O 11: Plasmonics and Nanooptics II: Microscopy

Time: Monday 15:00–18:00 Location: S054

Invited Talk O 11.1 Mon 15:00 S054

Principles of plasmonic imaging — •Angela Demetriadou[1] and Alexei Kornyshev[2] — [1]Blackett Laboratory, Imperial College London, Prince Consort Road, SW7 2AZ, London, United Kingdom — [2]Department of Chemistry, Imperial College London, Prince Consort Road, SW7 2AZ, London, United Kingdom

Plasmonic imaging exploits the evanescent nature of propagating surface plasmon polariton (SPP) waves to produce real-time images of sub-wavelength objects with high-precision. It is commonly used in biological sciences to track and image organelles in cells, such as DNA, mitochondria and virus molecules. The fast dynamics of intra-cellular processes enforce to keep the cells under their native state (i.e. label-free) and to be imaged in real-time, establishing plasmonic imaging as a powerful tool for mapping and understanding cellular behaviour. Additionally, it has been widely used to map the electro-catalytic activity of single nanoparticles with high spatial resolution and sensitivity.

Our theoretical model describes the electromagnetic process that forms the plasmonic image, and accurately predicts the image properties for particles of any composition and size. The intensity and shape of the plasmonic image is dominated by the SPP-induced natural modes. Hence, through the theoretical model, spectroscopic information can be extracted from recorded plasmonic images, expanding the capabilities of current plasmonic imaging techniques.

O 11.2 Mon 15:30 S054

Imaging the dynamics of plasmonic vortices — •Deirdre Kilbane[1], Anna-Katharina Mahro[1], Stefan Mathias[1], Grisha Spektor[2], Lior Gal[2], Meir Orenstein[2], Bettina Frank[3], Simon Ristock[3], Harald Giessen[3], Philip Kahl[4], Daniel Podbiel[4], Frank Meyer zu Heringdorf[4], and Martin Aeschlimann[1] — [1]Physics Department and Research Centre OPTIMAS, University of Kaiserslautern, Germany — [2]Department of Electrical Engineering, Technion, Haifa, Israel — [3]Fourth Physics Institute and Research Center SCoPE, University of Stuttgart, Germany — [4]Faculty of Physics and CENIDE, University of Duisburg-Essen, Germany

The formation of a plasmonic vortex (rotational flow around a phase singularity) can be achieved by selecting the spin of circularly polarized light, and the geometry of the illuminated metallic structure. We perform near-field imaging of the ultrafast dynamics of plasmonic vortices using time-resolved two photon photoemission electron microscopy (TR-PEEM). A broadband ultrashort pulse laser excites and probes surface plasmon polaritons (SPPs) with 100 as time step and 40 nm spatial resolution. Here we observe the sub-optical cycle spatiotemporal evolution of the dynamics in plasmonic Archimedes spirals (PAS) and plasmonic vortex lenses (PVL). These structures were fabricated by focused ion beam (FIB) milling into the surface of thin polycrystalline gold films and single crystalline, atomically flat gold flakes.

O 11.3 Mon 15:45 S054

Improving the lateral near-field confinement in a nanofocusing SNOM — •Simon F. Becker[1], Martin Esmann[1], KyungWan Yoo[1,2], Petra Gross[1], Ralf Vogelgesang[1], NamkYoo Park[2], and Christoph Lienau[1] — [1]Carl von Ossietzky Universität, Oldenburg, Germany — [2]Seoul National University, Seoul, Korea

The lateral near-field confinement in apertureless scanning near-field optical microscopy (SNOM) experiments is typically governed by the tip radius of the probe used. Here, we discuss how to improve the confinement beyond this limitation using a nanofocusing SNOM taper.

