

DS 28: [O] Plasmonics and Nanooptics IV (Joint Session DS/O/HL)

Time: Wednesday 15:00–17:45

Location: H32

DS 28.1 Wed 15:00 H32

Raman and Luminescence Enhancement Produced in Gap-mode Near-field Optical Microscopy — •DAI ZHANG, MARCUS SAKROW, KAI BRAUN, and ALFRED J. MEIXNER — Institute of Physical and Theoretical Chemistry, University Tübingen, Auf der Morgenstelle 8, Tübingen

Tip-enhanced near-field optical microscopy can produce spectroscopic images of molecular layers deposited on smooth Au-surfaces, of single molecules and of organic semi-conductor films with an optical resolution on the order of 10 nm. This is made possible by a sharp laser-illuminated Au-tip approaching as close as a few nanometers to the sample surface. The system behaves as an optical antenna for confining and enhancing the excitation field to a small sub-diffraction volume in the gap between the tip-apex and the surface and retrieving emitted or scattered photons to the far field for detection. In this way the signal emitted from the gap can be enhanced by several orders of magnitude with respect to the diffraction limited background signal by enhancing the excitation efficiency and the emission yield. In this contribution, a variety of enhancement mechanisms will be discussed.

DS 28.2 Wed 15:15 H32

Quantifying Excitation and Radiation Rate Enhancement Provided by Near-field Optical Antennas — •MIRIAM BÖHMLER, NICOLAI HARTMANN, CARSTEN GEORGI, and ACHIM HARTSCHUH — Department Chemie und Biochemie & CENS, Ludwig-Maximilians-Universität, 81377 München

Tip-enhanced near-field optical microscopy provides nanoscale optical resolution beyond the diffraction limit [1]. This is due to the highly confined enhancement of the optical fields at the tip apex, which locally increases both excitation and radiation rates. In our experiments we use sharp gold tips to probe the photoluminescence (PL) of individual single-walled carbon nanotubes. We show that excitation and emission enhancement can be distinguished by imaging the radiation pattern in the back focal plane of the microscope objective. The analysis is based on the characteristic radiation patterns generated by single dipoles with a particular orientation in 3D [2]. We first find that the PL emission of a nanotube can be described by an in-plane oriented dipole. In the presence of our tip, the radiation pattern is strongly modified and dominated by the signatures of a vertical dipole corresponding to the tip axis. This observation illustrates the spatial redirection of the emission by the tip acting as an optical antenna [3]. By comparing radiation patterns with and without the influence of our tip, we estimate excitation and radiation rate enhancement quantitatively.

[1] A. Hartschuh, *Angew. Chem. Int. Ed.* 47, 8178 (2008)[2] M. A. Lieb, *J. Opt. Soc. Am. B* 21, 1210 (2004)[3] T. H. Taminiau, *Nat. Photon.* 2, 234 (2008)

DS 28.3 Wed 15:30 H32

Pseudo-heterodyne scanning near-field optical microscope for surface plasmon detection with actively stabilized phase — •STEFAN GRIESING, ANDREAS ENGLISCH, and UWE HARTMANN — Experimental Physics, Saarland University, D-66123 Saarbrücken

Amplitude and phase of the evanescent surface plasmon field were detected by a pseudo-heterodyne scanning near-field optical microscope (SNOM). Light from a HeNe laser ($\lambda = 633\text{nm}$) and an argon ion (Ar) laser ($\lambda = 488\text{nm}$) was combined in a fiber coupler. One output channel of the coupler served as reference branch of an interferometer. A piezoelectric fiber stretcher in that branch was used for sinusoidal phase modulation. The other output of the coupler was linked to another coupler. By using different color filters, the two wavelengths were separated at the output channels. The HeNe laser was used for surface plasmon excitation under attenuated total internal reflection. The light from the Ar laser illuminated the sample under normal incidence, so that during the scanning process a constant phase from the Ar laser was detected. The SNOM tip detects both, surface plasmons excited by the HeNe laser and the spot from the Ar laser. The signal is combined in a third fiber coupler with the signal from the reference branch. At the output, the interference signal was wavelength-selectively detected by Si photodiodes and lock-in amplifiers. The signal from the Ar laser was used as input for a feedback-loop which modifies the

drive signal of the fiber stretcher. In this way, the phase fluctuations due to thermal and mechanical disturbances were compensated.

