O 87: Plasmonics and Nanooptics: Light-Matter Interaction, Spectroscopy I

Time: Thursday 15:00–17:30 Location: H4

O 87.1 Thu 15:00 H4

Nonlinear plasmonic near– and far fields in the context of Babinet's Principle — •Valentin Dichtl, Thorsten Schumacher, and Markus Lippitz — Experimental Physics III, University of Bayreuth

The third-order nonlinear material response of noble metals enables the formation of the third-harmonic near field around a plasmonic nanostructure [1]. The corresponding spatial emission pattern of the third-harmonic hot spots changes drastically when the fundamental wavelength is slightly tuned over a linear resonance of the nanorod.

However, third harmonic generation (THG) also leads to high temperatures in the structure and its surroundings. These temperatures are usually high enough to destroy more complex samples. Therefore, structures with the same emission properties but a higher ratio of THG to temperature are needed.

To overcome this, we are inspired by the Babinet principle. In this sense, a rod antenna can be replaced by a slit in a thin layer of gold. The surrounding gold should now dissipate heat more effectively than a single rod. This talk compares the (non-linear) emission properties of plasmonic nanostructures and their complementary counterparts, highlighting the applicability of Babinet's Principle.

[1] Wolf, D. *et al.* Shaping the nonlinear near field. *Nat. Commun.* 7:10361 (2016). doi: 10.1038/ncomms10361

O 87.2 Thu 15:15 H4

Beyond-dipole, self-consistent light-matter coupling in plasmonic cavities from first principles — •Franco Bonafe[1], Carlos Bustamante[1], Heiko Appel[1], and Angel Rubio[1,2] — [1]MPI for Structure and Dynamics of Matter, Hamburg, Germany — [2]Center for Computational Quantum Physics (CCQ), The Flatiron Institute, New York, USA

The theoretical treatment of light-matter coupling of molecules in complex electromagnetic (EM) environments is key for making progress in the area of modified matter properties in cavities and strong coupling. While several semiclassical methods can treat the coupling of molecules with EM environments, no method has yet considered the combined effects of self-consistent radiative coupling of molecule and environement, and beyond-electric-dipole interactions to account for the effects of inhomogeneous EM fields. This multiscale problem is now possible to address using our full minimal coupling Maxwell-TDDFT framework [1], implemented in the Octopus package. In this talk, we show how the effects of self-consistent light-matter coupling for different systems. On the one hand, we report the spectral Lamb-like shifts of plasmonic dimers described from first principles when radiation-reaction is considered. Moreover, we discuss the effects of transverse and longitudinal field inhomogeneity in the spectral properties of molecules in plasmonic environments described by a Drude response, including retardation effects. Finally, outlooks on self-consistent coupling of electrons, phonons and structured light are discussed. [1] F.P. Bonafé, A. Rubio, H. Appel et al. arXiv:2409.08959v2 (2024)

O 87.3 Thu 15:30 H4

Polarisation-dependent Mie void resonances — •Serkan Arslan[1], Shaban B. Sulejman[2], Sebastian Klein[1], Jonathan Haehner[2], Dominik Ludescher[1], Tim Davis[1,2], Lukas Wesemann[2], Ann Roberts[2], Harald Giessen[1], and Mario Hentschel[1] — [1]Univesity of Stuttgart, 4th Physics Institute, Germany — [2]University of Melbourne, School of Physics & TMOS, Australia

Over the last decade, researchers in nanophotonics have extensively studied the properties and applications of high refractive index dielectric nanoresonators that support subwavelength Mie resonances. Recently, it was demonstrated that similar resonances also occur in wavelength- and subwavelength-sized holes in high refractive index dielectrics, now known as Mie voids. These Mie voids efficiently confine light within air and support resonances spanning from the IR to UV wavelengths, thereby providing a new building block and expanding the parameter space of dielectric nanophotonics.

Until now, only circular Mie voids have been investigated. Here, we present the first study of non-isotropic Mie voids, including elliptical and rectangular shapes. We numerically and experimentally investigate the polarisation dependence of the resonances and reflection spectra, paving the way for polarisation-dependent structural colors and optical security features. Specifically, we demonstrate polarisation-dependent color patterns and micrometer-sized color prints. Anisotropic Mie voids also allow for the design of Mie void metasurfaces based on a generalised geometric phase.