We approach such a taper [1,2] to a thin, semitransparent gold film. Angle-resolved images of elastically scattered radiation transmitted through the film [3,4] show a steep increase in the detected signal over the last few nanometers. We compare the experimental results to finite element method simulations and find this steep increase to be a clear signature of gap plasmon formation in the gap between tip and sample. The lateral confinement of these fields is proportional to the geometric mean of tip-sample distance and tip radius. Hence, gap plasmons may enable decreasing the confinement to sizes typically found for interaction cross-sections of single quantum emitters. Combining this spectrally broadband effect with the background-free detection available with nanofocusing SNOM seems promising for nanospectroscopic investigations of dense and heterogeneous quantum emitter systems.

[1] M.I. Stockman, PRL 93, 137404 (2004); [2] S. Schmidt et al., ACS Nano 6, 6040 (2012); [3] M. Esmann et al., BJ Nano 4, 603 (2013); [4] S.F. Becker et al., (submitted).

O 11.4 Mon 16:00 S054

Coherent broadband nano-spectroscopy through plasmonic nanofocusing — •M. Esmann[1], S.F. Becker[1], H. Kollmann[1], J. Witt[1], K.W. Yoo[1,2], A. Chimeh[1], P. Gross[1], R. Vogelgesang[1], N.K. Park[2], and C. Lienau[1] — [1]Carl von Ossietzky Universität, Oldenburg, Germany — [2]Seoul National University, Seoul, Korea

Plasmonic nanofocusing microscopy enables broadband coherent light scattering spectroscopy with 5 nm spatial resolution. This is used to image optical near-fields around individual metallic nanoparticles. Conceptually, surface plasmon polaritons are launched by a grating coupler on a conical metallic taper and propagate towards the taper apex where they are transformed into highly confined near-fields [1-3]. Upon optical interaction with a sample, far-fields scattered from the apex are collected almost background free. Here, we use coherent white light to excite this isolated, spectrally broadband nano-lightsource at the apex of a monocrystalline gold taper. We then investigate both the spectral and spatial characteristics of optical near-fields around chemically synthesized gold nanorods. We find dipolar plasmon resonances with comparatively high Q-factors of up to 15. These are confirmed by far-field measurements. We simultaneously also image the corresponding near-field pattern in the spatial domain and achieve spatial resolution down to 5 nm. As these measurements are fully coherent, they can be straightforwardly extended to the investigation of coherences in biological or semiconducting systems.

[1] M.I. Stockman, PRL 93, 137404 (2004); [2] S. Schmidt et al., ACS Nano 6, 6040 (2012); [3] M. Esmann et al., BJNANO 4, 603 (2013).

O 11.5 Mon 16:15 S054

Probing and Controlling Electronic and Vibrational Coherences in Individual Carbon Nanotubes using Femtosecond Pulse Shaping Microscopy — •Veit Giegold[1,2], Richard Ciesielski[1,2], Nicolai F. Hartmann[3], Erik H. Haroz[3], Stephen K. Doorn[3], and Achim Hartschuh[1,2] — [1]Department Chemie and CeNS, LMU Munich, 81377 Munich — [2]Nanosystems Initiative Munich, 80799 Munich — [3]Los Alamos National Laboratory, New Mexico, 87545 United States

We probe and coherently manipulate the exciton population of individual semiconducting single-walled carbon nanotubes (SWCNTs) at room temperature using femtosecond laser pulse shaping microscopy [1,2]. In our experiment, the exciton state E_{11} of defect-doped (5,4)-SWCNTs is resonantly excited by a pair of phase-locked 20 fs laser pulses while the photoluminescence (PL) emission of the defect state E_{11}^D serves as reporter for the E_{11}-population. Tuning the pulse delay and relative carrier envelope phase results in damped PL intensity oscillations that can be described using the Bloch equations for a two-level system. We show that light absorption of individual SWCNTs can be controlled within their electronic dephasing time, ranging from 20 to 66 fs for different nanotubes. For longer pulse separations coherent radial breathing mode excitations with a period of 90 fs are observed.

[1] R. Hildner, D. Brinks, N.F. van Hulst, Nature Physics (2010).
[2] R. Ciesielski, V. Giegold, A. Hartschuh, et al., in prep.

