HL 11: Plasmonics and Nanooptics I: Light-Matter Interactions

Time: Monday 10:30–13:00 Location: TRE Ma

HL 11.1 Mon 10:30 TRE Ma

Nanoplasmonics from large-scale ab initio calculations: opposite trends in Ag and Na clusters — •Marc Barbry[1], Peter Koval[2], Natalia E. Koval[1], Javier Aizpurua[1], and Daniel Sánchez[1,2] — [1]Material Physics Center, San Sebastián, Spain — [2]Donostia International Physics Center, San Sebastián, Spain

An accurate description of electronic excitations is indispensable for understanding material properties and designing nanoscale devices. For instance, using large-scale TDDFT calculations, we have recently demonstrated the importance of taking into account the details of the atomic-scale structure [1] and the quantization of electron transport [2] in metal nanostructures in order to accurately describe their plasmonic properties. In this contribution we will compare the surface plasmon resonance of sodium and silver clusters within the same framework of *iterative* TDDFT [3]. Recent progress in our implementation made it possible to perform calculations of large clusters of diameters ranging from a few Å to 4–5 nm, counting up to 5000 silver atoms and using only modest computational resources (a 32-core node with 500GB RAM). With these new capabilities, we have characterized the size-scaling of the SPR frequency for both sodium and silver clusters. As expected these two materials show opposite behaviours that can be related to the different spill out of charge at the surface and to the additional screening created by the 4*d* electrons in silver.

[1] M. Barbry *et al. Nano Letters*, **15** (2015) 3410. [2] F. Marchesin *et al. ACS Photonics*, **3** (2016) 269. [3] P. Koval *et al. J. Phys.: Condens. Matter*, **28** (2016) 214001.

HL 11.2 Mon 10:45 TRE Ma

Mueller matrix investigation of gold gratings — •Meng Wang, Anja Löhle, Bruno Gompf, Martin Dressel, and Audrey Berrier — 1. Physikalisches Institut and Research Center SCoPE, Universität Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany

Surface plasmon polaritons (SPPs), dispersive collective electron excitations propagating along metal-dielectric interfaces leading to strong field confinement, are powerful ways to control the electromagnetic field. They foster promising applications, e.g., in sensing and nanophotonics. In this work, a plasmonic one-dimensional grating fabricated by evaporating gold thin films on an elastomer is investigated by Mueller Matrix (MM) spectroscopic ellipsometry, a powerful tool to characterize the interaction of nanostructured objects with polarized light. All 16 MM elements were measured in reflection. To evaluate the respective role of specific features, such as plasmonic modes, anisotropy, material absorption, or simply diffraction orders, the optical response was measured at various angles of incidence and different azimuthal orientations over a broad frequency range. Hence, the complex interactions resulting from the interplay of these excitations can be unravelled. Modelling of the optical properties based on a biaxial model completes the study. Anisotropy, SPPs, diffraction orders and inter-band transition are distinguished from their different dispersive behaviour and explain all MM features. This knowledge can help to better understand how to tailor the specific excitations and provides guidelines for the design of novel optical functionalities using nanostructured materials.

HL 11.3 Mon 11:00 TRE Ma

Pump-probe nanoscopy with NIR to deep THz radiation — •F. Kuschewski[1], S.C. Kehr[1], J. Döring[1], N. Awari[2], B. Green[2], S. Kovalev[2], M. Gensch[2], and L.M. Eng[1] — [1]Institut für Angewandte Physik, TU Dresden — [2]Helmholtz-Zentrum Dresden-Rossendorf

Recently, scattering-type scanning near-field infrared microscopy (s-SNIM) was successfully combined with pump-probe experiments pushing the temporal resolution of s-SNIM down to a few fs [1]. That combination offers spectacular possibilities to explore the dynamics of nanoscale physical devices, but usually shows a low optical contrast due to high background signals. We analyzed the frequency spectrum in pump-probe s-SNIM finding sidebands to the main carrier frequency that provide a much higher signal-to-noise ratio, as proven both by simulations and recent experiments [2]. Experimentally a greatly increased contrast in the 1st order sideband was found [2], that now has been extended up to the 5th order, applying s-SNIM to a structured gold/semiconductor sample. Note that our approach is applicable to the broad spectral range from visible to THz wavelengths.

[1] M. Wagner et al. *Nano Lett.* **14**, 894 (2014).
[2] F. Kuschewski et al. *Sci. Rep.* **5**, 12582 (2015).

