O 58: Plasmonics and Nanooptics II: Light-Matter Interaction and Spectroscopy I

Time: Wednesday 15:00–17:30 Location: WIL A317

O 58.1 Wed 15:00 WIL A317

Simultaneous Strong Coupling of the H- and J-Bands of Molecular Aggregates in Microcavities — •Roland Schäfer[1], Lukas Böhner[1], Manuela Schiek[2,3], Klaus Meerholz[1], and Klas Lindfors[1] — [1]Department für Chemie, Universität zu Köln, 50939 Köln, Germany — [2]LIOS & ZONA, Johannes Kepler Universität, 4040 Linz, Austria — [3]Institut für Physik, Carl von Ossietzky Universität Oldenburg, 26129 Oldenburg, Germany

We simultaneously strongly couple the H- and J-bands of aggregates of a merocyanine dye (HB238 [1]) to cavity photons. This is achieved by tuning the $3\lambda/2$- and 1λ mode of a planar microcavity to the spectral positions of the H- and J-bands, respectively. Strong coupling behavior is confirmed via the observation of Rabi-splitting of both bands in angle-resolved reflectivity spectra.

Due to the orientation of the aggregates J-polaritons can be observed with s- and p-polarized light, while the H-polaritons can only be accessed with p-polarized light and a non-normal angle of incidence. Therefore, our system supports two or four polaritons, depending on the polarization, making it an interesting platform to study strong light-matter coupling.

[1] Bürckstümmer, H., et. al., *Angew. Chem. Int. Ed.* **2011**, 50: 11628-11632.

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O 58.2 Wed 15:15 WIL A317

Two-Dimensional Electronic Spectroscopy of Strong Exciton-Surface Plasmon Polariton Coupling — •Daniel Timmer[1], Moritz Gittinger[1], Thomas Quenzel[1], Sven Stephan[1], Jennifer Zablocki[2], Arne Lützen[2], Jin-Hui Zhong[1], Martin Silies[1], Antonietta De Sio[1], and Christoph Lienau[1] — [1]University of Oldenburg, Germany — [2]University of Bonn, Germany

The creation of hybrid light-matter systems due to strong coupling is of major current interest for material science. Of special interest are dipolar interactions between molecular excitons (X) and plasmonic resonators, such as surface plasmon polaritons (SPP). Sufficiently strong coupling to vacuum field fluctuations allows for the coherent flow of energy between X and SPP, i.e. Rabi oscillations, forming new hybridized upper (UP) and lower (LP) polariton states [1]. We explore these strong coupling phenomena and especially their dynamics using two-dimensional electronic spectroscopy (2DES). Here, strong coupling predicts notorious "oscillating cross-peaks" between diagonal UP and LP peaks in a 2DES map. We investigate the polariton dynamics of a prototypical system for X-SPP coupling, a J-aggregate coated periodic gold slit array. In our measurements, the strongly coupled system indeed shows these hallmarks: Rabi oscillations of 2DES cross-peaks. Surprisingly, however, spatial modulations of the plasmon field seem to result in both strongly and weakly coupled excitons, which can then undergo coherent population transfer via the SPP. This conclusion is further supported by FDTD and Frenkel exciton model simulations.
[1]: Vasa, Parinda, et al., Nat. Photon 7.2, 128-132 (2013).

Topical Talk O 58.3 Wed 15:30 WIL A317

Phase-locked photon-electron interaction without a laser — •Nahid Talebi — Institute for Experimental and Applied Physics, Kiel University, 24107 Kiel, Germany

Ultrafast electron-photon spectroscopy in electron microscopes commonly requires ultrafast laser setups. Photoemission from an engineered electron source is used to generate pulsed electrons, interacting with a sample that is excited by the ultrafast laser pulse at a specified time delay. Thus, developing an ultrafast electron microscope demands the exploitation of extrinsic laser excitations and complex synchronization schemes. Here, we present an inverse approach based on cathodoluminescence spectroscopy to introduce internal radiation sources in an electron microscope. Our method is based on a sequential interaction of the electron beam with an electron-driven photon source (EDPHS) and the investigated sample. An electron-driven photon source in an electron microscope generates phase-locked photons that are mutually coherent with the near-field distribution of the swift electron. Due to their different velocities, one can readily change the delay between the photons and electrons arriving at the sample by changing the distance between the EDPHS and the sample. We demonstrate the mutual coherence between the radiations from the EDPHS and the sample by performing interferometry with a combined system of an EDPHS and a WSe_2 flake. We assert the mutual frequency and momentum-dependent correlation of the EDPHS and sample radiation, and determine experimentally the degree of mutual coherence of up to 27%.

