CPP 29: Plasmonics and Nanooptics III: Ultrafast and Nonlinear Phenomena (joint session O/CPP)

Time: Monday 15:00–18:00 Location: WIL A317

Invited Talk CPP 29.1 Mon 15:00 WIL A317

Probing Nanophotonic Modes and Optical Phase Shaping of Electron Beams in Ultrafast Transmission Electron Microscopy — •Armin Feist — IV. Physical Institute, University of Goettingen, 37077 Göttingen, Germany

Ultrafast transmission electron microscopy (UTEM) combines the versatile imaging, diffraction and spectroscopy capabilities of state-of-the-art TEM with femtosecond temporal resolution achieved by a laser-pump/electron-probe scheme [1,2]. The novel applications of UTEM include the study of coherent inelastic electron-light scattering (IELS) at laser-excited nanostructures [3,4].

Here, I will briefly introduce the UTEM methodology and show recent results of the Göttingen UTEM instrument, which features high coherence electron pulses generated from nanoscale field emitter tips [2]. Besides nanometer mapping of chiral optical near-fields and local plasmonic modes, IELS enables the transverse and longitudinal phase control of the free-electron wavefunction [4,5], with applications for coherent electron beam splitters and generating attosecond electron pulse trains. Furthermore, the phase-matched interaction of electrons with optical whispering gallery modes (WGMs) enables a strongly enhanced coupling and traces the ring-down of a dielectric microresonator [6].

[1] A. H. Zewail, Science **328**, 187 (2010). [2] A. Feist *et al.*, Ultramicroscopy **176**, 63 (2017). [3] Barwick *et al.*, Nature **462**, 902 (2009). [4] A. Feist *et al.*, Nature **521**, 200 (2015). [5] K. E. Priebe *et al.*, Nat. Photonics **11**, 793 (2017). [6]. O. Kfir *et al.*, arXiv:1910.09540 (2019).

CPP 29.2 Mon 15:30 WIL A317

Time-resolved and spatially-resolved mode dynamics within a plasmonic nanoslit cavity investigated by coherent two-dimensional nanoscopy — Sebastian Pres[1], Daniel Friedrich[2], Daniel Fersch[1], Enno Krauss[2], Bernhard Huber[1], Victor Lisinetskii[1], •Matthias Hensen[1], Bert Hecht[2], and Tobias Brixner[1] — [1]Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [2]NanoOptics & Biophotonics Group, Experimental Physics 5, University of Würzburg, Am Hubland, 97074 Würzburg, Germany

Recently, we revealed the impact of off-resonant eigenmodes on the local field dynamics of plasmonic hot-spots within a single nanoslit cavity with a spatial and temporal resolution of 10 nm and 20 fs, respectively, by combining photoemission electron microscopy and interferometric two-pulse sequences [1]. Here, we extend the excitation scheme of the nanoslit cavity to a three-pulse sequence using an LCD-based pulse shaper to perform two-dimensional spectroscopy with unprecedented spatial resolution [2]. Due to the nonlinearity of the photoelectron emission process higher-order signals are easily disclosed. These results are supported by quantum dynamical simulations in which the plasmonic nanoslit is modelled as a quantized harmonic oscillator instead of classical fields. Detecting such signal contributions will be helpful in future experiments to reveal the impact of many-body interactions in molecular or solid-state systems with nanometer resolution.

[1] M. Hensen et al., *Nano Lett.* 19, 4651 (2019)
[2] B. Huber et al., *Rev. Sci. Intrum.* 90, 113103 (2019)

CPP 29.3 Mon 15:45 WIL A317

Coherent two-dimensional nanoscopy on coupled plasmonic nanostructures — •Lysanne Monika Dietrich[1], Daniel Friedrich[2], Jessica Meier[2], Enno Krauss[2], Daniel Fersch[1], Raphael Wichary[1], Victor Lisinetskii[1], Matthias Hensen[1], Bert Hecht[2], and Tobias Brixner[1] — [1]Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [2]NanoOptics & Biophotonics Group, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

