O 11: Ultrafast and Nonlinear Plasmonics

Time: Monday 15:00–18:00 Location: MA 004

O 11.1 Mon 15:00 MA 004

Surface plasmon polariton induced shortening of the optical response in perforated gold films — •Felix Spitzer[1], Martin Pohl[1], Boris A. Glavin[2], Vladimir Belotelov[3], Ilya Akimov[1], Sachin Kasture[4], Arvind S. Vengurlekar[4], Achanta V. Gopal[4], and Manfred Bayer[1] — [1]Technische Universität Dortmund, Dortmund, Germany — [2]V.E. Lashkaryov Institute of Semiconductor Physics, Kiev, Ukraine — [3]Lomonosov Moscow State University, Moscow, Russia — [4]Tata Institute of Fundamental Research, Mumbai, India

Ultrafast relaxation dynamics of a laser-heated periodically perforated gold film are investigated using a femtosecond pump-probe technique with 30 fs pulses far from intrinsic gold resonances. The grating period is 595 nm with a thickness of 120 nm and 110 nm slit width. Changes of differential transmission are measured time- and angular-resolved and fitted afterwards using the nonequilibrium electron model to obtain the characteristic relaxation times. We observe, that the pump induced optical response becomes significantly shorter when the electrons in metal are excited with surface plasmon polaritons. These results uncover the role of thermal conductivity of electrons which becomes important in case of strongly inhomogeneous spatial distribution of hot carriers.

O 11.2 Mon 15:15 MA 004

Probing electron relaxation in single metal nanotips with terahertz-induced field emission — •Lara Wimmer, Georg Herink, Sergey V. Yalunin, and Claus Ropers — 4. Physical Institute - University of Göttingen, Göttingen, Germany

We implemented a streaking-type photoemission experiment at individual metal nanotips [1], employing single-cycle terahertz (THz) transients and femtosecond near-infrared (NIR) pulses. In a first scenario, the photoelectrons are generated by the NIR pulses and subsequently accelerated in the local THz electric field. Due to the high confinement of the THz-induced near-field at the tip apex, the photoelectrons leave the electric field in much less than one optical half-cycle [2]. This allows for a phase-resolved characterization of the THz near-field and for a control of the photoelectron kinetic energy spectra.

In a second set of measurements, we increase the local THz field strength to above 3 V/nm, inducing THz field emission [3]. Slightly below the threshold for cold field emission, the THz transient triggers the emission of NIR-excited hot electrons in the metal. The electron relaxation is quantitatively characterized by varying the relative time delay between the two pulses, and we find direct evidence for a confinement-induced prolongation of cooling times as compared with planar surfaces.

[1] Wimmer et al., Nature Physics 10, 432-436 (2014).
[2] Herink et al., Nature 483, 190-193 (2012).
[3] Herink et al., New Journal of Physics, in press (2014).

O 11.3 Mon 15:30 MA 004

Transient absorption spectroscopy on gold nanorods coated with molecular J-aggregates — •Thomas Simon[1,2], Dzimitry Melnikau[3], Alexander Urban[1,2], Jochen Feldmann[1,2], and Yury Rakovich[3] — [1]Chair for Photonics and Optoelectronics, Ludwig-Maximilians-Universität München, Amalienstr. 54, 80799 Munich, Germany — [2]Nanosystems Initiative Munich (NIM), Schellingstr. 4, 80799 Munich, Germany — [3]Centro de Física de Materiales (MPC, CSIC-UPV/EHU), Donostia International Physics Center (DIPC), Po Manuel de Lardizabal 5, Donostia-San Sebastian 20018, Spain

We have performed femtosecond transient absorption spectroscopy on cyanine based molecular J-aggregates covering the surface of gold nanorods. By changing the aspect ratio of the rods or choosing J-aggregates with different HOMO-LUMO transition energies, the coupling between excitonic and plasmonic resonances can be tuned and controlled. We have analyzed the nonlinear optical responses of purely excitonic, purely plasmonic and hybrid excitonic-plasmonic transitions. The transient behavior of the absorption changes at the hybrid transition frequencies gives further insight into coupling mechanisms between excitons and plasmons.

O 11.4 Mon 15:45 MA 004

Short-range surface plasmonics on atomically flat thin single gold nanocrystals: Electron emission from a 60 nm spot at 800 nm wavelength — •Bettina Frank[1], Philip Kahl[2], Thomas Weiss[1], Liwei Fu[3], Michael Horn von Hoegen[2], Frank Meyer zu Heringdorf[2], and Harald Giessen[1] — [1]4th Physics Institute and Research Center SCoPE, University of Stuttgart, Germany — [2]Faculty of Physics and Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, Duisburg, Germany — [3]Institute of Applied Optics, University of Stuttgart, Germany

We observe short-range surface plasmons in atomically flat single gold nanocrystals with a thickness in the 20-40 nm range. The crystals are grown electrochemically on atomically flat silicon wafers and structured with FIB-milled patterns on the surface. Sub-20 fs laser pulses at 800 nm in normal incidence geometry excite long-range as well as short-range surface plasmons. The latter ones possess plasmon wavelengths in the 140-180 nm range and exhibit considerably reduced propagation velocity. Using PEEM we can image these plasmons, statically as well as in a time-resolved fashion. Using circular grating patterns, nanofocusing down to 60 nm is experimentally observed.

