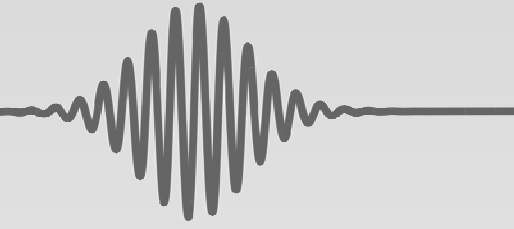


# UNO-4



4<sup>th</sup> International Workshop on Ultrafast Nanooptics

collocated with

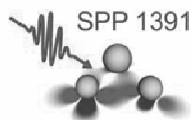
**COST Action MP1403 - Nanoscale Quantum Optics - Work Group Meeting  
Nonlinearities and Ultrafast Processes in Nanostructured Media**



October 18-22, 2015

Bad Dürkheim

**DFG**



**cost**  
EUROPEAN COOPERATION  
IN SCIENCE AND TECHNOLOGY

## Scope and Aim

We cordially welcome you in Bad Dürkheim to the 4<sup>th</sup> *International Workshop on Ultrafast Nanooptics* (UNO-4) collocated with the regular Meeting of Work Group 2 on *Nonlinearities and Ultrafast Processes in Nanostructured Media* of the COST Action MP1403 on *Nanoscale Quantum Optics*. In addition, this workshop serves as the final meeting of the DFG priority program 1391 *Ultrafast Nanooptics* and will follow the successful previous versions of this international workshop.

The aim of *Ultrafast Nanooptics* is the combination of nanooptics and ultrafast laser spectroscopy, a rapidly evolving field in physics and neighboring disciplines. As in the past, this workshop is dedicated to facilitate the exchange of knowledge between the two up to now rather separate fields of research and shall stimulate the interaction of different groups working in these research areas. 11 invited speaker together with 23 contributed talks and 25 posters will present and discuss the recent advances in this emerging fields. Topics will cover both theoretical and experimental aspects of nonlinearities and ultrafast processes in plasmonic nanostructures.

The COST action MP1403 on *Nanoscale Quantum Optics* serves as European platform to enhance networking and research collaboration in the rapidly evolving field of quantum nanooptics that has substantial overlap with the topics covered in the UNO workshop series. Actually, half of the workshop is dedicated to the COST work group meeting on *Nonlinearities and Ultrafast Processes in Nanostructured Media*.

The image shows two handwritten signatures in black ink. The signature on the left is 'W. Pfeiffer' and the signature on the right is 'M. Aeschlimann'. Both are written in a cursive, flowing style.

Walter Pfeiffer and Martin Aeschlimann

# Workshop Program

## Sunday, October 18, 2015

17:00 – 21:00      Arrival and Registration at Bad Dürkheim Kurpark-Hotel  
from 19:00          *SOCIAL GATHERING AND BUFFET SUPPER*

## Monday, October 19, 2015

7:45                *BREAKFAST*

8:50                **Opening remarks**

### **Session 1**

9:00                *Martin Aeschlimann*  
(University of Kaiserslautern, Germany)  
**Priority program 1391 on Ultrafast Nanooptics - Résumé**

9:25                *Walter Pfeiffer*  
(Bielefeld University, Germany)  
**Future developments and information about the  
COST action 1403 on Nanoscale Quantum Optics**

9:50                *F. Süßmann, L. Seiffert, S. Zherebtsov, V. Mondes, J. Stierle,  
M. Arbeiter, J. Plenge, P. Rupp, C. Peltz, A. Kessel, S. A. Trushin,  
B. Ahn, D. Kim, C. Graf, E. Rühl, M.F. Kling, T. Fennel*  
(MPI Garching, LMU Munich, Univ. Rostock, FU Berlin, Germany &  
POSTECH Pohang, Korea)  
**Field propagation-induced directionality of CEP-controlled  
photoemission from nanospheres**

10:15              *COFFEE BREAK*

### **Session 2**

11:00              *Mark I. Stockman* – invited  
(Georgia State University, USA)  
**Condensed matter in ultrafast and  
superstrong fields: attosecond phenomena**

11:50              *Lara Wimmer*, *Georg Herink, Oliver Karnbach,  
Sergey Yalunin, Claus Ropers*  
(University of Göttingen, Germany)  
**THz near-field streaking at metal nanotips**

## Monday, October 19, 2015

12:15 S. H. Chew, A. Gliserin, J. Schmidt, S. Nobis, H. Bian, E. Krauss, P. Geisler, F. Schertz, M. Kübel, B. Hecht, U. Kleineberg  
(LMU Munich, Univ. Würzburg, Surface-Concept GmbH, Germany)  
**Laser intensity effects in carrier-envelope phase-tagged time of flight-photoemission electron microscopy**

12:30 LUNCH

### Session 3

14:00 Ulrich Höfer - invited  
(University of Marburg, Germany)  
**Ultrafast dynamics of electrons in topological surface and metal/organic interface states**

14:50 D. Kilbane, A. K. Mahro, S. Mathias, G. Spektor, L. Gal, M. Orenstein, B. Frank, S. Ristock, H. Giessen, F. Meyer zu Heringdorf, M. Aeschlimann  
(Univ. Kaiserslautern, Univ. Stuttgart, Univ. Duisburg-Essen, Germany & Technion, Israel)  
**Near-field imaging of the dynamics of plasmonic vortices**

15:15 COFFEE BREAK

### Session 4

16:00 Daniel Podbiel, Philip Kahl, Bettina Frank, Harald Giessen, Frank Meyer zu Heringdorf  
(Univ. Stuttgart, Univ. Duisburg-Essen, Germany)  
**Plasmoemission: emission of electrons in a strong plasmonic field**

16:25 Carsten Reinhardt, Tobias Birr, Tim Fischer, Urs Zywiets, Boris Chichkov  
(Laser Zentrum Hannover e.V., Germany)  
**Ultrafast interactions of surface plasmon polaritons**

16:50 E. Oesterschulze, C. Gonzalez, M. Hartelt, D. Bayer, E. Ilin, M. Aeschlimann  
(Univ. Kaiserslautern, Germany)  
**Mie scattering probe for polarization resolved investigation of surface plasmons**

## Monday, October 19, 2015

- 17:15 *Ralf Vogelgesang, Martin Esmann, Simon F. Becker, KyungWan Yoo, Heiko Kollmann, Jan Vogelsang, Jörg Robin, Benedek J. Nagy, Péter Dombi, Daniel Rosenkranz, Manuela Schiek, Petra Groß, NamKyoo Park, Christoph Lienau*  
(Univ. Oldenburg, Germany & Seoul Nat. Univ., Korea & Wigner Res. Centr., Hungary)  
**Plasmonic nanofocusing: Gap plasmons, light scattering spectroscopy and electron generation**
- 17:40 **Poster Session**
- 19:00 DINNER
- 21:00 **Informal Discussion**

## Tuesday, October 20, 2015

7:45 BREAKFAST

### Session 5

9:00 Hayk Harutyunyan - invited  
(Emory University, USA)  
**Controlling the generation of hot electrons in hybrid plasmonic nanostructures**

9:50 Heiko Linnenbank, Yevgen Grynk, Jens Förstner, Stefan Linden  
(Univ. Bonn, Univ. Stuttgart, Univ. Paderborn, Germany)  
**Ultrafast nonlinear response of hybrid dielectric/plasmonic nanoantennas**

10:15 COFFEE BREAK

### Session 6

11:00 Melanie Müller, Lutz Waldecker, Roman Bertoni, Thomas Vasileiadis, Vasily Kravtsov, Markus Raschke, Alexander Paarmann,  
Ralph Ernstorfer - invited  
(FHI Berlin, Germany & University of Colorado, USA)  
**Photocurrents and structural dynamics in nanomaterials probed by femtosecond electron pulses**

11:50 J. Kern, A. Trügler, I. Niehues, J. Ewering, R. Schmidt,  
R. Schneider, S. Najmaei, A. George, J. Zhang, J. Lou,  
U. Hohenester, S. Michaelis de Vasconcellos, R. Bratschitsch  
(Univ. Münster, Germany & Univ. Graz, Austria & Rice University, USA)  
**Nanoantenna-enhanced light-matter interaction of atomically thin WS<sub>2</sub>**

12:15 Dominik Differt, Matthias Hensen, Walter Pfeiffer  
(Bielefeld University, Germany)  
**Time reversal versus adaptive optimization for spatiotemporal nanolocalization in a random nanoantenna**

12:40 LUNCH

### Session 7

14:00 Shinya Koshihara - invited, Keiki Fukumoto, Ken Onda  
(Tokyo Institute of Technology, JST-CREST, KEK, JST-PRESTO, Japan)  
**Visualization of ultrafast electron dynamics using time-resolved photoemission electron microscopy**

## Tuesday, October 20, 2015

- 14:50      *Thomas Pertsch, Carsten Rockstuhl*  
(Univ. Jena, KIT, Germany)  
**Nonlinear optical nano-antennas and nano-resonators  
from Lithium Niobate**
- 15:15      *Alfred J. Meixner, Anke Horneber, Frank Wackenhut, Xiao Wang  
and Dai Zhang*  
(University of Tübingen, Germany)  
**Interplay of plasmon and exciton decay excited  
by ultrashort laserpulses**
- 15:40      *Malte Großmann, Alwin Klick, Christoph Lemke, Ahnaf Zillohu,  
Jacek Fiotowski, Jacob Kjelstrup-Hansen, Mady Elbahri, Franz Faupel,  
Horst-Günter Rubahn, and Michael Bauer*  
(University of Kiel, Germany & Univ. of Southern Denmark, Denmark)  
**Light-triggered Control of Plasmonic Refraction and Group Delay by  
Photochromic Molecular Switches**
- 16:30      EXCURSION
- 19:00      DINNER
- 21:00      **Informal Discussion**

## Wednesday, October 21, 2015

07:45 *BREAKFAST*

### **Session 8**

8:50 Opening Remarks:  
COST action MP 1403 – Nanoscale Quantum Optics - WG2 Meeting  
**Nonlinearities and Ultrafast Processes in Nanostructured Media**

9:00 Steven Cundiff - *invited*  
(University of Michigan, USA)  
**Two Dimensional Coherent Spectroscopy of Epitaxial Quantum Dots**

9:50 Markus Krecik, Marten Richter  
(TU Berlin, Germany)  
**Proposal of using nanoplasmonics and coherent two-dimensional spectroscopy to dissect secular and non secular relaxation processes**

10:15 COFFEE BREAK

### **Session 9**

11:00 Giulio Cerullo - *invited*  
(Politecnico di Milano, Italy)  
**Tracking ultrafast energy flow in plasmonic nanostructures**

11:50 Markus Betz  
(TU Dortmund, Germany)  
**Active plasmonics with surface acoustic waves**

12:15 Polina Lisinetskaya, Roland Mitric  
(University of Würzburg, Germany)  
**Simulation and control of light propagation in ultrasmall noble-metal cluster arrays**

12:40 LUNCH

### **Session 10**

14:00 Javier García de Abajo - *invited*  
(ICFO-Institut de Ciències Fotoniques, Spain)  
**Graphene plasmonics**