Two-dimensional (2D) nanoscopy combines the femtosecond time resolution of coherent 2D spectroscopy with nanoscale selectivity: Instead of measuring optical diffraction-limited light fields, we detect photoemitted electrons with a spatial resolution < 5 nm utilizing aberration-corrected photoemission electron microscopy (AC-PEEM) [1]. Since 2D spectroscopy provides direct insights into couplings between single system constituents, we investigate coupled plasmonic nanostructures (i.e. "dolmen" structures) as a model system [2,3] to demonstrate the additional spatial resolution. We complement our experimental findings by simulated 2D spectra using the local response function as retrieved from finite-difference time-domain (FDTD) method.

[1] B. Huber et al., *Rev. Sci. Instrum.*, **90**, 113103 (2019).
[2] H. Yu et al., *ACS Nano*, **10**, 10373-10391 (2016).
[3] J.-S. Huang et al., *Nano Lett.*, **10**, 2105-2110 (2010).

CPP 29.4 Mon 16:00 WIL A317

THz streaking spectroscopy for mapping the transformation from far-fields to nanostructure near-fields — •Felix Sommer and Georg Herink — Physikalisches Institut EP VIII, Universität Bayreuth

Field-driven experiments at the nanoscale require the precise characterization of local ultrafast transients. We apply THz near-field streaking spectroscopy [1,2] to metallic nanotips to map the enhanced near-field at the apex.

The local waveforms are critical to various emerging ultrafast instruments for nanoscopic imaging and spectroscopy. The transformation of focused far-fields to nanoscale near-fields sensitively depends on either the individual nano- and macroscopic geometry and structure, involving effects of antenna resonances, plasmon excitations and excitation geometries [3,4]. In order to study the THz-response in the time-domain, we developed an optimized near-field streaking setup based on a strong-field THz source and present experimental data on the THz-waveform transformation.

[1] Wimmer et al., Nat. Phys. 10, 432-436 (2014).
[2] Wimmer et al., APL 111(13), 131102 (2017).
[3] Talebi et al., ACS nano 9(7), 7641-7648 (2015).
[4] Schröder et al., PRB 92(8), 085411 (2015).

CPP 29.5 Mon 16:15 WIL A317

Tunable strong coupling — •Daniel Friedrich, Benedikt Schurr, Heiko Gross, and Bert Hecht — NanoOptics & Biophotonics Group, Experimental Physics 5, University of Würzburg, Germany

Using a scanning plasmonic nano resonator we have shown recently how to achieve tunable strong coupling to colloidal CdSe quantum dots at ambient conditions. The developed system provides an elegant method to tune the coupling strength and can also be used as high resolution optical microscope. [1]

In the presentation we highlight the possibility to establish strong coupling with different kinds of emitters that can be placed on any (transparent) substrate. The emitters include a variety of colloidal semiconductor nanoparticles and platelets as well as TMDCs.

[1] H. Groß et al., Science Adv. 4, 3, eaar4906 (2018)

CPP 29.6 Mon 16:30 WIL A317

Strong coupling in planar metamaterials — •Manuel Gonçalves[1], Hayk Minassian[2], and Armen Melikyan[3] — [1]Ulm University, Ulm, Germany — [2]A. Alikhanyan National Laboratory (YerPHI), Yerevan, Armenia — [3]Russian-Armenian University, Yerevan, Armenia

Strong coupling is a regime of matter-radiation interaction in a resonant cavity where both the decay rate of the excited state of the matter and the loss rate of the radiation in the cavity are smaller than the coupling strength of matter with radiation. This is a special case of modified spontaneous emission. Fast decay of excited matter state and large cavity loss lead to the more probable Purcell enhancement of spontaneous emission. However, there is strong coupling beyond the quantum optical realm. In classical systems an interaction regime of coupled resonators with similar properties to the quantum case can be produced.

