O 14: Plasmonics and Nanooptics II: Light-Matter Interaction and Spectroscopy

Time: Monday 15:00–17:30 Location: MA 042

O 14.1 Mon 15:00 MA 042

Towards Isolated Attosecond Electron Trios — Germann Hergert, •Rasmus Lampe, Andreas Wöste, and Christoph Lienau — Institut für Physik, Carl-von-Ossietzky Universität, 26129 Oldenburg, Germany

Very recently, the formation of multi-electron number states in the laser-triggered photoemission from metal tips has created much attention [1,2]. These number-states show intriguing properties, like highly correlated electron energies and increasing nonlinearities in their power scaling. Here, we use, for the first time, few-cycle near-infrared driving pulse around 2000 nm for the creation of such number states. The low photon energy, 1/7 of the work function of gold, drastically increases the nonlinear order of the emission process, up to the 21st order for multiphoton-emission of a three-electron state.

Such extreme nonlinearities make the emission process exceptionally sensitive to minor changes in the driving field. We demonstrate this by monitoring the electron yield while varying the delay of a phase-locked pulse pair. We observe autocorrelations that are fully governed by an isolated central peak with 0.8 fs width. Only two side peaks with amplitudes < 25% are seen. This may enable the generation of isolated attosecond electron trios. We are currently studying the CEP dependence of the emission to gain further insight into the emission regime.

[1] S. Meier et al., Nat. Phys. 19, 1402 (2023)
[2] R. Haindl et al., Nat. Phys. 19, 1410 (2023)

O 14.2 Mon 15:15 MA 042

Soliton Formation Dynamics in a Microresonator Probed by Electron Energy Gain Spectroscopy — •F. Jasmin Kappert[1], Yujia Yang[2], Jan-Wilke Henke[1], Arslan S. Raja[2], Germaine Arend[1], Guanhao Huang[2], Armin Feist[1], Zheru Qiu[2], Rui Ning Wang[2], Aleksandr Tusnin[2], Alexey Tikan[2], Tobias J. Kippenberg[2], and Claus Ropers[1] — [1]Max Planck Institute for Multidisciplinary Sciences, Göttingen,Germany — [2]Institute of Physics, Swiss Federal Institute of TechnologyLausanne (EPFL), Switzerland

Ultrafast electron microscopes provide a powerful platform for investigating confined optical modes with high temporal and spatial resolution. Recently, integrated photonics has boosted the interaction strength of light with free electrons, enabling nanoscale spectroscopy of optical states with a continuous electron beam.

Here, we use continuous-beam electron energy gain spectroscopy (EEGS) to probe nonlinear optical excitations inside a microresonator [1]. Starting from single-color excitation, increasing the optical input power leads to the formation of dissipative Kerr solitons via various nonlinear comb states, evident in characteristic optical and EEG spectra. This scheme enables non-invasive probing of nonlinear optical dynamics with direct access to the intracavity field. Moreover, the interaction with solitons facilitates high-frequency spatiotemporal electron beam modulation.

[1] Y. Yang, et al., arXiv:2307.12142 (2023)

Topical Talk O 14.3 Mon 15:30 MA 042

Time-resolved interaction of the electron system with strong surface plasmon polariton fields — •Frank Meyer zu Heringdorf — Faculty of Physics, University of Duisburg-Essen, 47048 Duisburg, Germany

Time-resolved experiments often rely on femtosecond laser pulses to create a non-equilibrium situation in the electron system of a surface. The specifics of the created nonequilibrium hereby depends on the detailed spatiotemporal properties of the excitation field. It has been demonstrated that by controlling surface plasmon polaritons (SPPs) it is possible to create unique (even topologically non-trivial) polarization fields at surfaces. Experimental knowledge of the SPP vector field on a few hundred nm length- and a fs time-scale can be obtained using vector microscopy and polarimetry, where time-resolved, probe-polarization-dependent two photon photoemission data is obtained in a spectroscopic low energy electron microscope (SPE-LEEM). Using Archimedean spirals for excitation, strong SPP foci can be created that exhibit highly-nonlinear electron emission, and the ponderomotive interaction of the electron with the SPP field during the photoemission process can be used to quantify the field strength. Time- and angle-resolved experiments at a SPP focus clarify the intermediate states involved in the emission process and demonstrate that SPPs can coherently couple to electronic states in above-threshold electron emission.