14:50 Harald Giessen, Bettina Frank, Simon Ristok, Liwei Fu,  
Grisha Spektor, Philip Kahl, Frank Meyer zu Heringdorf,  
Deirdre Kilbane, Martin Aeschlimann  
(Univ. Stuttgart, Univ. Duisburg-Essen, Univ. Kaiserslautern, Germany)  
**Short-range surface plasmonics and its (sub-)femtosecond dynamics**



## Wednesday, October 21, 2015

15:15 COFFEE BREAK

### **Session 11**

- 16:00 Stephen K. Gray - invited  
(Argonne National Laboratory, USA)  
**Optically-Induced Entanglement in Hybrid Quantum Dot/Plasmonic Systems**
- 16:50 Alberto Comin, Richard Ciesielski, Alexander Bouhelier,  
Achim Hartschuh  
(LMU München, Germany & Univ. de Bourgogne, France)  
**Coherent control in single plasmonic nanostructures**
- 17:15 Daniela Wolf, Thorsten Schumacher, Markus Lippitz  
(University of Bayreuth, Germany)  
**Shaping the nonlinear near-field**
- 17:40 **Poster Session**
- 19:00 *DINNER*
- 21:00 **COST: Work Group 2 Discussion**

## Thursday, October 22, 2015

07:45      *BREAKFAST*

### **Session 12**

9:00      *M. Ethis de Corny, N. Chauvet, L. Olgeirsson, S. Waterman, M. Jeannin, G. Nogues, S. Huant, A. Bouhelier, J. Sharma, E. Dujardin, G. Bachelier - invited*

(Univ. Joseph Fourier, Univ. Bourgogne, CEMES - CNRS, France)

**Nonlinear responses in plasmonic nanostructures**

9:50      *Jeremy Butet, Kuang-Yu Yang, Gabriel Bernasconi, Olivier J. F. Martin*  
(EPFL, Switzerland)

**Second harmonic generation in plasmonic nanostructures:  
multiresonant nanoantennas and eigenmodes**

10:15      *COFFEE BREAK*

### **Session 13**

11:00      *Thomas Durt - invited*

(Institut Fresnel, Ecole Centrale de Marseille, France)

**The photon wave function: a bridge from Maxwell to  
Schroedinger, and beyond**

11:50      *Martin Aeschlimann, Tobias Brixner, Benjamin Frisch, Bert Hecht, Bernhard Huber, Matthias Hensen, Christian Kramer, Enno Krauss, Walter Pfeiffer, Martin Piecuch, Philip Thielen*

(Univ. Kaiserslautern, Univ. Würzburg, Bielefeld Univ., Germany)

**Coherent and long-range plasmon-assisted energy transfer  
between two plasmonic nanoantennas**

12:15      *Armin Feist, Katharina E. Echternkamp, Jakob Schauss, Sergey V. Yalunin, Sascha Schäfer, Claus Ropers*

(University of Göttingen, Germany)

**Coherent electron-light interaction in an ultrafast electron microscope**

12:40      *Closing Remarks*

13:00      *LUNCH*

*END OF WORKSHOP and WORK GROUP MEETING / DEPARTURE*



**Abstracts of oral Presentations** *(in chronological order)*

# Field propagation-induced directionality of CEP-controlled photoemission from nanospheres

F. Süßmann<sup>1,2</sup>, L. Seiffert<sup>7</sup>, S. Zherebtsov<sup>1,2</sup>, V. Mondes<sup>3</sup>, J. Stierle<sup>1</sup>, M. Arbeiter<sup>7</sup>, J. Plenge<sup>3</sup>,  
P. Rupp<sup>1,2</sup>, C. Peltz<sup>7</sup>, A. Kessel<sup>1</sup>, S.A. Trushin<sup>1</sup>, B. Ahn<sup>4,5</sup>, D. Kim<sup>4,5</sup>, C. Graf<sup>3</sup>,  
E. Rühl<sup>3</sup>, M.F. Kling<sup>1,2,4,6</sup>, and T. Fennel<sup>7</sup>

<sup>1</sup> *Max-Planck Institut für Quantenoptik, D-85748 Garching, Germany*

<sup>2</sup> *Physik Department, Ludwig-Maximilians-Universität München, D-85749 Garching, Germany*

<sup>3</sup> *Physical Chemistry, Freie Universität Berlin, D-14195 Berlin, Germany*

<sup>4</sup> *Physics Department, POSTECH, Pohang, Kyungbuk 790-784, Republic of Korea*

<sup>5</sup> *Max Planck Center for Attosecond Science, Pohang, Kyungbuk 790-784, Republic of Korea*

<sup>6</sup> *J.R. Macdonald Laboratory, Physics Department, Kansas-State University, Manhattan, KS, USA*

<sup>7</sup> *Institut für Physik, Universität Rostock, D-18059 Rostock, Germany*

Near-fields of non-resonantly laser-excited nanostructures enable strong localization of ultrashort light fields and have opened novel routes to fundamentally modify and control electronic strong-field processes [1, 2]. Harnessing spatiotemporally tunable near-fields for the steering of sub-cycle electron dynamics may enable ultrafast optoelectronic devices and unprecedented control in the generation of attosecond electron and photon pulses. Here we utilize unsupported sub-wavelength dielectric nanospheres to generate near-fields with adjustable structure and study the resulting strong-field dynamics via photoelectron imaging [3]. We demonstrate field propagation induced tunability of the emission direction of fast recollision electrons up to a regime, where non-linear charge interaction effects become dominant in the acceleration process. Our analysis supports that the timing of the recollision process remains controllable with attosecond resolution by the carrier-envelope phase, indicating the possibility to expand near-field mediated control far into the realm of high-field phenomena.

[1] M. Krüger et al., *Nature* **475**, 78-81 (2011)

[2] S. Zherebtsov et al., *Nature Phys.* **7**, 656-662 (2011)

[3] F. Süßmann et al., *Nat. Commun.* **6**, 7944 (2015)

# Condensed matter in ultrafast and superstrong fields: attosecond phenomena

Mark I. Stockman

*Center for Nano-Optics, Georgia State University, 29 Peachtree Center Ave, Atlanta, GA 30303, USA*

We present a new class of phenomena in condensed matter optics when a strong optical field  $\sim 1-3$  V/Å adiabatically (reversibly) changes a solid within optical cycle [1-7]. Such a pulse drives ampere-scale currents in dielectrics and controls their properties, including optical absorption and reflection, extreme UV absorption, and generation of high harmonics [8] in a non-perturbative manner on a 100-as temporal scale. Applied to a metal, such a pulse causes an instantaneous and reversible loss of the metallic properties. We will also discuss our latest theoretical results on graphene that in a strong ultrashort pulse exhibits unique behavior [9, 10]. New phenomena are predicted for buckled two-dimensional solids, silicene and germanine [11]. These are fastest phenomena in optics unfolding within half period of light. They offer potential for petahertz-bandwidth signal processing, generation of high harmonics on a nanometer spatial scale, etc.

- [1] M. Durach et al., Phys. Rev. Lett. **105**, 086803 (2010).
- [2] M. Durach et al., Phys. Rev. Lett. **107**, 086602 (2011).
- [3] A. Schiffrin et al., Nature 493, **70** (2013).
- [4] M. Schultze et al., Nature 493, **75** (2013).
- [5] V. Apalkov, and M. I. Stockman, Phys. Rev. B **88**, 245438 (2013).
- [6] V. Apalkov, and M. I. Stockman, Phys. Rev. B **86**, 165118 (2012).
- [7] F. Krausz, and M. I. Stockman, Nat. Phot. **8**, 205 (2014).
- [8] T. Higuchi, M. I. Stockman, and P. Hommelhoff, Phys. Rev. Lett. **113**, 213901 (2014).
- [9] H. K. Keldar, V. Apalkov, and M. I. Stockman, Phys. Rev. B **90**, 085313 (2014).
- [10] H. K. Keldar, V. Apalkov, and M. I. Stockman, Phys. Rev. B **91**, 045439 (2015).
- [11] H. K. Keldar, V. Apalkov, and M. I. Stockman, Phys. Rev. B **92**, 045413 (2015).

## **THz near-field streaking at metal nanotips**

Lara Wimmer, Georg Herink, Oliver Karnbach, Sergey Yalunin, Claus Ropers

*IV. Physical Institute, Georg-August University Göttingen, Friedrich-Hund-Platz 1, 37077 Göttingen*

We present a streaking experiment at single metal nanotips with ultrashort terahertz (THz) transients and femtosecond near-infrared (NIR) pulses. The photoelectrons are emitted by the NIR pulses and accelerated in the THz-induced near-field at the tip apex. The high localization and enhancement of the THz near-field allow for a direct mapping of the near-field strength onto the electron kinetic energy. The characteristic parameters of the near-field are determined by numerical simulations.

# **Laser intensity effects in carrier-envelope phase-tagged time of flight-photoemission electron microscopy**

S. H. Chew<sup>1</sup>, A. Gliserin<sup>1</sup>, J. Schmidt<sup>1</sup>, S. Nobis<sup>1</sup>, H. Bian<sup>1</sup>, E. Krauss<sup>2</sup>, P. Geisler<sup>2</sup>,  
F. Schertz<sup>3</sup>, M. Kübel<sup>1</sup>, B. Hecht<sup>2</sup>, U. Kleineberg<sup>1</sup>

<sup>1</sup> *Faculty of Physics, Ludwig-Maximilians-Universität München, 85748 Garching, Germany*

<sup>2</sup> *Nano-Optics and Biophotonics Group, Experimentelle Physik 5, Physikalisches Institut,  
Universität Würzburg, Würzburg D-97074, Germany*

<sup>3</sup> *Surface Concept GmbH, Staudingerweg 7, 55128 Mainz, Germany*

A time of flight-photoemission electron microscope is combined with a single-shot stereographic above-threshold ionization phase meter for studying attosecond control of electrons in tailored plasmonic nanostructures spatially and energetically via a carrier-envelope phase tagging technique. Recent carrier-envelope phase-resolved measurements of gold nanoparticles on gold plane and surface roughness from a gold film show an apparent carrier-envelope phase modulation with a period of  $\pi$ . This modulation is found to originate from an intensity dependence of the photoelectron spectra and the carrier-envelope phase measurement rather than from an intrinsic carrier-envelope phase dependence, which is supported by a correlation of this modulation with intensity fluctuations of our laser on a time scale of hundreds of milliseconds to seconds. We suggest intensity tagging should be taken into account in future phase-tagged experiments in order to detect small CEP-dependence from supported nanostructures.



# Ultrafast dynamics of electrons in topological surface and metal/organic interface states

Ulrich Höfer

*Department of Physics, Philipps University of Marburg, Germany*

Electron transfer processes at surfaces and interfaces play a crucial role in diverse fields of materials sciences. In this talk I will discuss surfaces of three-dimensional topological insulators and interfaces between metals and organic semiconductors. Time-resolved two-photon photoemission (2PPE), a method that combines femtosecond pump-probe techniques with photoelectron spectroscopy, can provide detailed information about the ultrafast dynamics of electrons excited into surface and interface-specific states of these systems. For investigations of metal/organic interfaces, PTCDA/Ag(111) has proven to be an excellent model system. It will be shown that the interface state, located between the Fermi level of the metal and the molecular LUMO of this and related systems, can efficiently mediate the electron transfer between the metal and the organic semiconductor. Three-dimensional topological insulators belong to a new class of materials which are characterized by an insulating bulk and a metallic topological surface state (TSS). The most remarkable properties of the TSS are its Dirac-cone-like energy dispersion and its chiral spin texture in  $k$  space which incorporates a protection against electron backscattering. It will be shown that mid-infrared pump pulses permit a direct excitation of the unoccupied TSS of  $\text{Sb}_2\text{Te}_3$  across the Dirac point. The optical coupling leads to an asymmetric transient population of the TSS in momentum space which in turn reflects a macroscopic electric surface current. By observing the decay of the asymmetric population with time-delayed UV probe pulses, we directly access the dynamics of the long-lived photocurrent and its topologically protected property.

