O 93: Plasmonics & Nanooptics VI: Near-Field Microscopy and Phenomena

Time: Friday 10:30–13:00 Location: H8

O 93.1 Fri 10:30 H8

Electron near-field circular dichroism — •Tyler Harvey, Jan-Wilke Henke, Ofer Kfir, and Claus Ropers — IV. Physical Institute: Solids and Nanostructures, University of Göttingen, Germany

Circular dichroism spectroscopy with visible light and x-rays has long been used to characterize chiral-structured materials, magnetic materials, and chiral electronic transitions. In this presentation, we demonstrate a nanometer-resolution circular dichroism technique that employs electrons to probe optical near fields. Electron near-field circular dichroism offers nanoscale insight into chirality.

Electrons can absorb or emit integer multiples of the photon energy from an optical field adjacent to a surface [1,2]. The strength of the coupling between the electron momentum and the optical field amplitude depends on the shape and optical properties of the surface. Because electron beams can be focused to sub-nanometer spots in modern electron microscopes, this interaction, called photon-induced near-field electron microscopy (PINEM) can be employed to image plasmonic modes and optical properties with nanometer spatial resolution. By illuminating a sample with left- and right-circularly polarized light and measuring the difference in coupling strength with electrons, we probe chiral optical near fields with nanometer spatial resolution. This technique may enable the investigation of chiral optical and electronic states in plasmonic nanostructures, molecules and atoms with sub-nanometer spatial resolution.

[1] B. Barwick et al., Nature 462 (2009) 902.
[2] A. Feist et al., Nature 521 (2015) 200.

O 93.2 Fri 10:45 H8

Vectorial near-field coupling — Martin Esmann[1,2], Simon F. Becker[2], Julia Witt[2], •Anke Korte[2], Abbas Chimeh[2], Jinxin Zhan[2], Jinhui Zhong[2], Ralf Vogelgesang[2], Gunther Wittstock[2], and Christoph Lienau[2] — [1]CNRS Centre de Nanosciences et de Nanotechnologies (C2N), 91220 Palaiseau, France — [2]Carl von Ossietzky University, 26111 Oldenburg, Germany

The coherent exchange of optical near-fields between neighboring dipoles is essential for the optical properties, quantum dynamics and thus for the function of many naturally occurring and artificial nanosystems[1,2]. These interactions are inherently nanometer-ranged and depend sensitively on relative orientation, spectral detuning and dephasing, i.e., on the vectorial properties of the coupled dipolar near-fields. This makes them challenging to analyze experimentally.

Here, we introduce plasmonic nanofocusing[3] spectroscopy to record coherent light scattering spectra with 5-nm spatial resolution[4] from a small dipole antenna, excited solely by evanescent fields and coupled to plasmon resonances in a single gold nanorod. We resolve mode couplings, resonance shifts and Purcell effects as a function of dipole alignment, and show how they arise from different vectorial components of the interacting near-fields. Our results pave the way to control optical properties and function of nanoscale systems by dipolar alignment.

[1] Zhang, Y. et al., Nature 531, 623 (2016).
[2] Scholes, G.D., et al., Nature Chemistry 3, 763 (2011).
[3] Stockman, M.I., PRL 93, 137404 (2004).
[4] Esmann, M., et al., arXiv:1801.10426 (2018).

O 93.3 Fri 11:00 H8

Anisotropic scattering from gold SNOM tips and its role in the near-field light scattering spectroscopy of single nanoparticles — •Abbas Chimeh[1], Anke Korte[1], Jinxin Zhan[1], Jinhui Zhong[1], Martin Esmann[2], Nahid Talebi[3], and Christoph Lienau[1] — [1]Universität Oldenburg — [2]Centre de Nanosciences et de Nanotechnologies, Paris — [3]Max-Planck Institute for Solid State Research, Stuttgart

Plasmonic nanofocusing of light using sharp conical gold tapers enables broadband coherent spectroscopy of optical near-fields around single nanoobjects with unprecedented spatial resolution . Such measurements probe how the coupling between optical near fields of tip and sample affects the light scattering spectra from the nanofocusing gold tip. Interestingly, they reveal a coupling of the sample near-fields to both longitudinal and transversal plasmonic resonances of the tip[1], allowing to unveil the vectorial nature of the near-field coupling. To understand these couplings, knowledge about the transverse resonance, corresponding to charge oscillations perpendicular to the tip axis, are needed. Here, we employed evanescent fields at a prism surface to isolate the transverse tip resonance. By polarization resolved scattering measurements, we proved the anisotropic scattering from the tip apex comprising a longitudinal broadband dipole resonance at ~800 nm and a transversal narrowband resonance at ~550 nm. This leads to a more realistic model of tip-sample coupling proposes a novel technique for broadband near-field spectroscopy in the visible spectral range.

[1] M. Esmann et al., arXiv:1801.10426 (2018).

