O 24: Plasmonics and Nanooptics III: Light-Matter Interaction and Spectroscopy

Time: Tuesday 10:30–13:00 Location: MA 042

O 24.1 Tue 10:30 MA 042

Identification of weak molecular absorption on single-wavelength s-SNOM images — •Iris Niehues[1,2], Lars Mester[3], Edoardo Vincentini[2], Daniel Wigger[4], Martin Schnell[2,5], and Rainer Hillenbrand[2,5,6] — [1]Institute of Physics, University of Münster, Germany — [2]CIC nanoGUNE BRTA, Donostia-San Sebastián, Spain — [3]attocubes systems AG, Munich-Haar, Germany — [4]Department of Physics, University of Münster, Germany — [5]IKERBASQUE, Basque Foundation for Science, Spain — [6]Department of Electricity and Electronics, UpV/EHU, Donostia-San Sebastián, Spain

Scattering-type scanning near-field optical microscopy (s-SNOM) and nanoscale IR point spectroscopy (nano-FTIR) allow for nanoscale optical mapping of manifold material properties. Both techniques are based on elastic light scattering at an atomic force microscope tip that is illuminated with monochromatic or broadband laser illumination. For dielectric samples, the near-field amplitude and phase signals of the tip scattered field reveal the local reflectivity and absorption, respectively. Importantly, absorption in s-SNOM imaging corresponds to a positive phase contrast relative to a non-absorbing reference sample. Interestingly, a negative phase contrast (NPC) can be observed when imaging a non-absorbing material on a highly reflecting substrate. We explore the origin of the NPC using representative test samples and demonstrate straightforward simple correction methods that remove the NPC and that allow for the identification of weak absorption contrasts. [Opt. Express 31(4), 7012 – 7022 (2023)]

O 24.2 Tue 10:45 MA 042

Near - field optical microscopy of complex plasmonic excitations — •Farid Aghashirinov, Anant Mantha, Florian Mangold, Julian Schwab, Bettina Frank, and Harald Giessen — 4-th Physics Institute, University of Stuttgart, Stuttgart, Germany

We experimentally and theoretically study surface plasmon polaritons (SPPs) on single crystalline gold platelets of both long- and short-range type. Short-range surface plasmon polaritons (SR-SPPs) have received less attention compared to long-range SPPs (LR-SPPs) due to high attenuation losses, which shortens their propagation length. To investigate the complex SPP near - field pattern on single crystalline gold platelet, we utilize a reflection s - SNOM combined with a tunable broadband laser source. We disentangle excitations coming both from the scanning tip and the gold platelet edges by applying a Fourier analysis method. This allows us to determine the SPP wavelength and furthermore identify hidden excitations that are covered by interference of other waves. Fourier filtering makes it possible to identify the propagation direction of short-range surface plasmon polaritons, as well as their propagation length. In addition, we explore the impact of platelet thickness on the short - range SPP wavelength, which in future will give us another tuning parameter for scaling and combining complex near - field optical microscopy with topological plasmonics.

O 24.3 Tue 11:00 MA 042

Near-field Fano spectroscopy of MaPbI3 nanoparticles — Jinxin Zhan[1], •Tom Jehle[1], Sven Stephan[1], Sam Nochowitz[1], Ekaterina Tiguntseva[2], Sergey Makarov[2], Juanmei Duan[1], Petra Gross[1], and Christoph Lienau[1] — [1]Universität Oldenburg, D-26129, Germany — [2]St. Petersburg, Russia

Dielectric nanoparticles particles have optical shape resonances that confine light on the nanoscale in localized modes with well-defined spatial field profiles. A particularly interesting example are halide perovskite nanoparticles, for which the coupling between excitons and Mie modes results in Fano lineshapes in the spectral domain [1]. Here, we use a new broadband, interferometric sSNOM technique [2] to probe the time dynamics of the local optical near-fields of such particles. We measure amplitude and phase of the scattered light field in a broad spectral range and with 10 nm spatial resolution. Direct Fourier transformation gives the time dynamics of the local electric field, recorded with sub-cycle resolution. We uncover biexponential near-field decays with a characteristic destructive interference dip after a few fs. In the spectral domain, this corresponds to a Fano resonance with an unual 2π phase jump. We show that this signature arises from the intereference between spectrally broad dipole and narrow quadrupole resonances of the particles. Our results give new insight into the optical properties of high-index, active semiconductor nanoparticles with intriguing applicatoins for nanoscale all-opticals switching and lasing. [1] Tiguntseva, E. Y., et al. Nano Lett. 2018, 18 (2), 1185-1190. [2] Zhan, J., et al. Advanced Photonics 2020, 2 (04).

