O 46: Plasmonics and Nanooptics I

Time: Wednesday 10:30–13:00 Location: TRE Phy

O 46.1 Wed 10:30 TRE Phy

Tailored nano-antennas for directional Raman studies of individual carbon nanotubes — •Nicola Paradiso, Fatemeh Yaghobian, Christoph Lange, Tobias Korn, Christian Schüller, Ruper Huber, and Christoph Strunk — Institut für Experimentelle und Angewandte Physik, University of Regensburg

We exploit patterned nano-antennas to investigate the Raman spectra of otherwise not optically detectable carbon nanotubes (CNTs). We demonstrate that a top–down approach is particularly promising when applied to CNTs, owing to the sharp dependence of the scattered signal on the angle between incident light polarization and CNT axis. In contrast to tip enhancement techniques, our method enables us to control the light polarization in the sample plane, locally amplifying and rotating the incident field in order to optimize the Raman signal. Such promising features are confirmed also by the numerical simulations presented here. From the analysis of the G modes we deduced that the CNT under study is semiconducting and chiral, and we estimated its diameter and chiral angle. Measurements on arrays with different orientation with respect to the CNT revealed that thin metal strips allow not only to magnify, but also to rotate the electromagnetic field, thus introducing a new degree of freedom for SERS applications. The relative ease of fabrication and alignment makes this technique suitable for the realization of integrated devices that combine scanning probe, optical, and transport characterization.

O 46.2 Wed 10:45 TRE Phy

Yttrium hydride nanoantennas for reconfigurable plasmonics — •Nikolai Strohfeldt[1], Andreas Tittl[1], Martin Schäferling[1], Frank Neubrech[1], Uwe Kreibig[2], Ronald Griessen[3], and Harald Giessen[1] — [1]Universität Stuttgart, Germany — [2]RTWH Aachen, Germany — [3]Vrije Universiteit Amsterdam, The Netherlands

A key challenge for the development of active plasmonic devices is the lack of materials with fully controllable plasmonic properties. In this work we demonstrate that a plasmonic resonance in top-down nanofabricated yttrium antennas can be completely and reversibly turned on and off using hydrogen exposure. We fabricate arrays of yttrium nanorods and optically observe in extinction spectra the hydrogen-induced phase transition between the metallic yttrium dihydride and the insulating trihydride. Whereas the yttrium dihydride nanostructures exhibit a pronounced particle plasmon resonance, the transition to yttrium trihydride leads to a complete vanishing of the resonant behavior. Our fully reversible plasmonic switch can be tuned over a wide wavelength range by simply varying the size of the nanostructures. Furthermore, we developed an analytical diffusion model with which we are able to explain the temporal behavior of the reconfiguration process observed in our experiments and gain information about the thermodynamics of our device. Thus, our nanorod system serves as a versatile basic building block for reconfigurable plasmonic devices ranging from reconfigurable perfect absorbers to active local heating control elements.

O 46.3 Wed 11:00 TRE Phy

Large-Area Low-Cost Resonant Nano-Antenna Enhanced SEIRA Substrate Using Interference Lithography — •Shahin Bagheri, Frank Neubrech, and Harald Giessen — 4th Physics Institute and Research Center SCoPE, University of Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany

We manufactured large-area plasmonic antenna arrays by laser interference lithography for surface-enhanced infrared absorption. Geometrical parameters of the homogeneous antennas can be precisely defined by changing the interference condition in each exposure process independent of the underlying wafer. Such antennas with tunable and high quality resonances from near- to mid-infrared (1 to 7 μm) are well suited as substrates for surface-enhanced infrared absorption studies. We show the broad applicability of these substrates by enhancing the infrared vibrational signals of a monolayer octadecanethiol and monitoring the ultraviolet degradation of a polymer via the decrease of its specific vibrational modes. Large-area and fast fabrication process make our approach suited as a low-cost sensing platform for a variety of different SEIRA real-world applications.

O 46.4 Wed 11:15 TRE Phy

Resonant surface-enhanced Raman scattering in nanoparticle dimer structures — •Matthias Hensen[1], Ingo Heesemann[2], Elina Oberlander[3], Adelheid Godt[2], Thomas Huser[3], and Walter Pfeiffer[1] — [1]Molecular and Surface Physics, Department of Physics, Bielefeld University, 33615 Bielefeld, Germany — [2]Organic Chemistry and Polymer Chemistry, Department of Chemistry, Bielefeld University, 33615 Bielefeld, Germany — [3]Biomolecular Photonics Group, Department of Physics, Bielefeld University, 33615 Bielefeld, Germany

Surface-enhanced Raman scattering (SERS) benefits from the local field enhancement in the vicinity of plasmonic nanoantennas. We experimentally investigate rodlike acetyl-protected dithiol molecules that specifically link two gold nanoparticles with a diameter of 40 nm and gap dimensions of about 1 nm. Raman spectroscopy using excitation between 473 nm and 785 nm wavelength reveals a resonant enhancement of the linker molecule related Raman signals for excitation around 650 nm. This coincides with the gold dimer resonance, as predicted by FDTD calculations, and we conclude that the resonant excitation of gold-nanoparticle dimers enhances the Raman signal. Furthermore, based on a quantitative analysis of particle and aggregate concentrations we derive Raman enhancement factors in comparison to dithiol molecules in acetone. About 100 linker molecules in the gap of a nanoparticle dimer contribute to the signal and a Raman enhancement factor of about 10^6 is obtained for resonant excitation.