O 11.6 Mon 16:30 S054

Confocal active interference scattering microscopy: A new approach to characterize single gold nanoparticles — •Otto Hauler — Institute of Physical Chemistry — Tübingen — Germany

Gold nanoparticles and their applications have attracted considerable research interest in recent times. By using confocal interference microscopy in combination with cylindrical vector beams it is possible to directly image the orientation and to detect the shape of single metal nanoparticles, with sizes well beyond the diffraction limit [1-4]. We present a newly developed method to further investigate the properties of these promising materials, the confocal active interference scattering microscope. This novel technique allows the measurement of the phase of the elastically scattered light. It furthermore enables the control of the excitation polarization, through the use of radially- and azimuthally-polarized laser modes. [1] A.V. Failla, H. Qian, H. Qian, A. Hartschuh, A. J. Meixner, Nano Lett. (2006), 6: 1374. [2]

F. Wackenhut, A.V. Failla, A.J. Meixner, Phys. Chem. Chem. Phys. (2013),15: 5407-5414. [3] F. Wackenhut, A.V. Failla, A.J. Meixner, Anal Bioanal Chem (2015), 407: 4029-4034. [4] F. Wackenhut, A.V. Failla, T. Züchner, M. Steiner, A.J. Meixner, Appl. Phys. Lett. (2012), 100: 263102.

O 11.7 Mon 16:45 S054

Polarization sensitive scanning near field optical microscope on polymer thin films — •Jens Brauer[1], Jinxin Zhan[1], Petra Gross[1], Christoph Lienau[1], Daniel Trefz[2], and Sabine Ludwigs[2] — [1]Carl von Ossietzky Universität, Oldenburg, Germany — [2]Institut für Polymerchemie, Stuttgart, Germany

Currently there is a high need to investigate the relationship between structure and functionality of novel, polymer-based organic solar cell materials. Specfically information about local fluctuations of their optical properties, largely affecting the charge carrier mobility is needed [1]. For this, an optical imaging method is required with a spatial resolution in the order of the domain size, typically a few nanometers, or even below.

Here we present a technique to use near field optical microscopy to fulfill these requirements. We use a Titanium:Sapphire laser focused through the sample onto a sharp gold tip and collect the scattered near-field signal from the tip in a backscattering geometry. By applying a modulation to the tip with tens of kilohertz and using a lock-in amplifier after detection to demodulate the signal at the n-th harmonic we can significantly increase the signal to noise ratio. Changing the direction of the linear laser polarization enables us to determine the molecular orientation with a resolution of about 10 nm. In this talk we present a first demonstration by applying the technique to ordered and unordered thin film polymer samples.

[1] Brinkmann, M. et al.; ACS Nano 6, pp 10319-10326 (2012)

O 11.8 Mon 17:00 S054

Nanoscale probing of optical near-fields by ultrafast transmission electron microscopy — •Armin Feist, Katharina E. Echternkamp, Murat Sivis, Sascha Schäfer, and Claus Ropers — 4th Physical Institute, University of Göttingen, Göttingen, Germany

Ultrafast transmission electron microscopy (UTEM) allows for the study of structural and electronic dynamics on nanometer length scales [1], as well as for the local probing of optical near-fields [2].

Here, we employ the inelastic scattering of swift electrons to characterize optical near-fields in metallic nanostructures. The experiments utilize a novel short-pulsed electron gun driven by localized photoemission from a nanotip emitter, yielding electron probes with a pulse duration of 300 fs and beam diameters down to 1.5 nm.

In the interaction with optical near-fields, the energy spectra of free electrons develop into a comb of spectral sidebands representing the absorption and emission of multiple photons [2]. The quantum coherence of this process is evidenced by the observation of multilevel Rabi oscillations in the sideband populations [3]. The interaction facilitates few nanometer spatial resolution in near-field imaging, as demonstrated by raster-scanning the focused electron probe across resonantly excited plasmonic nanostructures with feature sizes down to 5 nm.