O 24.4 Tue 11:15 MA 042

Direct programming of confined Surface Phonon Polariton Resonators with the plasmonic phase-change material In_3SbTe_2 — Lukas Conrads, Luis Schüler, Konstantin Wirth, Matthias Wuttig, and •Thomas Taubner — I. Institute of Physics (IA), RWTH Aachen University

Tailoring light-matter interaction is essential to realize nanophotonic components and can be achieved with surface phonon polaritons (SPhPs), an excitation of photons coupled with phonons of polar crystals. Ultra-confined resonances are observed by restricting the SPhPs to cavities. Phase-change materials (PCMs) enable non-volatile programming of these cavities based on a change in the refractive index [1]. Recently, the new plasmonic PCM In_3SbTe_2 (IST) was introduced which can be reversibly switched from an amorphous dielectric to a crystalline metallic state in the infrared to realize numerous nanoantenna geometries [2]. However, the application potential of IST to create and modify SPhP resonators has not been exploited yet. Here, we demonstrate direct programming of confined SPhP resonators by phase-switching IST on top of a polar silicon carbide crystal and investigate the strongly confined resonance modes with scanning near-field optical microscopy. Reconfiguring the size of the resonators themselves result in enhanced mode confinements up to a value of $\lambda/35$ [3]. This study is a first step towards rapid prototyping of reconfigurable SPhP resonators even with hyperbolic and anisotropic 2d materials.
[1] Wuttig et al., *Nat. Photon.* **11**, 465 (2017) [2] Heßler et al., *Nat. Commun.* **12**, 924 (2021) [3] Conrads et al. *arXiv:2310.12841* (2023)

O 24.5 Tue 11:30 MA 042

Observation of Anisotropic Phonon Polariton Propagation with Sum-Frequency Generation Microscopy — •Richarda Niemann[1], Sören Wasserroth[1], Gonzalo Álvarez-Pérez[2], Javier Martín-Sánchez[2], Pablo Alonso-Gonzáles[2], Martin Wolf[1], and Alexander Paarmann[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin — [2]Department of Physics, University of Oviedo, Oviedo 33006, Spain

Surface phonon polaritons (SPhPs) have been proven an interesting tool in the field of nanophotonics as they can be used in applications in the mid- to far-IR regime with lower losses than their plasmonic counterparts.[1] Commonly, propagating SPhPs are imaged using tip-based near-field techniques like scanning near-field optical microscopy which suffer from long image acquisition times due to the point-scanning approach and thus are often limited to only a few selected frequencies.[2] Here, we observe propagating phonon polaritons launched on an m-cut AlN substrate in a far-field approach by interferometric wide-field imaging. Our technique of sum-frequency generation (SFG) spectromicroscopy offers high spatial and spectral resolution at rapid acquisition speed.[3] We are able to extract the anisotropic dispersion of the propagating polaritons and compare them to numerical simulations.

[1] Caldwell et al., Nanophotonics 4, 1 (2015)
[2] Barnett et al., Appl. Phys. Lett. 120, 211107 (2022)
[3] Niemann et al., Appl. Phys. Lett. 120, 131102 (2022)

O 24.6 Tue 11:45 MA 042

Surface phonon polariton ellipsometry — •Giulia Carini, Richarda Niemann, Niclas Sven Müller, Martin Wolf, and Alexander Paarmann — Fritz Haber Institute, Berlin, Germany

Phonon polaritons are light-matter electromagnetic waves emerging in polar crystals from the hybridization of infrared impinging photons with IR-active phonon resonances. The light-matter coupling appears in the bulk polariton dispersion as an avoided crossing and gives rise to a spectral region - known as reststrahlen band (RB) - where the material optically behaves like a metal, only supporting evanescent surface waves, while light propagation inside the crystal is prohibited. The excitation of surface phonon polaritons (SPhPs) requires special experimental schemes to overcome the momentum mismatch between SPhPs and free-space radiation.

In our contribution, we investigate surface phonon polaritons at the interface between gallium phosphide (GaP) and air in momentum-domain by means of the Otto-type prism coupling geometry, a total internal reflection-based approach. Furthermore, we combine this technique with spectroscopic ellipsometry to enable the acquisition of both amplitude and phase information of the reflected waves along the whole RB of GaP. The adjustability of the prism-sample air gap width enables a systematic study of the ellipsometry parameters' dependence on the optical coupling efficiency. In particular, we show that combined observation of both ellipsometry parameters - amplitude and phase - provides a powerful toolkit for the detection of SPhPs, even in the presence of high optical losses.