O 46.5 Wed 11:30 TRE Phy

Resonance enhanced local thermionic emission — •Felix Becker, Dominik Differt, Matthias Hensen, Christian Strüber, and Walter Pfeiffer — Molecular and Surface Physics, Department of Physics, Bielefeld University, 33615 Bielefeld, Germany

In the interaction of intense laser pulses with nanostructures or optical antennas the electron emission is commonly discussed in the context of multi-photon processes and strong field phenomena. However, resonant excitation and the related field enhancement also significantly increase the locally absorbed energy density leading to a transient local heating of the electron gas. Thermionic emission dominates the electron yield for sufficiently strong excitation. This has been demonstrated recently for nanoantenna induced currents in metal-insulator-metal junctions and for photonic resonances in nanotextured amorphous silicon layers. Based on electron-electron and electron-phonon scattering rates transient electron energy distributions after short pulse excitation and the related thermionic emission over an internal tunnel barrier and for electron emission into the continuum are simulated. Variation of both scattering rates determines the ratio between sequential multiphoton photoemission and thermionic emission following the Richardson-Dushman equation. Despite the incoherent character of thermionic emission it can be exploited as a probe for this energy concentration process.

O 46.6 Wed 11:45 TRE Phy

Third harmonic spectroscopy of single Au- nanoantennas fabricated by helium ion beam milling — •Heiko Kollmann[1], Martin Esmann[1], Simon F. Becker[1], Xianji Piao[2], Chuong Huynh[3], Lars-Oliver Kautschor[3], Guido Bösker[3], Henning Vieker[4], Armin Gölzhäuser[4], Namkyoo Park[2], Martin Silies[1], and Christoph Lienau[1] — [1]University of Oldenburg, Germany — [2]Seoul National University, Korea — [3]Carl Zeiss Microscopy GmbH, Jena, Germany — [4]University of Bielefeld, Germany

Metallic nanoantennas are able to localize far-field electromagnetic waves in volumes of a fraction of their wavelength. Standard tools for fabricating these structures with sub-20-nm feature sizes are Electron Beam Lithography or Ga-based Focused Ion Beam (FIB) milling. Here, we combine Ga- and He-ion based milling (HIM) for the fabrication of gold bow-tie antennas with few-nanometer gap sizes. Using polarization-sensitive Third-Harmonic (TH) spectroscopy, we compare the nonlinear optical properties of single HIM-antennas with sub-6-nm gaps with those produced by Gallium-based FIB. We find a pronounced enhancement of the intensity and a greatly improved polarization contrast of the TH for He-ion produced antennas in comparison with state-of-the-art Ga-FIB antennas. Our experimental findings are strongly supported by FEM calculations and demonstrate electric field localization in the few-nanometer gap of the bow-tie antenna. This makes

He-ion beam milling a highly attractive and promising new tool for the fabrication of plasmonic nanoantennas with few-nanometer feature sizes.

O 46.7 Wed 12:00 TRE Phy

Tayloring the third harmonic response of plasmonic nanoantennas by incorporation of dielectric nanocrystals — •Bernd Metzger[1], Mario Hentschel[1,2], Thorsten Schumacher[1,2], Markus Lippitz[1,2], Xingchen Ye[3], Christopher B. Murray[3], Bastian Knabe[4], Karsten Buse[4], and Harald Giessen[1] — [1]4th Physics Institute and Research Center SCoPE, University of Stuttgart, Germany — [2]Max Planck Institute for Solid State Research, Stuttgart, Germany — [3]Department of Chemistry, University of Pennsylvania, Philadelphia, USA — [4]Fraunhofer Institute for Physical Measurement Techniques IPM, Freiburg, Germany

The electric field strength in the hot-spot of a plasmonic nanoantenna [1] can surpass the far-field amplitude by more than an order of magnitude, which caused scientists to envision enhanced conversion efficiencies for nonlinear optical effects [2]. Hence, we incorporate indium tin oxide (ITO) nanocrystals into the hot-spot of plasmonic gold gap-antennas and investigate the nonlinear third harmonic (TH) response of the hybrid nanostructure arrays. We find an enhancement by about a factor of two of the TH emission when comparing the hybrid to bare gap-antenna arrays, where no dielectric nanocrystals have been incorporated. Surprisingly, we find that the enhancement is not due to the third order susceptibility of the ITO nanocrystals, but due to the optical nonlinearity of the gold and predominantly due to an enhanced dipole moment of the plasmonic oscillations in the gold gap-antennas. [1] L. Novotny and N. van Hulst, Nature Photon. 5, 83 (2011). [2] M. Kauranen and A. V. Zayats, Nature Photon. 6, 737 (2012).

