O 24: Poster Session II: Plasmonics and nanooptics II

Time: Monday 13:30–15:30 Location: P

O 24.1 Mon 13:30 P

Direct observation of Surface Plasmon Polaritons on permalloy nanostructures with femtosecond photoemission electron microscopy — •Maximilian Paleschke and Wolf Widdra — Martin-Luther-Universität Halle-Wittenberg, Halle (Saale), Germany

Due to remarkable progress in nanofabrication on the one hand and magneto optical methods on the other, the vivid field of magnetoplasmonics has become more and more accessible by a variety of excitation and investigation techniques.

Although it is well established that surface plasmon polaritons (SPP) can be imaged by photoemission electron microscopy (PEEM) in silver nanostructures[1], similar observations have not been reported for ordinary ferromagnetic materials like iron or nickel. Here, we report on dichroism images in threshold photoemission facilitating a tuneable femtosecond laser setup. The dichroism images show clear edge-induced standing waves with sub-micrometer wavelength. Analyzing the observed beating pattern as well as the coupling of the photon's spin-angular momentum to the direction of the fringe fields hint to propagation characteristics exclusive to evanescent waves, such as SPPs.[2, 3] This implies the possibility that many materials with a high plasma frequency allow for excitation and experimental observation of SPPs via this method.

[1] M. Dabrowski *et al.*, Chem. Rev. **120**, 6247 (2020)
[2] A. Y. Bekshaev *et al.*, Nature Com. **5**, 8 (2014)
[3] Y. Dai *et al.*, ACS Photonics **6**, 2005 (2019)

O 24.2 Mon 13:30 P

Particle Plasmon Induced Electronic Excitations on Silicon and Tetracene Crystals — •Katharina Engster[1,2], Kevin Oldenburg[2], Chris Rehhagen[1,2], Stefan Lochbrunner[1,2], Karl-Heinz Meiwes-Broer[1,2], Sylvia Speller[1,2], and Ingo Barke[1,2] — [1]Institut für Physik, Universität Rostock, Germany — [2]Department Leben-Licht-Materie, Universität Rostock, Germany

Utilizing plasmonics of metal nanoparticles to locally excite organic semiconductors is promising with respect to transfer energy from a defined starting point over longer distances. Two-photon photoemission electron microscopy (2P-PEEM) enables observation of near-fields with high lateral resolution due to the increased photoemission yield. With excitation spectroscopy we show that the particles can be addressed individually by their distinct plasmonic properties [1]. This results in an enhanced excitation of both, the substrate and nearby organic structures. The coupling to the substrate was studied by analyzing the occurring surface photovoltage (SPV) on Si(100)-(2x1). The distance dependence from the particles reveals an enhanced plasmon induced charge carrier density in the vicinity of the nanoparticles. For tetracene nanostructures we observe a plasmonically assisted local triplet exciton density. We do not observe clear migration in these small structures, which we attribute to inhomogeneities of the molecule aggregate [2]. Therefore, we present first measurements on larger, solution grown tetracene microcrystals.

[1] K. Oldenburg et al., J. Phys. Chem. C 123 (2019).
[2] G. M. Akselrod et al., Nat. Commun. 5 (2014).

O 24.3 Mon 13:30 P

Combining GW-BSE and PCM approaches for the description of real time electronic dynamics of molecules close to a plasmonic nanoparticle: application to LiCN and p-nitroaniline (PNA) molecules. — •Margherita Marsili and Stefano Corni — Dipartimento di Scienze Chimiche, Università di Padova, via Marzolo 1, Padova, Italy

In the presence of a plasmonic nanoparticle (NP) the optical response of molecules is strongly modified. The theoretical modeling of this phenomenon is especially challenging due to the inherent multiscale nature of the system. Recently, the simulation of the simultaneous electronic dynamics of molecule and NP was achieved by combining a time-dependent configuration interaction approach for the molecule, and a description of the NP as a continuous dielectric. The model takes into account the reciprocal effect of the molecule and NP time-dependent polarization, and the coupling with an external electromagnetic field. We combine this approach with the description of the molecule at the GW-BSE level, which is suited to treat extended and charge-transfer systems. We apply this methodology to study Rabi oscillations of the ground and excited states population of a LiCN molecule, an ideal test system for the study of optical dipole switching. The molecule is set at increasing distances with respect to a plasmonic NP probing the local field enhancement and the strength of the mutual interaction. Moreover, the population and dipole dynamics of the prototypical push–pull PNA molecule in proximity of a tip-shaped NP is studied with the tip scanning the molecule at different positions.

O 24.4 Mon 13:30 P

Mechano-optical switching of a single molecule with doublet emission — Jiří Doležal[1], Pingo Mutombo[1], Dana Nachtigallová[2], Pavel Jelínek[1], •Pablo Merino[3], and Martin Švec[1] — [1]Institute of Physics, Czech Academy of Sciences, Praha, Czech Republic — [2]Institute of Organic Chemistry and Biochemistry of the Czech Academy of Sciences, Praha, Czech Republic — [3]Instituto de Ciencia de Materiales de Madrid, CSIC, Sor Juana Inés de la Cruz 3, E28049, Madrid, Spain