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O 11.5 Mon 16:00 MA 004

Nonlinear Plasmo-Emission of Electrons from Focused Surface Plasmon Polaritons — •Philip Kahl, Daniel Podbiel, Andreas Makris, Simon Sindermann, Michael Horn-von Hoegen, and Frank-J. Meyer zu Heringdorf — Faculty of Physics and Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, 47057 Duisburg, Germany

In a photoemission electron microscope with normal incidence light geometry we recently observed high-order photoemission of electrons from focused surface plasmon polaritons (SPPs) on a silver film. The SPPs are excited at concentric circular gratings, have convex wave fronts and propagate from opposite directions into the center of the inlying disc. By combining sub 20 fs laser pulses and a home-built Mach-Zehnder interferometer to a pump-probe experiment we are able to temporarily resolve the propagation of these SPPs, the formation of a standing wave at the center and the reversal of the wave front curvature of the SPPs. The fact that the circles have diameters larger than 40 microns guarantees that the laser is not present anymore when the SPPs reach the center. Hence, the electrons are emitted from the plasmonic field alone, which is why we call this effect plasmo-emission. We also captured energy-filtered photoelectron images and analyzed the kinetic energy spectra of the electrons originating from the standing wave fringes at the focal spot. The spectra indicate photoemission orders of up to five.

O 11.6 Mon 16:15 MA 004

Shaping and spatiotemporal characterization of sub-10-fs pulses focused by a high-NA objective — •Sebastian Goetz[1], Monika Pawlowska[1], Christian Dreher[1], Matthias Wurdack[1], Enno Krauss[2], Peter Geisler[2], Gary Razinskas[2], Bert Hecht[2,3], and Tobias Brixner[1,3] — [1]Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [2]Nano-Optics and Biophotonics Group, Experimentelle Physik 5, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [3]Röntgen Center for Complex Material Systems (RCCM), Am Hubland, 97074 Würzburg, Germany

For the investigation of ultrafast processes on the nanoscale by far-field radiation, the temporal resolution of femtosecond laser pulses has to be combined with the spatial resolution of a high-NA microscope. We combine confocal microscopy with phase and amplitude pulse shaping of few-cycle NIR pulses using an LCD pulse shaper in $4f$ single-pass geometry. By utilizing the pulse compression algorithm PRISM, a pulse length of 10 fs could be achieved at the focus position. The capability of the setup is demonstrated by investigating the influence on the diffraction-limited focus of a high-NA objective, LCD imperfections and spatio-temporal coupling. By scanning of gold nanorods through the focal plane, position-dependent reflections within the focus reveal the spectrally resolved intensity profile and the spatial dependency of the pulse arrival time. The ability to shape the light fields and the availability of high peak powers open the path to all-optical control of near-field propagation in the linear and nonlinear regimes.

O 11.7 Mon 16:30 MA 004

Local Nonlinear Spectroscopy of Single Gold Nanostructures — •Daniela Wolf, Thorsten Schumacher, and Markus Lippitz — Experimental Physics III, University of Bayreuth

While higher harmonics generation is a commonly known effect and is used in many applications, the origin of the higher harmonics light generated in plasmonic nanostructures is still not fully understood. Contrary to the luminescence from gold structures, higher harmonics generation is a coherent process so that interference effects come into play when more complex structures are considered. In our experiments, we investigate the response of different gold nanostructures upon excitation with infrared light. The homogeneous luminescence background as well as the interference effects in the third harmonic signal can be observed in spectrally resolved backfocal plane images. In combination with calculated interference patterns from a dipole model, this method allows conclusions about the spatial origin of THG.

O 11.8 Mon 16:45 MA 004

Second-harmonic generation with double resonant hybrid plasmonic/dielectric antennas — •Heiko Linnenbank[1], Yevgen Grynko[2], Jens Förstner[2], and Stefan Linden[1] — [1]Physikalisches Institut, Universität Bonn, Germany — [2]Institut für Elektrotechnik und Informationstechnik, Universität Paderborn, Germany

In several studies it has been shown that the excitation of plasmonic resonances in gold nanostructures with ultrashort light pulses can give rise to large second and third harmonic signals. Here, we demonstrate that the second harmonic generation efficiency of metallic nanostructures can be increased by depositing a dielectric material with a large second order nonlinear coefficient in the hot spots of the structures. By varying the geometry of the nanostructures and by performing nonlinear spectroscopy we rule out that the enhancement of the second harmonic signal is due to a trivial shift of the plasmonic resonance frequency caused by the incorporation of the dielectric material. Furthermore, we have performed nonlinear experiments with double-resonant metallic nanostructures that exhibit plasmonic resonances at both the frequency of the pump light and the generated second harmonic light. Compared to metallic nanostructures which are only resonant to the pump light, the double-resonant structures give rise to larger second harmonic signals. Moreover, they also allow to control the polarisation properties of the generated light.

