O 78: Plasmonics and Nanooptics 3

Time: Friday 10:30–12:30 Location: H3

O 78.1 Fri 10:30 H3

Reconfiguring magnetic resonances with the plasmonic phase-change material In_3SbTe_2 — •Lukas Conrads, Andreas Hessler, Konstantin Wirth, Matthias Wuttig, and Thomas Taubner — I. Institute of Physics (IA), RWTH Aachen University

For miniaturized active nanophotonic components, resonance tuning of nanoantennas is a key ingredient. Phase-change materials (PCMs) have been established as prime candidates for non-volatile resonance tuning based on a change in refractive index [1]. Currently, a novel material class of switchable infrared plasmonic PCMs, like In_3SbTe_2 (IST), is emerging. Since IST can be locally optically switched between dielectric (amorphous phase) and metallic (crystalline phase) states in the whole infrared range, it becomes possible to directly change the geometry and size of nanoantennas to tune their infrared resonances [2]. Here, crystalline IST split-ring resonators (SRRs) are directly optically written and reconfigured in their arm size to continuously tune their magnetic dipole resonances over a range of 2.4 μm without changing their electric dipole resonances. The SRRs are further modified into crescents and J-antennas, which feature more complex resonance modes dependent on the polarization of the incident light [3]. Our concepts are well-suited for rapid prototyping, speeding up workflows for engineering ultrathin, tunable, plasmonic devices for infrared nanophotonics, telecommunications, or (bio)sensing.

[1] Wuttig et al., *Nat. Photon.* **11**, 465 (2017) [2] Heßler et al., *Nat. Commun.* **12**, 924 (2021) [3] Heßler, Conrads et al. *ACS Photonics* **9**, 5 (2022)

O 78.2 Fri 10:45 H3

Impact of atomistic structure and dynamics on inelastic light scattering in a plasmonic picocavity — •Franco Bonafé[1], Shuyi Liu[2], Heiko Appel[1], Martin Wolf[2], Takashi Kumagai[2,3], and Angel Rubio[1] — [1]MPI for Structure and Dynamics of Matter, Hamburg, Germany — [2]Dpmt. of Physical Chemistry, Fritz-Haber Institute, Berlin, Germany — [3]Center for Mesoscopic Sciences, Institute for Molecular Science, Okazaki, Japan

Atomically sharp metallic tips can focus an electromagnetic field down to the sub-nanometer scale, leading to strong light-matter interactions in plasmonic “picocavities”. This atomic-scale field can be used in plasmon-enhanced spectroscopy, such as tip-enhanced Raman spectroscopy (TERS), which has enabled the visualization of optical properties at sub-molecular resolution. However, a full microscopic understanding of the interplay of structural relaxation, bonding, near-fields and vibrations has not been achieved.

In this work, we combine experimental observations of inelastic light scattering from a single silver adatom in a plasmonic picocavity controlled by a low-temperature scanning tunneling microscope (STM), with ab initio real-time electron dynamics simulations. The experiment demonstrates a dramatic enhancement of Raman scattering that occurs upon the formation of a quantum point contact. We model possible geometries for the plasmonic tips and compute the vibrational modes, electronic current and near-fields localized in the vicinity of the single adatom. These simulations reveal a crucial role of the atomistic structural relaxation in the optical response in a plasmonic nanocavity.

O 78.3 Fri 11:00 H3

Vector Polarimetry - Measuring Electrical Fields on Surfaces — •Alexandra Rödl[1], David Janoschka[1], Pascal Dreher[1], Alexander Neuhaus[1], Bettina Frank[2], Timothy Davis[1,2,3], Michael Horn-von Hoegen[1], Harald Giessen[2], and Frank-J. Meyer zu Heringdorf[1] — [1]Faculty of Physics and Center for Nanointegration, Duisburg-Essen, University of Duisburg-Essen, 47048 Duisburg, Germany — [2]4th Physics Institute, Research Center SCoPE, and Integrated Quantum Science and Technology Center, University of Stuttgart, 70569 Stuttgart, Germany — [3]School of Physics, University of Melbourne, Parkville, Victoria 3010 Australia

