O 96: Plasmonics and Nanooptics: Light-Matter Interaction, Spectroscopy II

Time: Friday 10:30–13:00 Location: H4

O 96.1 Fri 10:30 H4

Enhancing light-matter interaction through inverse design of optical devices. — •Carlos Bustamante[1], Mark Svendsen[2], Franco Bonafé[1], Burak Gurlek[1], Maxim Sukharev[3], Abraham Nitzan[4], and Angel Rubio[1] — [1]MPSD, Hamburg, Germany — [2]Niels Bohr Institute, University of Copenhagen, Denmark — [3]Department of Physics, Arizona State University, USA — [4]Department of Chemistry, University of Pennsylvania, USA

Light-matter interaction plays a crucial role in processes such as spontaneous emission, energy transfer and polaritonic formation. This interaction is sensitive to alterations in the electromagnetic environment which can be caused by the presence of optical materials. In classical optics, changes in the topology of optical materials can lead to the fabrication of optical devices tailored towards specific characteristics, using inverse design methodologies like density-based topology optimization (TO). This work presents the application of optical devices, derived from TO, that can be used to modify light-matter interaction among molecules. To achieve this, we have implemented a TO algorithm that can solve Maxwell equations in the frequency domain on a 2D grid. These tailored devices enhance locally one component of the transverse electric field obtained from classical emitters. In subsequent semiclassical simulations, Maxwell equations were classically propagated with the emitters replaced by pentacene molecules using the quantum mechanical simulation software DFTB+. The results highlight the significant potential of optical devices produced by TO to influence the above-mentioned processes.

O 96.2 Fri 10:45 H4

Squeezing few-cycle light pulses in space and time in the gap of a nanoplasmonic resonator — •Tom Jehle, Sam S. Nochowitz, Juanmei Duan, and Christoph Lienau — Universität Oldenburg, D-26129, Germany

The spatial confinement of light to (sub-)nanometer spot sizes in the gap of a nanoparticle-on-mirror resonator or in the gap of a tunnelling junction has led to dramatic advances in nanosensing [1] and tip-enhanced Raman Spectroscopy [2]. So far, the time dynamics of the fields emitted from such nanocavities have achieved little attention and only recently the time structure of a Terahertz field scattered from the junction of an STM tip could be recorded [3]. Here, we introduce a broadband interferometric scattering-type SNOM technique [4] to reconstruct amplitude and phase of light scattered from a sharp gold taper acting as a near-field probe. Direct Fourier transform gives the time structure of the electric field. We distinguish the near-field scattered from the apex and that emitted by spatially well separated scattering mode of the taper. The apex field decays within 13 fs, a decay time mainly given by the radiative damping of the apex mode. Upon approaching the tip to a gold surface, we observe a 3.5-fold reduction in the decay time to less than 4 fs: coupling to the image dipole drastically increases the radiative damping in the tip-surface junction. Our results pave the way towards linear and nonlinear ultrafast oscilloscopy with nm/fs resolution. [1] R. J. Chikkardy et al., Nature 535, 127 (2016); [2] R. Zhang et al., Nature 531, 623 (2016); [3] T. Siday et al., Nature 629 (2024); [4] J. Zhan et al., Nano Lett. 2024

O 96.3 Fri 11:00 H4

Full-wave simulations of core-shell nanoparticle investigation by tapping mode near-field optical microscopy — •Dario Siebenkotten, Dinghe Dai, Richard Ciesielski, Arne Hoehl, and Bernd Kästner — Physikalisch-Technische Bundesanstalt, Abbestr. 2-12 10587 Berlin

Core-shell nanoparticles are important in applications such as optoelectronics, biosensing, and medicine, where their unique optical and geometric properties play a critical role in functionality. Quantifying the properties of both, the core and the shell, is crucial to understand the nanoparticle's ultimate functionality. However, optical investigation of their properties, while widely employed, is generally diffraction-limited and thus unsuitable for the investigation of individual nanoparticles, as they only provide averaged ensemble information. Scattering-type scanning near-field optical microscopy (s-SNOM) promises access to a single nanoparticle's size and optical properties through the use of an atomic force microscopy tip as an optical antenna, which confines the sampling fields to nanoscale dimensions. To explore the single nanoparticle s-SNOM response, we model different core-shell nanoparticles using Finite Element Modelling, revealing complex resonance-antiresonance behaviour in dependence of their geometrical and optical properties. We further explore the origin of the emergent antiresonance through the use of Fourier-demodulation of the probe tapping [1], closely mimicking the experiment. [1] Mooshammer et al., ACS Photonics 7, 344-351 (2020)

O 96.4 Fri 11:15 H4

Near-field microscopy of complex polaritonic excitations — •Farid Aghashirinov[1], Bettina Frank[1], Harald Giessen[1], Andrea Manchini[2], Lin Nan[2], Giacomo Venturi[2], Nicola Melchioni[2], and Antonio Ambrosio[2] — [1]4-th Physics Institute, University of Stuttgart, Stuttgart, Germany — [2]Vectorial Nano-Imaging, Istituto Italiano di Tecnologia, Milano, Italy

