O 83: Poster Session - Plasmonics and Nanooptics: Light-Matter Interaction, Spectroscopy

Time: Wednesday 18:15–20:00 Location: P2/EG

O 83.1 Wed 18:15 P2/EG

Steps towards higher harmonics pump-probe spectroscopy — •Julian Obermeier, Thorsten Schumacher, and Markus Lippitz — University of Bayreuth, Germany

In the last decade the field of nonlinear plasmonics has been growing rapidly. Many fascinating experiments have shown the nonlinear emission of complex nanostructures, methods to enhance the emission and that the nonlinear near field of nanostructures can be shaped remotely. Yet, also some fundamental questions are still unanswered, for example, how nonlinear plasmonic effects are connected to the band structure and especially the electronic occupation function. A well known way to cause time-dependent modifications of both is intense pulsed optical pumping. Interband absorption followed by Auger recombination as well as rethermalized electron distributions both lead to a transiently drastically increased electron temperature.

Here we investigate the influence of hot electrons on the nonlinear optical response of plasmonic particles and structures. This will not only give us new insight in the nonlinear processes themselves but also allow us to deterministically alter them. We present our higher harmonics pump-probe setup and experimental results of pump induced changes in the third harmonic emission of plasmonic structures.

O 83.2 Wed 18:15 P2/EG

Measuring Hot-Electron Dynamics by Pump-Probe Spectroscopy — •Fabian Paul, Julian Obermeier, Christoph Schnupfhagen, Thorsten Schumacher, and Markus Lippitz — University of Bayreuth, Germany

After excitation of a plasmonic structure with an ultrashort laser puls an athermal occupation function of electrons is generated that rethermalizes to a Fermi-Dirac distribution within about 15 fs. During the next tens of femtoseconds the most energetic electrons relax due to Coulomb interactions between each other and at last on a scale of several picoseconds interacting with the lattice structure relax back to thermal equilibrium. Due to the enormous improvement of super continuum lasers and pulse shapers in the last years, it is possible to generate femtosecond laser pulses and shape them nearly unlimited, so that one can easily do pump-probe experiments.

With this techniques it is now possible to measure the decay of these high energetic electrons on ultrafast timescales. We are using in the first step plasmonic nano-structures to increase the signal. The results will improve our understanding of the microscopic origin of plasmonic nonlinearities.

O 83.3 Wed 18:15 P2/EG

Nanoparticle-on-mirror systems for strong coupling with emitters — •Christoph Schnupfhagn, Thorsten Schumacher, and Markus Lippitz — Experimental Physics III, University of Bayreuth, Germany

Plasmonic nanostructures allow to increase the interaction with light by resonant oscillations of the conduction electrons. Over the last years, the nanoparticle-on-mirror geometry has received increasing interest, where plasmonic nanoparticles are placed above a metallic substrate. If the gap size between particle and substrate is on the order of only a few nanometers, the systems couple and form a cavity mode. In the literature, mode volumes as small as 40 nm^3 have been realized. When incorporating emitters into the gap, this allows for strong coupling between emitter and the plasmonic mode. Here, we will present results on the spectroscopy of colloidal gold nanoparticles on top of crystalline gold flakes. Furthermore, we will give an outlook how emitters could be positioned in the cavity.

O 83.4 Wed 18:15 P2/EG

Coupling single quantum dots to plasmonic waveguide structures — •Michael Seidel[1], Gerhard Schäfer[1], Peter Schnauber[2], Armando Rastelli[3], Stephan Reitzenstein[2], and Markus Lippitz[1] — [1]Experimental Physics III, University of Bayreuth, Bayreuth, Germany — [2]Institute of Solid State Physics, Technische Universität Berlin, Berlin, Germany — [3]Institute of Semiconductor and Solid State Physics, Johannes Kepler University, Linz, Austria

Integrated plasmonic nanocircuits are highly promising building blocks for future quantum optical applications. In combination with self-assembled quantum dots as stable, bright and narrow-band single-photon sources, ultra-compact nanocircuits operating below the diffraction limit can be designed. A crucial aspect is the coupling of the quantum dot emission into plasmonic waveguide modes. In this work, we propose a plasmon–quantum dot hybrid structure which offers a highly efficient coupling scheme due to enhanced light-matter-interaction. We perform numerical simulations to optimize the coupling efficiency and show first experimental steps towards fabricating the proposed structure.