Non-linear photoemission microscopy has been established as an excellent tool to investigate nano-optical fields at surfaces, in particular the fields of surface plasmon polaritons (SPPs). In a pump-probe experiment with femtosecond laser pulses we excite SPPs at grooves that are ion-milled into a Au platelet. The electric field of the probe-laser pulse interferes coherently with the electric field of the SPP and electrons are liberated through a nonlinear process by the combined field at the surface. The contrast depends on the alignment of the probe polarization and the orientation of the in-plane component of the SPP’s electric field. Using a set of different polarizations for the probe laser pulse while keeping the same excitation conditions, one can reconstruct the in-plane component of the electric field of the SPP. The out-of-plane field component is calculated by Maxwell’s equations to reconstruct the full electric vector field. Here, we measure and image complex electric vector fields of SPPs and analyze their topology.

O 78.4 Fri 11:15 H3

Spatially-resolved THz near-field spectroscopy — •Moritz B. Heindl[1], Nicholas Kirkwood[2], Tobias Lauster[3], Julia A. Lang[1], Markus Retsch[3], Paul Mulvaney[2], and Georg Herink[1] — [1]Experimental Physics VIII, University of Bayreuth, Germany — [2]ARC Centre of Excellence in Exciton Science, School of Chemistry, University of Melbourne, Australia — [3]Physical Chemistry I, University of Bayreuth, Germany

Spectroscopic access to ultrafast electric waveforms is critical to the understanding of plasmonic and field-driven nonlinear phenomena, yet, microscopic measurements still present a grand challenge. Here, we present a fluorescence-based field microscope for imaging ultrafast THz near-field evolutions employing the quantum-confined Stark-effect in semiconductor quantum dots [1,2]. This Quantum-Probe Field Microscopy (QFIM) scheme [3] allows for detection of strongly confined near-fields in three-dimensional structures. Using QFIM, we demonstrate spatially-resolved near-field spectroscopy of single THz resonators and propagating THz excitations inside wave-guiding structures.

[1] Hoffmann, M. C. et al. Appl. Phys. Lett. 97, 231108 (2010)
[2] Pein, B. C. et al. Nano Lett. 17, 5375-5380 (2017)
[3] Heindl, M. B. et al. Light Sci. Appl. 11, 5 (2022)

O 78.5 Fri 11:30 H3

Mode-selective imaging and control of nano-plasmonic near-fields — •Murat Sivis[1,2], Hugo Lourenço-Martins[1,2], Andre Geese[2], Tyler R. Harvey[2], Thomas Danz[1,2], Radwan M. Sarhan[3], Matias Bargheer[3], Armin Feist[1,2], and Claus Ropers[1,2] — [1]Max Plank Institute for Multidisciplinary Sciences, Göttingen, Germany — [2]4th Physical Institute - Solids and Nanostructures, University of Göttingen, Germany — [3]Institut für Physik und Astronomie, Universität Potsdam, Potsdam, Germany

Electron energy-loss spectroscopy (EELS) in a transmission electron microscope allows for the study of optical excitations in plasmonic nanostructures with sub-nanometer spatial resolution. While EELS is a powerful tool, it can only provide information about the spontaneous losses in a system with limited spectral resolution (10-100 meV in the most advanced microscopes). Recent developments in ultrafast transmission electron microscopy overcome these limitations and enable the probing of laser-excited modes by using photon-induced near-field electron microscopy (PINEM). Here, we demonstrate how PINEM can be used to measure the modal structure of the optical response of individual plasmonic systems (metal nano-triangle-resonators) at the nanoscale. Using our boundary element method (BEM)-based data analysis, we extract the magnitude and relative phase of each plasmonic mode from optical near-field maps. This approach opens a route to study the influence of the laser polarization, wavelength and incidence angle on the population of each mode, as well as the control of complex mode patterns created by multicolor fields.

