O 13: Plasmonics and nanooptics: Light-matter interaction, spectroscopy I

Time: Monday 15:00–18:15 Location: MA 041

O 13.1 Mon 15:00 MA 041

Ultrafine-Dust Detection Based on Surface Enhanced Infrared Absorption Using Bowtie Nanoantennas — •Christian Huck[1], Michael Tzschoppe[1], Rostyslav Semenyshyn[2], Frank Neubrech[1], and Annemarie Pucci[1] — [1]Kirchhoff-Institute for Physics, Heidelberg Universtity, Germany — [2]4th Physics Institute and Research Center SCoPE, University of Stuttgart, Germany

In the last years, surface-enhanced infrared absorption (SEIRA) has been established as a powerful method for chemical structure analysis of thin molecular layers. Much less attention has so far been payed to other potential fields of application, such as the detection and characterization of small particles, *e.g.* ultrafine-dust particles. Here, we report on an experimental study, which shows, that deeply subwavelength sized particles can be detected and chemically analyzed by SEIRA. Our study is based on bowtie shaped apertures etched in a thin Au layer, used to enhance phononic excitations[1] of individual silica particles. As it turned out, the bowtie geometry features several advantages for the detection of ultrafine-dust particles, which we want to detail during the talk. Backed up by numerical calculations, we show that a detection limit in terms of a particle diameter of as low as 20 nm can be achieved, corresponding to a ratio to the wavelength of $\lambda^2/d^2 > 250000$. Our approach offers the possibility to analyze infrared vibrations from tiniest particles and thus paves the way toward SEIRA-based fine dust sensing devices.

[1] Huck *et al.* Opt. Express 24, 25528-25539 (2016)

O 13.2 Mon 15:15 MA 041

Continuous-Wave Multiphoton Photoemission from Plasmonic Nanostars — •Murat Sivis[1], Nicolas Pazos-Perez[2], Renwen Yu[3], Ramon Alvarez Puebla[2,4], F. Javier García de Abajo[3,4], and Claus Ropers[1] — [1]4th Physical Institute - Solids and Nanostructures, Georg-August University, Göttingen, Germany — [2]Department of Physical Chemistry and EMaS, Universitat Rovira i Virgili,Tarragona, Spain — [3]Institut de Ciencies Fotoniques, The Barcelona Institute of Science and Technology, Castelldefels, Spain — [4]Institució Catalana de Reserca i Estudis Avançats, Barcelona, Spain

Highly nonlinear optical photoemission from tailored plasmonic nanostructures, as used in localized electron sources, is typically restricted to ultrafast laser pulse excitation due to the required high local intensities. Here, we demonstrate localized 3-photon photoemission from chemically-synthesized plasmonic gold nanostars under continuous-wave illumination at sub-MWcm^{-2} incident intensities. Intensity- and polarization-dependent measurements confirm the nonlinear photoemission from the nanostar tips with feature sizes smaller than 5 nm, which facilitate near-field-intensity enhancement factors exceeding 1000. Our results highlight the possibilities for new designs of nanoscale coherent electron sources, with potential applications in microscopy, spectroscopy and sensing.

O 13.3 Mon 15:30 MA 041

Single Plasmonic Particle Chiral Spectroscopy — •Julian Karst, Mario Hentschel, Nikolai Strohfeldt, and Harald Giessen — 4th Physics Insitute and Research Center SCoPE, University of Stuttgart

Chirality plays a crucial role in our everyday lives. Virtually all studies are performed on large ensembles of chiral objects, which obstructs the contribution of the individual particle. Hence, it is highly desirable to study individual chiral objects. Here, we show that we are able to measure reproducibly circular dichroism spectra in the visible spectral range of left and right handed C4 symmetric single plasmonic oligomer. We utilize darkfield scattering spectroscopy as well as correlative scattering electron microscopy in order to study the influence of structural arrangement and shape on the optical properties of a single plasmonic oligomer. Our two-layered oligomers are fabricated by top-down electron beam lithography to offer a high degree of structural control. We find that even minute structural differences, which we cannot resolve in scanning electron microscopy, manifest themselves in clear differences in the chiroptical response. Our results play an important role for further investigations and optimizations in design, fabrication, and measurements of single plasmonic enantiomers, but also on the way towards single chiral molecule sensing.