DS 28.4 Wed 15:45 H32

Scattering near-field microscopy in the THz with a free-electron laser — •HANS-GEORG VON RIBBECK^{1,2}, MARC TOBIAS WENZEL¹, RAINER JACOB², and LUKAS M. ENG¹ — ¹Institut für Angewandte Photophysik, TU Dresden, 01062 Dresden, Germany — ²Institut für Ionenstrahlphysik und Materialforschung, Forschungszentrum Dresden - Rossendorf, 01314 Dresden

We present scattering-type scanning near-field optical spectroscopy (s-SNOM) investigations, successfully operated in the THz range with a wavelength independent spatial resolution of 150 nm. Our microscopy set-up bases on a true noncontact atomic force microscope (nc-AFM) combined with the free-electron laser (FEL) source at the Forschungszentrum Dresden-Rossendorf. This laser provides tunability from 30 - 250 μm . We were able to record, for the first time ever, s-SNOM signatures with a FEL at wavelengths ranging from 30 μm to 180 μm (10 - 1.67 THz). In addition to the near-field dependent optical signals we also demonstrate the imaging capabilities of our THz-s-SNOM. Image scans were performed on a specially designed test structure consisting of a topography-free composite of a polymer/gold sample. On such samples, topography independent strong optical material contrast could be demonstrated at 150 μm wavelength. We achieve a resolution of better than 150 nm corresponding to better than 1/1000.

DS 28.5 Wed 16:00 H32

Ultrahigh temporal and spatial resolution imaging of second harmonic fields in random zinc oxide nanostructure arrays — •MANFRED MASCHECK¹, SLAWA SCHMIDT¹, MARTIN SILIES¹, TAKASHI YATSUI², MOTOICHI OHTSU², DAVID LEIPOLD³, ERICH ROLDS³, and CHRISTOPH LIENAU¹ — ¹Carl-von-Ossietzky-Universität, Oldenburg — ²University of Tokyo — ³Technische Universität Ilmenau

Among the most prominent signatures of the weak (Anderson) localization of light in random dielectric media are an enhanced coherent backscattering and the localization of electromagnetic fields in both space and time. Light localization is particularly relevant in nanophotonic materials, where it can give rise to strong field localization and greatly enhanced optical nonlinearities.

Here, we explore this light localization in densely packed, random arrays of ZnO nanoneedles with tip diameters of less than 20 nm. A phase-stabilized pair of 6 fs laser pulses is focused to its diffraction limit of $1\ \mu\text{m}^2$ onto the ZnO needles.

The generated surface second harmonic (SH) spectra are detected as a function of the lateral position of the laser focus and the delay between the two pulses. We observed a pronounced spatial localization of the SH signal in hot spots of less than 300 nm dimension. We show, by retrieving the time structure of the localized electric field from interferometric FROG traces, that light is stored in these hot spots for more than 100 fs. Such nanoneedle arrays therefore present a highly interesting new model system for exploring the rich physics of weakly localized light fields.

