-resolution measurement of attosecond materials responses in electron microscopy, without a need for electron density bunching, and offers fascinating new possibilities to image local attosecond and phase-resolved responses on the nanometer scale.

[1] B. Barwick, et al. Nature 462, 902 (2009). [2] L. Piazza et al. Nat Commun 6, 6407 (2015). [3] D. Nabben, et al. Nature 619, 63 (2023). [4] T. Bucher et al., arXiv:2305.04877 (2023). [5] J. H. Gaida, et al. arXiv:2305.03005 (2023).

O 7.4 Mon 11:15 MA 043

Ultrafast near-field scanning optical oscilloscopy — •Juanmei Duan, Tom Jehle, Sam Nochowitz, and Christoph Lienau — Universität Oldenburg, D-26129, Germany

Metallic, dielectric and hybrid nanoparticles offer exciting opportunities to localize, manipulate and switch light on the nanoscale. A direct measurement of the local electric field at the surface of the nanostructures is challenging however, since these fields are often localized on exceedingly short length and time scales. While experiments such as attosecond photoelectron emission microscopy or phase-resolved photon-induced near-field electron microscopy have been proposed, direct time-resolved measurements are still lacking. Here, we describe and demonstrate a new experimental technique, ultrafast near-field oscilloscopy, to probe coherent optical near-fields in the time with nanometer spatial resolution. For this, amplitude and phase of the local near-field scattered by a sharp metal taper are recorded in a broad spectral range and on a time scale that is faster than the tip modulation period. This allows us to record spectra as a function of tip-sample distance, the key to probe tip-sample coupling experimentally. Direct Fourier transform of the scattering spectra gives the local near-field dyanamics with sub-cycle temporal and nanometer spatial resolution. We demonstrate the versatility of this new approach by probing near-fields of dielectric and semiconducting nanoparticles, as well as different localized and propagating plasmon mode of metal nanostructures.

O 7.5 Mon 11:30 MA 043

Determination of the transient absorbance during ultrafast laser-matter interaction by applying ultrafast imaging ellipsometry — •Markus Olbrich, Pflug Theo, Lungwitz Philipp, and Alexander Horn — Laserinstitut Hochschule Mittweida, Hochschule Mittweida, Technikumplatz 17, 09648 Mittweida

Applying ultrafast imaging metrology combined with modeling the ablation represents a promising way to comprehend the interaction of ultrafast laser radiation with matter fundamentally. Accurately simulating the absorption of the laser radiation is challenging due to the complexity of the needed models and the temperature- and density-dependencies of all included parameters. To solve this, determining the absorbed energy by ultrafast imaging ellipsometry neglects the necessity of using an optical model.

In this study, a 150 nm thick gold film, including an adhesive layer of 25 nm chromium on a glass substrate is irradiated by single-pulsed ultrafast laser radiation (pump radiation: pulse duration $\tau_H = 40$ fs, wavelength $\lambda = 800$ nm, peak fluence $H_0 = 5.0\,\mathrm{J/cm^2}$). Ultrafast imaging ellipsometry reveals the complex refractive index at the probe wavelengths 700 nm and 900 nm within 10 ps post-pump irradiation. The complex refractive index for the pump radiation at $\lambda = 800$ nm is calculated by linear interpolation afterward. Subsequently, the absorbed energy of the pump radiation is derived and included in the model. The simulations incorporating the experimentally determined absorbed energy yield a much better fit to the experimental results than uncorrected simulations.

O 7.6 Mon 11:45 MA 043

Photon-Induced Near-Field Interaction in Ultrafast Point-Projection Electron Microscopy — •Germann Hergert[1], Andreas Wöste[1], Martin Silies[1], Dong Wang[2], Petra Gross[1], and Christoph Lienau[1] — [1]Institut für Physik, Carl-von-Ossietzky Universität, 26129 Oldenburg, Germany — [2]Institut für Werkstofftechnik, TU Ilmenau, 98693, Germany

Photon-induced electron microscopy (PINEM) utilizes the coherent interaction of swift electrons (10-100keV) with optical near-fields for high-resolution imaging of nano-sized systems. Transferring PINEM to significantly lower electron energies (<100eV) increases the interaction time between electrons and near-fields and may enhance coupling to nano-confined optical modes. However, this interaction has not been

demonstrated due to high momentum mismatch at such low energies.

This study presents the first PINEM-like interactions in ultrafast point-projection electron microscopy (UPEM) with 100eV electrons [1]. Plasmonic nanofocusing generates 30fs electron pulses from a sharp gold taper that are accelerated by a -100V bias voltage towards an optically pumped Yagi-Uda antenna milled inside a 13nm thick free-standing gold film. The 3D momentum of the transmitted electrons is detected with a time-of-flight delay-line detector. A spread in the electron's momentum distribution is observed, originating from their coupling to both - longitudinal and transverse - electric field components of the antenna. This paves the way for spatial and temporal characterization of vectorial near fields in UPEM.