O 11.9 Mon 17:00 MA 004

Frequency doubling with second harmonic resonant plasmonic nanostructures — •Heiko Linnenbank and Stefan Linden — Physikalisches Institut, Universität Bonn, Germany

Second harmonic generation from lithographically defined metallic nanostructures has been a topic of large interest in recent years. In order to take advantage of local field enhancement effects, the samples are typically designed such that one of the plasmonic modes of the respective metallic nanostructure is resonant to the incident pump wave. In contrast, the second harmonic signal usually is not resonant to one of the higher order plasmonic modes. Here we show that also the opposite scheme can be employed for enhanced second harmonic generation. For this purpose, we have performed nonlinear spectroscopic experiments with different metallic nanostructures where rather the second harmonic light drives a plasmonic resonance than the pump field. Our findings can be explained in the framework of the anharmonic oscillator model where the characteristic parameters are extracted from linear extinction measurements.

O 11.10 Mon 17:15 MA 004

Nonlinear plasmonics of aluminum nanoantennas resonant at the second harmonic frequency — •Lili Gui, Jaco Fuchs, Bernd Metzger, Dominik Floess, Maxim Nesterov, and Harald Giessen — 4th Physics Institute and Research Center SCoPE, University of Stuttgart, 70569 Stuttgart, Germany

Localized surface plasmon resonances can boost significantly the optical nonlinearity of metallic nanoparticles due to the strongly confined and enhanced electromagnetic near-field. Although some double-resonant and multi-resonant configurations have been employed in order to achieve large nonlinear conversion efficiencies, there has been so far no quantitative investigation to illustrate the role of the plasmonic resonance of the nanoantenna at the harmonic frequency. It is still controversial whether the contribution of the harmonic resonance is adverse, as a loss mechanism due to more absorption, or beneficial, due to higher radiation efficiency.

Through comprehensive experimental studies of the second-harmonic spectroscopy of aluminum nanorods resonant at the second harmonic, we demonstrate that the contribution of the harmonic resonance is higher with regards to increased radiation efficiency than to increased absorption loss, with a clear evidence of larger overall second-harmonic-generation efficiency. The nonlinear response of the nanoantennas can be well explained by an analytic anharmonic oscillator model and nicely reproduced with finite-element simulations.

O 11.11 Mon 17:30 MA 004

Applying the Otto geometry to observe second-harmonic generation from a surface plasmon resonance — •Karsten Pufahl[1], Jan Heckmann[1], Nicolai B. Grosse[1], Liuyang Sun[2], John Sipe[3], Xiaoqin Li[2], and Ulrike Woggon[1] — [1]Institut für Optik und Atomare Physik, Technische Universität Berlin, Germany — [2]Department of Physics, University of Texas-Austin, USA — [3]Department of Physics, University of Toronoto, Canada

The Otto geometry enables the probing of material surfaces using evanescent fields having well-defined k-vectors. We have demonstrated this technique in the nonlinear optical regime to investigate how the excitation of a surface-plasmon (SP) resonance leads to enhanced second-harmonic generation (SHG) from a silver surface. The analysis of SHG in k-space allows one to infer SP propagation length and the enhancement of the near-field, while being free to vary the air gap which sets the SP-to-photon coupling strength. In contrast to the Kretschmann geometry where one is limited to investigating thin films at a fixed coupling strength, the Otto technique has the additional advantage that surface wave phenomena can also be explored in bulk and opaque samples.

O 11.12 Mon 17:45 MA 004

Electron tunneling mediated amplification of radiative plasmon decay — •xiao wang, kai braun, dai zhang, heiko peisert, hilamar adler, thomas chassé, and alfred meixner — Institute of physical and theoretical chemistry, University of Tübingen, Auf der Morgenstelle 18, 72076 Tübingen, Germany

Here we demonstrate electron tunneling mediated amplification of photoluminescence (PL) from an Au-Au junction in a combined tip enhanced near-field optical and scanning tunneling microscope. Luminescence spectra were collected from the same Au-Au junction with different bias voltages with and without laser illumination. Without optical excitation the luminescence spectra show typical plasmon modes in the spectral range from 700-1050 nm excited by inelastic electron tunneling. Under laser illumination at 634 nm, at low bias voltages the PL is dominated by the radiative decay of the laser excited electron-hole pairs from the sp/d interband transition with a single band at 690 nm. At higher bias voltages, the luminescence increases dramatically, showing both the band from electron-hole recombination and the plasmon-modes from inelastic tunneling. The increase of the luminescence from inelastic tunneling is more than an order of magnitude and is attributed to the laser induced hot-electron population closely above the Fermi-level to inelastic tunneling having a higher radiative decay rate than the conducting electrons from the sp-band.