We investigate directional near-infrared (NIR) plasmon polaritons (PPs) in mechanically exfoliated bulk molybdenum oxide dichloride (MoOCl2) flakes on a glass substrate with gold disk launchers positioned on top of the flakes, through experimental and theoretical methods [1]. MoOCl2, a natural van der Waals crystal, exhibits low-loss, in-plane hyperbolic PPs across the visible and NIR ranges, and displays a unique anisotropic optical response based on the polarization of the incident light. When light polarization aligns with the flake's shortest axis, MoOCl2 behaves optically metallic, while aligning with the longest axis reveals a dielectric behaviour. To experimentally study the hyperbolic PPs in MoOCl2, we use a near-field excitation and detection approach. Gold disks serve as excitation sources, scattering incident light to couple it with the material's polaritonic modes, while a reflection scattering-type scanning near-field optical microscope (s-SNOM) enables subwavelength imaging. For tunable wavelength illumination, we use a broadband laser from Stuttgart Instruments (SI), which, when combined with s-SNOM, allows us to investigate these modes across a wide wavelength range, capturing detailed insights into their behavior and propagation. [1] G. Venturi et al., Nat Commun 15, 9727 (2024)

O 96.5 Fri 11:30 H4

Discovery of phonon-polaritonic skyrmions: Transition from bubble- to Néel-type — •Florian Mangold[1], Enrico Baù[2], Lin Nan[2], Julian Schwab[1], Thorsten Gölz[2], Andrea Mancini[2], Bettina Frank[1], Andreas Tittl[2], and Harald Giessen[1] — [1]4th Physics Institute, Research Center SCoPE, and IQST, University of Stuttgart, Germany — [2]Chair in Hybrid Nanosystems, Nano-Institute Munich, Ludwig-Maximilians Universität München, Germany

Optical skyrmions constitute an emerging and rapidly developing field in the domain of solid-state physics and photonics, allowing for control over topological light textures through light-matter interactions. However, until now their practical applications have been severely limited by inherent restrictions of materials with high optical losses and a lack of tunability between different topological properties. We leverage the steep dispersion of silicon carbide thin films to image surface phonon polariton skyrmion lattices via near-field microscopy and experimentally demonstrate topological tuning between Bubble- and Néel-type skyrmions, a unique advantage that polar dielectrics offer over most existing approaches. The ability to control and manipulate these skyrmion lattices through small changes (<10%) in excitation wavelength allows for tailoring the size of individual skyrmions by a factor of 4. Tuning topological properties has been a long-standing goal in the field of optics. Our results expand the tunability of skyrmions in evanescent waves and are a starting point to investigate other topological features in phononic systems such as merons, skyrmion bags and other complex structured light fields.

O 96.6 Fri 11:45 H4

Tracking Non-Equilibrium Electron Distributions to Propel Chemical Dynamics at Surfaces — •Henry T. Snowden[1], Reinhard J. Maurer[1], Markus Uehlein[2], and Baerbel Rethfeld[2] — [1]University of Warwick, Coventry, UK — [2]RPTU Kaiserslautern-Landau, 67663 Kaiserslautern, Germany

A mechanistic understanding of ultrafast light-matter interactions with surfaces and nanoparticles is invaluable for the study of ultrafast dynamics at surfaces. Immediately after a laser pulse, non-equilibrium electrons are generated in metals which rapidly relax to a thermal dis-

tribution that follows the two-temperature model. Here, we study the longevity of electronic non-equilibrium which depends on the electronic structure and other materials properties. We simulate the temporal evolution of non-equilibrium electron distributions with the Athermal Electron Model (AthEM), which explicitly considers electronic nonequilibrium, a laser term and electron scattering in the relaxation time approximation. Equipped with this approach, we investigate how athermal electron-hole pair distributions differ in various materials configurations such as intermetallic interphases, and nanoparticles. We will use this information to discuss the requirements to create long-lived non-equilibrium distributions from which high-energy charge carriers can be harvested.

O 96.7 Fri 12:00 H4

Collective Behavior of SiC Phonon Polariton Resonators — •Richarda Niemann[1,2], Sören Wasserroth[1], Guanyu Lu[2], Christopher R. Gubbin[3], Martin Wolf[1], Simone de Liberato[3], Joshua D. Caldwell[2], and Alexander Paarmann[1] — [1]Fritz Haber Institute, Berlin, Germany — [2]Vanderbilt University, Nashville, TN, USA — [3]University of Southampton, Southampton, UK

Structuring polar materials with microresonators enables light confinement to sub-wavelength scales and the emergence of localized phonon polaritons (PhPs). Previous studies on PhP single resonators and 2D resonator arrays investigated the coupling behavior of resonators with their surrounding [1,2]. Here, we bridge the gap between a single resonator and a 2D array by studying subdiffractional SiC microresonators on a SiC substrate in 1D arrays with sumfrequency generation (SFG) spectro-microscopy [3,4]. The high spatial resolution on the order of the lateral size of the microresonators allows us an in-depth analysis of the build-up of collective modes in 1D arrays with sizes from one to several resonators. We observe different optical responses for in-phase and out-of-phase excitation showing the importance of phase relationships for coupling between the resonators. By investigating arrays with different periods, we gain information about the coupling mechanisms.