O 13.4 Mon 15:45 MA 041

Wavelength-dependent Third Harmonic Generation in Plasmonic Gold Nanoantennas: Role of the d-band — •Joachim Krauth, Mario Hentschel, and Harald Giessen — 4^{th} Physics Institute and Research Center SCoPE, University of Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany

Plasmonic gold nanoantennas are highly efficient nanoscale nonlinear light converters. The nanoantennas provide large resonant light interaction cross sections as well as strongly enhanced local fields. The frequency conversion, however, takes places inside the gold volume and is thus ultimately determined by the microscopic gold nonlinearity. While the influence of the nanoantenna geometry has been studied in great detail, only little attention has been paid to the microscopic material nonlinearity. Here we show that the microscopic third-order nonlinearity of gold is in fact a resonant one by virtue of interband transitions between the d- and sp-bands. Utilizing a large set of resonant nanoantennas and an optical parametric oscillator as broadband tunable light source, we show that the radiated third harmonic (TH) signals significantly increase as soon as the TH becomes resonant with allowed interband transitions. With the help of an anharmonic oscillator model and independent reference measurements on a gold film we can unambiguously demonstrate that the observed TH increase is related to a strongly wavelength-dependent microscopic third-order gold nonlinearity. This additional tuning parameter allows further manipulation and optimization of nonlinear nanoscale systems and thus renders the investigation of other plasmonic materials highly intriguing.

O 13.5 Mon 16:00 MA 041

Nanoscale hydrogenography on individual magnesium nanoparticles — •Florian Sterl, Heiko Linnenbank, Tobias Steinle, Florian Mörz, Nikolai Strohfeldt, and Harald Giessen — 4th Physics Institute and Research Center SCoPE, University of Stuttgart, Germany

MgH_2 is considered a promising candidate for solid-state hydrogen storage, owing to its high hydrogen content. It can also be considered a model system for other energy storage materials, such as magnesium-based alloys. While the hydrogen diffusion mechanism in Mg at the 100 μm length scale has been investigated extensively using hydrogenography, in which the optical contrast between a metal and its hydride is exploited, an observation of this process at sub-diffraction limit length scales in single Mg nanoparticles has not been carried out so far.

In order to characterize this process at the nanoscale, we employ scattering type scanning near-field optical microscopy (s-SNOM). In this method, the local scattering ability of a nanoparticle is recorded along with its topography, enabling us to record the local material composition as well as topographical changes during hydrogen absorption and desorption. By combining s-SNOM imaging with dark-field scattering spectroscopy, we observe the distribution of metallic Mg and dielectric MgH_2 during both hydrogen absorption and desorption of Mg nanodisks. Our measurement results indicate a strong influence of the crystallinity, as individual Mg crystallites appear to hydrogenate independently from one another. In the future, this method can also be applied to other hydrogen-storage materials.

O 13.6 Mon 16:15 MA 041

Scanning Tunneling Microscopy Induced Luminescence of Bimodal Exciton-Plasmon Emitters — Pablo Merino[1,3], Anna Rosławska[1], Christoph Grosse[1,4], •Christopher Leon[1], Klaus Kuhnke[1], and Klaus Kern[1,2] — [1]Max Planck Institute for Solid State Research, Heisenbergstraße 1, 70569, Stuttgart, Germany. — [2]Institut de Physique, École Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland. — [3]present address: Instituto de Ciencia de Materiales de Madrid, CSIC, c/Sor Juana Inés de la Cruz 3, E28049, Madrid, Spain. — [4]present address: Nanophotonics Centre, Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, United Kingdom.

Scanning tunneling microscopy induced luminescence (STML) is a unique method to locally excite and observe excitons and plasmons in an electroluminescent material, leading to the implementation of

a combined exciton-plasmon light source. The light emission from a proof-of-concept buckyball-based system is characterized at the nanometer scale and the relative contributions of light originating from excitons and plasmons can be reproducibly tuned by STM parameters under exclusion of any tip modification. This ability is intimately related to an understanding of intrinsic properties of excitons and plasmons, namely, their formation and decay mechanisms in conjunction with their associated timescales.