DS 28.6 Wed 16:15 H32

Second Harmonic Imaging of Gold Nanocones with a Parabolic Mirror Microscope — •ANKE HORNEBER¹, DAI ZHANG¹, MONIKA FLEISCHER², MARCUS SACKROW¹, KAI BRAUN¹, DIETER P. KERN², and ALFRED J. MEIXNER¹ — ¹Institute of Physical Chemistry, Eberhard Karls University Tübingen, Germany — ²Institute of Applied Physics, Eberhard Karls University Tübingen, Germany

Second harmonic generation (SHG) is a valuable technique for the visualization of surfaces. Our confocal optical microscope uses a parabolic mirror with a smaller focus than objective lenses in air and the largest possible signal collection angle. We use higher order laser modes which are often called cylindrical vector beams for the investigation of complex plasmonic structures. The longitudinal polarization component in a radially polarized laser beam focus can excite the longitudinal plasmon of a gold cone most efficiently[1]. We present first SHG images of gold nanocones with an optical resolution below 300 nm. We are developing a flexible setup that will allow non-linear experiments as well

as near-field measurements. Such a setup will provide further opportunities like non-linear near-field investigations and the combination of a high spatial resolution with ultrafast dynamics.

[1] M. Fleischer et al., APL, 93 (2008) 111114.

DS 28.7 Wed 16:30 H32

Nanosphere Lithography of Sub-50 nm Plasmonic Structures — •JUN ZHAO, BETTINA FRANK, and HARALD GIESSEN — Universität Stuttgart, Deutschland

Nanosphere lithography is a powerful and fast fabrication technique for periodic large-area metallic nanostructures of different and complex shapes [1]. We use tilted-angle-rotation thermal evaporation onto the monolayers of close-packed polystyrene nanospheres to fabricate metamaterial samples of up to 1cm^2 .

With this fabrication technique we can prepare sub-50 nm plasmonic structures such as pentamers, hexamers, and also ring structures with a centered or off-centered disk in the middle, which is useful for investigation of Fano-type plasmon resonances and future sensors.

The optical response of our structures was measured in reflectance geometry with FTIR-microscopy. The measurements show good agreement with our simulations. We also performed stacking experiments of split-ring resonators, which can be arranged in an ordered or twisted stereometamaterial fashion [2].

[1] J. M. C. Gwinner, E. Koroknay, L. Fu, P. Patoka, W. Kandulski, M. Giersig, and H. Giessen, *Small* 5, 400 (2009) [2]. N. Liu, H. Liu, S. Zhu, and H. Giessen, *Nature Photonics* 3, 157 (2009)

DS 28.8 Wed 16:45 H32

Gold nanocone probes for near-field scanning optical microscopy — •BASTIAN ZEEB, CHRISTIAN SCHÄFER, PETER NILL, MONIKA FLEISCHER, and DIETER P. KERN — Institute of Applied Physics, University of Tübingen, Auf der Morgenstelle 10, 72076 Tübingen

Apertureless near-field scanning optical microscopy (ANSOM) provides the possibility to collect simultaneously high-resolution topographical and sub-diffraction limited optical information from a surface. When optically excited, the scanning probes act as optical antennae with a strong near-field enhancement near the tip apex. Spatial resolution and optical near-field enhancement depend strongly on the properties and geometry of the scanning probe - in particular on very sharp tip radii. Various possibilities for fabricating good antennae have been pursued. Most commonly, scanning probes consist of electrochemically etched gold wires which are sharp but not well-defined in geometry.

We present two different approaches for ultra sharp and well-defined antennae based upon fabricating gold nanocones with a tip radius smaller than 10 nm which can be used in ANSOM [e.g. M. Fleischer et al., *Appl. Phys. Lett.* 93, 111114 (2008)]. A transfer process is presented that can be used to attach single gold nanocones to non-metallic probes such as sharp glass fiber tips. Alternatively, new processes are presented to fabricate cones directly on pillars of different materials such as silicon or bismuth, which can be applied to cantilever tips for ANSOM scanning applications.