O 24.7 Tue 12:00 MA 042

Ultra-confined THz hyperbolic phonon polaritons in $HfSe_2$ — Ryan Kowalski[1], Gonzalo Alvarez-Perez[2], Niclas S. Müller[3], Maximilian Obst[4], Giulia Carini[3], Saurabh Dixit[1], Katja Diaz-Granados[1], Aditha S. Senarath[1], Lukas M. Eng[4], Martin Wolf[3], Thomas G. Folland[5], Pablo Alonso-Gonzalez[2], Susanne C. Kehr[4], •Alexander Paarmann[3], and Joshua D. Caldwell[1] — [1]Vanderbilt University, Nashville, TS, USA — [2]University of Oviedo, Oviedo, Spain — [3]Fritz Haber Institute of the Max Planck Society, Berlin, Germany — [4]Institute of Applied Physics, TUD Dresden University of Technology, Dresden, Germany — [5]The University of Iowa, Iowa City, IA, USA

Hyperbolic phonon polaritons have recently attracted tremendous attention due to their ability to confine infrared- to terahertz light to deeply subwavelength dimensions. So far, however, high-quality polaritons in the THz region have only been reported for very few naturally hyperbolic materials.[1] Here, we experimentally demonstrate ultraconfined hyperbolic phonon polaritons in the 3-5 THz spectral range in ultrathin flakes of the van der Waals crystal $HfSe_2$,[2] with confinement factors of up to ≈80 below free space wavelength. Additonally, we also observe a unique case of strong coupling between the hyperbolic mode with the intrinsic epsilon-near-zero polariton emerging from the out-of-plane longitudinal phonon resonance within the hyperbolic band.

[1] T. de Oliveira, et, Adv. Mat. 33, 2005777 (2021).
[2] R.A. Kowalski, Adv. Opt. Mat. 10, 2200933 (2022).

O 24.8 Tue 12:15 MA 042

THz Cavities for Investigation of Strong Light-Matter Coupling — •Michael S. Spencer, Maximilian Frenzel, Joanna M. Urban, and Sebastian F. Maehrlein — Fritz Haber Institute of the Max Planck Society, Berlin, Germany

Next-generation materials science aims for precise and ultrafast manipulation of structural properties and fundamental excitations to alter material properties on-demand, in search of emergent, enhanced, or even hidden states of matter. A novel pathway for achieving these goals is through modulation of light-matter coupling, experimentally achieved using electromagnetic cavities. Modification of low-energy material properties using Terahertz (THz) cavities is a nascent research focus, enabled in recent years by the availability of high-intensity THz radiation sources.

Here, we present our recent development of new Fabry-Pérot cavity designs for experimental realization of THz cavities. These designs allow for full tunability of the THz cavity resonance frequencies, while simultaneously allowing a sample to be positioned at a field-maximum, boosting light-matter interaction. By making use of electrooptic sampling, we measure the full cavity mode spectrum in amplitude and phase. Our experimental and analytical results can be generalized to complex, multi-layered cavities, allowing for a rigorous description of samples beyond the thin-layer approximation. Furthermore, we will experimentally demonstrate how the practical design of such double-cavities provides a unique avenue for simple and direct investigations of light-matter interactions in the THz and mid-infrared spectral regions.

O 24.9 Tue 12:30 MA 042

Generative inverse design of functional molecules for plasmonic nanodevices — •Zsuzsanna Koczor-Benda, Shayantan Chaudhuri, Joe Gilkes, and Reinhard J. Maurer — University of Warwick, Coventry, United Kingdom

The interaction between molecules and strongly confined electromagnetic fields at metallic nanostructures results in extreme enhancement of molecular spectroscopic signals. This effect can be utilized in new nanoscale devices such as molecular terahertz (THz) detectors. However, to achieve high efficiency, molecules with highly specialized properties are required. We explore how quantum chemistry and machine learning methods can provide good candidate molecules for these applications. In particular, we investigate a promising new THz detection technique based on frequency upconversion by molecular vibrations. By screening databases containing millions of molecules, a two-orders-of-magnitude improvement of spectral intensity can be achieved. Generative machine learning provides a route for going beyond existing molecular databases, to instead design new functional molecules by biasing towards the desired properties. We discuss how recent developments in inverse property-driven design open the way for the targeted generation of molecules for THz detection.

O 24.10 Tue 12:45 MA 042

A microscopic approach for active plasmonics in THz-pumped metal nanoparticles — •Jonas Grumm[1], Robert Salzwedel[1], Holger Lange[2], and Andreas Knorr[1] — [1]Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Technische Universität Berlin, Berlin, Germany — [2]Institut für Physikalische Chemie, Universität Hamburg, Hamburg, Germany

The optical response of metal nanoparticles is dominated by the formation of collective electronic resonances, forming localized plasmons. To gain insight into their dynamics and for switching applications, it would be beneficial to influence the plasmonic resonance actively.

Here, we present theoretical models for an active tuning of the plasmonic resonance in optical spectra by using strong THz fields. We discuss theoretical descriptions with different complexity, ranging from a numerical solution of kinetic equations for the electron dynamics to coarse-grained hydrodynamic descriptions of metal nanoparticles. In all cases, we merge the electron dynamics and Maxwell's equations to obtain self-consistent solutions. A renormalization of the optical metal nanoparticle plasmon resonance by the THz field can be realized by THz-induced spatial gradients.