# CPP 35: Plasmonics III (joint session O/CPP)

Time: Wednesday 10:30–13:15 Location: H8

CPP 35.1 Wed 10:30 H8

**Plasmon-assisted resonant electron tunneling in a scanning tunneling microscope junction** — Shuyi Liu[1], Martin Wolf[1], and •Takashi Kumagai[1,2] — [1]Fritz-Haber Institute of the Max-Planck Society — [2]JST-PRESTO

Plasmon-induced phenomena have attracted increasing attention due to diverse applications in nanoscale science and technology [1]. Plasmonic nanocavities play a particularly important role because of their ability to confine light to nanometric volumes and generate a strong field enhancement. We report plasmon-assisted resonant electron tunneling from an Ag or Au tip to field emission resonances (FERs) of a Ag(111) surface induced by CW laser excitation of a scanning tunneling microscope (STM) junction at visible wavelengths [2]. As a hallmark of the plasmon-assisted resonant tunneling, we observe a downshift of the first peak in the FER spectra by a fixed amount equal to the incident photon energy. STM-induced luminescence measurement for the Ag and Au tip reveals the clear correlation between the laser-induced change in the FER spectra and the plasmonic properties of the junction. Our results clarify a novel resonant electron transfer mechanism in a plasmonic nanocavity. References: [1] M. L. Brongersma, N. J. Halas, P. Nordlander, Plasmon-induced hot carrier science and technology. Nat. Nanotechnol. 10, 25-34 (2015). [2] S. Liu, M. Wolf, T. Kumagai, Phys. Rev. Lett. in print, 10.1103/PhysRevLett.121.226802.

CPP 35.2 Wed 10:45 H8

**Investigation of plasmon assisted light emission from heteroepitaxial system of Co islands on Cu(111) by scanning tunneling microscopy** — •Vibhuti Rai[1], Kevin Edelmann[1,2], Lars Wilmes[1], Lukas Gerhard[1], and Wulf Wulfhekel[1,2] — [1]Institut für Nanotechnologie, Karlsruher Institut für Technologie, 76344 Eggenstein-Leopoldshafen, Germany — [2]Physikalisches Institut, Karlsruher Institut für Technologie, 76131 Karlsruhe, Germany

Scanning tunneling microscopy (STM) with optical access allows systematic and controlled investigation of light emission from nanometer-sized structures[1]. Here, plasmon mediated light emission from a heteroepitaxial system of bilayer and trilayer Co islands on Cu(111) is studied in ultra-high vacuum at low temperature (4.4 K). Electrical spectroscopy and optical spectroscopy were performed on an image size of 40 x 40 nm where an optical spectrum was taken at every point of a 160 x 160 grid. These measurements show the effect of geometric cavity alteration and the associated resonance shift, influence of dielectric constant of the sample and the tip, and modulation of emission intensity by Friedel oscillation. Interestingly, the results indicate that the difference in the yield of inelastic tunneling dominates over the difference of the gap plasmon resonance.

[1] K. Edelmann et al. Rev. Sci. Instrum. accepted for publication.

CPP 35.3 Wed 11:00 H8

**Photon super-bunching from a metal-metal tunnel junction** — Christopher C. Leon[1], Anna Rosławska[1], •Abhishek Grewal[1], Olle Gunnarsson[1], Klaus Kuhnke[1], and Klaus Kern[1,2] — [1]Max-Planck-Institut für Festkörperforschung, Stuttgart, Germany — [2]Institut de Physique, École Polytechnique Fédérale de Lausanne, Switzerland

Generating correlated photon pairs at the nanoscale is a prerequisite to creating highly-integrated optoelectronic circuits that perform quantum computing tasks based on heralded single-photons. Here, we report on the observation of bunched light emission from a generic tunnel junction under DC-bias. Using LT-STM coupled with Hanbury Brown-Twiss interferometer we probe the dynamics of the photon stream emitted by a metal-metal junction. The inelastic tunneling events of single electrons produce a plasmonic emission whose bunching factor can be 17 (super-bunching) when measured with 53 picosecond instrument resolution. Spectral filtering indicates that two photons of energy higher and lower than half the tunneling electron energy participate in bunching.