[1] A.H. Zewail, Science, **328**, 187 (2010).
[2] B. Barwick *et al.*, Nature, **462**, 902 (2009).
[3] A. Feist *et al.*, Nature, **521**, 200 (2015).

O 11.9 Mon 17:15 S054

Coherent spectroscopy of single metallic nanostructures — •Martin Silies[1], Heiko Kollmann[1], Martin Esmann[1], Julia Witt[2], Gunther Wittstock[2], and Christoph Lienau[1] — [1]AG Ultraschnelle Nano-Optik, Institut für Physik, Carl von Ossietzky Universität Oldenburg, Germany — [2]AG Wittstock, Institut für Chemie, Carl von Ossietzky Universität Oldenburg, Germany

Metallic nanostructures exhibit strong optical resonances and enhanced optical near-fields, enabling the transfer of far-field radiation onto subwavelength scales. Even so, the mismatch of the optical absorption or scattering cross-section of a single nanostructure and the free space wavelength makes far-field spectroscopic investigations challenging. Further, single nanostructures with geometric dimensions of less than 50nm require highly sensitive and almost background-free spectroscopic methods. For this, modulation-based methods such as focus or Spatial Modulation Spectroscopy (SMS)[1] have been proven to be able to quantitatively measure the scattering and absorption cross section simultaneously [2]. Here, we present a combined approach of a commonly used confocal SMS setup and broadband VIS-IR Fourier Transform spectroscopy to measure the extinction cross section of single nanostructures in the time domain. We show polarization-resolved spectra of single chemically synthesized gold nanorods resonant in the near infrared. An extension of the approach to the study of hybrid nanostructures and to time-resolved phenomena is discussed.

[1] A. Arbouet et al., PRL 93, 127401 (2004)
[2] M. Husnik et al., PRL 109, 233902 (2012)

O 11.10 Mon 17:30 S054

Plasmon-mediated circularly polarized luminescence of GaAs in a scanning tunneling microscope — Svenja Mühlenberend, Markus Gruyters, and •Richard Berndt — Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, 24098 Kiel, Germany

The electroluminescence from *p*-type GaAs(110) in a scanning tunneling microscope has been investigated at 6 K. Unexpectedly high degrees of circular polarization have often been observed with ferromagnetic Ni tips and also with paramagnetic W and Ag tips. The data is interpreted in terms of two distinct excitation mechanisms. Electron injection generates intense luminescence with low polarization. Plasmon-mediated generation of electron-hole pairs leads to less intense emission, which, however, is highly polarized for many tips.

O 11.11 Mon 17:45 S054

Plasmonic Activation of Platinum Clusters for Photocatalytic Reactions Detected by STM — Sarah Wieghold[1], Lea Nienhaus[2,3], Marian D. Rötzer[1], Fabian Knoller[1], Florian F. Schweinberger[1], Joseph W. Lyding[2], Ulrich Heiz[1], Martin Grübele[2,3], and •Friedrich Esch[1] — [1]Chemistry Dept. and CRC, TU München, Lichtenbergstr. 4, 85748 Garching, Germany — [2]Beckman Institute, University of Illinois, Urbana — [3]Department of Chemistry, University of Illinois, Urbana, Illinois 61801

Their unique structural, optical and electronic properties make small metal clusters prime candidates for catalytic applications, especially under mild reaction conditions such as in photocatalysis. Due to their small cross sections for the interaction with light and due to low surface coverages, an indirect, support-mediated photoactivation mechanism is most efficient. We imaged this activation at the level of individual $Pt_{>35}$ clusters supported on a thin, structured gold film. The film shows a strong plasmonic interaction with visible light that is optimized to work in a back-illumination geometry. We used a scanning tunneling microscope to map the tunneling current modulation induced by light at 532 nm. When tunneling into unoccupied states of the film, we detect a light-induced current increase that is enhanced on the clusters. In this way, we image the plasmonic coupling of the clusters to the gold support with nanometer resolution. This activation leads indeed to an enhanced catalytic activity, as we demonstrate for the oxidative decomposition of methylene blue.