We show how coupled planar optical resonators can be fabricated and present their optical properties. For some configurations the classical analogue of the quantum strong coupling is achieved. Furthermore, higher degrees of interaction between resonators can be achieved and an analogy with the ultrastrong coupling regime of quantum optical systems can be established.

CPP 29.7 Mon 16:45 WIL A317

Quantum Coherent Interference Paths in Interaction of Single-Electron Wavepackets with Light — •Nahid Talebi[1] and Christoph Lienau[2] — [1]Institute für Experimentelle und Angewandte Physik, Christian Albrechts Universität zu Kiel, D-24118 Kiel, Deutschland — [2]Institut für Physik and Center of Interface Science, Carl von Ossietzky Universität Oldenburg, D-26111 Oldenburg, Germany

Ultrafast coherent processes imposed by laser beams into a traveling electron wavepacket in an electron microscope modulate the phase and amplitude of the electron wavefunction (Near-Field Mediated Electron-Light Interactions, Springer Nature, Switzerland (2019)). As a result, electrons can be either accelerated (Nat. Phys. 14 121-5 (2018)), or diffracted (Proc. Camb. Phil. Soc. 29 297-300 (1923)). Particularly the latter, the so-called Kapitza-Dirac effect (KDE), is within the scope of current work. Here, with the aid of first-principle numerical calculations, we describe the physics of KDE. Moreover, we generalize the KDE into a combination of standing-wave and travelling-wave platforms (New J. Phys. 21 093016 (2019)). We show that by virtue of such generalizations, novel quantum-coherent interference paths are formed. Two competing parts of the interaction Hamiltonian, i.e., photon-absorption and emission processes as well as ponderomotive potential, contributes to these interference effects. These interference paths can be controlled by means of either laser or electron wavepacket parameters. Our investigations open up new directions in the domain of matter-wave interferometry.

CPP 29.8 Mon 17:00 WIL A317

Strong-Field Angle-Resolved Plasmoemission Spectroscopy — •Pascal Dreher, David Janoschka, Jan-Henrik Herrig, Michael Horn-von Hoegen, and Frank-J. Meyer zu Heringdorf — Faculty of Physics, University of Duisburg-Essen, Germany

Photoemission in strong fields has received a lot of attention in the past and the coherent interaction of electrons with the field after the emission process has been revealed, i.e. using plasmonic near-field enhancement at nanotips. The behavior of electrons within the solid, however, can as well become dominated by the influence of strong fields, and the respective electron dynamics is expected to be fundamentally different from the electronic equilibrium. Clarifying the dominant interaction processes in this case ultimately requires electronic state resolution as well as precise control over the exact field.

We combine nanofocusing of femtosecond surface plasmon polariton (SPP) pulses on flat surfaces with photoemission electron microscopy to achieve angle-resolved plasmoemission spectroscopy (ARPLES) in the strong-field regime. The emission of electrons from the Au(111) Shockely surface state into SPP-dressed electron final states by the absorption of up to seven SPP quanta is observed. The ponderomotive energy that the emitted electrons acquire within the strong plasmonic nanofocus is determined and provides us with a direct measure for the transient SPP field strength in the focus point.

CPP 29.9 Mon 17:15 WIL A317

Nonlinear plasmon-exciton coupling enhances sum-frequency generation from a Au/ZnO nanohybrid — •Jin-Hui Zhong[1], Jan Vogelsang[2], Jue-Min Yi[1], Dong Wang[3], Lukas Wittenbecher[2], Sara Mikaelsson[2], Anke Korte[1], Abbas Chimeh[1], Cord L. Arnold[2], Peter Schaaf[3], Erich Runge[3], Anne L'Huillier[2], Anders Mikkelsen[2], and Christoph Lienau[1] — [1]Carl von Ossietzky University, 26111 Oldenburg, Germany — [2]Lund University, SE-221 00 Lund, Sweden — [3]Technische Universität Ilmenau, 98693 Ilmenau, Germany