O 46.8 Wed 12:15 TRE Phy

Gold strip gratings for surface enhanced infrared spectroscopy — Tao Wang[1], •Tobias W. W. Mass[1], Vu Hoa Nguyen[2], Andreas Buchenauer[2], Uwe Schnakenberg[2], and Thomas Taubner[1,3] — [1]I. Institute of Physics (IA), RWTH Aachen University — [2]Institute of Materials and Electrical Engineering I, RWTH Aachen University — [3]Fraunhofer Institute for Laser Technology (ILT)

Metallic nanoantennas efficiently couple light into a region of subwavelength size. Antenna arrays designed for surface enhanced infrared absorption spectroscopy enable the detection of molecular vibration with high sensitivity [1,2]. In contrast to the cost-intensive and low-throughput fabrication of nanostructures e.g. via e-beam lithography, gold strip gratings fabricated with standard optical UV-lithography offer great potential for future commercial applications. The grating structures are analysed using FTIR-spectroscopy and exhibit grating resonances depending on the period and the refractive index of the environment. Accordingly, the grating resonances can be tuned over a broad spectral range in order to match and enhance the absorption bands of interest. We demonstrate enhancement factors of more than 6000 and significantly amplify the well defined absorption band of a 30 nm thick PMMA layer at about 5.8 μm [3].

[1] Adato et al. *PNAS* **2009** 106(46), 19227-19232.
[2] Neubrech et al. *Phys.Rev.Lett.* **2008** 101(15), 157403.
[3] Wang et al. *Opt. Express* **2013** 21(7), 9005-9010.

O 46.9 Wed 12:30 TRE Phy

Distinction between horizontal and vertical plasmonic modes in 3D nanostructures — •Monika Fleischer[1], Julia Fulmes[1], Christian Schäfer[1], Dieter Kern[1], Mikael Renault[2], Anne-Laure Baudrion[2], and Pierre-Michel Adam[2] — [1]Eberhard Karls University Tübingen, 72076 Tübingen, Germany — [2]University of Technology Troyes, 10000 Troyes, France

The dark-field scattering or extinction spectra of plasmonic nanostructures, such as e.g. nanorods, exhibit distinct peaks that indicate the resonances of the localized surface plasmon polaritons excited in the structures [1]. For three-dimensional nanostructures, vertical modes may occur as well. These are however rarely reported due to the predominant excitation and detection of in-plane modes. Here gold nanocone antennas with different aspect ratios are fabricated by two strategies using top-down lithography [2,3], and their plasmon resonance spectra in the far-field are analyzed by single-particle dark field scattering spectroscopy [4,5], variable angle UV-vis extinction spectroscopy, and total internal reflection spectroscopy. The study shows that both in-plane and out-of-plane modes of the 3D nanostructures can be excited, and that their relative spectral positions can be individually engineered via the nanocone geometry [3]. The experiments are compared with simulations for the same geometries.

[1] W. Gotschy et al., Opt. Lett. 21, 1099 (1996); [2] M. Fleischer et al., Nanotechnol. 21, 065301 (2010); [3] J. Fulmes et al., in preparation (2013); [4] C. Sönnichsen et al., New J. Of Physics 4, 93 (2002); [5] C. Schäfer et al., Nanoscale 5, 7861 (2013).

O 46.10 Wed 12:45 TRE Phy

Higher multipolar mode contributions to nonlinear nanoantenna response — Jan Renger[1], •Pablo M. de Roque[1], Nikos Fayard[1,2], Marta Castro-Lopez[1], Nicolo Accanto[1], and Niek F. van Hulst[1] — [1]ICFO-Institut de Ciencies Fotoniques, 08860 Castelldefels (Barcelona), Spain — [2]Institut Langevin, ESPCI ParisTech, France

Nanoantennas, such as metallic nanorods featuring localized surface plasmon resonances (LSPR), are attractive for application such as optical sensing of chemicals or biomedical markers because of their wide spectral-tunability and strong mode confinement and enhancement. The latter one together with the intrinsically strong nonlinearity of metals allows these antennas to act as coherent nano light sources. For a short gold nanorod, the dipolar LSPR occurring in the visible spectral range will dominate the optical response. This resonance rapidly shifts towards the near infrared for a longer nanorod and additional multipolar modes contribute in the visible and NIR wavelength range. We identified the resonance conditions for the different multipolar modes by tracking the linear and nonlinear optical properties of nanorods for increasing lengths and to demonstrate that dark and bright multipolar modes of metallic nanorods contribute equally to the nonlinear optical emission via two-photon photoluminescence (TPPL) and second harmonic generation (SHG).