O 52: Photonics and Nanooptics II: Nonlinear Response

Time: Wednesday 10:30–13:15 Location: S051

O 52.1 Wed 10:30 S051

Nonlinear Plasmonic Sensing — •Martin Mesch, Bernd Metzger, Mario Hentschel, and Harald Giessen — 4th Physics Institute and Research Center SCoPE, University of Stuttgart, Germany

We introduce the concept of *nonlinear* plasmonic sensing, relying on third harmonic generation from simple plasmonic nanoantennas. Due to the nonlinear conversion process we observe a larger sensitivity upon a local change in the refractive index as compared to the commonly used linear localized surface plasmon resonance sensing. Refractive index changes as small as 10^{-3} can be detected. In order to determine the spectral position of highest sensitivity, we perform linear and third harmonic spectroscopy on plasmonic nanoantenna arrays, which are the fundamental building blocks of our sensor. Furthermore, simultaneous detection of linear and nonlinear signals allows quantitative comparison of both methods, providing further insight into the working principle of our sensor. While the signal-to-noise ratio is comparable, nonlinear sensing gives about seven times higher signal levels. Our scheme can be extended to other nonlinear processes such as second harmonic generation and sum frequency generation. This opens a new avenue in plasmonic sensing.

O 52.2 Wed 10:45 S051

Investigation of Plasmonic Modes of Gold Tapers — •Surong Guo[1], Nahid Talebi[1], Wilfried Sigle[1], Christian Knipl[1], Martin Esmann[2], Simon Becker[2], Ralf Vogelgesang[2], Christoph Lienau[2], and Peter van Aken[1] — [1]Max Planck Institute for Solid State Research , Heisenbergstraße 1, D-70569 Stuttgart, Germany — [2]Carl von Ossietzky Universität Oldenburg, Ammerländer Heerstraße 114-118, D-26129 Oldenburg, Germany

Plasmonic tapers have been studied intensively due to the ability of adiabatically coupling the propagating surface plasmon polaritons along their shaft to the nanolocalized plasmons at their apex. Therefore, they can find applications in the fields of sub-diffraction-limit nanofocusing, ultrafast photoemission, and near-field optical microscopy.

We investigate the plasmonic modes of three-dimensional single crystalline gold tapers by means of electron energy loss spectroscopy and numerical calculation. We observe discrete higher-order azimuthal plasmonic modes of the gold taper with an opening angle of ~45° with energy dispersions roughly proportional to the inverse local radius. The importance of phase-matching between electron field and radiative taper modes in mesoscopic structure is demonstrated [1]. We further systematically study the changes in the dispersion of higher-order plasmonic modes of gold tapers versus the opening angle of the taper, both experimentally and theoretically.

[1] N. Talebi et al, ACS Nano, 2015, 9 (7), 7641-7648.

O 52.3 Wed 11:00 S051

Nonlinear Emission of Electrons in a Strong Plasmonic Field — Daniel Podbiel[1], Philip Kahl[1], Bettina Frank[1], Harald Giessen[2], and •Frank Meyer zu Heringdorf[2] — [1]Faculty of Physics and CENIDE, University of Duisburg-Essen, 47048 Duisburg, Germany — [2]4th Physics Institute and Research Center SCoPE, University of Stuttgart, 70550 Stuttgart, Germany

Observing surface plasmon polaritons (SPPs) in a photoemission electronmicroscope (PEEM) is possible via nonlinear photoemission if ultra-short laser pulses (<20fs) of a suitable wavelength are directed onto the surface of a plasmonic material. We study the time-resolved propagation and interaction of SPPs by means of a direct conceptual visualization of the SPPs in a "normal incidence geometry". This experimental setup allows us to observe transient phenomena that exist for only a few femtoseconds during the coherent interaction of the ultrashort SPP pulses. In focusing structures for SPPs we find an unexpected time-signature of the nonlinear photoemission signal at the focus point that must be explained by emission of electrons from the SPP alone. The energy distribution of these 'plasmoelectrons' shows that the SPP fields are sufficiently high to make nonlinear photoemission pathways of higher orders the dominant contribution to the PEEM signal.

O 52.4 Wed 11:15 S051

Coherent control of photoemission from a single nanotip in a two-color scheme — •Michael Förster[1,2], Timo Paschen[1], Michael Krüger[1,2], Florian Libisch[3], Christoph Lemell[3], Georg Wachter[3], Thomas Madlener[3], Joachim Burgdörfer[3], and Peter Hommelhoff[1,2,4] — [1]Department Physik, Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), Staudtstrasse 1, 91058 Erlangen — [2]Max-Planck Institut für Quantenoptik, Hans-Kopfermann-Str. 1, 85748 Garching — [3]Institute for Theoretical Physics, Vienna University of Technology, Wiedner Hauptstr. 8-10/E136, 1040 Wien — [4]Max-Planck Institut für die Physik des Lichts, Günther-Scharowsky-Str. 1/Geb. 24, 91058 Erlangen

We discuss coherent control of photoemission from a tungsten tip in a Brumer-Shapiro scheme. We observe that photoemission from the nanotip induced by a femtosecond laser displaying many multiphoton orders can be strongly enhanced or reduced by the presence of a weak second harmonic, depending on the phase between the laser pulses. With optimized parameters the phase-dependent contrast in the total emitted current can exceed 90%. Time-resolved data shows that emission takes place on femtosecond time scales. With the help of spectrally-resolved measurements and density functional theory calculations we interpret our observations in terms of interfering emission pathways.