# Near-field imaging of the dynamics of plasmonic vortices

D. Kilbane<sup>1</sup>, A. K. Mahro<sup>1</sup>, S. Mathias<sup>1</sup>, G. Spektor<sup>2</sup>, L. Gal<sup>2</sup>, M. Orenstein<sup>2</sup>, B. Frank<sup>3</sup>, S. Ristock<sup>3</sup>,  
H. Giessen<sup>3</sup>, F. Meyer zu Heringdorf<sup>4</sup>, M. Aeschlimann<sup>1</sup>

<sup>1</sup> *Physics Department and Research Centre OPTIMAS, University of Kaiserslautern, Germany*

<sup>2</sup> *Department of Electrical Engineering, Technion, Haifa, Israel*

<sup>3</sup> *Fourth Physics Institute and Research Center SCoPE, University of Stuttgart, Germany*

<sup>4</sup> *Faculty of Physics and CENIDE, University of Duisburg-Essen, Germany*

Depending on the spin angular momentum of circularly polarized light, and the geometry of the illuminated metallic structure we can shape plasmonic near fields to form a vortex - a rotational flow around a phase singularity [1]. By combining interferometric time-resolved two photon photoemission and photoemission electron microscopy (ITR-2PPE PEEM) [2,3] we perform near-field imaging of plasmonic vortices. A broadband ultrashort pulse laser excites and probes the plasmonic dynamics with 100 as time step and 40 nm spatial resolution. Here we observe the spatiotemporal evolution of vortices in plasmonic Archimedes spirals (PAS) and vortex lenses (PVL).

[1] C.-D. Ku, W.-L. Huang, J.-S. Huang and C.-B. Huang, *IEEE Photon. J.* **5** 4800409 (2013)

[2] A. Kubo, K. Onda, H. Petek, Z. Sun, Y. S. Jung and H. K. Kim, *Nano Lett.* **5** 1123 (2005)

[3] M. Bauer, C. Wiemann, J. Lange, D. Bayer, M. Rohmer and M. Aeschlimann, *Appl. Phys. A* **88** 473 (2007)

## **Plasmoemission: emission of electrons in a strong plasmonic field**

Daniel Podbiel<sup>1</sup>, Philip Kahl<sup>1</sup>, Bettina Frank<sup>2</sup>, Harald Giessen<sup>2</sup>, Frank Meyer zu Heringdorf<sup>1</sup>

<sup>1</sup> *Faculty of Physics and CENIDE, University of Duisburg-Essen, 47048 Duisburg, Germany*

<sup>2</sup> *4th Physics Institute and Research Center SCoPE, University of Stuttgart, 70550 Stuttgart, Germany*

Observing surface plasmon polaritons (SPPs) in a photoemission electronmicroscope (PEEM) is possible via nonlinear photoemission if ultra-short laser pulses (<20fs) of a suitable wavelength are directed onto the surface of a plasmonic material. We study the time-resolved propagation and interaction of SPPs by means of a direct conceptual visualization of the SPPs in a "normal incidence geometry". This experimental setup allows us to observe transient phenomena that exist for only a few femtoseconds during the coherent interaction of the ultrashort SPP pulses. In focusing structures for SPPs we find an unexpected time-signature of the nonlinear photoemission signal at the focus point that must be explained by emission of electrons from the SPP alone. The energy distribution of these 'plasmoelectrons' shows that the SPP fields are sufficiently high to make nonlinear photoemission pathways of higher orders the dominant contribution to the PEEM signal.

# Ultrafast interactions of surface plasmon polaritons

Carsten Reinhardt, Tobias Birr, Tim Fischer, Urs Zywietz, Boris Chichkov

*Laser Zentrum Hannover e.V., Hollerithallee 8, 30419 Hannover, Germany*

We present studies on ultrafast interactions of multiple coherently excited surface plasmon-polaritons (SPPs) and light beams in nanoparticle and waveguiding systems and on plane metal films. SPPs are excited by local scattering of laser light on surface nanostructures consisting of polymeric or metallic ridges, grooves, and nanoparticles. SPP interaction and scattering effects are investigated by temporally resolved leakage radiation microscopy. The interference of SPPs inside dielectrically-loaded SPP waveguides as well as the interference of SPPs with additional light fields is used for tracking the propagation of ultrashort SPP pulses excited by 60 fs laser pulses at a central wavelength of 800 nm. We demonstrate ultrafast scattering of propagating SPPs on regions of metal films pumped by additional laser pulses. In a time-delay pump-probe experiment it is shown that this SPP scattering occurs on the time scale of the pulse duration. Additionally, the interactions of coherently excited SPPs and light beams in complex dielectric waveguides and with laser printed isolated spherical metallic or silicon nanoparticles are demonstrated. Coherent control of SPP and light interactions further allows for the construction of ultrafast low-power SPP switches, transistors, and all-optical gate structures. Results on cascading of these plasmonic elements are presented and an all-plasmonic half adder is demonstrated.

# Mie scattering probe for polarization resolved investigation of surface plasmons

E. Oesterschulze<sup>1</sup>, C. Gonzalez<sup>2</sup>, M. Hartelt<sup>2</sup>, D. Bayer<sup>2</sup>, E. Ilin<sup>1</sup>, M. Aeschlimann<sup>2</sup>

<sup>1</sup> *University of Kaiserslautern, Physics and Technology of Nanostructures, Physics Department,  
Erwin Schrödinger Strasse 46, D-67663 Kaiserslautern*

<sup>2</sup> *University of Kaiserslautern, Department of Physics and Research Center OPTIMAS,  
Erwin Schrödinger Strasse 46, D-67663 Kaiserslautern*

We present a cantilever based near field probe with integrated Mie scattering dielectric silicon dioxide microsphere (MSDM) for near-field optical imaging as well as femtosecond spectroscopy applications. In contrast to the state of the art transmissive near-field probes, the MSDM reveals a transmission of almost unity known from far-field microscopy configuration. For proper handling the microsphere is integrated at the apex of a conventional pyramidal aperture tip carried by an atomic force microscopy cantilever. It proved to be mechanically robust during the scanning process even if operating it in the contact mode. The spherical symmetry provides on one hand a well defined mechanical contact point with the sample irrespective of its inclination angle to the sample surface. On the other hand, the symmetry of the device preserves the polarization of light proving to be useful for the investigation of the polarization dependent behavior of plasmonic nanostructures. The device has also shown to be able to be an excellent tool for the study of surface plasmons on Au thin films when measuring the dispersion relation of SPPs, bringing results with an excellent agreement with similar experiments performed with PEEM. The high transmission combined with low dispersion renders spectroscopic investigations on the femtosecond time scale with a moderate lateral resolution. Moreover, second autocorrelation experiments on a BBO-crystal reveals a time resolution well below 100fs at 190nm spatial resolution.

# Plasmonic nanofocusing: Gap plasmons, light scattering spectroscopy and electron generation

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Apertureless scattering-type scanning near-field optical microscopy (SNOM) conventionally operates with some form of modulation and demodulation technique to eliminate the overwhelmingly strong far-field optical signal background. Such an approach renders SNOM *spectroscopy* a relatively slow, serial measurement technique. To establish parallel, multi-channel SNOM spectroscopy, a very promising approach is the concept of nanofocusing in single crystalline metallic tapers [1,2]. Surface plasmon polaritons are excited on a grating coupler and result in strongly confined near-fields at the taper apex without much background excitation of surrounding volumes.

We apply nanofocusing SNOM to spectrally resolve the field distribution around individual plasmonic rod antennas. An AFM image of chemically grown rods is acquired simultaneously with maps of the intensity scattered in different wavelength intervals. These maps exhibit a pronounced intensity pattern with spatially confined minima at the rod ends.

Initial characterization of the SNOM taper was done by measuring optical transmission approach curves over a gold film in combination with a  $k$ -space imaging technique [3]. At very small tip-sample separation the transmitted intensity sharply increases. Numerical simulations attribute this feature to the formation of an intense gap-mode between tip and sample, which is laterally confined to a length scale proportional to the geometric mean of the gap separation and the effective apex radius [4]. Hence, this extremely short ranged interaction has the potential to further improve the spectroscopic imaging resolution of nanofocusing SNOM down to the single nanometer regime.

Finally, we present photoelectron emission from the apex of a gold taper illuminated with few-cycle near-infrared laser pulses [5]. Compared to direct apex illumination, we find a fifty-fold increase in electron yield. Point-projection microscopic imaging of Ag-nanowires is performed and spatial localization of the electron emission to a nanometer-sized region is demonstrated.

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# **Controlling the generation of hot electrons in hybrid plasmonic nanostructures**

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The interaction of light and matter in noble metal nanosystems is governed by the collective oscillation of their conduction electrons, called surface plasmons. After excitation, these resonances experience ultrafast damping through radiative and non-radiative losses, i.e. absorption. The latter occurs through coupling to electronic inter- and intraband transitions in the metal, creating a highly non-thermal distribution of hot electrons. The electron population then decays through electron–electron interactions, creating thermalized hot electron distribution within a few hundred femtoseconds, followed by a further relaxation via electron–phonon scattering on the timescale of a few picoseconds. In the spectral domain, these processes can be observed by tracking the changes of the plasmonic resonance of the nanostructure due to the modification of the dielectric constant of the metal at different timescales. In this talk I will discuss strategies of enhancing hot electron generation in plasmonic hot spots and will show how their ultrafast response can be controlled both in temporal and spectral domain. I will present our recent results on hot electron generation in metal/oxide nanostructures and show how the geometry, the composition of the nanostructure and the excitation wavelength can drastically influence the ultrafast dynamics. The ability to enhance and control the generation of energetic electrons through specifically designed plasmonic nanostructures can be used in applications where hot electron generation is beneficial, such as in solar photocatalysis, photodetectors and nonlinear devices.