O 58.4 Wed 16:00 WIL A317

Layer-resolved resonance intensity of evanescent polariton modes — Nikolai C. Passler[1], Giulia Carini[1], Dmitry N. Chigrin[2,3], and •Alexander Paarmann[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin — [2]DWI - Leibniz-Institut für Interaktive Materialien, Aachen — [3]Institute of Physics (1A), RWTH Aachen University, Aachen

Most nanophotonic approaches rely on polaritons to confine light to the nanoscale. In particular heterostructures of strongly anisotropic polar crystals have been of key interest recently, where the twist angles between different anisotropic layers key be leveraged to create and control exotic polaritonic states [1]. To guide the designs of such multi-parameter systems, low-cost simulation tools are invaluable. Common approaches like employing the reflection coefficient under evanescent excitation enable analysis of the polariton resonances without the need to specify the exact experimental arrangement, but lack layer-specific information. Here [2], we introduce a formalism based on a 4x4 transfer matrix algorithm [3] and energy flow analysis using the Poynting vector [4], that provides full depth-resolved information even for evanescent wave excitation. We illustrate the power of the approach by analysing a state-of-the-art example of twisted bilayer molybdenum trioxide [1].

[1] G. Hu, et al., Nature 582, 209 (2020).
[2] N.C. Passler, et al., arXiv preprint, arXiv:2209.00877 (2022).
[3] N.C. Passler & A. Paarmann, JOSA B 34, 2128 (2017).
[4] N.C. Passler, et al., PRB 101, 165425 (2020).

O 58.5 Wed 16:15 WIL A317

Observation of multi-quantum phenomena in molecular films strongly coupled to surface plasmons using ultrafast laser spectroscopy — •Simon Büttner[1], Matthias Hensen[1], Katja Mayershofer[1], Maximilian Rödel[2], Jens Pflaum[2], and Tobias Brixner[1] — [1]Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [2]Experimental Physics VI, University of Würzburg, Am Hubland, 97074 Würzburg, Germany

Multi-quantum phenomena such as exciton–exciton annihilation can be used to study exciton diffusion [1], which is a key process in optoelectronic devices like solar cells. Recently, our group has developed a method to isolate different multi-quantum signals using transient absorption (TA) spectroscopy. This novel method makes it possible to filter out the interaction of a distinct number of quasiparticles and their time evolution. Here, we use this method together with a new ultrafast laser setup that allows us to shape and detect femtosecond laser pulses at a repetition rate of 100 kHz. As a model system we investigate a thin Zinc phthalocyanine (ZnPc) film on a surface-plasmon-supporting gold substrate. Strong coupling between ZnPc excitons and surface plasmons leads to the formation of plexcitons [2], and thus to a delocalization of charge carriers, which is of particular interest for increasing charge carrier mobilities. In this work we focus on the influence of the strong coupling on the quasiparticle diffusion.
[1] P. Malý et al., *Chem. Sci.* 11, 456 (2020)
[2] M. Rödel et al., *J. Phys. Chem. C* 126, 4163-4171 (2022)

O 58.6 Wed 16:30 WIL A317

Resonant imaging of infrared light confinement using phonon polaritons in sub-diffractional 4H-SiC nanostructures — •Richarda Niemann[1], Sören Wasserroth[1], Guanyu Lu[2], Christopher R. Gubbin[3], Martin Wolf[1], Simone de Liberato[3], Joshua D. Caldwell[2], and Alexander Paarmann[1] — [1]Fritz Haber Institute of the Max Planck Society, Berlin, Germany — [2]Vanderbilt University, Nashville, USA — [3]University of Southampton, UK

Confinement of light to deeply sub-wavelength scales can be achieved using polaritons spatially confined to sub-diffractional nanostructures.

Next to well-established plasmon polariton approaches, phonon polaritons supported in nanostructures made from polar dielectric crystals are a promising tool for the infrared spectral range [1].

Here, we study localized phonon polariton resonances in sub-diffractional 4H-SiC nanostructures using infrared super-resolution sum-frequency generation (SFG) microscopy [2], employing our institute's infrared free-electron laser [3]. With this technique we achieve high sensitivity to optical field enhancement associated with the spatial confinement of light. Simultaneously it is providing sub-diffractional imaging resolution, naturally enabling to study polaritons on their intrinsic length scales.

