# O 80: Plasmonics and Nanooptics IV: Light-Matter Interaction and Spectroscopy III

Time: Thursday 15:00–17:15 Location: CHE 89

O 80.1 Thu 15:00 CHE 89

**Energy and momentum distribution of surface plasmon-induced hot carriers** — •Christopher Weiss[1], Michael Hartelt[1], Pavel N. Terekhin[1], Tobias Eul[1], Anna-Katharina Mahro[1], Benjamin Frisch[1], Eva Prinz[1], Baerbel Rethfeld[1], Benjamin Stadtmüller[1,2], and Martin Aeschlimann[1] — [1]Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, Germany — [2]Institute of Physics, Johannes Gutenberg University Mainz, Germany

Are the spectroscopic properties of plasmon- and photon-induced hot carriers fundamentally different? Electrons excited at the bulk plasmon resonance, for example, exhibit a distinct spectroscopic signature [1]. However, bulk plasmons are rather unsuited for technological applications compared to surface plasmons.

To answer the initial question for surface plasmons, we separate the energy and momentum distributions of surface plasmon polariton (SPP)-induced hot electrons from those of photoexcited electrons in a two-color time-resolved photoemission electron microscopy experiment.

In our experiment, these SPP-induced hot electrons show only small momenta parallel to the surface and a narrow energy distribution close to the Fermi energy [2]. This result clearly distinguishes them spectroscopically from their photoinduced counterparts.

[1] Reutzel et al., Phys. Rev. Lett. 123 (2019), 017404

[2] Hartelt et al., ACS Nano 15, 12 (2021), 19559–19569

O 80.2 Thu 15:15 CHE 89

**Lorentz Microscopy of Optical Fields** — •John H. Gaida[1,2], Hugo Lourenço-Martins[1,2], Sergey V. Yalunin[1,2], Armin Feist[1,2], Murat Sivis[1,2], Thorsten Hohage[3], F. Javier García de Abajo[4,5], and Claus Ropers[1,2] — [1]MPI for Multidisciplinary Sciences, Göttingen, Germany — [2]4th Physical Institute, University of Göttingen, Germany — [3]Institute of Numerical and Applied Mathematics, University of Göttingen, Germany — [4]ICFO-Institut de Ciencies Fotoniques, Castelldefels (Barcelona), Spain — [5]ICREA-Institució Catalana de Recerca i Estudis Avançats, Barcelona, Spain

In electron microscopy, detailed insights into nanoscale optical properties of materials are gained by spontaneous inelastic scattering leading to electron-energy loss and cathodoluminescence. Photon-induced near-field electron microscopy (PINEM) allows for mode- and polarization-selective imaging based on stimulated scattering in the presence of external sample excitation. Through this process a spatial phase profile inherited from the optical fields is imprinted onto the wave function of the probing electrons. Here, we introduce Lorentz-PINEM for the full-field, non-invasive imaging of complex optical near fields at high spatial resolution. We use energy-filtered defocus phase-contrast imaging and iterative phase retrieval to reconstruct the phase distribution of interfering surface-bound modes on a plasmonic nanotip. Our approach is universally applicable to retrieve the spatial phase of nanoscale fields and topological modes.

[1] John H. Gaida, *et al.*, 2022, Preprint at Research Square *doi.org/10.21203/rs.3.rs-2150760/v1.*

**Topical Talk** O 80.3 Thu 15:30 CHE 89

**Topological Plasmonics and Plasmonic Twistronics: Skyrmions, Merons, Quasicrystals, and Sykrmion Bags** — •Harald Giessen[1], Bettina Frank[1], Pascal Dreher[2], David Janoschka[2], Frank Meyer zu Heringdorf[2], Julian Schwab[1], Tim Davis[1,4], Kobi Cohen[3], Shai Tsesses[3], and Guy Bartal[3] — [1]4. Physikalisches Institut, Universität Stuttgart, Germany — [2]CENIDE, University of Duisburg-Essen, Germany — [3]Technion, Haifa, Israel — [4]University of Melbourne, Australia

We present ultrafast vectorial electric field measurements of plasmonic skyrmions, merons, quasicrystalline structures, and skyrmion bags. 10 nm spatial resolution, sub-fs temporal resolution, and vectorial properties of the surface plasmon field on single crystalline, atomically flat gold surfaces are presented. Our techniques combine ultrafast vector PEEM and interferometric s-SNOM. Our tailored model systems reveal a rich set of topological phenomena, reaching even into 4-dimensional topology.