O 83.5 Wed 18:15 P2/EG

Step towards Two Dimensional Electronic Spectroscopy (2DES) of a single molecule using fluorescence based detection scheme — •Sanchayeeta Jana, Christoph Schnupfhagn, and Markus Lippitz — Experimental Physics III, University of Bayreuth, Bayreuth, Germany

2DES is an ultrafast spectroscopic technique which can give us the information about the couplings between the molecular energy levels due to the added dimensionality. This technique uses two replicas of an ultrashort laser pulse with controllable delay between them to excite the sample and a third pulse probes the excited sample after some time. But performing this kind of experiments on a single molecule is extremely difficult since the absorption cross section of a single molecule is much smaller than the probe volume resulting very low signal to background noise ratio. To overcome this, fluorescence based detection scheme can be used where fluorescence from the excited state is measured using an additional 4th pulse.

In this work, we describe the experimental challenges for such an experiment on a single molecule and propose the convenient procedure to overcome those.

O 83.6 Wed 18:15 P2/EG

Quantifying quantum dot fluorescence in the near-field — •Niklas Stenger, Thorsten Schumacher, Jonas Albert, and Markus Lippitz — University Bayreuth, Chair Experimental Physics III

The spatial resolution in the far field is limited by diffraction. To overcome this limitation, atomic force microscopy can be used. We present a powerful tool to investigate the light-matter interaction on the nanoscale: an AFM combined with a time-correlated single photon counting setup to measure fluorescence as well as the probe topography. The AFM tip used to investigate the tip-induced quenching of quantum dots in the near-field. The influence of factors like tip geometry, tip-probe distance and wavelength on the fluorescence signal will be presented.

O 83.7 Wed 18:15 P2/EG

Spatially-resolved hyper spectral single quantum dot strong coupling — •Benedikt Schurr, Daniel Friedrich, Heiko Gross, and Bert Hecht — NanoOptics & Biophotonics Group, Experimental Physics 5, University of Würzburg, Germany

Strong coupling single emitters and plasmonic nano resonators at ambient conditions opens new prospects for applications of quantum technologies. In the past decade multiple types of photonic cavities and emitters were investigated. [1] However only recently it became possible to directly tune the coupling strength by means of scanning probe technology. [2]

Here, we report a qualitative hyperspectral analysis of strong coupling between single colloidal CdSe quantum dots and plasmonic nanoresonators fabricated at a scanning probe tip. When using precise helium ion beam milling to achieve sub 10 nm light localization, we observe an increased Rabi splitting of up to 160 meV. The larger splitting in combination with high-resolution scanning allows us to record spatially resolved maps of the coupling strength between the emitter and the plasmonic resonator mode.

[1] O. Bitton et al., Nanophotonics 8 (4), 559-575 (2019)
[2] H. Gross et al., Sci. Adv. 4, eaar4906 (2018)

O 83.8 Wed 18:15 P2/EG

Massive parallel heterogeneous assembly of single-crystalline Ag and Au spheres — •Daniel Schletz[1], Johannes Schultz[2], Martin Mayer[1], Anja Maria Steiner[1], Axel Lubk[2], Tobias

König[1], and Andreas Fery[3] — [1]Leibniz-Institut für Polymerforschung Dresden e.V., Germany — [2]Leibniz-Institut für Festkörper- und Werkstoffforschung Dresden, Germany — [3]Department of Physical Chemistry of Polymeric Materials TU Dresden, Germany

In pursuit of new optical properties, different materials and particle morphologies have been investigated in detail. Combining multiple of these building block types promises novel properties with respect to the single use thereof. Due to the nature of combinatorics, introducing more than a single building block leads to an exponential increase in parameter space, unable to be explored by established linearly scaling fabrication methods.