# Ultrafast nonlinear response of hybrid dielectric/plasmonic nanoantennas

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An important goal of the project “Ultrafast nonlinear optical response of metallic nanostructures: Collective effects and hybrid materials” is to boost the intrinsic nonlinear response of dielectric nanoparticles by employing the huge local field enhancement inherent to plasmonic nanostructures. To characterize the nonlinear response of the hybrid structures consisting of metallic nanoantennas and dielectric nanoparticles the technique of second harmonic generation spectroscopy is employed. For this purpose a frequency tunable femtosecond light source is developed. This light source is based on optical parametric generation and amplification with a double-pass geometry in a single macroscopic lithium niobate crystal. The device delivers more than two watts of tunable near-infrared radiation at 42 MHz repetition rate with pulse durations down to 200 fs. By combining the results of linear extinction measurements and SHG spectroscopy it is revealed that an increase of the SHG efficiency of plasmonic nanoantennas is obtained by filling their feed gaps with a dielectric nanoparticle. However, this enhancement of the SHG signal is independent of the nonlinear properties of the dielectric nanoparticles. Our experiments show that the intrinsic nonlinear response of plasmonic nanoantennas is several orders of magnitude higher than that of nonlinear dielectric nanoparticles and can be further boosted by employing double resonant nanoantenna geometries. To gain a deeper understanding of double resonant plasmonic nanostructures, plasmonic nanostructures are investigated which are resonant for the second harmonic light only. We observe an enhancement of the second harmonic efficiency even in the absence of resonances for the pump light. This property can be understood in the framework of the anharmonic oscillator model. Furthermore this study indicates that the general symmetry selection rules for second harmonic generation can be also applied to plasmonic nanostructures.



# Photocurrents and structural dynamics in nanomaterials probed by femtosecond electron pulses

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We report a novel concept for femtosecond point-projection microscopy (fsPPM) and femtosecond low-energy electron diffraction (fsLEED) utilizing laser-triggered electron emission from metal nanotips as pulsed electron source, delivering either divergent or collimated femtosecond single-electron wave packets in the sub-kV energy range. Due to the large scattering cross-section of low-energy electrons and their high sensitivity to electric fields, such electrons represent sensitive probes for the investigation of ultrafast currents and electric fields in nanoobjects as well as of atomic structure in low-dimensional materials with femtosecond time resolution. We demonstrate the visualization of ultrafast photo-induced currents in III-V semiconductor nanowires [1]. To further advance the temporal and spatial resolution in fsPPM, we realized plasmon-driven sub-10 femtosecond electron emission from a gold nanotip as femtosecond electron point source. Electron emission is triggered by the optical near field at the tip apex after laser excitation and subsequent nanofocusing of a surface plasmon polariton [2]. Structural dynamics in nanoscale materials, in particular few-layer semiconducting transition metal dichalcogenides and gold nanoparticles, are investigated with femtosecond transmission electron diffraction [3].

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## Nanoantenna-enhanced light-matter interaction of atomically thin WS<sub>2</sub>

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Atomically-thin transition metal dichalcogenides (TMDCs) are promising two-dimensional materials for optical and opto-electronic devices, because they exhibit an optical band gap in the visible regime. Monolayers absorb more than 10% of the light at their excitonic resonance and show photoluminescence. However, the absorption length of TMDC monolayers is extremely short and the photoluminescence quantum yield of  $10^{-3}$  is low. Therefore, strategies are needed to optimize the light-matter interaction. We present a hybrid system consisting of a plasmonic nanoantenna and atomically-thin WS<sub>2</sub> layer [1]. The antennas are single-crystalline gold nanorods, functionalized by a molecular layer. The nanorods are drop-cast onto a monolayer of tungsten disulphide (WS<sub>2</sub>), which was grown by chemical vapor deposition (CVD) on SiO<sub>2</sub>/Si substrate. Whereas the PL emission from the WS<sub>2</sub> monolayer alone shows no polarization dependence and is homogenous across the flake, a clear enhancement is observed at the position of the nanoantennas. By matching both excitation and emission polarization an overall PL enhancement of up to 11 is observed. The fact that the PL intensity strongly depends on the excitation as well as emission polarization indicates that absorption as well as emission are increased. This effect is due to the enhanced optical near-field created by the longitudinal plasmon resonance of the nanoantenna. The tailored hybrid nanoantenna-monolayer system lights the way to efficient photodetectors, solar cells, light emitting and conceptually new valleytronic devices based on two-dimensional materials.

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# **Time reversal versus adaptive optimization for spatiotemporal nanolocalization in a random nanoantenna**

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Spatiotemporal nanolocalization of ultrashort pulses in a random scattering nanostructure via time reversal and adaptive optimization is studied for two positions separated by only a tenth of the wavelength. The nanostructure is composed of core-shell nanoparticles placed randomly surrounding the target positions. The time reversal scheme achieves selective nanolocalization only by chance if the incident radiation can couple efficiently to dipolar local modes interacting with the target/emitter particle. Even embedding the structure in a reverberation chamber fails improving the nanolocalization. In contrast, the adaptive optimization reliably yields nanolocalization of the radiation and allows a highly selective excitation of either target position. This demonstrates that random scattering structures are interesting multi-purpose optical nanoantennas to realize highly flexible spatiotemporal optical near-field control.

# **Visualization of ultrafast electron dynamics using time-resolved photoemission electron microscopy**

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We constructed a TR-PEEM with 180fs and 90nm time and spatial resolutions, respectively. This compact facility makes possible to directly image the photo-generated electron dynamics in semiconductor on nm and fs scales. Carrier transport properties relating to device performance, carrier lifetime, drift velocity and mobility, are investigated.

# Nonlinear optical nano-antennas and nano-resonators from Lithium Niobate

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The notion of nano-antennas has altered our perception how to tame light. In most cases, nano-antennas are made from metallic structures to induce a resonant response at one or multiple wavelengths while being able to enhance light-matter-interactions. Here, we exploit nano-antennas to enhance the nonlinear optical response. For this purpose, the nano-antennas are in most cases supported by LiNbO<sub>3</sub>; being a strong  $\chi^{(2)}$ -nonlinear material. To harvest the potential of optical nanoantennas, it is required to have means at hand to tune their optical response. On the one hand, quantifying the response in terms of multipole moments allows to tailor how the nano-antenna scatters light. On the other hand, the near field enhancement and the efficient conversion of energy at multiple wavelengths are other criteria. In this contribution, we demonstrate the applicability of such methodology, both in theory and experiment, to study optical nano-antennas that enhance the nonlinear response from LiNbO<sub>3</sub>. For this purpose, isolated metallic nano-antennas are of interest whose geometry is tuned. We also consider nano-antennas to be embedded into an optical environment that enforces their optical response. An example is the surrounding of an electric dipolar nanoantennas with an annular metallic grating. Here, the grating serves to enhance the local field in the central antenna. In the cause of time it has been appreciated that optical nano-resonators can be also made directly from LiNbO<sub>3</sub>. The material possesses a high permittivity, which allows inducing strong resonances in spherical or cylindrical objects. Moreover, periodically patterning the material at length scales comparable to the wavelength allows to take advantage of multiply coherently scattered light. This enables the exploitation of photonic crystal effects to observe a notable nonlinear response. An overview across our activities in the last years in the context of this stream of research will be presented.

## Interplay of plasmon and exciton decay excited by ultrashort laser pulses

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Gold nanostructures (or nanoantennas) exhibit usually strong plasmon resonances, depending on their geometry and on the coupling of particles which influence significantly their emission properties. While scattering of coupled plasmonic systems has been in the focus during the last years studies of the photoluminescence (PL) emission of gold nanostructures excited by ultrashort laser pulses depends also on band-structure of these materials is by far less common. We will report on our recent studies aiming at a deeper insight the interplay between plasmons and excitons of gold nanostructures. In particular, we have investigated the linear and non-linear PL emission from various plasmonic systems like nanoparticles, sharp tips and nanotriangles [1-3] excited with radially or azimuthally polarized 100 fs laser pulses in combination with either diffraction limited confocal optical microscopy or tip enhanced scanning near-field optical microscopy.

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# Light-triggered Control of Plasmonic Refraction and Group Delay by Photochromic Molecular Switches

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Photochromic molecules have been successfully used in the past to achieve reversible control of optical transmission [1] and plasmonic damping [2] at metal-dielectric interfaces. In this paper we provide experimental evidence that in the same manner also the propagation of surface plasmon polaritons (SPP) can be steered. Switchable metal-dielectric interfaces are prepared from a gold substrate coated with polystyrene films doped with spirophenanthrooxazine molecules. Changes in the SPP dispersion relation at illumination with light of different wavelength are measured using photoemission electron microscopy [3] evidencing a substantial and reversible switching of SPP group and phase velocity [4]. The results imply the realization of nonvolatile plasmonic switching units providing new and complex functionalities. Based on this concept we realize a switchable plasmonic lens as a first demonstrator for potential applications. Reversible focus control is experimentally demonstrated yielding changes of about 5% of the total focal length.

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# Two Dimensional Coherent Spectroscopy of Epitaxial Quantum Dots

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Two-dimensional coherent spectroscopy has the ability to make size resolved measurements in an inhomogeneously broadened ensemble of nano-objects where the inhomogeneous distribution of resonance energies arises from the size fluctuations of the objects [1]. This ability results from correlating the frequencies during two time periods, via a two-dimensional Fourier transform. This concept will be used to make measurements on “natural” quantum dots, which occur in thin quantum wells due to intrinsic well-width fluctuations, and used to measure the size dependence of the exciton-phonon and exciton-exciton interaction [2]. It will then be used to study InAs self-organized quantum dots, which display biexcitonic side bands. The results show that the biexciton binding energy appears to be independent of dots size within the inhomogeneous distribution, whereas single dot studies showed significant variation. By using a pre-pulse to coherently prepare the ensemble, clear Rabi oscillations are observed for the individual size groups. The results show that both ground-state to exciton and exciton to biexciton Rabi oscillations are occurring simultaneously due to the use of broadband pulses.

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# **Proposal of using nanoplasmonics and coherent two-dimensional spectroscopy to dissect secular and non secular relaxation processes**

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Relaxation or coherence conversion caused by system bath interaction, like electron-phonon interaction, is often described sufficiently by secular processes and the Markovian approximation. Only conversion processes, where the involved excitonic populations or coherences are nearly resonant, are included in secular processes. For many treatments, non-secular process can be often neglected. However interesting Non-Markovian signatures potentially involve secular as well as non secular processes. The dissection and study of non secular and secular processes is routinely carried out in theory, however the identification of a pure non secular experimental observable was pending. This is especially important for testing various Non-Markovian theories. We suggest an experimental signal, which we includes only contributions from non secular processes in an idealized case. We use a combination of coherent spectroscopy and nanoplasmonics to detect non secular relaxation processes in a quantum dot. The optical pulses at the quantum dot are dynamically controlled by the nanoplasmonic structure, to have either a vanishing field or field gradient at the quantum dot position. This results in a control of optical selection rules, which we use to detect Liouville pathways including only non secular processes with coherent spectroscopy. This will open the door to study and test nonsecular processes in experiments against e.g. elevated Non-Markovian theories. Furthermore we also discuss the robustness of the protocol to error in the dynamical control of optical selection rules.

# Tracking ultrafast energy flow in plasmonic nanostructures

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A very recent novel branch of plasmonics is aimed at the investigation of the nonlinear optical phenomena taking place in metal nanostructures (NS), and which can be exploited for a new generation of devices with application to all-optical switching and sensing. To address the challenging task of designing such nonlinear nanoplasmonic devices, it is necessary to quantitatively understand and model the nonlinear optical phenomena taking place on the ultrafast timescale in metallic NS, which occur on a hierarchy of timescales, ranging from tens of fs to hundreds of ps. An ultrashort pump pulse impinging on the NS excites free electrons in a non-thermal distribution, with an excess energy per unit volume that is subsequently rapidly released, by electron–electron scattering, to the thermalized electronic population, heating it up to a higher temperature. Subsequently, electron–phonon scattering causes the electronic gas to cool down and the lattice to increase its temperature, until a long-living equilibrium temperature is achieved. A much slower phonon–phonon scattering process causes the excess energy originally delivered to the nano-object to flow into the environment. If the timescale of the lattice heating is much shorter than the period of mechanical vibrational modes of the NS, coherent oscillations can be launched which modulate the extinction of the nanostructure. The period and damping of these oscillations provide information on the geometrical properties, the speed of sound and the Young modulus at the nanoscale. This presentation will introduce several examples of ultrafast spectroscopy of metal nanostructures, from thin films to metal-dielectric heterostructures, individual colloidal and lithographed nanoparticles finally and self-assembled supracrystals.

