HL 43: Plasmonics and Nanooptics V: Light-Matter Interaction

Time: Tuesday 14:00–16:00 Location: TRE Ma

HL 43.1 Tue 14:00 TRE Ma

Scanning near-field optical microscopy with inline-homodyne detection — •Jens Brauer, Jinxin Zhan, Petra Gross, and Christoph Lienau — Carl von Ossietzky University, Oldenburg, Germany

Apertureless near-field optical microscopy is a good choice when it comes to optically investigating nanostructures with a size of only a few nanometer and is nowadays widely used in different geometries.

For most of the setups there remains the challenge of distinguishing the desired near-field signal from the very large optical background. To overcome this problem the tip-sample distance often is modulated at some tens of kilohertz and the detection signal is demodulated at higher harmonics. As Knoll & Keilmann [1] have shown theoretically the ratio of near-field to optical background improves with increasing demodulation frequency. However, also the signal amplitude at higher harmonics is decreasing dramatically. Therefore the use of interferometers, either in a homodyne or heterodyne detection scheme, is often used to amplify the near-field signal [2].

As a downside the contrast in the SNOM images critically depends on the stability of the interferometer. Here we present how to disentangle near field from background radiation in the 1st to 4th harmonic signal by measuring approach curves in an inherently stable in-line homodyne detection scheme and we give an outlook to ongoing spectroscopically resolved SNOM measurements.

[1] B. Knoll & F.Keilmann, Optics Communications, 182(4) (2000)
[2] Ocelic et al.,Applied Physics Letters, 89(10) (2006)

HL 43.2 Tue 14:15 TRE Ma

Vectorial near-field coupling on the nano scale — •M. Esmann, S.F. Becker, J. Witt, G. Wittstock, R. Vogelgesang, and C. Lienau — Carl von Ossietzky Universität, 26111 Oldenburg, Germany

Dipole-dipole coupling is ubiquitous in nanoscale systems [1,2] leading to optical modes and coherent dynamics which sensitively depend on dipole configuration. To simultaneously study such systems on 5nm length scales and over a wide spectral range, we implemented a novel type of Scanning Near-Field Optical Microscope (SNOM): Grating-coupled Surface Plasmon Polaritions (SPPs) are adiabatically nanofocused [3] at the 10nm sized apex of a metallic nanotaper resulting in a bright, spatially isolated and spectrally broad nano light source, which acts as a probe for background-free near-field spectroscopy.

Here, we apply this technique to a prototypical system formed by dipolar coupling of the nanotaper apex to small 10×40nm gold nanorods. Upon systematic variation of coupling strength we find clear signatures of coupling-induced spectral shifts and broadening of plasmonic resonances with dramatic variations on few-nanometer length scales. We argue that our approach presents a fundamentally new way to interrogate dipole-dipole coupling in nanosystems in the spatial-, spectral- and temporal domain providing full access to coupling energies, mode profiles and the associated coherent dynamics.

[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., BJNANO 4, 603 (2013). [5] Becker, S.F. et al., ACS Photonics 3, 223 (2016).

HL 43.3 Tue 14:30 TRE Ma

Modulation of extraordinary optical transmission through nanohole arrays using ultrashort laser pulses — Kellie Pearce[1,2], Benjamin Duschner[1], Robin Dehde[1], Florian Richter[1], and •Ulf Kleineberg[1,2] — [1]Ludwig-Maximilians-Universität München, Am Coulombwall 1, 85748 Garching, Germany — [2]Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Str. 1, 85748 Garching, Germany

We perform pump-probe measurements of the transmission through subwavelength hole arrays in a gold film on ITO using 16-fs laser pulses. By tuning the delay between the pulses, we can modify the contribution from different transmission resonances, and observe plasmon dephasing and decay. Simulations using a 3D FDTD solver show that the modulation of the transmission is due to the change in the intensity and distribution of fields on both interfaces.

HL 43.4 Tue 14:45 TRE Ma

Non-linear imaging of individual plasmonic nanostructures using phase-shaped femtosecond laser pulses — •Veit Giegold, Richard Ciesielski, Alexander Biewald, Tobia Mancabelli, Alberto Comin, and Achim Hartschuh — Department Chemie and CeNS, LMU Munich, 81377 Munich

We studied the second harmonic (SH) and near-degenerate four-wave (FWM) mixing response of individual plasmonic nanostructures using a confocal femtosecond pulse shaping setup. We find that the two different signals are maximized for different spectral phases of the laser pulse. We attribute this to different phase terms in the second- and third-order susceptibility of the nanostructures, respectively. We show that by using optimized phase profiles, the contrast of confocal SH and FWM images can be manipulated. This phase-enhanced nonlinear imaging can provide further insight into properties of plasmonic nanostructures.