O 93.4 Fri 11:15 H8

Polarization in scattering near-field optical microscopy at resonant excitation — •Hamed Aminpour[1], Susanne Kehr[1], Michael Klopf[2], and Lukas Eng[1] — [1]Technische Universität Dresden, Germany — [2]Helmholtz-Zentrum Dresden-Rossendorf, Germany

Scattering-type near-field optical microscopy (s-SNOM) is a promising technique that allows to obtain optical information of samples on the nanometer length scale. Specifically, the response of molecular vibrations, phonons, and excitons in s-SNOM is particularly sensitive to the polarization direction of the incident optical field. To date, most published works focus on out-of-plane polarized s-SNOM at non-resonant excitation, only. Recently, however, excitation and detection of both in- and out-of-plane polarized local fields was demonstrated, by breaking the axial symmetry of the near-field probe [1].

In this presentation, we investigate an elegant alternative in order to achieve in-plane polarized resonant sample excitation of mid-infrared phonon modes [2]. To backup our experiments, we report here on a COMSOL simulation of the scattered near field, investigated by varying the following parameters: Angle and polarization direction of the incident light; tip-sample distance; sample permittivity as a function of wavelength. We compare these results with polarization-dependent s-SNOM measurements on $SrTiO_3$ in the spectral range from 13 to 16 μm, demonstrating the large potential of polarization-sensitive s-SNOM for the nanoscopic material analysis.

[1] K.-D. Park et al., Nano Lett. 18, 2912(2018)
[2] S. C. Kehr et al., Synchrotron Rad. News 30, 31(2017)

O 93.5 Fri 11:30 H8

Polarization-dependent resonant near-field spectroscopy in the mid- to far-infrared regime — •Lukas Wehmeier[1], Tobias Nörenberg[1], Denny Lang[2], Susanne C. Kehr[1], and Lukas M. Eng[1] — [1]Technische Universität Dresden, Germany — [2]Helmholtz-Zentrum Dresden-Rossendorf (HZDR), Germany

Resonant infrared (IR) near-field spectroscopy provides a highly material-specific response with sub-wavelength spatial resolution of ∼10 nm. Here, we provide the infrared near-field response of selected paraelectric and ferroelectric materials (i.e. $SrTiO_3$, $LiNbO_3$, and $BiFeO_3$) that we investigated via scattering scanning near-field optical microscopy (s-SNOM), using the mid- to far-infrared free-electron laser (FEL) at HZDR. For these materials, we demonstrate that resonant near-field excitation is possible for both s- and p-polarized incident IR beams, which is particularly interesting for the nanoscopic investigation of anisotropic materials. We explored, for instance, the different near-field resonances of $BiFeO_3$, finding characteristic s-SNOM enhancement for three consecutive phonon modes in the 10 to 60 μm wavelength range. Utilizing the same material, we present the first realization of a superlens within the so-called THz gap, demonstrating a fascinating application of sub-wavelength imaging capabilities.

O 93.6 Fri 11:45 H8

Comparison of two approaches to near field spectroscopy on a Sb2S3 film — •Jinxin Zhan[1], Jens Brauer[1], Wei Wang[2], Lukas Schmidt-Mende[2], Petra Gross[1], and Christoph Lienau[1] — [1]Ammerländer Heerstr. 114-118, Oldenburg — [2]Universitätsstr. 10, 78457 Konstanz

Fundamental understanding of structure and function of nanomaterials is of great significance for investigating their optical and electric properties, and for further applications in corresponding fields. Optical spectroscopy on a nanometer length scale is one promising approach to obtain such insights. Here we employ scattering-type near-field scanning optical microscopy (NSOM) to probe the local spectrum of a Sb2S3 film, a very promising candidate for solar cell applications due to its high absorption coefficient and suitable bandgap. In our NSOM

setup, the near field gold probe, is modulated with a frequency of 27 kHz to suppress background by demodulating the scattered optical signal at the second or third order harmonics. We measure the near field spectrum by two approaches. First, the tip is excited by a broadband pulse laser and the scattered light is recorded by a monochromator and a fast camera with up to 210 kHz line readout. Alternatively, Fourier transform spectroscopy combined with a lock-in amplifier is employed. We perform a comprehensive comparison between two methods and discuss the local spectrum information drawn from the near field spectra.

O 93.7 Fri 12:00 H8

Revisiting the Dipole Model for a Thermal Infrared Near-Field Spectroscope — •Florian Herz[1], Zhenghua An[2], Susumu Komiyama[3], and Svend-Age Biehs[1] — [1]Institut für Physik, Carl von Ossietzky Universität, 26111 Oldenburg, Germany — [2]State Key Laboratory of Surface Physics and Key Laboratory of Micro and Nano Photonics Structures (Ministry of Education), Department of Physics, Fudan University, Shanghai 200433, Peoples Republic of China — [3]Department of Basic Science, The University of Tokyo, Komaba 3-8-1, Meguro-ku, Tokyo 153-8902, Japan

I will present our work on determining the scattered near-field and directly emitted power of a heated spherical nanoparticle above a sample within the framework of fluctuational electrodynamics using the dipole approximation. Additionally, for a configuration of a nanoparticle heated with respect to its environment, I show that the scattered power of the near field of the sample is strictly zero. Hence, applied on the interpretation of near-field imaging setups like thermal infrared near-field spectroscopy not only the scattering of the near field but also the direct emission of the tip has to be considered.