In an innovative approach, heterogeneous single particle lines of single-crystalline Ag and Au spheres are manufactured in a massive parallel approach by stochastic confinement assembly. By assembling billions of particles in one step, millions of combinations are generated. These ensembles are then studied in detail by electron energy loss spectroscopy (EELS) to reveal their plasmonic properties and to gain insight into heterogeneous coupling efficiencies.

O 83.9 Wed 18:15 P2/EG

Nano-scale Analysis of Surface Phonon Polaritons in $SrTiO_3$ using a scattering-type Scanning Near-field Optical Microscope — •Daniel Wendland[1], Julian Barnett[1], Marc Rose[2], Felix Gunkel[2], Regina Dittmann[2], and Thomas Taubner[1] — [1]Institute of Physics (IA) RWTH Aachen — [2]Peter Grünberg Institut, Forschungszentrum Jülich GmbH

Metasurfaces can arbitrarily manipulate light through resonant structures. Many metasurfaces are based on plasmonic materials, but phonon-based metasurfaces are on the rise, benefiting from the excitation of Surface Phonon Polaritons (SPhP) which can offer low losses and high propagation lengths [1]. $SrTiO_3$ (STO) could be an interesting candidate, because it supports strong phonon modes in its undoped state as well as nonvolatile resistive switching, which gives control over the local doping level. This switching process is reversible and could allow rewritable metasurfaces. To estimate the viability for SPhP-based resonator metasurfaces, we will investigate the SPhP behaviour in STO for different resonator geometries and doping levels. The latter allows us to gain a better understanding of plasmon-phonon-coupling [2], which is fundamental for this type of resonators. Here Scattering-type Scanning Near-field Optical Microscopy is applied to obtain nanoscale IR spectra and images. Using these results, we aim to predict the behaviour of STO resonator arrays and the viability of resistive switching for phonon-based metasurfaces.
[1] J. D. Caldwell et al., Nanophotonics, 4, 44 (2015)
[2] M. Lewin et al., Adv. Funct. Mater. 28, 1802834 (2018).

O 83.10 Wed 18:15 P2/EG

Towards Plasmonic Transistors - Plasmon damping on J-aggregated Squaraine dye coated gold films — •Christoph Bennenhei[1], Moritz Gittinger[1], Sven Stephan[1], Jennifer Zablicki[2], Arne Lützen[2], Manuela Schiek[1], Christoph Lienau[1], and Martin Silies[1] — [1]Carl von Ossietzky Universität Oldenburg — [2]Rheinische Friedrich-Wilhelms-Universität Bonn

Surface plasmon polaritons (SPPs) generated in metallic waveguides provide the possibility to manipulate light below the diffraction limit and are hence a first building block towards all-optical switching on the nanoscale. For ultrafast all-optical switching, coupling of the SPP modes to highly nonlinear excitonic materials [1] is necessary. We present J-aggregated squaraine dye (SQ) thin films as a promising candidate, since it shows a broad spectral resonance tunable by a thermal annealing process [2]. Here, we investigate power-dependent photobleaching of 10nm spin-coated ProSQ-C16 films in a confocal reflection spectroscopy setup, in a second step, we study the damping of the SPP modes propagating along the metal-SQ film interface by measuring the amount of light that outcoupled from pairs of grating couplers with varying distances. We find a significantly reduced decay length of the SPP for the SQ-metal interface around the excitonic resonance compared to an air-metal interface. High oscillator strength and photostability of SQ motivate future investigations of both optical pumping to invert damping rates as well as coherent energy transfer between SQ and SPPs. [1] P. Vasa, et al., Nature Photonics, 7 (2), pp 128-132 (2013) [2] M. Schulz, et al, Nat Commun 9, 2413 (2018)

O 83.11 Wed 18:15 P2/EG

Plasmoemission from a Plasmonic Nanofocus — •Jan-Henrik Herrig, Pascal Dreher, David Janoschka, Ping Zhou, Michael Horn-von Hoegen, and Frank Meyer zu Heringdorf — Faculty of Physics, University of Duisburg-Essen, Germany

Nonlinear photoemission from metal surfaces can be manipulated by strong plasmonic fields that are present during the emission process. Electron emission exclusively from such strong plasmonic fields (plasmoemission) can be distinguished from photoemission by spatio-temporal separation [1]. Obtaining the necessary field intensities requires efficient excitation of strong plasmonic fields.