We show that mechanisms such as mechanical instabilities at the tip apex, electron co-tunneling, and electronic detection artifacts can be excluded, confirming the non-triviality of the emission. The $1e^- \rightarrow n\gamma$ process is promoted by the localized Purcell effect in conjunction with optical nonlinearities due to inversion symmetry breaking at the tunnel junction. The results suggest that an optoelectronic component useful for quantum computing can be miniaturized to the atomic scale.

CPP 35.4 Wed 11:15 H8

**Simulating ultrashort light pulses in STM tunnel junctions** — •Alexander Neef[1], Dominik Peller[2], Rupert Huber[2], and Jascha Repp[2] — [1]Fritz-Haber-Institute of the MPG, D-14195, Berlin,Germany — [2]Fakultaet fuer Physik, Universitaet Regensburg, D-93040 Regensburg, Germany

Combining ultrafast lightwave control and scanning tunneling miscroscopy (STM) recently opened the door to atomic-scale femtosecond imaging [1,2]. In lightwave STM, a THz field transient is coupled into the tunneling junction to apply an ultrashort bias voltage. The near-field waveform at the tip apex controls single-electron tunneling with combined femtosecond temporal and sub-Å spatial precision. Exploiting this process, single-molecule THz vibrations could be resolved directly in space and time [2]. The mechanism of sub-cycle tunneling control crucially depends on the exact shape of the near-field waveform in the junction, which is determined by interactions of the incoupled terahertz pulses with tip and substrate. To understand the effect of these near-field interactions on the voltage pulses in the junction, we carried out finite element simulations in the frequency domain. The resulting near-field waveforms coincide with experimental results. To further optimize the near-field transients, we study different geometries of the junction.

[1] T. L. Cocker et al., Nature Photon. 7, 620-625 (2013)
[2] T. L. Cocker, D. Peller, P. Yu, J. Repp, and R. Huber, Nature 539, 263 (2016).

CPP 35.5 Wed 11:30 H8

**Advances with Attosecond Electron Pulse Trains in Ultrafast Transmission Electron Microscopy** — •Thomas Rittmann[1], Katharina E. Priebe[1], Christopher Rathje[1,2], Sascha Schäfer[1,2], Sergey V. Yalunin[1], Thorsten Hohage[3], Armin Feist[1], and Claus Ropers[1] — [1]4th Physical Institute - Solids and Nanostructures, University of Göttingen, Germany — [2]Institut für Physik, University of Oldenburg, Germany — [3]Institut für Numerische und Angewandte Mathematik, University of Göttingen

In an ultrafast transmission electron microscope (UTEM), inelastic scattering between a free-electron beam and strong optical near fields [1] allows for a coherent manipulation of the electron quantum state. In this mechanism, the optical field imprints a sinusoidal phase modulation on the electron wave function, which, after subsequent dispersive propagation, results in a temporal electron density modulation [2].

Here, we employ a second electron-light interaction at varied propagation distances with accurately controlled phase delay, and reconstruct the temporal shape of the electron density at each distance with our quantum state tomography algorithm 'SQUIRRELS' [3]. We demonstrate the compression of electron pulses into trains of attosecond bursts and explore the improvement of pulse durations by minimizing phase averaging effects. Such pulse trains will promote new forms of ultrafast electron microscopy with attosecond resolution.