HL 11.4 Mon 11:15 TRE Ma

Photon emission from plasmonic tunnel junctions including a microscope tip and metallic quantum wells — •Tomoki Sueyoshi, Peter-Jan Peters, and Richard Berndt — Institut für Experimentelle und Angewandte Physik, CAU Kiel, Germany

Radiative decay processes involving tunneling electrons are studied by probing light emission from metallic quantum-well (QW) junctions in a scanning tunneling microscope. The electronic states of Pb QWs on Ag(111) are controlled with the number of Pb layers and identified by scanning tunneling spectroscopy. The luminescence from the tip-QW-metal junctions involves radiative decay of tip-induced localized plasmons excited by inelastic tunneling electrons. The observed optical spectra reflect the plasmon density of states in the junctions. In addition, intense emission is observed from transitions of tunneling electrons between unoccupied QW states. These results demonstrate plasmon-assisted luminescence induced by tunneling electrons in the QW tunnel junctions.

HL 11.5 Mon 11:30 TRE Ma

Temporal dynamics of nanowire based lasers — •Robert Röder[1], Themistoklis Sidiropoulos[2], Rupert F. Oulton[2], and Carsten Ronning[1] — [1]Institut für Festkörperphysik, Friedrich-Schiller-Universität Jena, Germany — [2]Imperial College London, UK

Reinforced work in the field of nanophotonics for on-chip integrated optical components is urgent to provide novel approaches for optical data processing which can circumvent the forthcoming limitations of conventional electronic circuits. Here, II-VI compound semiconductor nanowires (NW) provide robust high optical gain and show beneficial Fabry-Pérot resonator properties allowing low threshold NW lasing at room temperature [Geburt et al, Nanotechnology 23, 365204 (2012)] and even continuous wave emission [Röder et al, Nano Letters 13, 3602 (2013)]. Yet, optical confinement in semiconductor NWs, and thus there size, is diffraction limited. However, plasmonic lasers using semiconductor NWs as gain medium have generated significant interest, since the optical mode size in these systems is far below the vacuum wavelength. By exploiting the natural non-linearity of the laser process itself, we investigate the laser dynamics of both photonic [Röder et al, Nano Lett. 15, 4637 (2015)] and plasmonic NW lasers [Sidiropoulos et al., Nat. Phys. 10, 870 (2014)], which are of high interest for concepts of ultrafast optical switching, nanosensing and nanospectroscopy.

HL 11.6 Mon 11:45 TRE Ma

Using plasmonic nanoantennas to read out the orbital angular momentum of light — •Richard M. Kerber[1], Jamie M. Fitzgerald[2], Sang Soon Oh[2], Ortwin Hess[2], and Doris E. Reiter[1,2] — [1]Institut für Festkörpertheorie, Universität Münster, Wilhelm-Klemm-Straße 10, 48149 Münster, Germany — [2]Department of Physics, The Blackett Laboratory, Imperial College London, South Kensington Campus, London SW7 2AZ, United Kingdom

The orbital angular momentum of light has recently been recognized as a new degree of freedom to encode information in communication technology. One way to determine the orbital angular momentum of light is to destroy the helical wavefront of the light beam. We here propose an alternative way to read out the orbital angular momentum of light using plasmonic nanoantennas by converting the phase information into spectral information without losing the phase properties of the beam. Using the excitation of bright and dark modes of a plasmonic nanoantenna, which exhibit different resonant wavelengths, the determination of the value of orbital angular momentum of the light beam becomes possible. Exemplary considering rotation-symmetrical nanorod antennas we show that their scattering cross-section is sensitive to the value of orbital angular momentum combined with the polarization of an incident orbital angular momentum light beam. For the simulation of the scattering cross-section we use the boundary element method and further predict the orbital angular momentum dependence of the excited modes with an analytical line antenna model.

HL 11.7 Mon 12:00 TRE Ma

Temporal characteristics of intense tunable pulses from an in-

frared free-electron laser — •Riko Kiessling, Sandy Gewinner, Wieland Schöllkopf, Martin Wolf, and Alexander Paarmann — Fritz-Haber-Institut der MPG, Berlin

For the generation of coherent radiation in the infrared region, the free-electron laser (FEL) posseses unique properties regarding intensity, wavelength tunability and short pulse durations. Covering the spectral range from 3 to 50 μm, the IR FEL at the Fritz Haber Institute is dedicated to (non-)linear spectroscopy of vibrational resonances in molecules, clusters and solid matter.