O 52.5 Wed 11:30 S051

Above threshold ionization of Rydberg electrons localized to a gold nanotip — •Jörg Robin[1], Jan Vogelsang[1], Benedek J. Nagy[2], Petra Gross[1], and Christoph Lienau[1] — [1]Carl von Ossietzky Universität, 26129 Oldenburg — [2]Wigner Research Centre for Physics, H-1121 Budapest

Metallic nanotips are model systems to study nanometre and femtosecond electron dynamics and provide the possibility for ultrafast electron microscopy. Evidence of strong-field phenomena has been observed by one-colour photoemission of electrons from metallic nanotips [1-3], while two-colour photoemission has established the existence of surface states on metallic films [4]. Here, we report femtosecond two-colour photoemission of electrons from a gold nanotip. We observe long-lived wave packets of Rydberg electrons bound to their own image potential. These intermediate bound states facilitate above-threshold ionization similar to atomic systems and give access to a cold, ultrafast, nanolocalized electron source. [1] Krüger, M. et al. Nature 475, 78 (2011) [2] Herink, G. et al. Nature 483, 190 (2012) [3] Piglosiewicz, B. et al. Nat. Photon. 9, 37 (2014) [4] Höfer, U. et al. Science 277, 1480 (1997)

O 52.6 Wed 11:45 S051

Third harmonic efficiency scaling with plasmonic antenna length — •Maxim Nesterov, Mario Hentschel, Bernd Metzger, Harald Giessen, and Thomas Weiss — 4th Physics Insitute and Research Centre SCoPE, University of Stuttgart, 70550 Stuttgart, Germany

Nonlinear interaction of light with plasmonic structures leads to higher-order harmonic generation, such as second-order harmonics in non-centrosymmetric media, and third-order harmonics in centrosymmetric systems. Metallic antennas are characterized by a high field concentration at resonances, resulting in a relatively strong nonlinear optical response. The enhancement of higher-order harmonic generation depends on the size and shape of antenna, and a strong nonlinear signal can be achieved with a proper design.

We study scaling of the third harmonic generation with rod antenna length numerically for a single antenna, as well as for array configurations. The third-harmonic intensity is found to be increasing as a twelfth power of an antenna length in the model based on Miller's rule. We discuss applicability of the Miller's rule in the context of the origin of the nonlinear susceptibility in the metals. An analytical model has been developed to support the numerical results.

O 52.7 Wed 12:00 S051

Nonlinear chiral plasmonics: Quantitative modeling of the third-harmonic response of 3D chiral plasmonic nanoantennas — •Lili Gui, Xinghui Yin, Mario Hentschel, and Harald Giessen — 4th Physics Institute and Research Center SCoPE, University of Stuttgart, 70569 Stuttgart, Germany

Optical activity exists generally in nature, as many biomolecules such as proteins are chiral. Chiral plasmonic nanostructures are highly in-

teresting since they are potentially ideal and sensitive platforms for biosensing due to the strong superchiral near fields and their own giant chiroptical effects. Exploration of nonlinear chiroptical effects in chiral plasmonic structures is even more desired since the nonlinear chiroptical effects are typically orders of magnitude higher than the linear optical counterparts. Here we study the third-order nonlinear chiroptical effects of 3D chiral structures consisting of corner-stacked gold nanorods. Compared to second-order effects, third-order effects are more immune to fabrication defects and surface roughness. We experimentally investigate the third-harmonic spectroscopy with left- and right-circularly polarized fundamental light, respectively. The third-order chiroptical responses can be well understood when we utilize a coupled anharmonic oscillator model considering the phase retardation of light interaction with the 3D gold nanorods. This model is instructive to guide a practical design of plasmonic chiral structures for a giant third-harmonic-generation circular dichroism (THG-CD) effect.

O 52.8 Wed 12:15 S051

Shaping the Nonlinear Near-Field of Single Gold Nanostructures — •Daniela Wolf[1,2], Julian Obermeier[1], Thorsten Schumacher[1], and Markus Lippitz[1] — [1]Experimental Physics III, University of Bayreuth, Universitaetsstr. 30, 95447 Bayreuth, Germany — [2]Max Planck Institute for Solid State Research, Heisenbergstr. 1, 70569 Stuttgart, Germany

Light scattering at plasmonic nanoparticles and their assemblies has led to a wealth of applications in metamaterials and nanooptics. While the shaping of fields around nanostructures is widely studied, the influence of the field inside the nanostructures is often overlooked. The linear field distribution inside the structure taken to the third power causes third-harmonic generation, a nonlinear optical response of matter. Here we demonstrate by a far-field Fourier imaging method how this simple fact can be used to shape fields around already a single particle alone. We employ this scheme to switch the third-harmonic emission from a single point source to two spatially separated but coherent sources, as in Young's double slit assembly. Finally, we present some recent results on more advanced structures combining nonlinear plasmonics and waveguiding.