[1] B. Barwick *et al.*, Nature **462**, 902-906 (2009)
[2] A. Feist *et al.*, Nature Physics **12**, 1000-1004 (2016)
[3] K. Priebe *et al.*, Nature Photonics **11**, 793-797 (2017)

CPP 35.6 Wed 11:45 H8

**A versatile setup utilizing shaped optical pulses and time-resolved photoemission electron microscopy to disentangle the ultrafast local response of nanostructured surface systems** — •Sebastian Pres[1], Bernhard Huber[1], Daniel Fersch[1], Enno Krauss[2], Daniel Friedrich[2], 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]Nano-Optics & Biophotonics Group, Experimentelle Physik 5, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

The possibility to disentangle local field dynamics on nanometer length scales is an important prerequisite for the exploration of interactions between nanostructures and nearby quantum systems.
We combine time-resolved aberration-corrected photoemission electron microscopy, enabling sub-10 nm spatial resolution, with a widely tun-

able laser source generating sub-20 fs excitation pulses at 1 MHz repetition rate. Phase-stable pulse sequences are formed by liquid-crystal-based pulse shaping and characterised by Fourier-transform spectral interferometry. A detailed knowledge of each pulse sequence's amplitude and phase structure during the measurement allows to quantitatively analyse the influence of the pulse shape and laser spectrum on resulting time-resolved multidimensional spectroscopy signals. Using coherent 2D nanoscopy [1] we investigate local field dynamics within a plasmonic nanoslit resonator.

[1] M. Aeschlimann et al., *Nat. Photonics*, Vol. 9 (2015)

CPP 35.7 Wed 12:00 H8

**Dynamic imaging of plasmonic nanostructures with an ultrafast point-projection electron microscope** — •Germann Hergert[1], Andreas Wöste[1], Jan Vogelsang[1], Dong Wang[2], Petra Gross[1], and Christoph Lienau[1] — [1]Institut für Physik, Carl von Ossietzky Universität, 26129 Oldenburg, Germany — [2]Institut für Werkstofftechnik, TU Ilmenau, 98693, Germany

The motion of electrons inside metallic nanostructures defines their optical properties. The study of this electron motion requires microscopes with few-fs time and nm-spatial resolution. Ultrafast electron microscopy (UEM) is a promising approach towards this goal, combining the spatial resolution of electron microscopes with the temporal resolution of ultrashort laser pulses. So far, the mesoscopic distance between sample and electron emitter limits the temporal resolution in UEM to 100fs.

We solve this problem with a novel electron source in form of a conical gold taper, without direct illumination of the apex. Light is coupled to surface plasmons on the shaft of the tip, which propagate towards the apex, where they cause electron emission.

Implementing this source in our ultrafast point-projection microscope allows minimal sample-emitter distances and therefore enhanced temporal resolution of 20fs [1]. We use this microscope to observe the ultrafast expansion of a photoemitted electron cloud inside a plasmonic nanoresonator in real space, and in addition, we observe a streaking of the probing electrons by the photoemitted charges.

[1] J. Vogelsang et al., Light: Science & Applications 7, 55 (2018)

CPP 35.8 Wed 12:15 H8

**Quantum Pathway Interference between Surface Plasmon Polariton and Photon** — •David Janoschka, Pascal Dreher, Michael Horn- von Hoegen, and Frank J. Meyer zu Heringdorf — Faculty of Physics and CENIDE, University of Duisburg-Essen, Duisburg, Germany

It is well known that the coherent superposition of light and surface plasmon polaritons (SPPs) at a surface leads to an interferometric mixture of both their electromagnetic fields. At metal surfaces, electrons can be liberated from this mixture by a nonlinear electron emission pathway. In pump probe experiments, the time dependence of the fields has been used to image the propagation of SPPs in a photoemission electron microscope (PEEM). Strictly speaking, the contrast in the microscope arises from the absorption of either photons, SPPs, or both. Here we use Fourier techniques to disentangle the different contributions of photons and SPPs to the electron emission. In addition to the individual contributions of photons and SPPs we find emission pathways that can only be explained by a quantum interference of SPP and photon.