[1] Wöste, A. et al., Nano Lett. 23, 5528-5534 (2023)

O 7.7 Mon 12:00 MA 043

Spectroscopic and Interferometric Sum-Frequency Imaging of Strongly Coupled Phonon Polaritons in SiC Metasurfaces — •Niclas S. Mueller[1], Richarda Niemann[1], Sören Wasserroth[1], Guanyu Lu[2], Martin Wolf[1], Joshua D. Caldwell[2], and Alexander Paarmann[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [2]Department of Mechanical Engineering, Vanderbilt University, Nashville, USA

Phonon polaritons enable waveguiding and localization of infrared light with extreme confinement and low losses. Here, we introduce sum-frequency spectro-microscopy as a tool to image phonon polaritons in infrared metasurfaces with sub-wavelength spatial resolution and highly-resolved spectral resonance information.[1] This is implemented by resonantly exciting polaritons using a tunable infrared free-electron laser and wide-field microscopic detection of the upconverted light. We employ this technique to image hybridization and strong coupling of localized and propagating surface phonon polaritons in metasurfaces of SiC micropillars. Spectro-microscopy allows us to measure the polariton dispersion simultaneously in momentum space by angle-dependent resonance imaging, and in real space by polariton interferometry. Notably, we directly visualize how strong coupling affects the spatial localization of polaritons, inaccessible with conventional spectroscopic techniques. We further observe the formation of edge states at excitation frequencies where strong coupling prevents polariton propagation into the metasurface.

[1] R. Niemann, N.S. Mueller et al. arXiv 2311.13284 (2023)

O 7.8 Mon 12:15 MA 043

Highly polarization dependent formation of polaritons due to molecular alignment — •Roland Schäfer, Philipp Weitkamp, Dirk Hertel, Klaus Meerholz, and Klas Lindfors — Department für Chemie, Universität zu Köln, 50939 Köln, Germany

Spin cast films of HB238 [1] aggregate, forming both a J- and H-like transition that have a high polarization dependence, because the J-transition is located in the substrate plane, while the H-transition is located in the surface normal. In our previous work we have shown that both transitions can be simultaneously strongly coupled to planar microcavites, forming four polaritons. The two H-polaritons are highly polarization dependent.[2]

By aligning the aggregates along one direction, we achieve strong polarization dependence for both the J- and H-transition, enabling us to control the strong coupling behavior of both transitions by polarization.

[1] Bürckstümmer, et al., *Angew. Chem. Int. Ed.* **2011**, 50: 11628-11632.

[2] Schäfer, et al., *ACS Photonics*, in press.

O 7.9 Mon 12:30 MA 043

On-Chip Strong Coupling Device based on a slit nanoresonator — •Benedikt Schurr[1], Patrick Pertsch[1], Jin Qin[1], Saeid Asgarnezhadzorgabad[2], Alagesan Subramaniango[3], Monika Emmerling[1], Artur P. Podhorodecki[3], Ortwin Hess[2], and Bert Hecht[1] — [1]NanoOptics & Biophotonics Group, Experimental Physics 5, University of Würzburg, Am Hubland, 97074 Würzburg, Germany — [2]School of Physics and CRANN Institute, Trinity College Dublin, Dublin 2, Ireland — [3]Wroclaw University of Science and Technology, Department of Experimental Physics, wyb. Stanislawa Wyspianskiego 27, 50-370 Wroclaw, Poland

Strong coupling (SC) of light and matter states at ambient conditions is of interest in quantum technologies. In previous work, we demonstrated ambient condition SC between single quantum dots and a plasmonic slit resonator at a scanning probe tip [1,2]. To gain stability and scalability we have now developed an on-chip platform. We fabricate plasmonic slit resonators on a glass substrate located at the tip of a first electrode structure opposed by a second electrode. We then use dielectrophoresis to attract & position single quantum dots at the nanoresonator. This combination of techniques provides additional degrees of freedom as well as the opportunity to apply electric fields to tune the system.

[1] Groß, H. et al., Sci. Adv.4, eaar4906 (2018)

[2] Friedrich, D. et al., arXiv:2305.06909 (2023)

O 7.10 Mon 12:45 MA 043

Plasmon-mediated coherent population oscillations in molecular aggregates — •Daniel Timmer[1], Moritz Gittinger[1], Thomas Quenzel[1], Sven Stephan[1], Yu Zhang[2], Marvin Schumacher[3], Arne Lützen[3], Martin Silies[1], Sergei Tretiak[2], Jin-Hui Zhong[1], Antonietta De Sio[1], and Christoph Lienau[1] — [1]University of Oldenburg, Germany — [2]Los Alamos National Laboratory, USA — [3]University of Bonn, Germany

Strong coupling between plasmonic nanostructures and excitonic quantum emitters is a powerful way to alter material properties on the nanoscale. Dipolar coupling of excitons to vacuum field fluctuations of surface plasmon polaritons (SPPs) leads to a periodic energy transfer (Rabi oscillations) and the creation of hybridized polariton states. Often, experiments are limited to linear spectroscopy and time-domain studies of the energy transfer are lacking. We investigate the coherent dynamics of J-aggregate excitons that strongly couple to structured SPP fields of a periodic nanoslit array using two-dimensional electronic spectroscopy (2DES) [1]. Strong coupling predicts that "oscillating cross-peaks" should appear between diagonal polariton peaks. Our experiment indeed resolves these Rabi oscillations. Their analysis reveals that they reflect a coherent transfer between different, spatially separated excitons. The structured SPP field creates two classes of excitons that either couple strongly or weakly. Our study shows that 2DES allows to directly access the quantum dynamics of the strongly couples system, revealing a new and unexpected plasmon-mediated energy transport. [1]: D. Timmer, et al. arXiv:2307.14708 (2023).