We employed amplified laser pulses to excite surface plasmon polaritons in Archimedean spirals [2], which transiently form a diffraction-limited nanofocus if illuminated by circularly polarized light of the appropriate handedness. Plasmonic field intensities on the order of 10^{13} W/cm^2 were obtained at the Au(111) surface. The energetic distribution of the emitted electrons reveals the interaction with the plasmonic field.

[1] Podbiel *et al.*, Nano Letters 2017, 17(11): 6569-6574.
[2] Spektor *et al.*, Science 2017, 355(6330): 1187-1191.

O 83.12 Wed 18:15 P2/EG

Far-infrared sum-frequency generation wide-field microscopy — •Richarda Niemann, Riko Kiessling, Christopher Winta, Sören Wasserroth, Yujin Tong, Martin Wolf, and Alexander Paarmann — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

Super-resolution microscopy typically makes use of nonlinear-optical effects, such as multi-photon absorption or stimulated emission depletion microscopy, to overcome the diffraction limit of optical beams. In the infrared spectral range, infrared-visible sum-frequency generation (SFG) can provide vibrational contrast with a spatial resolution ultimately limited by the visible SFG wavelength [1]. In particular for far-infrared SFG, this would enable imaging resolution well below $\lambda_{IR}/100$.

As a proof-of-concept, we recently demonstrated SFG microscopy beyond infrared wavelength of 10 μm for surface phonon polariton resonance imaging [2], using a scanning-focus method. In the current work, we introduce our new approach of SFG wide-field microscopy [1]. Specifically for high-intensity, low-repetition rate lasers, the usage of wide-field nonlinear imaging efficiently avoids otherwise ubiquitous sample damage problems. Here, the laser focal size is adapted to the field-of-view of the imaging system, reducing the laser fluence without deteriorating the microscopes resolution [1].
[1] Hoffmann et al.,Rev. Sci. Instrum, 3221 (2002)
[2] Kiessling et al., ACS Photonics, (2019)

O 83.13 Wed 18:15 P2/EG

Coupling of quantum emitters to localized photonic modes in disordered structures — •Patrick Folge, Mohsen JaniPour, Ruben Pompe, Felix Fenner, and Walter Pfeiffer — Fakultät für Physik, Universität Bielefeld, 33615 Bielefeld, Germany

We investigate the coupling between quantum emitters to localized photonic modes in disordered dielectric waveguide structures. Localization of electromagnetic fields facilitates enhanced light matter interaction as it is commonly used in photonic crystal cavities. Periodic placement of holes in a two-dimensional waveguide forms photonic crystals with photonic band gaps that enable the formation of defect induces localized modes. Here we focus on disordered placement of these holes in a dielectric waveguide to achieve localization. Therefore we perform FDTD field simulations to calculate the Purcell factor, which can quantifies the light matter interaction. And we investigate the spectral features of the modes in the disordered structures. Thereby we find Purcell factors up to 25.

O 83.14 Wed 18:15 P2/EG

Low temperature scanning tunneling microscope light emission. — Liv Hornekaer and •Richard Balog — Aarhus University, Aarhus, Denmark

I will present modifications implemented in a Createc type low temperature scanning tunneling microscope (LT-STM) situated at iNano, Aarhus University, Denmark, expanding the capabilities of the setup towards tip-induced light emission experiments. Here, the light generated by tunneling electrons is collected by optics located nearby the STM head and then spectrally resolved and detected by a high efficiency LN2 cooled detector. The overall light detection system is optimized for the VIS-NIR range (400 nm - 1000 nm). The design and preliminary STM-light emission experiments on an Au(111) substrate and graphene covered SiC (0001) will be presented, demonstrating the full potential of the setup to reveal complex opto-electronic properties of the samples.