[1] Gubbin et al., Journal of Appl. Phys. 131, 030901 (2022)
[2] Niemann et al., Appl. Phys. Lett. 120, 131102 (2022)
[3] Schöllkopf et al., Proc. SPIE 9512, 95121L (2015)

O 58.7 Wed 16:45 WIL A317

Nano-scale Plasmonic Su-Schrieffer-Heeger Chains — •Benedikt Schurr[1,4], Philipp Grimm[1], Tobias Helbig[2], Tobias Hofmann[2], Lukas Wehmeier[3], Felix G. Kaps[3], Thorsten Feichtner[1,4], Monika Emmerling[1], Susanne C. Kehr[3], Lukas Eng[3,4], Ronny Thomale[2,4], and Bert Hecht[1,4] — [1]NanoOptics & Biophotonics Group, Experimental Physics 5, University of Würzburg, Am Hubland, 97074 Würzburg, Germany — [2]Institute for Theoretical Physics and Astrophysics, University of Würzburg, Am Hubland, 97074 Würzburg, Germany — [3]Institut für Angewandte Physik, Technische Universität Dresden, 01062 Dresden, Germany — [4]Würzburg-Dresden Cluster of Excellence ct.qmat, University of Würzburg, Am Hubland, 97074 Würzburg, Germany

The tight-binding Su-Schrieffer-Heeger (SSH) model describes 1D periodic chains of resonators with alternating coupling strengths. For certain configurations, topologically protected collective states occur at the edges. Here, we propose a plasmonic chain SSH-system with alternating few-nanometer gaps, fabricated from mono-crystalline Au-microplatelets by means of He-ion beam milling. Full FDTD simulations show the occurrence of edge modes for such geometries in real space. The frequency at which edge states occur can be fully controlled via particle size and the next-neighbour coupling via the gaps. Scattering scanning near-field optical microscopy (sSNOM) experiments provide first evidence for the presence of topologically protected edge states.

O 58.8 Wed 17:00 WIL A317

Directional leaky polaritons in anisotropic crystals — Xiang Ni[1], •Giulia Carini[2], Weiliang Ma[3], Enrico M. Renzi[1], Emanuele Galiffi[1], Sören T. Wasserroth[2], Martin Wolf[2], Peining Li[3], Alexander Paarmann[2], and Andrea Alù[1] — [1]CUNY, New York, USA — [2]FHI, Berlin, Germany — [3]HUST, Wuhan, China

For quite a few years now, natural hyperbolic materials have been attracting significant attention due to their ability of engaging strong interactions between light and their IR active phonon resonances. The hybridized light-matter quasiparticles arising in their bulk, known as hyperbolic phonon polaritons, display large momenta and highly directional propagation stemming from their open topology.

In our contribution, we demonstrate a new class of directional polaritons supported in the lower reststrahlen band of calcite, that feature lenticular isofrequency contours. These novel polaritons, dubbed Leaky Polaritons (LPs), arise in the type-I in-plane hyperbolic region as hybridized states between extraordinary surface-bound and ordinary propagating bulk modes. Despite their closed topology, they support highly directional, long-range, sub-diffractive propagation at the interface. Their dispersion curve also crosses the free-space light cone, providing radiative far-field coupling to both sides of the interface.

To observe the features of LPs experimentally, we employed polariton spectroscopy, far-field probing and near-field imaging, revealing their lenticular dispersion curve, their high directionality, their long lifetime and real-space propagation.

O 58.9 Wed 17:15 WIL A317

Anticrossing of a plasmonic nanoresonator mode and a single quantum dot at room temperature — •Daniel Friedrich[1], Jin Qin[1], Benedikt Schurr[1], Tommaso Tufarelli[2], Heiko Gross[1], and Bert Hecht[1] — [1]NanoOptics & Biophotonics Group, Experimental Physics 5, University of Würzburg, Germany — [2]School of Mathematical Sciences and Centre for the Mathematics andTheoretical Physics of Quantum Non-Equilibrium Systems, Universityof Nottingham, United Kingdom

At ambient conditions strong coupling (SC) is achieved due to the extremely small mode volume of plasmonic nanoresonators and the broadband spectral overlap between emitter and nanocavity. This results in ultrafast energy transfer which overcomes dephasing. Normal mode splittings in luminescence spectra of single quantum systems coupled to plasmonic nanoresonators have been reported and exploited to estimate the light-matter coupling strength g. However, there is only sketchy evidence for the hallmark of single-emitter strong coupling, the anticrossing of emitter and cavity resonances. Here, we exploit the light-induced oxygen-dependent blue-shift of individual CdSe/ZnS semiconductor quantum dots to tune their transition energy across the weakly radiative resonance of a scanning plasmonic slit resonator. The observed anticrossing in photoluminescence spectra recorded as a function of time provide clear proof of SC as well as a solid measure for the single-emitter coupling strength consistent with classical field simulations and a quantum model including dissipation.