Here, we present the first cross-correlation measurements of the ps FEL pulses with a synchronized fs fiber oscillator via sum-frequency generation in nonlinear media. The observed FEL pulse intensity envelope confirms a lasing behavior typical for oscillator-type FELs [1]. Also, the influence of the FEL cavity detuning on the temporal shape, duration and fluctuation of the IR pulse is discussed. Using a differential cross-correlation scheme [2] will allow the exact determination of the absolute timing between accelerator-based and table-top optical pulses, which enables time-resolved studies of FEL-induced transient processes in solid-state systems.

[1] Knippels et al., PRL 83, 1578 (1999)
[2] Schulz et al., Nat. Commun. 6, 5938 (2015)

HL 11.8 Mon 12:15 TRE Ma

Tip modified fluorescence — •Jonas Albert and Markus Lippitz — Experimental Physics III, University of Bayreuth, Germany

In Near Field Microscopy the near field interaction is used to overcome the diffraction limit. We utilise the near-field interaction to modify fluorescence of our samples by the presence of the near-field probe, a metal coated AFM-tip. First experiments on CdSe nanocrystals show significant changes in the fluorescence lifetime dependent on the distance between crystal and a gold-tip.

Furthermore the optical fields at the near-field probe are confined in a space much smaller than the wavelength and therefore showing a high field gradient on the nanometer scale. We are using the optical field gradients to enhance dipole forbidden transitions in coherent excitonic systems, such as wire-like molecular aggregates.

HL 11.9 Mon 12:30 TRE Ma

Strongly Enhanced Mid-Infrared Second Harmonic Generation from Weak Optical Phonon Modes — •Alexander Paarmann, Ilya Razdolski, Sandy Gewinner, Wieland Schöllkopf, and Martin Wolf — Fritz-Haber-Institut, Berlin, Germany

Mid-infrared second harmonic generation (SHG) spectroscopy [1] provides a unique access to optical phonon resonances in the nonlinear-optical response of polar dielectrics, representing an alternativ spectroscopic approach to established techniques like infrared or Raman spectroscopy. Specifically, the SHG signal is sensitive not only to resonances in the nonlinear susceptibility, but also to the local electromagnetic fields determined by the linear optics. Here, we demonstrate how this intertwined sensitivity can be exploited for enhancement of the SHG specfically for weak oscillators.

Our experiments employ intense, tunable and narrowband mid-infrared pulses from a free-electron laser to acquire SHG excitation spectra from 4H-SiC with the c-axis of the uniaxial crystal in the surface plane [2]. Depending on the crystal azimuthal angle, we observe sharp enhancement of the SHG yield at the frequency of a weak zone-folded mode that exists in 4H-SiC due to the layer stacking along the c-axis. Perspectives are discussed on how to use this effect to detect periodic lattice distortions in strongly correlated insulators.

[1] Paarmann et al., Appl. Phys. Lett. 107, 081101 (2015), [2] Paarmann et al., Phys. Rev. B 94, 134312 (2016)

HL 11.10 Mon 12:45 TRE Ma

A near field study on the transition from localized to propagating plasmons on 2D nano-tips — •Thorsten Weber[1,2], Thomas Kiel[3], Stephan Irsen[2], Kurt Busch[3,4], and Stefan Linden[1] — [1]Physikalisches Institut, Universität Bonn, Nußallee 12, D-53115 Bonn, Germany — [2]Electron Microscopy and Analytics, Center of Advanced European Studies and Research, Ludwig-Erhard-Allee 2, D-53175 Bonn, Germany — [3]Institut für Physik, Humboldt-Universität Berlin, Newtonstraße 15, D-12489 Berlin, Germany — [4]Max-Born-Institut, Max-Born-Straße 2A, D-12489 Berlin, Germany

Plasmonic nano-structures have the unique capability to concentrate light in nanometric volumes. One approach to use this capability is based on resonant plasmonic nanostructures like rod nanoantennas or split ring resonators. In these resonant structures, hot spots of the electromagnetic field are created by localized particle plasmons. Another approach is utilizing non-resonant structures, such as nano-tips, on which propagating surface plasmons are excited at the wider end and travel towards the tip's apex.

Here, we report on a near field study of two-dimensional plasmonic gold nano-tips using electron energy loss spectroscopy in combination with scanning transmission electron microscopy, as well as discontinuous Galerkin time-domain calculations. With increasing nanotip size, we observe a transition from localized particle plasmons on resonant nano-tips to non-resonant propagating surface plasmons on large nano-tips. Furthermore we demonstrate that nano-tips with a groove cut can support both localized and propagating plasmons.