O 80.4 Thu 16:00 CHE 89

**Direct imaging of photonic band-edge states in golden-angle Vogel spirals using photoemission electron microscopy** — Martin Aeschlimann[1], Felix Fenner[2], Tobias Brixner[3], Benjamin Frisch[1], Patrick Folge[4], Michael Hartelt[1], •Matthias Hensen[3], Thomas H. Loeber[5], Walter Pfeiffer[2], Sebastian Pres[3], and Bernd Stannowski[6] — [1]Fachbereich Physik and Research Center OPTIMAS, TU Kaiserslautern, 67663 Kaiserslautern, Germany — [2]Fakultät für Physik, Universität Bielefeld, 33615 Bielefeld, Germany — [3]Institut für Physikalische und Theoretische Chemie, Universität Würzburg, 97074 Würzburg, Germany — [4]Angewandte Physik, Universität Paderborn, 33098 Paderborn, Germany — [5]Nano-Structuring-Center, 67663 Kaiserslautern, Germany — [6]5PVcomB, Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, 12489 Berlin, Germany

Golden-angle Vogel spirals, as deterministic aperiodic structures, support isotropic photonic band gaps and have interesting applications. Especially localized modes, such as band-edge states, are essential for tailored light–matter interaction. Here we report imaging of such modes using photoemission electron microscopy (PEEM). Tunable ultrashort light pulses excite them in golden-angle Vogel spirals that were fabricated by focused-ion-beam (FIB) milling of an a-Si:H layer. The local near-field leads to electron emission which is detected spatially resolved. The demonstration of FIB-textured a-Si:H as photonic material and the ability of PEEM mode imaging offers means to spatiotemporally resolve mode dynamics and to perform nanospectroscopy.

O 80.5 Thu 16:15 CHE 89

**Short-range plasmonic skyrmions** — •Bettina Frank[1], Simon Mangold[1], Harald Giessen[1], Pascal Dreher[2], Frank Meyer zu Heringdorf[2], and Stefan Kaiser[1] — [1]4. Physikalisches Institut, Universität Stuttgart, Germany — [2]CENIDE, University of Duisburg-Essen

We utilize ultrathin, atomically flat, single crystalline gold flakes which compress the plasmon wavelength to lambda_air/7. Using hexagonal boundaries that are carved with focused gold ions, we demonstrate plasmonic skyrmions, both using PEEM as well as s-SNOM. The electromagnetic field is structured on a 140 nm scale, which offers opportunities for topological light-matter interaction on a deep subwavelength scale.

O 80.6 Thu 16:30 CHE 89

**Phase-resolved mapping of plasmonic resonances at nanostructures** — •Tim Dauwe[1,2], Hugo Lourenco-Martins[1,2], Murat Sivis[1,2], Jakob Hagen[1,2], and Claus Ropers[1,2] — [1]Max Planck Institute for Multidisciplinary Sciences, 37077 Göttingen, Germany — [2]IV. Physical Institute, University of Göttingen, 37077 Göttingen, Germany

In this contribution, we study resonant plasmonic nanostructures by a combination of inelastic electron-light scattering techniques. We investigate metallic nanorods using electron-energy loss spectroscopy (EELS) and photon-induced near-field electron microscopy (PINEM), elucidating spectrally-dependent mode profiles and symmetries using spontaneous and stimulated interactions. We explore multicolor excitation and nonlinear wave mixing as contrast mechanisms accessing spatial phase profiles and local nonlinear response.

O 80.7 Thu 16:45 CHE 89

**Shortcut to Self-Consistent Light-Matter Interaction and Realistic Spectra from First Principles** — •Christian Schäfer[1,2] and Göran Johansson[1] — [1]Department of Microtechnology and Nanoscience, MC2, Chalmers University of Technology, 412 96 Göteborg, Sweden — [2]Department of Physics, Chalmers University of Technology, 412 96 Göteborg, Sweden

Nanoplasmonic and optical cavity environments provide a novel handle to non-intrusively control materials and chemistry. We introduce here a simple approach to how an electromagnetic environment can be efficiently embedded into state-of-the-art electronic structure methods, taking the form of radiation-reaction forces [1]. We demonstrate that this self-consistently provides access to various optical effects and provides an efficient path to connect vastly different system scales [2]. As an example, we illustrate its seamless integration into time-dependent density-functional theory and its application to polaritonic chemistry with virtually no additional cost, presenting a convenient shortcut to self-consistent light-matter interactions.

[1] C. Schäfer and G. Johansson, PRL 128, 156402 (2022).
[2] C. Schäfer, J. Phys. Chem. Lett. 2022, 13, 6905-6911.

O 80.8 Thu 17:00 CHE 89

**Microscopic description of the optical response in metal nanoparticles** — •Jonas Grumm, Robert Salzwedel, Malte Selig, and Andreas Knorr — Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Technische Universität Berlin, Berlin, Germany

The optical response of metal nanoparticles is dominated by the formation of collective electronic resonances, so-called localized plasmons.

Here, we present a microscopic approach to their temporal dynamics based on the self-consistent treatment of the microscopic Boltzmann transport equations and the macroscopic Maxwell's equations for the electromagnetic fields. The corresponding numerical simulations describe the formation, propagation and thermalization dynamics of plasmons via the coupled electron-phonon dynamics in the self-consisting solved optical field. The formalism allows nonlinear optical processes to be included in the description.