O 52.9 Wed 12:30 S051

An ultrafast nanotip electron gun triggered by grating-coupled surface plasmons — •Benjamin Schröder, Murat Sivis, Reiner Bormann, Sascha Schäfer, and Claus Ropers — 4th Physical Institute - Solids and Nanostructures, University of Göttingen, Friedrich-Hund-Platz 1, 37077 Göttingen

We present recent results on the photoelectron emission from metal nanotips induced by propagating surface plasmon polaritons (SPPs) excited in a grating-coupler on the tip shaft [1-3]. Nanofocusing of the SPPs induces multiphoton photoemission with high efficiency. The nanostructure is inserted in a modified field-emitter electron gun geometry controlling the electrostatic environment of the tip. In this way, a site-selective control of the electron emission is achieved, and we can disentangle electrons emitted from the apex from those originating at the grating or shaft. Both the control of the transverse beam properties and the local extraction fields enabled by the gun design will be beneficial in future ultrafast transmission electron microscope applications [4].

[1] C. Ropers, C.C. Neacsu, T. Elsaesser, M. Albrecht, M.B. Raschke, and C. Lienau, Nano Lett. 7, 2784 (2007).

[2] S. Berweger, J.M. Atkin, X.G. Xu, R.L. Olmon, and M.B. Raschke, Nano Lett. 11, 4309 (2011).

[3] J. Vogelsang, J. Robin, B.J. Nagy, P. Dombi, D. Rosenkranz, M. Schiek, P. Groß, and C. Lienau, Nano Lett. 15, 4685 (2015).

[4] B. Schröder, M. Sivis, R. Bormann, S. Schäfer, and C. Ropers, accepted for puplication in Appl. Phys. Lett. (2015).

O 52.10 Wed 12:45 S051

Plasmon dynamics on nanoporous gold particles revealed by strong field photoemission — Germann Hergert[1], •Jan Vogelsang[1], Jörg Robin[1], Petra Gross[1], Dong Wang[2], Peter Schaaf[2], Erich Runge[2], and Christoph Lienau[1] — [1]Carl von Ossietzky Universität, Oldenburg, Germany — [2]Technische Universität Ilmenau, Ilmenau, Germany

Nanoporous gold particles or "nanosponges" are particles with a diameter of hundreds of nanometers, perforated with randomly arranged pores with diameters in the 10-nm range. They have recently attracted attention as templates for surface-enhanced Raman sensing as they combine several advantageous properties, such as multiple plasmon resonances in the visible or NIR spectral range, a high surface-to-volume ratio, and a high density of catalytic sites [1].

Here we investigate the plasmon dynamics of such particles using ultrafast photoemission microscopy. When using 14-fs excitation pulses at 1600 nm for excitation of single nanosponges, we observe long-lived plasmon oscillations as a function of the pulse delay. Introducing a new model for the optical properties of these particles, we show that our results reveal the existence of long-lived plasmon hotspots with lifetimes of several tens of fs on the surface of the sponge. This plasmon localization could be of key importance for enhancing the sensing capability of such particles.

[1] C. Vidal et al, ACS Photonics 2, 1436 (2015)

O 52.11 Wed 13:00 S051

Suppression of radiative damping and enhancement of second harmonic generation in bull's eye nanoresonators — •Vladimir Smirnov[1], Jue-min Yi[1], Xianji Piao[2], Jiho Hong[2], Heiko Kollmann[1], Martin Silies[1], Wei Wang[1], Petra Gross[1], Ralf Vogelgesang[1], Namkyoo Park[2], and Christoph Lienau[1] — [1]University of Oldenburg, Germany — [2]Seoul National University, Korea

Bull's eye (BE) nanoresonators, consisting of a central aperture in an Au film surrounded by a series of concentric circular grooves, are highly interesting structures in the context of manipulating light propagation [1] and also optimizing the coupling to quantum emitters [2]. Using broadband linear spectral interferometry and ultrahigh time resolution spectroscopy, we study the temporal response and the local field enhancement of such BE nanoresonators. We find surprisingly long lifetimes of the extended resonator eigenmodes of more than 35 fs [3]. Furthermore, by replacing the central circular hole with an annular ring void, we obtain 50-times higher second harmonic generation efficiency, illustrating the efficient field enhancement and confinement possible in BE nanoresonators. The combination of spatial light concentration with high quality factors has high potential for sensing and coherent control of light-matter interactions on the nanoscale.

[1] H. J. Lezec et al., Science 297, p. 820 (2002); [2] A. G. Curto et al., Science 329, p. 930 (2010); [3] J. Yi et al, submitted to ACS Nano (2015)