15 min. break

O 13.7 Mon 16:45 MA 041

Localization of photonic modes in optimised disordered amorphous silicon thin films — Martin Aeschlimann[1], •Felix Becker[2], Tobias Brixner[3], Benjamin Frisch[1], Michael Hartelt[1], Matthias Hensen[3], Thomas H. Loeber[4], Walter Pfeiffer[2], Sebastian Pres[3], Bernd Stannowski[5], and Helmut Stiebig[2] — [1]Fachbereich Physik and Research Center OPTIMAS, TU Kaiserslautern, Erwin-Schrödinger-Str. 46, 67663 Kaiserslautern — [2]Fakultät für Physik, Universität Bielefeld, Universitätsstr. 25, 33615 Bielefeld — [3]Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg — [4]Nano-Structuring-Center, Erwin-Schrödinger-Str. 13, 67663 Kaiserslautern — [5]Helmholtz-Zentrum Berlin, PVcomB, Schwarzschildstr. 3, 12489 Berlin

Tailored disordered nanostructures that feature long-living photonic modes are employed to enhance local light scattering, light localization, and absorption. We demonstrate the controlled fabrication of nanotextured a-Si:H absorber layers using focused ion beam milling of planar ZnO substrates and PECVD. This allows studying the light absorption in nanotextured absorber layers with custom designed topographies. Light trapping and absorption in these samples is investigated by time- and energy-resolved PEEM. We observe both, field induced multiphoton photoemission from the scattered light fields in the absorber layer as well as nanolocalized photonic modes featuring highly nonlinear thermionic electron emission. The obtained map of photonic modes and absorption patterns is compared to FTDT simulations.

O 13.8 Mon 17:00 MA 041

Anomalous Kerr effect in hybrid Bi-YIG/Au magneto-plasmonic nanostructures — •Spiridon D. Pappas, Philipp Lang, and Evangelos Th. Papaioannou — Fachbereich Physik und Landesforschungszentrum OPTIMAS, Technische Universität, 67663 Kaiserslautern, Germany

The magneto-optical effects exhibited by ferromagnetic materials can be enhanced by the strong localization of light, enabled by adjacent plasmonic resonances [1,2]. In this work, we reveal the anomalous enhancement of the longitudinal magneto-optical Kerr effect (L-MOKE) in the ferrimagnetic dielectric bismuth substituted yttrium iron garnet (Bi-YIG), induced by localized surface plasmons (LSPs) in gold nanoparticles (AuNPs). In order to reveal the resonant frequency of the LSPs, we used optical spectrophotometry. A Bi-YIG sample containing no nanoparticles was also used as a reference. The magneto-plasmonic response was studied with the aid of a spectroscopic MOKE setup operating in the longitudinal mode, equipped with a filtered supercontinuum laser source. The experimental results reveal an anomalous enhancement of the L-MOKE signal (Kerr rotation, Kerr ellipticity) for the sample containing AuNPs close to the LSP resonances. In order to gain qualitative insight into the mechanism of the exhibited anomalous L-MOKE, the Near Field enhancement close to the LSP frequencies was simulated computationally.

[1] V. I. Belotelov et al., Nat. Nanotechnol. 6, 370 (2011).
[2] M. Rollinger et al., Nano Lett. 16, 2432 (2016).

O 13.9 Mon 17:15 MA 041

Visualization of photoemission quantum pathways via photon-plasmon spin-orbit mixing — •Eva Prinz[1], Grisha Spektor[2], Deirdre Kilbane[3], Anna-Katharina Mahro[1], Michael Hartelt[1], Meir Orenstein[2], and Martin Aeschlimann[1] — [1]Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, Germany — [2]Department of Electrical Engineering, Technion - Israel Institute of Technology, Israel — [3]School of Physics, University College of Dublin, Ireland

Transformation of light carrying spin angular momentum (SAM) to an optical field vortex carrying angular momentum (OAM) has been of wide interest in recent years [1]. We measured the interaction between 3D light carrying axial SAM and 2D plasmon-polariton vortices carrying high order transverse OAM [2].