Phthalocyanine and derived metal complexes on thin insulating layers studied by scanning tunneling microscope-induced luminescence (STML) offers an excellent playground for tuning their excitonic and electronic states by Coulomb interaction and to showcase their high environmental sensitivity.[1] Copper phthalocyanine (CuPc) has an open-shell electronic structure and its lowest-energy exciton is a doublet which brings interesting prospects in its application for optospintronic devices. Here, we demonstrate that the excitonic state of a single CuPc molecule can be reproducibly switched by atomic scale manipulations permitting precise positioning of the molecule on the NaCl ionic crystal lattice.[2] Using a combination of STML, AFM and ab-initio calculations, we show the modulation of electronic and optical bandgaps, and the exciton binding energy in CuPc by tens of meV. We explain this effect by spatially-dependent Coulomb interaction occurring at the molecule-insulator interface, which tunes the local dielectric environment of the emitter.References:[1] J. Doležal et al., Nano Letters, 19 8606 (2019).[2] J. Doležal et al., ACS Nano, 14, 8931 (2020).

O 24.5 Mon 13:30 P

Vibrational Heating of a Single C60 Molecule in a Current-Carrying Plasmonic Picocavity — •Borja Cirera[1], Chenfang Lin[1], Shuyi Liu[1], Yair Litman[2], Alaa Akkoush[2], Martin Wolf[1], Mariana Rossi[2], and Takashi Kumagai[1,3] — [1]PC Department Fritz Haber Institute, Berlin, Germany — [2]MPI for Structure and Dynamics of Matter, Hamburg, Germany — [3]IMS, Okazaki, Japan

Vibrational heating in single-molecule junctions under non-equilibrium conditions is of fundamental importance in molecular electronics. Here we report on single-molecule thermometry of fullerene (C60) in a current-carrying plasmonic picocavity by combining tip-enhanced Raman spectroscopy (TERS) with low-temperature scanning tunneling microscopy (STM). Thanks to the exceptional sensitivity of TERS, intense Stokes and anti-Stokes Raman bands can be observed from single C60, enabling direct access to heating of individual vibrational modes. The precise gap-distance control in STM allows performing single-molecule thermometry in two distinct regimes, where the molecule is in contact with one or two electrodes (tip/surface), respectively. In both cases, optically induced heating is dominant for low bias voltages. At high bias voltages, the vibrational population is increased by injecting electrons into the LUMO of C60 in tunneling conditions. Upon contacting with both electrodes, the molecule strongly hybridizes with metallic states and vibrational pumping is dominated by inelastic electron scattering due to a much larger current density.

O 24.6 Mon 13:30 P

Plasmon-excited near-field luminescence of semiconductor quantum dots — •Vlastimil Křápek, Petr Dvořák, Lukáš Kejík, Michal Kvapil, Petr Liška, Jan Krpenský, and Tomáš Šikola — Brno University of Technology, Brno, Czech Republic

On-chip integration of light sources would benefit from near-field handling of the emission with a subwavelength spatial resolution. Here we present a fully near-field photoluminescence study of semiconductor quantum dots, with a surface plasmon interference device (SPID) used for the excitation and an aperture-type scanning near-field optical microscope (SNOM) combined with a spetrometer for the collection.

The SPID consists of an opaque metallic layer with thin subwave-

length slits fabricated using focused-ion-beam milling. When illuminated from bottom, a near-field standing wave forms at the top interface [1,2,3] where it excites quantum dots deposited directly at the top interface.

We demonstrate the plasmon-excited near-field luminescence of CdSe/ZnS quantum dots with rather a weak effect of the excitation mechanism on the spectral profile of the emitted light. This makes the plasmon-excited luminescence a suitable tool for the on-chip integration of semiconductor light sources, as well as a characterization technique with the subwavelength spatial resolution.

[1] P. Dvořák et al., Nano Lett. **13**, 2558 (2013).
[2] P. Dvořák et al., Opt. Express **25**, 16560 (2017).
[3] P. Dvořák et al., Nanoscale **45**, 21363 (2018).

O 24.7 Mon 13:30 P

Hot electrons masquerading as plexcitons in J-aggregate/plasmonic nanoparticle systems — •Victoria C. A. Taylor[1], Sara Núñez-Sánchez[2], and Thomas A. A. Oliver[3] — [1]Fritz Haber Institute of the Max Planck Society, Faradayweg 4-6, 14195 Berlin, Germany — [2]Universidade de Vigo, Departamento de Química Física, Campus Universitario As Lagoas, Marcosende, 36310 Vigo, Spain — [3]School of Chemistry, University of Bristol, Bristol, BS8 1TS, UK

Surface plasmon polaritons (SPPs) can produce localised high electric field strengths. Through strong coupling with quantum emitters, they can create hybrid states with novel and promising properties. Excitons in J-aggregates, coupled with SPPs on metallic nanoparticles, are ideal systems for studying these effects. The hybrid quasiparticles that form in these systems under strong coupling are sometimes referred to as plexcitons (plasmon/exciton). Several studies have investigated the dynamics of these plexcitons and report lifetimes on the order of picoseconds, which is in strong contrast to the tens of femtoseconds lifetimes that are typically associated with SPPs.

In this work, we investigate the ultrafast transient absorption response of a TDBC J-aggregate/hollow gold nanoshells (HGNs) hybrid system. Through our excitation wavelength dependence measurements, and comparison with the response of the uncoupled HGNs, we show that the observed picosecond signal does not result from long-lived plexcitons; rather, it is caused by the hot electron population that remains after the plexciton has dephased.