Reference: Florian Herz, Zhenghua An, Susumu Komiyama, and Svend-Age Biehs: Revisiting the Dipole Model for a Thermal Infrared Near-Field Spectroscope, in Phys. Rev. Applied 10, 044051 (2018).

O 93.8 Fri 12:15 H8

Hyperspectral nano-imaging applied to s-SNOM enabled by compressed sensing — •Georg Ulrich[1], Bernd Kästner[1], Franko Schmähling[1], Andrea Hornemann[1], Arne Hoehl[1], Mattias Kruskopf[1], Klaus Pierz[1], Markus B. Raschke[2], Gerd Wübbeler[1], and Clemens Elster[1] — [1]Physikalisch-Technische Bundesanstalt, Braunschweig and Berlin, Germany — [2]Department of Physics, Department of Chemistry, and JILA, University of Colorado, Boulder, 80309, USA

Scattering-type scanning near-field optical microscopy (s-SNOM) enables to circumvent the diffraction limit known from classical optics. Ultra-broadband synchrotron radiation from the Metrology Light Source (MLS) provides infrared-radiation suited for performing nano-FTIR spectroscopy [1]. However, for many applications such as mapping of biological samples hyperspectral imaging is required, resulting in a large number of spectra and therefore long acquisition time. Here we will present results from applying compressed-sensing, providing both rapid and sensitive spatio-chemical nano-imaging [2]. By reducing the number of sampling points to $1/9^{\text{th}}$ we are on the route for further enhancing the compression rate to record large data cubes. [1] P. Hermann, et al., Opt. Express. 22, 17948 (2014) [2] B. Kästner, et al., Opt. Express. 26, 18115 (2018)

O 93.9 Fri 12:30 H8

Near-Field Spectroscopy of Nanoscale Molecular Aggregates — Xing Gao and •Alex Eisfeld — MPI-PKS Dresden

When molecules are assembled into an aggregate, their mutual dipole-dipole interaction leads to electronic eigenstates that are coherently delocalized over many molecules. Knowledge about these states is important to understand the optical and transfer properties of the aggregates. Optical spectroscopy, in principle, allows one to infer information on these eigenstates and about the interactions between the molecules. However, traditional optical techniques using an electromagnetic field which is uniform over the relevant size of the aggregate cannot access most of the excited states because of selection rules.

We demonstrate that by using localized fields one can obtain information about these otherwise inaccessible states. As an example, we discuss in detail the case of local excitation via radiation from the apex of a metallic tip, which allows also scanning across the aggregate.The resulting spatially resolved spectra provide extensive information on the eigenenergies and wave functions.

[1] J. Phys. Chem. Lett. 9, 6003 (2018)

O 93.10 Fri 12:45 H8

Limitations of the kinetic theory to describe the near-field heat exchanges in many-body systems — •Christoph Kathmann[1], Riccardo Messina[2], Philippe Ben-Abdallah[2], and Svend-Age Biehs[1] — [1]Institut für Physik, Carl von Ossietzky Universität, Oldenburg, Deutschland — [2]Institut d'Optique, CNRS, Université Paris-Saclay, Palaiseau, Frankreich

Kinetic theory based on solving Boltzmann equation is widely used to calculate heat fluxes in complex many-body systems on the nanoscale[1]. We compare this approach to an exact Landauer approach based on fluctuating electrodynamics[2] for a simple many-body system, namely a onedimensional chain consisting of nanoparticles. We find that the kinetic approach generally fails to predict heat fluxes in such a system, for polar nanoparticles we observe poor spectral agreement and a different scaling with the distance between the particles while for metallic nanoparticles the kinetic approach completely fails[3]. References: [1] J. Ordonez-Miranda, L. Tranchant, S. Gluchko, und S. Volz, Energy transport of surface phonon polaritons propagating along a chain of spheroidal nanoparticles, Phys. Rev. B 92, 115409 (2015). [2] R. Messina, M. Tschikin, S.-A. Biehs, und P. Ben-Abdallah, Fluctuation-electrodynamic theory and dynamics of heat transfer in systems of multiple dipoles, Phys. Rev. B 88, 104307 (2013). [3] C. Kathmann, R. Messina, P. Ben-Abdallah, S.-A. Biehs, Limitations of kinetic theory to describe near-field heat exchanges in many-body systems, Phys. Rev. B 98, 115434 (2018).