The interaction is mediated via two-photon absorption on a gold surface, imprinting the resulting angular momentum mixing into matter by excitation of electrons that are photo-emitted into vacuum and detected by PEEM. We show experimentally and theoretically that the absorptive nature of this interaction leads to both single and double photon-plasmon angular momentum mixing processes which correspond to different quantum pathways of the electron excitation.

[1] Bliokh et al., Nature Photonics 9 (2015)
[2] Spektor et al., Science 355 (2017)

O 13.10 Mon 17:30 MA 041

Electron-induced photon emission above the quantum cutoff due to time-energy uncertainty — •Kira Kolpatzeck, Philip Kapitza, Ebru Ekici, Christian A. Bobisch, and Rolf Möller — Faculty of Physics, Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, 47057 Duisburg, Germany

We have studied the light emission resulting from inelastic tunneling processes in the tip/sample junction between a silver covered PtIr tip and an Ag(111) surface with a low-temperature scanning tunneling microscope at 80 K. The observed light emission between the visible and the near infrared range carries information about the underlying inelastic processes. It has been studied around the cutoff at $h\nu = eV_T$.

A detailed analysis of the emission spectra normalized to a reference spectrum recorded at high voltages shows emission tails exceeding the energy eV_T provided by the the applied voltage V_T. For a satisfying explanation the consideration of thermal broadening of the electron Fermi distribution is insufficient. However, a correct description is found if a finite lifetime of the excited states in the range of 30-80 fs, causing a natural linewidth broadening, is included [1].

[1] E. Ekici, P.Kapitza, C. A. Bobisch, R. Möller, Opt. Lett. 42, 4585-4588 (2017).

O 13.11 Mon 17:45 MA 041

Measuring the Orbital Angular Momentum of Plasmonic Vortex Fields — •Pascal Dreher[1], David Janoschka[1], Norman Dünne[1], Bettina Frank[2], Timothy J. Davis[2,3], Harald Giessen[2], Michael Horn-von Hoegen[1], and Frank Meyer zu Heringdorf[1] — [1]Faculty of Physics and Center for Nanointegration (CENIDE), University of Duisburg-Essen, Duisburg, Germany — [2]4th Physics Institute and Stuttgart Center of Photonics Engineering (SCoPE), University of Stuttgart, Stuttgart, Germany — [3]School of Physics, University of Melbourne, Parkville, Victoria, Australia

The ability of light to carry orbital angular momentum in addition to its spin angular momentum has been thoroughly investigated in the past. Recent efforts have shown that the combination of these angular momenta can be utilized to excite surface plasmon polaritons with helical wavefronts, i.e. plasmonic vortices. The angular momentum of the exciting light is completely transferred to the plasmon. Further angular momentum can be transferred to the plasmonic field in the form of a topological charge provided by helical nanostructures, e.g. Archimedean spirals. We will report on the imaging of plasmonic vortex fields employing pump-probe photoemission electron microscopy (PEEM) with sub-femtosecond time resolution. We show that the angular momentum state of plasmonic vortices can be extracted from time-integrated PEEM micrographs, by analyzing the plasmonic field distribution for Archimedean spirals with topological charges from one to ten. The presented results agree with the predictions of the Bessel theory for plasmonic vortices.

O 13.12 Mon 18:00 MA 041

Circular dichroism in laser induced electron emission from metallic nanohelixarrays — •Daniel Nürenberg[1], Andrew G. Mark[2], Peer Fischer[2], and Helmut Zacharias[1] — [1]Physikalisches Institut & Center For Soft Nanoscience, Münster, Germany — [2]Max Plank Institut für intelligente Systeme,Stuttgart, Germany

We investigate the electron emission from metallic nanohelices on silicon triggered by fs laser radiation from an optical parametric chirped pulse amplifier. We find a strong asymmetry regarding to left and right circularly polarized excitation in the electron yield in accordance to the handedness of the helices. The emission can be interpreted in terms of a non-equilibrium heating of the electron gas by surface plasmon excitation. Simulations of the surface plasmons correlate the polarization dependence of the photosignal to the local field enhancement on the nanostructures.