Nanohybrids of plasmon/quantum emitter system can dramatically enhance coherent harmonic generation, often resulting from the coupling of fundamental plasmonic fields to higher-energy, electronic or excitonic transitions of quantum emitters. The ultrafast optical dynamics of such hybrids have been rarely explored. Here, we study those dynamics by interferometrically probing nonlinear optical emission from individual porous gold nanosponges infiltrated with zinc oxide (ZnO) emitters. Few-femtosecond time-resolved photoelectron emission microscopy reveals multiple long-lived localized plasmonic hot spot modes at the surface of single nanosponges, resonant in a broad spectral range. The hot spot near-field couples to the ZnO excitons, enhancing sum-frequency generation, and boosting resonant excitonic emission. The quantum pathways of the coupling are further uncovered from a two-dimensional spectrum correlating fundamental plasmonic excitations to nonlinearly driven excitonic emissions.

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fluctuation-modulated third harmonic deep ultraviolet emission from randomly disordered Si nanograss — •Juemin Yi[1], Dong wang[2], Jinhui Zhong[1], Peter Schaaf[1], Erich Runge[3], and Christoph Lienau[1] — [1]Carl von Ossietzky Universität, Institut für Physik and Center of Interface Science, 26129 Oldenburg — [2]TU Ilmenau, Institut für Werkstofftechnik und IMN MacroNano, 98693 Ilmenau — [3]TU Ilmenau, Institut für Physik und IMN MacroNano, 98693 Ilmenau

It is well known that Si nanograss or black Si greatly enhances light absorption in a broad spectral range and thus is of relevance for solar cell application. Very little is known about their nonlinear optical properties. For that, we have utilized a broadband and few-cycle ultrafast laser to generate coherent third harmonic (TH) emission from randomly disordered Si nanograss. The TH signal is two-order-of-magnitude stronger than that from bulk Si surfaces. For individual hot spots, the TH enhancement even reaches a factor of the order of 1000. A statistical analysis of the TH intensity fluctuations demonstrates that multiple light scattering within the randomly disorder medium is the cause of those fluctuations. A phase-locked and collinear pulse pair excitation proves the coherent nature of the deep ultraviolet emission. The interferometric frequency-resolved autocorrelation (IFRAC) microscopy shows that the deep ultraviolet pulse is around 10fs. Our findings identify random Si nanograss as the most promising candidates for generating ultrashort deep ultraviolet pulses.

CPP 29.11 Mon 17:45 WIL A317

Electron-Driven Photon-Sources for Spectral Interferometry using Electron Microscopes — Nika van Nielen[3], Mario Hentschel[2], Harald Giessen[2], Albert Polman[3], and •Nahid Talebi[1] — [1]Institute für Experimentelle und Angewandte Physik, Christian Albrechts Universität zu Kiel, D-24118 Kiel, Deutschland — [2]4th Physics Institute and Research Center SCoPE, University of Stuttgart, Pfaffenwaldring 57, 70550 Stuttgart, Germany — [3]Center for Nanophotonics, AMOLF, Science Park 104, 1098 XG Amsterdam, The Netherlands

Electron beams can inelastically interact with matters, and as a result, photons are emitted to the far-field. Mechanisms of electron-induced radiations are numerous, covering Cherenkov radiation, transition radiation, and plasmon-induced radiation, to name a few (Rev. Mod. Phys. 82, 209 (2010)). Here, we report on metamaterial- and photon-sieve-based structures for tailoring the electron-induced emission (Nature communications 10 (1), 599 (2019)). By engineering the dispersion of the interface optical modes, radiation relaxation channels for the plasmon polaritons supported by our engineered thin film are formed, leading to an enhanced electron-induced radiation. We also particularly emphasize on the generation of chiral light using plasmonic chains of nanohole arrays. Moreover, we outline our efforts towards realization of combined electron-photon spectroscopy techniques for investigating nanooptical systems in electron microscopes, using electron-driven photon sources (Scientific Reports 6, 33874 (2016)).