HL 43.5 Tue 15:00 TRE Ma

Modeling of higher harmonic generation with the Fourier modal method using adaptive coordinates — •Josselin Defrance, Martin Schäferling, Maxim L. Nesterov, and Thomas Weiss — 4th Physics Institute and Research Centers SCoPE, University of Stuttgart, Germany

Metallic nano-structures can concentrate light into sub-wavelength volumes resulting in strong nonlinear responses. The influence of the geometry of such nano-systems, however, is not fully understood. Modeling these effects will not only help to optimize the nonlinear response of plasmonic nano-structures, but will also provide a better understanding of these phenomena. Nowadays, different numerical methods to model the interaction of electromagnetic fields with matter exist. The so-called Fourier modal method offers a fast and highly accurate calculation of far-field responses.

It has been shown that the Fourier modal method can be extended in order to calculate the generation of higher harmonics [1]. We have combined this method with adaptive spatial resolution and adaptive coordinates [2], thus providing an efficient and accurate numerical method for the precise analysis of the nonlinear optical responses of complex plasmonic geometries.

[1] T. Paul et al., J. Opt. Soc. B, Vol. 27, Issue. 5, pp. 1118 (2010).
[2] T. Weiss et al., Opt. Express 17, 8051 (2009).

HL 43.6 Tue 15:15 TRE Ma

Tracking Plasmon Propagation by Terahertz-Streaking at Metal Nanotips — •Lara Wimmer, Benjamin Schröder, Murat Sivis, Georg Herink, and Claus Ropers — IV. Physical Institute - Solids and Nanostructures, University of Göttingen, Germany

We resolve the propagation time of surface plasmon polaritons (SPP) traveling along the shaft of gold nanotips [1] employing Terahertz (THz) near-field streaking spectroscopy [2, 3]. Surface plasmon polaritons are launched by femtosecond near-infrared (NIR) pulses in a grating coupler at the shaft of the nanostructure, 50 *m from the tip end. At the apex, the nanofocused SPPs induce the (multiphoton) emission of photoelectrons [1], which are streaked in the locally enhanced near-field of a THz transient incident on the tip. Reference spectrograms are obtained by direct NIR-excitation of the apex. The observed temporal shift between both streaking spectrograms provides a direct measure of the group delay [4] in SPP propagation from the grating to the tip apex.

[1] Schröder et al., Phys. Rev. B 92, 085411 (2015).
[2] Wimmer et al., Nature Physics 10, 432-436 (2014).
[3] Herink et al., New Journal of Physics 16, 123005 (2014).
[4] Kravtsov et al., Opt. Lett. 38, 1322-1324 (2013)

HL 43.7 Tue 15:30 TRE Ma

Relaxation dynamics of plasmonic hot-carriers in gold nanoparticles — •Emanuele Minutella[1,2], Florian Schulz[1], and Holger Lange[1,2] — [1]Physikalische Chemie, Universität Hamburg, Germany — [2]The Hamburg Centre for Ultrafast Imaging, CUI

Using light to drive or control chemical or physical processes is a very interesting field of research. One problem is to guide the light's energy to the specific place where reaction takes place. Due to their plasmonic properties noble metal nanoparticles have the ability to collect very efficiency photons and convert them into hot-electron-hole-pairs. As a result of size and photon energy dependent transient absorption measurements we found a binary kinetic for the relaxation times of the hot electrons. Depending on the photon energy either interband or intraband excitations are possible. We found faster decay rates for the intraband relaxation than for the interband. These results can help to improve possible hot-carrier devices.

HL 43.8 Tue 15:45 TRE Ma

Coherent phase-resolved near-field spectroscopy of single gold nanorods enabled by self-interference in an ultrasharp gold taper — Martin Esmann, Simon F. Becker, •Abbas Chimeh, Ralf Vogelgesang, and Christoph Lienau — University of Oldenburg

Plasmonic nanofocusing of light on an ultrasharp gold taper enables spectroscopic analysis of optical near-fields around individual metallic nanoparticles [1]. In order to characterize the ultrafast time-dynamics of these near-fields, the scheme should be extended to include the measurement of phase-resolved spectra. To this end, a stable inline interferometer is implemented in such a taper and plasmonic-nanofocusing spectroscopy is combined with spectral interferometry. In this method, surface plasmon polaritons are launched by a coupler on a pyramidal metallic taper and propagate towards the taper apex where they form a confined nanometer-scale light source [1]. After optical interaction with a sample, a part of the incident field is reflected backwards to the taper and interferes with the incident field on it. The interference signal is then collected from a scatterer on the shaft and spectrally resolved. Here, we implemented this method for the first time and use it to record local broadband optical spectra of single gold nanorod with 10x40 nm dimensions. We demonstrate phase-coherent mapping of optical near-field with sub-10 nanometer spatial resolution and provide a detailed analysis how near-field spectra are generated in this novel and highly sensitive interferometric measurement scheme.

[1] S. Schmidt et al., ACS Nano 6, 6040 (2012).