CPP 35.9 Wed 12:30 H8

**Revealing local mode dynamics within a plasmonic nanoslit cavity by time-resolved photoemission electron microscopy** — Bernhard Huber[1], Daniel Friedrich[2], Enno Krauss[2], Sebastian Pres[1], Philipp Grimm[2], Daniel Fersch[1], Julian Lüttig[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

Plasmonic cavities are known for supporting discrete modes upon resonant excitation with light. The associated characteristic pattern of intense electromagnetic hot-spots can be exploited to enhance light-matter interaction and to enable strong coupling of distinct cavities over a micrometer distance [1] or to strongly couple single excitons and plasmons at room temperature [2]. Here, we disentangle the local field dynamics of individual hot-spots within a nanoslit resonator with a spatial resolution of $< 10$ nm by combining time-resolved photoemission electron microscopy (PEEM) and a 1 MHz NOPA system. Interestingly, we detect local differences of rather global properties such as the $Q$-factor and resonance frequency. By using the concept of quasinormal modes we explain these notable local differences, which will be experienced by, e.g., quantum emitters, with a non-negligible influence of adjacent resonator modes.

[1] M. Aeschlimann et al., *Light Sci. Appl.* 6, e17111 (2017)
[2] H. Groß et al., *Sci. Adv.* 4, eaar4906 (2018)

CPP 35.10 Wed 12:45 H8

**Direct optical excitation of dark plasmons for hot electron generation.** — •Dominik Höing[1], Niclas Müller[2], Florian Schulz[1], Stephanie Reich[2], and Holger Lange[1] — [1]Institut für Physikalische Chemie, Universität Hamburg, Sedanstraße 19, VG1-045, 20146 Hamburg — [2]Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, Raum 1.2.42, 14195 Berlin

Plasmonic gold nanoparticles (AuNP) are of great interest because of their ability to generate hot charge carriers, which can play a key role in photoinduced catalysis [1]. Previous studies have shown that the generation of hot carriers depends on whether or not the photon energy is larger than the threshold for interband transitions while exciting at the plasmon resonance seems to have a low effect [2]. A possible explanation might be a significant contribution of radiative energy losses during the plasmon decay. Dark plasmon modes, which can be observed in AuNP multilayers, do not couple to the far field and might allow to reduce such losses [3]. An experimental access to the efficiency of hot carrier generation is transient absorption spectroscopy. It has proven to be a reliable method as it allows assessing the initial temperature of the electrons after excitation and thermalization [4]. In our contribution we investigate the effect of radiative losses on the excitation of hot electrons, by comparing the hot electron dynamics in AuNP mono- and bilayers.

[1] ACS Cent. Sci. 2017, 3, 482-488 ; [2] J. Phys. Chem. Lett. 2017, 8, 19, 4925-4929 ; [3] ACS Photonics 2018, 5, 10, 3962-3969 ; [4] Chem. Rev. 2011, 111, 3858-3887.

CPP 35.11 Wed 13:00 H8

**Angle-resolved plasmoemission from strong SPP fields** — •Pascal Dreher, David Janoschka, Michael Horn-von Hoegen, and Frank Meyer zu Heringdorf — Faculty of Physics and Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, Lotharstrasse 1-21, 47057 Duisburg, Germany

Recent efforts to observe strong-field phenomena in photoemission from metal surfaces have utilized the local enhancement of optical near-fields in nanostructures such as nanotips. On flat metal surfaces high field intensities can be achieved by femtosecond surface plasmon polariton (SPP) pulses. Here we exploit the spatio-temporal nanofocusing of SPPs in Archimedean vortex lenses [1] to achieve particularly strong plasmonic near fields with well-known field distributions on flat Au(111) surfaces. A spectroscopic photoemission electron microscope is employed to detect the electrons which are emitted from the surface by the simultaneous absorption of up to seven SPP quanta. In angle-resolved plasmoemission spectra (ARPLES) we observe signatures which can be attributed to above-threshold plasmoemission from the Au(111) Shockley surface state into SPP-dressed free electron states. The ponderomotive energy that the emitted electrons gain within the strong plasmonic nanofocus is determined from spatially-resolved plasmoemission spectra. The ponderomotive energy provides us with a direct measure for an absolute value of the transverse electric field strength of the SPP in the focus point.

[1] Spektor G, et al., Science 355, 1187 (2017)