DS 28.9 Wed 17:00 H32

Space Charge Effects in Photoemission Electron Microscopy — •JAN VANIS, NIEMMA M. BUCKANIE, PING ZHOU, DIETRICH VON DER LINDE, MICHAEL HORN-VON HOEGEN, and FRANK-J. MEYER ZU HERINGDORF — University Duisburg-Essen, Germany

We use the combination of a spectroscopic Photoemission Electron Microscope (PEEM) with femtosecond (fs) laser pulses to investigate

samples with high spatial and temporal resolution. For proper imaging, the laser pulses must have sufficiently low intensity to minimize the space charge. Here, we report on experiments with a regenerative Ti:sapphire amplifier system. The laser setup generates fs-pulses ($\lambda = 800\text{ nm}$, i.e., a photon energy of $E = 1.55\text{ eV}$). The system has a variable repetition rate up to 250 kHz. We studied Ag islands which have been grown *in-situ* by self-assembly on Si(111) surfaces. We used the fundamental and the 4th harmonic ($E = 6.2\text{ eV}$) of the amplifier system to evaluate space charge effects in PEEM. The space charge effect is reflected in an energetic broadening of the electron distribution and, at higher laser fluence, in a loss of focus. Insertion of apertures into the electron path provides a way to change the electron density in different areas of the microscope and locate in which section of the PEEM the space charge effect is dominant. The influence of the laser energy, wavelength, and repetition rate on the energy spectra and the image distortions will be discussed.

DS 28.10 Wed 17:15 H32

Characterization of Single Gold Nanoparticles Using Confocal Interference Microscopy in Combination with Higher Order Laser Modes — •FRANK WACKENHUT¹, TINA ZÜCHNER¹, ANTONIO VIRGILIO FAILLA^{1,2}, and ALFRED J. MEIXNER¹ — ¹Eberhard-Karls-Universität Tübingen, Institut für Physikalische und Theoretische Chemie, Tübingen, Germany — ²Max-Planck-Institut für Entwicklungsbiologie, Tübingen, Germany

By using confocal interference microscopy in combination with higher order laser modes it is possible to directly image the orientation and to detect the shape of single metal nanoparticles, with sizes well beyond the diffraction limit [1,2]. Metal nanoparticles can be imaged by detecting both their luminescence or the elastically scattered light. In the scattering detection mode the visualized pattern strongly depends on the local environment, e.g. the refractive index of the surrounding medium [3]. With this technique we are also able to observe and quantify dynamics in the motion of single nanoparticles [4]. Ongoing measurements show that this technique might be utilized for studying the wavelength dependence of the polarizability tensor of a single gold nanorod.

[1] A.V. Failla, H. Qian, H. Qian, A. Hartschuh, A. J. Meixner (2006), *Nano Lett.* 6, 1374. [2] T. Züchner, A. V. Failla, A. J. Meixner (2008), *J. Microsc.* 229, 337. [3] T. Züchner, A. V. Failla, M. Steiner, A. J. Meixner (2008), *Opt. Expr.* 16, 14635. [4] T. Züchner, F. Wackenhut, A. V. Failla, A. J. Meixner (2009), *Appl. Surf. Sci.* 255, 5391.

DS 28.11 Wed 17:30 H32

Polarization dependence of the optical response of individual metallic nanostructures and arrays — •RETO GIANNINI¹, YASIN EKINCI^{1,2}, and JÖRG F. LÖFFLER¹ — ¹Laboratory of Metal Physics and Technology, Department of Materials, ETH Zurich, 8093 Zurich, Switzerland — ²Laboratory of Micro and Nanotechnology, Paul Scherrer Institute, 5232 Villigen PSI, Switzerland

The optical response of designed metallic nanostructures depends strongly on the properties of the exciting light beam, and especially on the polarization relative to the characteristic axis of the nanostructures. To address this dependence, we established an experimental setup in the visible wavelength range that allows us to measure the optical response of individual metallic nanostructures and arrays with varying incident wave vectors and polarization. The possible excitation covers the whole range from normal excitation (relative to the surface of the substrate) to parallel excitation. The latter is realized using objective-based TIRF. In this presentation, we explain the principle of our setup and the optical response of various Au-nanostructures, such as pillars and dimers under varying wave vectors and polarization.