## Active plasmonics with surface acoustic waves

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In my talk, I will discuss the impact of coherent surface acoustic waves (SAWs) on plasmonic devices. These high-frequency mechanical distortions can be driven all-electrically using conventional radio-frequency electronics. In a first step, we demonstrate a SAW driven converter of light into surface plasmon polaritons. In essence, an otherwise unstructured metal thin film is deformed by surface-bound acoustic waves traveling on a piezoelectric substrate underneath. This spatially periodic corrugation enables to overcome the wavevector mismatch between free-space radiation and surface-bound electro-magnetic modes. This concept is demonstrated for gold thin films on a  $\text{LiNbO}_3$  chip. In particular, interdigital transducers generate  $\sim 500$  MHz surface acoustic waves of  $\sim 1$  nm surface ripple on the substrate and the deposited metal film. For near-infrared light of 950 nm wavelength we observe a 0.01% efficiency for the excitation of surface plasmon polaritons. As a next step, we explore the influence of radio-frequency SAWs travelling across a commensurable, static gold grating. Such a structure constitutes a paradigm for a pre-defined injector for surface plasmon polaritons. Here, the electro-mechanically induced, dynamic surface deformation strongly modulates the launcher's coupling characteristics on sub-nanosecond timescales. The modulation of the efficiency is as large as 2% and is monitored in real time with a stroboscopic technique utilizing SAWs synchronized to an optical pulse train.

# **Simulation and control of light propagation in ultrasmall noble-metal cluster arrays**

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We present a theoretical approach for simulation of light propagation in nanosized noble-metal cluster arrays. Within the approach the array is represented by a Hamiltonian with the coupling elements calculated based on transition dipole moments for arrays with well-separated subunits and transition charges in the opposite case. The proposed approach is applied to simulation of absorption spectra and electron dynamics in  $\text{Ag}^{3+}$  arrays and in porphyrin-based  $\text{Ag}^4$  arrays. Additionally, a possibility of optimal control of light propagation in a T-shaped structure consisting of ultrasmall silver clusters is investigated.

# Graphene plasmonics

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Graphene plasmons have recently attracted much attention because of their excellent electrical tunability and extreme sub-wavelength confinement, which enable applications such as fast light modulation, improved biosensing, and quantum optics in robust solid-state environments. In this presentation, we review recent theoretical and experimental advances in these directions and explore further possibilities, such as sensing at the single-molecule level, electrical detection of single plasmons, and ultrafast transient plasmonic phenomena.

## Short-range surface plasmonics and its (sub-)femtosecond dynamics

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We use single crystalline gold flakes on atomically flat silicon substrates to generate ideally suitable metals for plasmon propagation. By electrochemical means, the thickness is tunable from a few tens to over 100 nm. Using sub-20 fs laser pulses around 800 nm, we excite surface plasmons, whose dynamics can be observed using time-resolved two-photon excitation electron emission (PEEM).

Plotting the dispersion of surface plasmons in a thin gold slab on silicon, one finds that excitation at 800 nm can lead to extreme wavelength reduction due to the dispersion slope of over five. Using focused ion beam for cutting rings with appropriate periodicity into the samples (see left image), we can excite concentric surface plasmons that create a nanofocus of only 60 nm width for 800 nm excitation. Using Archimedean spirals with broken n-fold radial symmetry, it is possible to excite surface plasmons with angular orbital momentum on the gold flakes. This leads in case of 4-fold symmetry to cloverleaf-type nanofoci on the order of 100 nm, which rotate during four optical cycles by 360 degrees.

Using two-pulse experiments with a subwavelength-stabilized Michelson interferometer, it is possible to observe the dynamics of the surface patterns with a (sub-)femtosecond resolution, thus giving insight into the dynamics of the nanofocus formation as well as on the plasmonic spin-orbit coupling.

# Optically-Induced Entanglement in Hybrid Quantum Dot/Plasmonic Systems

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Quantum dynamics calculations are carried out on model Hamiltonians corresponding two, three, and four quantum dot systems interacting with a dissipative, plasmonic system that is subjected to ultrafast laser pulses. We show how the pulses can be used to create multiple entanglements among the dots from an initially cold systems.

## Coherent control in single plasmonic nanostructures

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In this contribution we present a new way to visualise coherent control of plasmonic resonances in single metal nanostructures with sub-diffraction dimensions. The approach makes use of the extremely high localisation accuracy that can be achieved in far-field nanoscopy. Here we apply it to the detection of the position of the second harmonic generation (SHG) and non-linear photoluminescence from gold nanostructures. Due to the high signal levels and their temporal stability the center-of-mass of these signals can be tracked with sub-nanometer resolution. We demonstrate that the center-of-mass can be controlled within a single nanostructure by chirping the spectral phase of the excitation pulse. We attribute this to the control of the interference of plasmonic resonances with different spatial profiles within the laser spectrum.



## Shaping the nonlinear near-field

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Linear scattering and absorption of plasmonic nanoparticles and their assemblies have led to a wealth of application in metamaterials and nanooptics. Almost any desired field distribution can be realized by well-chosen arrangements of multiple particles. While the shaping of fields around nanostructures is widely studied, the influence of the field inside nanostructures is often overlooked. The linear field distribution inside the structure taken to the third power causes third-harmonic generation, a nonlinear optical response of matter. Here we demonstrate how this simple fact can be used to shape complex fields around already a single particle alone. We employ this scheme to switch the third-harmonic emission from a single point source to two spatially separated but coherent sources, as in Young's double slit assembly. We envision applications as diverse as coherently feeding antenna arrays or optical circuits and optical spectroscopy of spatially extended electronic states.

## **Nonlinear responses in plasmonic nanostructures**

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Transferring the concepts of nonlinear optics down to the nanoscale is highly desirable for practical applications, but it suffers from the intrinsic weakness of the nonlinear responses. In this context, plasmonics is foreseen to play a major role thanks to the associated field enhancement and spatial tailoring of the near-field maps. For example, two photon luminescence (TPL) has been widely used at the single particle level to infer the field localization at the nanoscale, becoming a key tool for investigating the plasmonic antennas. Thought, as we have recently demonstrated, this technique fails in providing a clear view on another popular nonlinear response, the second harmonic generation (SHG). Using a specific particle geometry (a gold nanoprism with a triangular base) we will evidence the main differences between SHG and TPL but also between the two mechanisms involved in the SHG process. In addition, we will discuss different strategies to enhance the nonlinear efficiency and show how double resonance condition (resonance at the fundamental and the harmonic frequency) can be achieved using aluminum nanostructures. We will present our latest results obtained on a new experimental and simulation platform we have developed at the Néel Institute.

## **Second harmonic generation in plasmonic nanostructures: multiresonant nanoantennas and eigenmodes**

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During this presentation, we provide a full understanding of the mechanisms that lead to SHG in multiresonant plasmonic structures. By combining experiments on aluminum plasmonic nanostructures with surface integral equation simulations, we have investigated the SHG modulation from double resonant nanoantennas (DRAs) with a broad variety of geometries emphasizing the role played by the mode coupling in the nonlinear conversion enhancement. In a second part, we will show that SHG can also provide information on the underlying modes supported by a given plasmonic nanostructure. Indeed, a combination of modes at the fundamental frequency can generate SH waves supported by modes that cannot be excited at the fundamental frequency, the so called dark modes. SHG can then be used to measure the radiation pattern of those dark modes. If the fundamental frequency matches a resonance of the structure, the high field enhancement will yield a high SHG. Additionally, if the SH frequency matches another higher frequency mode, the SH emission will be stronger and thus more easily measurable as in the case of the DRAS. This will be very useful for the design of practical applications as nonlinear plasmonic sensing.

# **The Photon Wave Function: a bridge from Maxwell to Schroedinger, and beyond**

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When few photons are present, the photon wave function is a useful tool for understanding as well as exploring the predictions of QED. In certain circumstances (for instance when the state of light is coherent in the sense of Glauber), it is formally nearly equivalent to a classical, Maxwell field, but in other circumstances it exhibits a surprising behaviour. For instance, during the process of spontaneous emission of light by an excited atom, the photon wave function slightly violates Einsteinian causality, in accordance with Hegerfeldt's theorem. Our goal is to give an overview of these topics, and to discuss related departures from the Fermi golden rule that were never observed so far but constitute an exciting challenge for experimentalists.

# Coherent and long-range plasmon-assisted energy transfer between two plasmonic nanoantennas

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Short-range coherent energy transfer dynamics e.g. in light harvesting complexes and their implications from a control systems perspective has been discussed vividly. Here, the long-range energy transfer mechanism between two coupled plasmonic whispering gallery nanoantennas [1] in an elliptical cavity [2] is investigated by optical excitation with (shaped) sequences of ultrashort pulses centered at 800 nm. Only one gold antenna is excited selectively when the structure is illuminated under grazing incidence. Plasmon-enhanced time-resolved photoemission microscopy [3] detected at both nano-antennas reveals periodic energy transfer on a micrometer length scale in the hybridized antenna-cavity system. Exposing single quantum emitters to the antenna-induced field enhancement these systems might serve for future applications involving weak, or even strong, interaction of well-separated single quantum systems.

[1] E. J. R. Vesseur et al., *Nano Lett.* **9**, 3147-3150 (2009)

[2] D. T. Schoen et al., *Nano Lett.* **13**, 188-193 (2013)

[3] M. Aeschlimann et al., *Science* **333**, 1723-1726 (2011)

## Coherent electron-light interaction in an ultrafast electron microscope

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We implemented the first ultrafast transmission electron microscope (UTEM) [1] based on a laser pump/electron probe technique that operates with a nanoscale laser-driven photocathode. Here, we employ the advanced beam properties of the UTEM to study quantum coherent electron-light interactions. In our experiments, a tightly focused, pulsed electron beam traverses the optically excited near-field of a nanostructure. The spatial confinement of the near-field allows for an otherwise forbidden coupling between the free electrons and photons, leading to strongly modulated electron kinetic energy spectra. Field strength dependent Rabi oscillations in the sideband population reveal the quantum coherence of the process [2]. Free propagation over a few mm will reshape the electron wave function into a train of attosecond pulses, in principle allowing for the study of attosecond phenomena in electron microscopy. In an advanced experiment, we control the free electron momentum superposition states with two spatially separated interaction sites, in some analogy to the Ramsey method for atomic clocks.

[1] A. H. Zewail, *Science* 328, **187** (2010).

[2] A. Feist, K. E. Echternkamp, J. Schauss, S. V. Yalunin, S. Schäfer, and C. Ropers, *Nature* **521**, 200-203 (2015).