O 78.6 Fri 11:45 H3

Tailoring of nonlinear metasurfaces using sampling-based optimization — •David Hähnel, Jens Förstner, and Viktor Myroshnychenko — Paderborn University, Theoretical Electrical Engineering, Warburger Str. 100, 33098 Paderborn, Germany

Various efficient methods for the design and optimization of linear metasurfaces have already been developed in the past [1]. Nowadays, attempts are being made to use these methods also for the design and optimization of nonlinear metasurfaces, which is a quite complex task due to the nonlinear processes involved. Here we present the design and optimization of all-dielectric nonlinear metasurfaces using a simple sampling method combined with Monte Carlo simulation, demonstrating that the use of sophisticated optimization methods is not necessarily required. Furthermore, this combination provides an effective approach in the optimization of nonlinear metasurfaces with a dynamic problem, such as a varying number of optimization parameters that are unknown in advance. We apply this method combination in the optimization of a nonlinear beam deflector metasurface for the third harmonic, which consist of an array of elliptical silicon disks and obtained an significant improvement in the radiated intensity compared to literature results [2]. [1] Chen, S. et al., Advanced Optical Materials 2018, 6, 1800104. [2] Lei Wang et al., Nano Lett. 2018, 18, 6, 3978-3984.

O 78.7 Fri 12:00 H3

Polarization selective investigation of plasmonic Bloch modes with dark-field spectroscopy — •Maximilian Johannes Black and Nahid Talebi — Institute for Experimental and Applied Physics, Kiel University, 24118 Kiel, Germany

Plasmonic and photonic crystals are widely used to mold the flow of light. Owing to its symmetry even the seemingly simple structure of a periodic square lattice of holes within a thin gold film shows a broad variety of plasmonic Bloch modes that are distinguished by the momenta and polarization. Therefore, the response of the system to the excitation is formed by a superposition of these modes. In this work we use polarization-selective optical dark-field microscopy to decompose these modes. Dark-field microspectroscopy is used to enhance the resolution and to suppress the background illumination, whereas linear polarizers both in the illumination and the detection path enable the selection of Bloch modes by polarization. We find that the dominating signal is formed by the selective excitation and detection of radiating magnetic moments. These and a variety of other modes are visualized by hyperspectral imaging. In the far-field reciprocal space interference fringes are resolved, indicating the propagation of the observed modes along the surface of the plasmonic crystal. Our results prove the possibility to thoroughly investigate plasmonic Bloch modes using dark-field microscopy, adding its versatility to the range of methods for measuring light-matter interactions.

O 78.8 Fri 12:15 H3

A Ginzburg Landau model for femtosecond charge-density wave dynamics at the atomic scale — •Kurt Lichtenberg[1], Mohamad Abdo[1,2], Shaoxiang Sheng[1], Luigi Malavolti[1,2], and Sebastian Loth[1,2] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Max Planck Institute for Solid State Research, Stuttgart, Germany

Charge-density waves (CDWs) feature collective excitations that can be observed as an oscillatory response of the electron system of a material to a fast optical stimulus [1]. At the same time, scanning tunneling microscopy (STM) measurements reveal highly localized interactions with atomic defects [2]. THz pump-probe spectroscopy in the STM reveals highly heterogeneous dynamics with spatial variations down one unit cell of the CDW. To improve the understanding of such local CDW dynamics, we developed an empirical model that is motivated by former approaches [3,4] and based on time-dependent Ginzburg Landau Theory. The combination of this theory with THz-STM measurements establishes a possibility to study the interplay between collective CDW dynamics with atomic pinning sites.

[1] M.-A. Méasson et. al, PRB 89, 060503(R) (2014)
[2] C. J. Arguello et. al, PRB 89, 235115 (2014)
[3] W. L. McMillan, PRB 12, 1187-1196 (4) (1975).
[4] G. Grüner, Density Waves in Solids. Perseus Publishing - Cambridge, Massachusetts, (2000).