[1] Gubbin et al., Phys. Rev. B, 95 (2017)
[2] Chen et al., ACS Photonics, 1 (2014)
[3] Niemann et al., Appl. Phys. Lett. 120, 131102 (2022)
[4] Niemann et al., Adv. Mater. 2024, 36, 2312507

O 96.8 Fri 12:15 H4

High-Throughput Phonon Polariton Materials Discovery from First Principles — •Elena Gelzinyte[1], Giulia Carini[1], Niclas S. Mueller[1], Martin Wolf[1], Karsten Reuter[1], Johannes T. Margraf[2], Alexander Paarmann[1], and Christian Carbogno[1] — [1]Fritz-Haber-Institut der MPG, Berlin — [2]University of Bayreuth

Phonon Polaritons (PhPs), quasi-particles that arise from strong coupling between infrared photons and optical lattice vibrations, are promising in nanophotonic applications for highly directional and confined light propagation with low optical loss [1]. However, little is still known about the trends in material space that drive the emergence and characteristics of PhPs. To describe these trends, we compute the permittivity function [2] as the basis for describing PhP dispersion. We employ Kubo's linear-response theory, where the harmonic and anharmonic phonon properties as well as Born effective charges are used to model the involved couplings. This approach is validated on materials with well-characterised PhP behaviour, such as h-BN, MoO_3 and β-Ga_2O_3, and then applied to 5,000 materials from the JARVIS database [3]. By analysing the trends emerging from these data, we identify qualitative metrics that capture the material's ability to support PhPs. Finally, we highlight some examples and outliers of this high-throughput screening process.

[1] E. Galiffi *et al.*, *Nat. Rev. Mater.* **9**, 9 (2024).
[2] M. Born & K. Huang, *Dynamical Theory of Crystal Lattices* (1954).
[3] K. Choudhary *et al.*, *npj Comput. Mater.* **6**, 1 (2020).

O 96.9 Fri 12:30 H4

Electron Relaxation in Noble Metals: Microscopic Theory of Orientational Relaxation, Thermalization and Cooling — •Jonas Grumm and Andreas Knorr — Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Technische Universität Berlin, Berlin, Germany

The excitation of electrons in noble metals by light fields with high intensities results in non-equilibrium electron occupations. The relaxation back to thermodynamic equilibrium, proceeding on scales from a few femtoseconds to several picoseconds, depends strongly on these excitation conditions: Typically, during the first tens of femtoseconds after the excitation, momentum polarization induced by the distinct polarization direction of the light field dominates the electron gas.

To address the relaxation dynamics of this momentum polarized state, we solve a kinetic momentum-resolved Boltzmann equation with focus on electron-phonon interaction and introduce orientational relaxation as first temporal step in a sequence of relaxation processes followed by thermalization and cooling. Applying a self-consistent treatment of Maxwell's equations and material dynamics we are able to present results for the optical and thermal properties of plasmonic nanostructures after linear and non-linear optical excitations. Thereby, Pauli-blocking non-linearities in the electron-phonon interaction modify the relaxation dynamics for strong excitations and effect the optical response of the electron gas.

O 96.10 Fri 12:45 H4

Sum-Frequency Generation Spectro-Microscopy in Barium Titante — •Dorothee Mader[1], David Pesquera[2], Saptam Ganguly[2], Richarda Niemann[1,3], Martin Wolf[1], Sebastian F. Maehrlein[1], Jose Santiso[2], and Alexander Paarmann[1] — [1]Fritz Haber Institute of the Max Planck Society, Berlin, Germany — [2]Catalan Institute of Nanoscience and Nanotechnology, Barcelona, Spain — [3]Vanderbilt University, Nashville, USA

Ferroelectric Barum Titanate (BTO) can be switched by excitation of phonons in the infrared (IR), enabeling the applications in future optical technologies.[1] Here, we investigate the resonance behavior of BTO in the IR by employing infrared-visible (IR-VIS) sum-frequency generation (SFG) spectro-microscopy using the free-electron laser of the Fritz Haber Institute.[2,3] As a second-order nonlinear technique, this method is sensitive to the material's symmetry. Additionally, as it is employed resonantly, it can reveal vibrations, local structure and bonding of the material. We demonstrate resonant ferroelectric imaging using SFG spectro-microscopy for bulk BTO and 500 nm thin BTO membranes.

[1] M. Kwaaitaal et al., Nat. Photonics 82(3), 2731, (2024).
[2] R. Niemann et al., Appl. Phys. Lett. 120, 131102, (2022).
[3] D. Mader et al., J. Chem. Phys. 161, 094706 (2024).