## Abstracts of Poster Presentations (*sorted by poster number*)

#	Presenting Author	Title
1	<i>Felix Becker</i>	Nanoantenna supported thermionic current injection in metal-insulator-metal junctions
2	<i>Markus Betz</i>	Quantum interference control of electrical currents in GaAs microstructures: physics and spectroscopic applications
3	<i>Tobias Birr</i>	Ultrafast surface plasmon-polariton logic gates and half-adder
4	<i>Soo Hoon Chew</i>	Current progress on probing and control of near-field dynamics using photoemission electron microscopy
5	<i>Benjamin Frisch</i>	Coherent and long-range plasmon-assisted energy transfer between two plasmonic nanoantennas
6	<i>Cristian González</i>	Investigation of the dispersion relation of SPPs with Mie scattering probes in the optical and near infrared range
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9	<i>Alwin Klick</i>	Amplitude and phase of surface plasmon polaritons excited at step edges
10	<i>Aleksander Kovačević</i>	Inducing nanoparticles and periodic nanostructures on thin metal films by low-fluence femtosecond beam
11	<i>Martin Lehr</i>	Time of flight momentum microscopy of plasmon assisted photoemission
12	<i>Polina Lisinetskaya</i>	Methods for simulation of light propagation in small metal cluster arrays
13	<i>Anna-Katharina Mahro</i>	Time- and space-resolved imaging of gold nanostructures
14	<i>Martin Piecuch</i>	MANTIS – A novel XUV-IR pump probe photoemission scheme for the investigation of attosecond dynamics
15	<i>Daniel Podbiel</i>	Exploring Plasmoemission of Electrons in NI-PEEM
16	<i>Simon Ristok</i>	Bottom-up synthesis of large area single-crystalline atomically flat gold flakes with single crystalline silver cubes as single quantum emitter nanoantennas
17	<i>Carsten Rockstuhl</i>	Nonlinear optical plasmonic nanoantennas from Lithium Niobate
18	<i>Benjamin Schröder</i>	Nanotip surface plasmons: real-space imaging using EELS and grating-coupled nonlinear photoemission
19	<i>Thorsten Schumacher</i>	The optimal antenna for nonlinear spectroscopy of weakly and strongly scattering nanoobjects
20	<i>Martin Silies</i>	Light localization in disordered nanostructures - Giant fluctuations of Second Harmonic and electron emission
21	<i>Vladimir Smirnov</i>	Suppression of radiative damping and enhancement of second harmonic generation in bull's eye nanoantennas
22	<i>Judith Specht</i>	Applications of nanoplasmonics: Full quantum statistics of many emitter spasers and spin dependent reconstruction scheme of exciton wave functions
23	<i>Philip Thielen</i>	Long-Lived Coherence in the Metal-Organic-Hybrid Cobalt/Alq <sub>3</sub>
24	<i>Felicitas Walter</i>	Controlling the excitation of localized plasmonic resonances by strong near-field coupling
25	<i>Xiao Wang</i>	Plasmon and exciton decay in nanostructures excited with ultrashort pulses formed by cylindrical vector laser beams

## Poster 1:

### Nanoantenna supported thermionic current injection in metal-insulator-metal junctions

Felix Becker<sup>1</sup>, Detlef Diesing<sup>2</sup>, Dominik Differt<sup>1</sup>, Adelheid Godt<sup>3</sup>, Ingo Heesemann<sup>3</sup>,  
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In the interaction of intense laser pulses with nanostructures or optical antennas the electron emission is commonly discussed in the context of multi-photon processes [1] and strong field phenomena [2-5]. The latter process facilitates the opportunity to implement the scheme of “lightwave electronics” [6], in which electronic motion is coherently steered by the electric field itself. Although this scheme is studied at the metal-vacuum interface of nanostructures, it is not yet demonstrated for electrons remaining inside a nanodevice.

Few-cycle laser pulse injected currents in metal-insulator-metal junctions might fill this gap. Recently, it has been shown that plasmonic resonances at structural defects on the top electrode of the heterosystem facilitate the injection of charge carriers above the tunnel barrier of the oxide [7]. Here, we demonstrate the prearranged injection of charge carriers by exploiting the near field enhancement of gold nanoparticle antennas on the top electrode. In contrast to multi-photon and strong field interaction, we identify strongly localised thermionic emission as the dominating process for current injection. Experimental evidence for the transition from multi-photon excitation to thermionic emission is supported by finite-difference time-domain simulations and time-resolved calculations of the excited electronic distribution function. Time-resolved heat diffusion calculations of the electron gas model the spacial temperature distribution and thus the transient thermionic injection current. The results serve as an onset to overcome incoherent obstacles on the way to coherently controlled nanodevices on the sub-femtosecond time scale.

[1] J. Lehmann et al., Phys. Rev. Lett. **85**, 2921–2924 (2000)

[2] R. Bormann et al., Phys. Rev. Lett. **105**, 147601 (2010)

[3] P. Dombi et al., Opt. Express **18**, 24206–24212 (2010)

[4] M. Krüger et al., Nature **475**, 78–81 (2011)

[5] B. Piglosiewicz et al., Nature Photon. **8**, 37-42 (2014)

[6] E. Goulielmakis et al., Science **317**, 769-775 (2007)

[7] D. Differt et al., Appl. Phys. Lett. **101**, 111608 (2012)



## **Poster 2:**

### **Quantum Interference Control of Electrical Currents in GaAs Microstructures: Physics and Spectroscopic Applications**

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We present a comprehensive study of coherently controlled charge currents in electrically contacted GaAs microdevices. Currents are generated all-optically by phase-related femtosecond pulse pairs (fundamental/second harmonic) and are often linked to the third order optical nonlinearity. Here, we first focus on elevated irradiances where absorption saturation and ultimately the onset of Rabi oscillations contribute to the optical response. In particular, we identify clear departures of the injected current from the third order response. Theoretical simulations for the coherently controlled current based on the semiconductor Bloch equations agree well with the experimental trends. We then move on to investigate spectroscopic applications of the quantum interference control technique. In particular, we implement a versatile scheme to analyze the phase structure of femtosecond pulses. It relies on phase-sensitive current injection driven by two time-delayed portions of the pulse pair. Most strikingly, the group velocity dispersions of both the fundamental and second harmonic components can be unambiguously determined from a simple Fourier transform of the resulting current interferogram. Finally, we aim to use femtosecond pulse pairs to demonstrate a theoretically proposed scheme for all-optical current detection in thin GaAs membranes. However, we find the signal to be superimposed by second harmonic generation related to the electric field inducing the current. As a result, the currents' signature cannot be unambiguously identified.

### **Poster 3:**

## **Ultrafast surface plasmon-polariton logic gates and half-adder**

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In 1965, Moore verbalized his law about doubling of the packing density of circuits in electronic devices. However, this downsizing combined with an increase of operational speed is facing physical limits. On the other hand, optical fibers are used since decades but they have much bigger dimensions than electronic devices and are still using electronic front and back ends. Solving this mismatch would offer the possibility to strongly increase computational capacities. One possibility is the use of surface plasmon-polaritons as carriers of information. For this purpose several groups recently investigated the properties of different plasmonic structures in terms of the usage as plasmonic logic elements. These published approaches contain limitations since either random structures have been used or the system under consideration suffers from severe damping leading to extremely short propagation lengths. In this work, dielectric plasmonic waveguides are used for the construction of logic gates for near infrared light. They were investigated experimentally regarding their efficiency depending on their geometrical properties. These measurements, obtained by leakage radiation microscopy, have been compared with FDTD simulations of the same structures. The results were used to design plasmonic half adders which functionality has been demonstrated experimentally. These investigations are an important step in the development of optical technologies which could lead to a next photonic era.

## **Poster 4:**

### **Current progress on probing and control of near-field dynamics using photoemission electron microscopy**

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<sup>2</sup> *Max-Planck-Institut für Quantenoptik, 85748 Garching, Germany*

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Here we present the concept of carrier-envelope phase-tagged time of flight-photoemission electron microscopy and the recent experimental results. We discuss the laser intensity effects on the current phase-resolved measurements of gold nanoparticles on gold plane and surface roughness from a gold film. The apparent carrier-envelope phase modulation with a period of  $\pi$  is found to originate from an intensity dependence of the photoelectron spectra and the phase potato rather than from an intrinsic carrier-envelope phase dependence. We propose next steps such as the implementation of intensity tagging and improved plasmonic sample to probe and control near-field dynamics from supported nanostructures.

## Poster 5:

### Coherent and long-range plasmon-assisted energy transfer between two plasmonic nanoantennas

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Matthias Hensen<sup>5</sup>, Christian Kramer<sup>2</sup>, Enno Krauss<sup>4</sup>, T. Löber<sup>7</sup>, Walter Pfeiffer<sup>5</sup>,  
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Hybridization of localized plasmonic resonances and propagating surface plasmon-polaritons (SPP) facilitates to combine the benefits of two worlds: Longevity and coherent light concentration. This combination has recently been demonstrated in random plasmonic resonators [1]. Here, we present experimental and theoretical results on a prearranged system that allows long-distance ( $> 2\lambda_0$ ) coupling between two plasmonic nanoantennas [2] mediated by an extended SPP mode in an elliptic all-metallic cavity [3]. The system, qualitatively described by three coupled differential equations, is prepared on single crystalline gold flakes [4] allowing plasmonics on atomically flat surfaces. First experiments on temporal dynamics of plasmonic excitation with time-resolved PEEM confirm the coherent back and forth transfer of energy between widely separated nanoantennas. Moreover, we show theoretically that the system obeys the phenomenon of impedance matching: By tuning the dissipation of the cavity it is possible to periodically switch on the excitation at one antenna while the opposite antenna remains dark. Besides applications of the here presented system in long-range routing of plasmonic excitations the cavities might even serve for interesting experiments involving quantum systems coupled to incorporated nanoantennas.

[1] M. Aeschlimann et al., *Science* 333, 1723-1726 (2011)

[2] E. J. R. Vesseur et al., *Nano Lett.* 9, 3147-3150 (2009)

[3] D. T. Schoen et al., *Nano Lett.* 13, 188-193 (2013)

[4] X. Wu et al., *Cryst. Res. Technol.* 50, 595–602 (2015)

## **Poster 6:**

### **Investigation of the dispersion relation of SPPs with Mie scattering probes in the optical and near infrared range**

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We present a cantilever based near field probe with integrated Mie scattering dielectric silicon dioxide microsphere (MSDM) for near-field optical imaging as well as femtosecond spectroscopy applications. In contrast to the state of the art transmissive near-field probes, the MSDM reveals a transmission of almost unity known from far-field microscopy configuration. For proper handling the microsphere is integrated at the apex of a conventional pyramidal aperture tip carried by an atomic force microscopy cantilever. It proved to be mechanically robust during the scanning process even if operating it in the contact mode. The spherical symmetry provides on one hand a well defined mechanical contact point with the sample irrespective of its inclination angle to the sample surface. On the other hand, the symmetry of the device preserves the polarization of light proving to be useful for the investigation of the polarization dependent behavior of plasmonic nanostructures. The device has also shown to be able to be an excellent tool for the study of surface plasmons on Au thin films when measuring the dispersion relation of SPPs, bringing results with an excellent agreement with similar experiments performed with PEEM . The high transmission combined with low dispersion renders spectroscopic investigations on the femtosecond time scale with a moderate lateral resolution. Moreover, second autocorrelation experiments on a BBO-crystal reveals a time resolution well below 100 fs at 191 nm spatial resolution.