O 87.4 Thu 15:45 H4

Using strong coupling to control plasmonic catalysis — •Jakub Fojt, Paul Erhart, and Christian Schäfer — Department of Physics, Chalmers University of Technology, Gothenburg, Sweden

Plasmonic excitations decay within femtoseconds, leaving nonthermal (often referred to as "hot") charge carriers behind that can be injected into molecular structures to trigger chemical reactions that are otherwise out of reach – a process known as plasmonic catalysis. In this contribution, we demonstrate that strong coupling between resonator structures and plasmonic nanoparticles can be used to control the spectral overlap between the plasmonic excitation energy and the charge injection energy into nearby molecules. Our atomistic description couples real-time density-functional theory self-consistently to an electromagnetic resonator structure via the radiation-reaction potential. Control over the resonator provides then an additional knob for nonintrusively enhancing plasmonic catalysis, here more than 6-fold, and dynamically reacting to deterioration of the catalyst – a new facet of modern catalysis.

O 87.5 Thu 16:00 H4

Photoemission electron microscopy of Exciton-Polaritons in thin WSe_2 waveguides — •Tobias Eul[1], Victor DeManuel-Gonzalez[1], Miwan Sabir[1], Florian Diekmann[1], Kai Rossnagel[1,2,3], and Michael Bauer[1,2] — [1]Institute of Experimental and Applied Physics, Kiel University, 24098 Kiel, Germany — [2]Kiel Nano, Surface and Interface Science KiNSIS, Kiel University, 24118 Kiel, Germany — [3]Deutsches Elektronen-Synchrotron DESY, Ruprecht Haensel Lab, 22607 Hamburg, Germany

Exciton-polaritons emerging from the interaction of photons and excitons in the strong coupling regime are intriguing quasi-particles for the potential exchange of energy during light-matter interaction processes such as light harvesting. This coupling causes an energy anti-crossing in the photon dispersion centered around the exciton resonance, i.e. a Rabi-splitting between a lower and upper energetic branch. The size of this splitting correlates to the coupling strength between the exciton and the photonic modes. In this work, we investigate this coupling between excitons and photonic waveguide modes excited simultaneously in thin-film flakes of the transition-metal dichalcogenide WSe_2. Using a Photoemission electron microscope, we are able to extract the dispersion of the TE- and TM-modes propagating through these flakes as well as extract the energy splitting. Ultimately, our findings precipitate the investigation of the propagation of exciton-polaritons in the time-domain via time-resolved photoemission.

O 87.6 Thu 16:15 H4

Surface-mediated ultra-strong cavity coupling of two-dimensional itinerant electrons — Christian Eckhardt[1,2], Andrey Grankin[3], Dante Kennes[2,1], Michael Ruggenthaler[1], Angel Rubio[1], •Michael Sentef[4,1], Mohammad Hafezi[3], and Marios Michael[1] — [1]Max Planck Institute for the Structure and Dynamics of Matter, Hamburg — [2]Institut für Theorie der Statistischen Physik, RWTH Aachen University — [3]Joint Quantum Institute, Department of Physics, University of Maryland — [4]Institute for Theoretical Physics, University of Bremen

Engineering phases of matter in cavities requires effective light-matter coupling strengths that are on the same order of magnitude as the bare system energetics, coined the ultra-strong coupling regime. For models of itinerant electron systems, which do not have discrete energy levels, a clear definition of this regime is outstanding to date. Here we argue that a change of the electronic mass exceeding 10% of its bare value may serve as such a definition. We propose a quantitative computational scheme for obtaining the electronic mass in relation to its bare vacuum value and show that coupling to surface polariton modes can induce such mass changes. Our results have important implications for cavity design principles that enable the engineering of electronic properties with quantum light.

O 87.7 Thu 16:30 H4

Photoheating nanoscale Pd to temperatures exceeding attached Au nanoparticle antennas — Felix Stete[1], Shivani Kesarwani[2], Charlotte Ruhmlieb[2], Florian Schulz[2], Marc Herzog[1], Holger Lange[1,2], and •Matias Bargheer[1,3] — [1]Universität Potsdam — [2]Universität Hamburg — [3]Helmholtz-Zentrum Berlin

In the non-equilibrium following optical excitation, energy transfer processes at the nanoscale can exhibit extraordinary and surprising phenomena such as heat transport without heating and dominant phonon heat transport in the nobel metal gold. Here we present transient absorption experiments on a systematic and well-characterized series of gold nanoparticles with a Pd satellite shell. Modeling the fluence dependence of the entire series of hybrid structures with a fixed set of thermophysical parameters shows that we can create hybrid nanophoto-catalysts that concentrate light energy into the catalytically active Pd. We can make a few Pd satellites efficiently collect nearly all photothermal energy deposited by plasmonically enhanced absorption in gold and heat up the Pd by 180 K while the gold core remains cold. Consequently, highly active catalytic sites emerge which can enhance light driven molecular transformations. The tailoring and modeling of such unexpected nanoscale energy transfer phenomena is backed up by ultrafast x-ray diffraction experiments that provide a material-specific direct measure of the crystal lattice response to energy in phonons and electrons of each material in heterostructures and nanocomposites.