O 14.4 Mon 16:00 MA 042

Electron driven photon sources for engineered generation of light propagating in Free Space — •Masoud Taleb and Nahid Talebi — Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, Germany

Optical beams with angular momentum have been intensively investigated for offering an additional degree of freedom for information transfer or manipulating the chiral degrees of freedom in matter. Such beams are generally realized via metasurfaces, photon sieves, or structured lenses. However, most of them lack key properties in controlling the polarization, directionality and intensity of the light which is generated and directed from a localized radiation sources such as electron beam induced emission. Previously, we have realized electron-driven photon sources which are fabricated by specific orders of nanoholes in a thin layer of gold (Nano Lett. 2020, 20, 8, 5975*5981). Here, we further elucidate on a variety of degrees of freedom in our design principle to both control the polarization and the shape of the generated light. A moving electron at the kinetic energy of 30 kV interacts with the system acting as a broadband source of optical excitation. Coupled plasmon polaritons propagate on the lattice forming a chain plasmon polaritons, that further radiates to the far field and generates a vortex beam. This phenomenon is experimentally studied utilizing cathodoluminescence spectroscopy and angle-resolved mapping.

O 14.5 Mon 16:15 MA 042

Tuning Disorder-Driven Localization of Plasmons in Random Assemblies of Gold Nanoparticles — •Mohammed Fayis Kalady[1,2], Johannes Schultz[1], Kristina Weinel[1,3], Daniel Wolf[1], and Axel Lubk[1,4] — [1]Leibniz Institute for Solid State and Materials Research (IFW) Dresden, Helmholtzstraße 20, 01069 Dresden, Germany — [2]Indian Institute Of Technology (IIT) Delhi, Hauz Khas, New Delhi, Delhi 110016, India — [3]Federal Institute of Materials Research and Testing (BAM), Unter den Eichen 87, 12205 Berlin, Germany — [4]Institute of Solid State and Materials Physics, Dresden University of Technology, Haeckelstraße 3, 01069 Dresden, Germany

It is well-known that assemblies of plasmonic NPs support hybridized modes of localized surface plasmons, which delocalize in geometrically well-ordered arrangements. Here, we study the hybridization behaviour in geometrically completely disordered systems of Au NPs fabricated by a newly developed e-beam synthesis method that facilitates the production and arrangement of NPs of different sizes and distances. Employing state-of-the-art STEM Electron Energy Loss Spectroscopy in combination with numerical simulations, we reveal the spatial and spectral distribution of the coupled LSP modes. We show, amongst others, that their disorder-driven localization behaviour crucially depends on the thickness of NPs, exhibiting a transition in the energy dependence of the localization at a critical thickness of about 10 nm. Employing numerical simulations, we discuss this behaviour with respect to the size and distance distribution of the NPs.

O 14.6 Mon 16:30 MA 042

Exploring the limits of refractive index sensing using Mie voids — •Micha Kappel, Serkan Arslan, Michelle Pfahl, Mario Hentschel, and Harald Giessen — 4th Physics Institute and Research Center SCoPE, University of Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany

The research of resonant phenomena using dielectric nanostructures, described by Mie's scattering theory, has enable new technologies in the field of nanophotonics. Resonant structures such as Mie voids have recently been realized, but have not been characterized in terms of their sensing potential.

To investigate their resonant features, the reflectance spectra are recorded via microspectroscopy. By inducing known refractive index changes inside the Mie voids by temperature change or liquid mixtures, the spectral shift of the resonances can be tracked by comparing the spectra at each refractive index. Comparing the resonances positions to a linear fit yields us the smallest measurable refractive index change.

For a 10 by 10 array of Mie voids with an overall volume of 11 fL, the smallest measurable refractive index change Δn is 0.00029 RIU. For a single Mie void with a volume of 520 aL, it is possible to detect a refractive index change Δn of 0.0049 RIU.