## Poster 7:

### Anderson localization of light in randomly nanotextured layers as a new paradigm for the design of highly efficient thin-film absorbers

Martin Aeschlimann<sup>1</sup>, Tobias Brixner<sup>2,3</sup>, Dominik Differt<sup>4</sup>, Ulrich Heinzmann<sup>4,5</sup>, Matthias Hensen<sup>4</sup>, Bernhard Huber<sup>2</sup>, Christian Kramer<sup>2</sup>, Florian Lükermann<sup>4,5</sup>, Pascal Melchior<sup>1</sup>, Walter Pfeiffer<sup>4</sup>, Martin Piecuch<sup>1</sup>, Christian Schneider<sup>1</sup>, Helmut Stiebig<sup>4,5</sup>, Christian Strüber<sup>6</sup>, and Philip Thielen<sup>1,7</sup>

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<sup>2</sup> *Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany*

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Unraveling the localization mechanisms of light in disordered nanostructures is of great interest in a number of applications, including photovoltaics and thermoelectrics. The efficient use of natural resources and physical constraints in photovoltaic devices require thin but efficient absorbers. Although already exploited in commercial devices, the enhancement mechanism for devices with nanotextured interfaces is still subject to debate. Here, we demonstrate the existence of localized photonic states in nanotextured amorphous silicon layers as used in commercial thin-film solar cells using coherent light scattering and coherent two-dimensional nanoscopy [1]. Resonant absorption in these Anderson localized photonic modes accounts for enhanced absorption in the long-wavelength cutoff region giving rise to interesting opportunities for future highly effective absorber layer designs based on this localization mechanism.

- [1] M. Aeschlimann, T. Brixner, D. Differt, U. Heinzmann, M. Hensen, C. Kramer, F. Lükermann, P. Melchior, W. Pfeiffer, M. Piecuch, C. Schneider, H. Stiebig, C. Strüber, and P. Thielen, *Nature Photon.* **9**, 663 (2015).

**Poster 8:**

**Ultrafast sum-frequency and second-harmonic generation  
in plasmonic nanostructures**

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Optical second-order nonlinear spectroscopy has been a subject of investigations for over two decades [1,2]. Nevertheless, its theoretical understanding is still emerging. Based on a hydrodynamic description, featuring nonlinear as well as nonlocal characteristics, we present time-domain computations of three-wave-mixing signals, in particular second-harmonic and sum-frequency signals, from plasmonic nanoantennas.

- [1] J. E. Sipe, V. C. Y. So, M. Fukui, and G. I. Stegeman. Phys. Rev. B, **21**:4389–4402, May 1980.
- [2] J. A. Maytorena, W. L. Mochán, and B. S. Mendoza. Phys. Rev. B, **57**:2580–2585, Jan 1998.

## Poster 9:

### Amplitude and phase of surface plasmon polaritons excited at step edges

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We present a combined theoretical and experimental study on the laser-induced excitation of surface plasmon polaritons (SPPs) at defined step edges representing one of the most basic light-SPP coupling geometries. The calculations are performed within an integral equation approach for the solution of two-dimensional scattering problems [1], whereas in the experimental part we utilized two-photon photoemission electron microscopy (2P-PEEM) [2]. Analysis of the data provides information on the laser-SPP coupling efficiency of the step edges and the phase of the excited SPP. As an important parameter determining the coupling efficiency we identify the ratio between step height  $H$  and excitation wavelength  $\lambda$ . For specific values of  $H/\lambda$  a substantial suppression of light-SPP coupling is observed. At the same time the phase of the SPP undergoes an overall shift by  $\pi$ . Based on the calculations it is possible to assign the changes in coupling efficiency and phase to the interference of incident and reflected laser field in the presence of the step edge.

[1] De la Cruz, S. et al., *Physica Status Solidi (B)* **249(6)**, (2012)

[2] Lemke, C. et al., *Applied Physics B* **116** (2013)



## **Poster 10:**

### **Inducing nanoparticles and periodic nanostructures on thin metal films by low-fluence femtosecond beam**

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In many fields, like semiconductor technology, optics, chemistry, mechanics, magnetism, electricity, the applications of thin films play an important role. Various types of coatings for protection, diffusion barriers, filtering, reflection/antireflection, sensing, waveguiding, decorative and other purposes are just some to mention. Structuring of thin films can enhance their characteristics. From the other side, nanoparticles are of great interest due to their position between bulk materials and atomic/molecular structures. The interaction of femtosecond laser beam with thin films can have various outcomes, in which the generation of nanoparticles and of surface periodic structures gains more interest. We have used low-fluence femtosecond beam of various wavelengths and exposures to thin film samples: Al on Si, CrVN, multilayer Al/Ti on Si and multilayer Ni/Pd on Si. Having excellent mechanical characteristics (wearing, corrosion resistance), some samples have prospective applications in holography. The exposures yielded the generation of nanoparticles and the formation of periodic surface structures (LIPSS). The processes of liquid or solid-state dewetting could be responsible for regrouping of generated nanoparticles and nanoparticle clusters, while LIPSS is most probably induced by the occurrence of the surface plasmon polariton. The presence of the sublayer, depending on its optical characteristics, strongly influences both the shape and the quality of the LIPSS.

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## **Poster 11:**

### **Time of flight momentum microscopy of plasmon assisted photoemission**

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The photoemission properties of a Au nano triangular array have been investigated using a time-of-flight-momentum microscope (ToF-PEEM), equipped with a state-of-the-art Surface Concept delay line detector (DLD). The nano triangular array has been fabricated by deposition of a gold film on a self-assembled colloidal sphere structure. The triangles can be resonantly excited by Ti:sapphire femtosecond laser pulses. The sample is illuminated from the backside at normal incidence. The dependence of the electron momentum distribution on the light polarization has been investigated. The energy spectra of the emitted electrons show unexpectedly high energies of up to 4 eV.

## **Poster 12:**

### **Methods for simulation of light propagation in small metal cluster arrays**

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We present a theoretical approach for the simulation of the optical response and electric field propagation in ordered arrays constructed of small noble-metal clusters including the effect of a support. In such systems the size of an array subunit is comparable or larger than the separation between neighboring subunits. In order to describe the interaction between individual constituents we use the transition charge method. The electronic population dynamics in the array under an external laser pulse action is simulated by numerical integration of the time-dependent Schrödinger equation with the coupled array Hamiltonian. The results are employed to represent each subunit of the array by a system of time-dependent partial point charges and the spatio-temporal electric field distribution is evaluated by means of classical electrodynamics methods. The time-dependent partial charges are determined based on the stationary partial and transition charges obtained in the framework of the ab initio TDDFT. In order to treat large arrays constructed of many constituents the approximate self-consistent iterative approach presented in [1] is modified to include transition-charge-based interaction between subunits. The developed methods are used to study the optical response of  $\text{Ag}_3^+$  arrays, to control the light propagation in a T-shaped structure constructed of seven  $\text{Ag}_8$  clusters, and to simulate the spatio-temporal electric field distribution in a ring constructed of ten porphyrin- $\text{Ag}_4$  subunits under the action of circularly-polarized laser pulse.

[1] P. Lisinetskaya and R. Mitrić, Phys. Rev. B **89**, 035433, 2014.

## **Poster 13:**

### **Time- and space-resolved imaging of gold nanostructures**

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Knowledge of fundamental light-matter interactions such as the local response of nanostructures to incident light is necessary for designing plasmonic devices. This response is determined on the nanometer scale (below the diffraction limit of light). We therefore use a photoemission electron microscope (PEEM) to image the near-field distribution. To gain information about the local response of our gold nanostructures, we apply phase and time resolved two photon photoemission techniques. We use a phase-stabilized Mach-Zehnder-interferometer based on Pancharatnam phase [1,2] in combination with a broadband ultrashort pulse laser and PEEM. This setup provides a temporal precision in the pump-probe delay of 30 as thus allowing the dynamics of plasmon excitations in different nanostructures to be compared in real time. Under resonant excitation the phase-resolved autocorrelations from these nanostructures contain information about their local response.

[1] S. Pancharatnam, Proc. Indian Natl. Sci. Acad., A **44**, 247-262 (1956)

[2] M.U. Wehner et al., Opt. Lett. **22**, 1455-1457 (1997)

**Poster 14:**

**MANTIS – A novel XUV-IR pump probe photoemission scheme for the investigation of attosecond dynamics**

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Reconstruction of attosecond beating by interference of two-photon transitions (RABITT) is a powerful and well-established method for attosecond pulse characterization. Furthermore the method proved to be very useful for the investigation and the understanding of phase shifts in core-level photoemission spectra. However, as RABITT makes use of the energy comb of high-harmonics in an attosecond pulse train (APT), one collects multiple overlapping photoemission spectra, which renders the interpretation and investigation of phase shifts in valence-band spectroscopy a hard task. Here we present an extension of the RABITT scheme (named MANTIS) by combining it with a method derived from 2D-spectroscopy. We combine the APT with two IR-laser pulses and use the concept of phase cycling to select specific coherence pathways which results in clean high-resolution phase resolved photoemission spectra. We show that this method will open up a new and powerful route to study attosecond dynamics in atoms, molecules and materials.

## **Poster 15:**

### **Exploring plasmoemission of electrons in NI-PEEM**

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We use time-resolved normal-incidence photoemission electron microscopy (NI-PEEM) to study surface plasmon polariton (SPP) propagation, interference, and focussing. We analyzed the probing contrast in NI-PEEM and found that it is caused by the in-plane component of the SPPs electric field. We demonstrate that the measurement signatures in NI-PEEM (arising due to time-integration) can be easily explained with the help of ‘space-time’ diagrams. From an analysis of the signatures we can conclude that SPP-induced emission of electrons, referred to as ‘plasmoemission’, is governed by the out of-plane component of the SPPs electric field. To achieve higher plasmoemission yields, we studied focussing of SPPs at a Fresnel-type grating coupler as well as in circular grating couplers. At the focus points of circular grating couplers multiplasmon plasmoemission processes up to an order of six could be observed. Furthermore an SPP-induced material transport occurs, and leads to the formation of nanostructures, at which localized electron emission take place. Kinetic energy spectra of the emitted electrons indicate a transition into a different electron emission regime.

## **Poster 16:**

### **Bottom-up synthesis of large area single-crystalline atomically flat gold flakes with single crystalline silver cubes as single quantum emitter nanoantennas**

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When using quantum emitters such as NV centers in diamond, quantum dots, or dye molecules as effective photon sources, it is often necessary to control certain characteristics of the emitted radiation, e.g., the emission rate and direction. A promising approach is the coupling of the emitters to a plasmonic nanoantenna. In this work, we fabricate plasmonic nanopatch antennas using exclusively bottom-up methods. The antennas consist of silver nanocubes which are positioned over the atomically flat surface of a gold microplate. Both Ag nanocubes and Au microplates are single-crystalline and are separated by a dielectric polymer spacing layer. The plasmonic resonance of the antennas can be tuned by varying the size of the Ag nanocubes and the thickness of the spacing layer. The further steps include the embedding of fluorescent dye molecules into the gap of the nanopatch antenna, followed by fluorescence enhancement and lifetime measurements. Investigating this system can help us understanding the prerequisites to expand the range of emitters also to NV centers and quantum dots.