O 87.8 Thu 16:45 H4

Investigation of light-matter coupling in tight binding models — •Jonas Reimann[1,2], Michael Ruggenthaler[1,2], and Angel Rubio[1,2,3] — [1]Max Planck Institute for the Structure and Dynamics of Matter, Luruper Chaussee 149, 22761 Hamburg, Germany — [2]Center for Free-Electron Laser Science, Luruper Chaussee 149, 22761 Hamburg, Germany — [3]Center for Computational Quantum Physics, The Flatiron Institute, New York, NY, USA

The theoretical and mathematical foundations for light-matter interactions in a continuum theory are well-established. With the Pauli-Fierz Hamiltonian as the basic building block there is a starting Hamiltonian with all the required properties to study systems containing both matter and light. This can for example be used in the arising field of cavity material engineering where the interplay of light and matter is utilized to design new material properties.

How to imprint these mathematical properties onto tight-binding models describing such materials is at the center of a long-ongoing discussion. A widely deployed approach in this context is the Peierls substitution, coupling light to the hopping elements of the matter Hamiltonian. In this contribution we investigate the reliability of the Peierls substitution in terms of fundamental light-mattter coupling properties within the context of cavity material engineering.

O 87.9 Thu 17:00 H4

Revealing the crystallization dynamics of phase-change materials in vicinity of metallic nanostructures with multiphysics simulations — •Luis Schüler[1,2], Lukas Conrads[2], Sebastian Meyer[2], Yingfan Chen[2], Lina Jäckering[2], Matthias Wuttig[2], Thomas Taubner[2], and Dmitry Chigrin[1,2] — [1]DWI - Leibniz Institute for Interactive Materials, Aachen — [2]I. Institute of Physics (IA), RWTH Aachen University, Aachen

Optical metasurfaces composed of metallic or dielectric scatterers (meta-atoms) promise a powerful way of tailoring light-matter interactions. Phase-change materials (PCMs) are prime candidates for non-volatile resonance tuning of metasurfaces based on a change in refractive index. Precise resonance control can be achieved by locally applying laser pulses to crystallize a PCM, modifying the dielectric surrounding of meta-atoms. However, the complex crystallization kinetics of PCMs in the vicinity of metallic meta-atoms have not been studied yet. Here, we investigate metallic dimer antennas on top of the PCM $Ge_3Sb_2Te_6$ and address these nanoantennas with laser pulses to crystallize the PCM below. Our study reveals inhomogeneous crystallization caused by the absorption and heat conduction of the metallic nanoantennas. A self-consistent multiphysics model, including electromagnetic, thermal, and phase-transition processes, is employed to simulate the crystallization and predict the resulting resonance shift of the antennas. This model enables the optimization of the laser parameters and the geometry of the meta-atoms to achieve an optimal resonance shift, thereby improving the efficiency of metasurfaces.

O 87.10 Thu 17:15 H4

How accurate is the pole expansion of the scattering matrix? — •Elias Fösleitner[1], Adrià Canós Valero[1], Egor Muljarov[2], and Thomas Weiss[1] — [1]Department of Theoretical Physics, University of Graz, Graz, Austria — [2]School of Physics and Astronomy, Cardiff University, Cardiff, United Kingdom

Optical metasurfaces are flat arrangements of nanostructures with different subwavelength sizes and orientations, which allow tailoring the light propagation in a layer of subwavelength thickness. Such metasurfaces are often described using their resonant states. These states, also known as quasi-normal modes, serve as the foundation for the pole expansion of the optical scattering matrix, providing a more efficient and insightful alternative to conventional full-wave simulations. Existing formulations of such an expansion have, however, limitations, particularly in terms of accuracy, efficiency, and convergence. The aim of this study is to compare different approaches for pole expansions and identify their advantages and limitations. Moreover, we discuss how the choice of basis functions of the scattering matrix influences its complex pole structure and the resulting pole expansion. Overall, these findings will allow for a faster prediction of optical properties by choosing the best suited resonant expansion and also provide additional insight that is necessary for many applications such as nanophotonic sensors.