Using dielectric Mie voids we have realized the refractive index sensor the smallest sensing volume in the range of hundreds of attoliters. By using this technology, new applications in the field of biology and medicine could become possible, such as detecting neurotransmitters from single vesicles.

O 14.7 Mon 16:45 MA 042

Emission from propagating Bloch surface wave polaritons — •Sebastian Henn, Andreas Müller, Marius Grundmann, and Chris Sturm — Felix-Bloch-Institut für Festkörperphysik, Fakultät für Physik und Erdsystemwissenschaften, Uni Leipzig, Leipzig, Deutschland

Bloch surface waves (BSW) correspond to photonic modes propagating along the interface between a dielectric multilayer and the ambient. Due to their evanescent field on both sides of the surface they cannot couple to the electromagnetic far-field of the ambient yielding high lateral propagation lengths in the surface plane. This can be exploited to obtain long-range polaritons by strongly coupling BSW to excitons. In this contribution we demonstrate emission from propagating exciton-polariton states in ZnO. They are detected by diffractive outcoupling using line gratings, produced by electron-beam lithography, in the sample surface. By means of Fourier-imaged photoluminescence we are able to investigate the influence of the grating geometry on the emission behaviour and show propagation lengths of up to 30 μm for states of the lower polariton branch. This is makes Bloch surface wave polaritons interesting for on-chip polaritonic applications.

O 14.8 Mon 17:00 MA 042

Spatially resolved nonlinear plasmonics — •Florian Mangold, Johannes Schust, Mario Hentschel, Bettina Frank, and Harald Giessen — 4th Physics Institute, Research Center SCoPE, and IQST, University of Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany

The nonlinear behavior of plasmonic nanoantenna arrays can be investigated using nonlinear spectroscopy. In our contribution we introduce nonlinear spatially resolved spectroscopy, which is capable of imaging the k-space behavior and the spectral behavior of nonlinear nanoantenna signals, as well as to spatially resolve the nonlinear signal of single nanoantennas.

These additional abilities give us the possibility to spatially resolve the THG signal of gold nanoantenna arrays and investigate the homogeneity of the antenna field. Furthermore, we are able to spatially resolve the THG emission centers of the third-order mode and observe their response to tuning the wavelength over the resonance.

In addition, we discovered that by increasing the laser intensity, certain antennas in our array became exceptionally bright. By correlating our spatially resolved nonlinear image with structural SEM data, we can prove that these bright antennas have deformed into a peanut shape. Thus our NSRS setup enables the investigation of the nonlinear self-enhancement process of nanoantennas under intense laser heating. We also simulated these peanut shaped antennas and could observe that these 3-dimensional structures lead to the enhancement of the nonlinear emission.

O 14.9 Mon 17:15 MA 042

Mie Void Metasurfaces — •Michelle Pfahl, Benjamin Reichel, Serkan Arslan, Mario Hentschel, and Harald Giessen — 4th Physics Institute and Research Center SCoPE, University of Stuttgart, Pfaffenwaldring 57, 70569, Stuttgart, Germany

The field of nanooptics has developed into a subject of large interest as it addresses the manipulation of light on this microscopically small scale. Metasurfaces, which are based on the interaction of optical resonators with light, offer novel possibilities for compact optics replacing conventional optical devices as well as offering novel functionalities. Dielectric Mie voids have emerged as a new building block for these metasurfaces. Spherical air inclusions in high index materials allow to resonantly confine light, circumventing material loss. Thus, these structures show pronounced resonances even in the UV spectral range. We take the first step to implement Mie void based metasurfaces by realizing a blazed grating. Using microspectroscopy in the real as in the spatial Fourier domain, as well as carrying out diffraction experiments with monochromatic light, we confirm the asymmetric diffraction behavior of our gratings. Furthermore, we implemented an analytical model which reproduces our results well. Mie voids promise further interesting applications for metasurfaces, such as reflective lenses, which could even work in the deep UV spectral range.