## **Poster 17:**

### **Nonlinear optical plasmonic nanoantennas from Lithium Niobate**

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Optical nanoantennas, or photonic nanostructures in more general, can be used to shape the propagation characteristics of light as well as its localization. Both aspects can be controlled to a notable extent spectrally selective. This renders photonic nanostructures ideal candidates to mediate and to enhance nonlinear optical effects and to enhance the interaction of light with quantum emitters at the nanoscale. Here, we report in a comprehensive manner on our contributions to the field of optical nanoantennas, both dielectric and metallic, their tunability, and their ability to affect nonlinear processes. We are interested in LiNbO<sub>3</sub> as the nonlinear material. Nonlinear processes in such material tend to be enhanced when considering two distinguished scenarios. First, nanoantennas themselves can be combined in a suitable manner with LiNbO<sub>3</sub>. Second, the resonant photonic nanostructure itself is made from LiNbO<sub>3</sub>. When considering nanoantennas as additional elements to enhance the nonlinear processes, we studied on the one hand metallic disc antennas. Their resonant response is reinforced when surrounded by annular metallic gratings and light is much better localized and enhanced. On the other hand, metallic nanowires with a suitably chosen termination have been considered. Using such material platform allows to tune multiple resonances independent to each other, being a prerequisite to drive parametric nonlinear processes highly efficient. When considering resonant photonic nanostructures made from LiNbO<sub>3</sub>, we studied isolated nanoparticles or nanowires but also photonic crystal based architectures. The work is done both experimentally and theoretically/numerically. The methodology developed in our work will be presented at the poster. The methodology had also been used to study problems in the context of ultrafast nanooptics in close collaboration with other partners from the SPP. An overview across these collaborations will be presented as well.



## **Poster 18:**

### **Nanotip surface plasmons: real-space imaging using EELS and grating-coupled nonlinear photoemission**

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In this contribution, we present our latest results concerning the excitation of surface plasmon polaritons (SPPs) on metal nanotips. First, SPPs on a straight conical gold taper are imaged and spectrally analyzed by electron energy loss spectroscopy within a scanning transmission electron microscope. We observe standing wave patterns which are ascribed to reflections of the SPPs at the apex of the taper. The experimental findings are in a very good agreement with numerical computations and analytical calculations. In a second experiment, SPPs are excited with a resonant grating coupler on the shaft of a gold tip and accompanied photoemission is measured and analyzed. In our setup, we use a modified field-emitter gun assembly to control the electrostatic environment around nanotip and demonstrate that grating excitation remotely triggers multiphoton photoemission from the nanometer size apex.

**Poster 19:**

**The optimal antenna for nonlinear spectroscopy of weakly and strongly scattering nanoobjects**

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Optical nanoantennas, i.e., arrangements of plasmonic nanostructures, promise to enhance the light-matter interaction on the nanoscale. Especially nonlinear optical spectroscopy of single nanoobjects would profit from such an antenna, as nonlinear optical effects are already weak for bulk material, and become almost undetectable for single nanoobjects. We investigate the design of optical nanoantennas for transient absorption spectroscopy in two different cases: the mechanical breathing mode of a metal nanodisc and the quantum-confined carrier dynamics in a single CdSe nanowire. In the latter case, an antenna with a resonance at the desired wavelength optimally increases the light intensity at the nanoobject. In the first case, the perturbation of the antenna by the investigated nanosystem can not be neglected and off-resonant antennas become most efficient.

## **Poster 20:**

### **Light localization in disordered nanostructures - giant fluctuations of second harmonic and electron emission**

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Localization of light as an ubiquitous phenomena occurs in all kinds of disordered materials including randomly arranged arrays of dielectric or metallic scatterers. However, a direct comparison of the localization in dielectric and metallic samples for the same dimensions of the scattering particles is lacking. Here, we measure the localization of near-infrared light and of localized surface plasmons in randomly arranged dielectric and metallic nanoneedles of the same length, diameter and density. Coherent, ultra-broadband Second Harmonic microscopy and ultrashort, multi-photon photoemission electron microscopy are used to probe the spatial and temporal mode distribution in both ZnO and Au-coated ZnO needle arrays. Intensity fluctuations of the SH for the dielectric and of the multi-photon-induced electron emission for the metal-covered needles are taken as an indicator for the localization of electric and plasmonic fields within the arrays. Additionally, by using interferometric autocorrelation techniques, life times of several femtoseconds of the photon and the plasmon mode are measured, therefore exhibiting the central characteristics for weak localization of light in an array of randomly distributed dielectric and metallic scatterers.

## **Poster 21:**

### **Suppression of radiative damping and enhancement of second harmonic generation in bull's eye nanoantennas**

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We report a drastic enhancement of the damping time of surface plasmon polaritons in bull's eye nanostructures to more than 35 fs. This is achieved by tailoring the groove depth and by enlarging the coherence length of the confined plasmon field to more than 10  $\mu\text{m}$ . Experimentally, this is demonstrated by probing the plasmon dynamics at the field level using broadband spectral interferometry. The experimental results are supported by numerical simulations performed using finite element method. We demonstrate that the nanoantennas allow us to efficiently confine the incident field inside the central aperture of the bull's eye structure and to tailor its local optical nonlinearity by varying the aperture geometry. In particular we achieve a 50-time enhancement in second harmonic generation efficiency by replacing the central circular hole with an annular ring structure. This allows us to demonstrate the efficient localization of long-lived SPP fields inside nanoapertures by interferometric frequency-resolved autocorrelation (IFRAC). Such a light localization in a nanoresonator with high quality factor has high potential for sensing and coherent control of light-matter interactions on the nanoscale.

## **Poster 22:**

### **Applications of nanoplasmonics: Full quantum statistics of many emitter spasers and spin dependent reconstruction scheme of exciton wave functions**

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The spatial confinement and control of electromagnetic excitations on a subwavelength scale plays a key role in nanooptics and quantum plasmonics. Optically active (hybrid) systems composed of several coupled constituents are promising candidates for optoelectronic applications. Nanoplasmonic emitters allow for an electric field localization below the diffraction limit. For instance, there is a need for externally pumped nanoscale sources of different plasmon statistics. The considered composite system consists of a metal nanoparticle driven by a gain medium formed of externally pumped quantum emitters, e.g., quantum dots or dye molecules. We study the spaser using a non-perturbative, numerically exact solution of the full quantum statistics of the multi quantum emitter - cavity laser problem. This way, the statistical properties of plasmon generation and exciton distribution can be accessed and different operating regimes are characterized for varying numbers of quantum emitters. As a possible application of localized nanooptical fields, we propose a quantum state tomography protocol for reconstructing the hybrid wave functions of coupled quantum emitters, now extended to the case including different spin states of the quantum emitters. To achieve this, a four-wave mixing technique is combined with field localization supplied by nanoplasmonic structures together with pulse-shaped fields. Therefore, one pulse of the pulse sequence applied to the hybrid system is localized at a specific quantum emitter and its polarization is controlled. We study a system composed of two coupled semiconductor quantum dots which form new, delocalized exciton states due to Förster resonance energy transfer between the two quantum emitters. Applying the suggested reconstruction scheme to coherent two-dimensional spectra allows to investigate the internal structure of the interacting single-exciton and two-exciton states and to explore their spin dependent properties.

## Poster 23:

### Long-lived coherence in the metal-organic-hybrid Cobalt/Alq<sub>3</sub>

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In order to exploit the great potential of hybrid metalorganic interfaces for future applications, an understanding of where and how charge transport takes place is crucial. Here we investigate the coherent electron dynamics in optically pumped (400 nm) molecular states of the metalorganic complex tris(8-hydroxyquinolinato)aluminium (Alq<sub>3</sub>) [1] deposited on a Cobalt surface by means of time-resolved photoemission spectroscopy. We observe a coherence signal in interferometric autocorrelation traces when probing the excited-state manifold of Alq<sub>3</sub> with 800nm laser pulses. Desorption of the molecules down to one monolayer is achieved by heating of the sample, determining that the observed molecular feature originates from the first monolayer of Alq<sub>3</sub>. In two-dimensional nanoscopy spectra [2], two excited electronic states are identified with an energy spacing of about 77 meV. Their linewidth is 11 meV and 48 meV, respectively, corresponding to coherence lifetimes of about 370 fs and 87 fs. Measuring the kinetic energy of the photoelectrons, these features exist over the entire accessible energy range of about one eV. We explain the observed features by long-lived coherent excited states of the adsorbate that decay among others via coupling to excited electrons in the substrates giving rise to a photoemission signal that depends linearly on the 800 nm laser intensity. The appearance of such narrow spectral features indicates that electronic excitations in an individual adsorbate state can be surprisingly long-lived and thus can play an important role in determining charge transfer efficiencies at the metal-hybrid interface.

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## **Poster 24:**

### **Controlling the excitation of localized plasmonic resonances by strong near-field coupling**

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A direct control of the light-matter interaction and therefore the modification of the light propagation is a major challenge in optical research. In particular the weak interaction of light with active materials requires propagation distances of at least several wavelengths. Plasmonic nanostructures on the other side have shown the potential to strongly interact with light and generate high optical density of states. Here, we demonstrate how a coherent excitation of orthogonal eigenmodes in a plasmonic meta-atom can be controlled by a strong near-field coupling effect. The excitation of strongly coupled plasmonic modes allows therefore to utilize noninterfering far-field excitation of different polarization at the same time. We will show how the excitation of normal modes in the coupled system can be used to obtain a coherent control of the localized plasmon modes without having the problem with the short life time of the plasmonic system or the interference of the excitation beam. For our demonstration we utilize the strong nonlinearities in plasmonic system to detect the excitation strength of the localized plasmons by the emission of the third-harmonic signal in a particular polarization state. By introducing a phase shift into two orthogonal excitations beams we are able to control the excitation which build up by the coupling of different nanorod within the unit-cell of the material. Our results give a more complete understanding of the linear and nonlinear optical properties of collective excitations in strongly coupled metallic meta-atoms and metamaterials and demonstrate the potential of utilizing the near-field coupling for coherent processes.

## **Poster 25:**

### **Plasmon and exciton decay in nanostructures excited with ultrashort pulses formed by cylindrical vector laser beams**

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Noble metal nanostructures, such as gold or silver nano spheres and nano rods, exhibit strong plasmon resonances in the visible spectral range, which influence significantly their photoluminescence (PL) emission properties. While scattering of coupled plasmonic systems has been in the focus during the last years, PL emission of nanostructures excited by ultrashort laser pulses depends also on the band-structure of these materials. Here we present our recent studies aiming at a deeper insight into the interplay between plasmons and excitons of gold nanostructures. In particular, we have investigated the linear and non-linear PL emission from various plasmonic systems like nanoparticles, sharp tips and nanotriangles [1-4] excited with radially or azimuthally polarized 100 fs laser pulses in combination with either diffraction limited confocal optical microscopy or tip enhanced scanning near-field optical microscopy. Furthermore, we are extending our studies to other nanoscale systems, such as the exciton decay in semiconductor nanowires excited by ultrashort focused cylindrical vector ultrashort laser pulses.

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