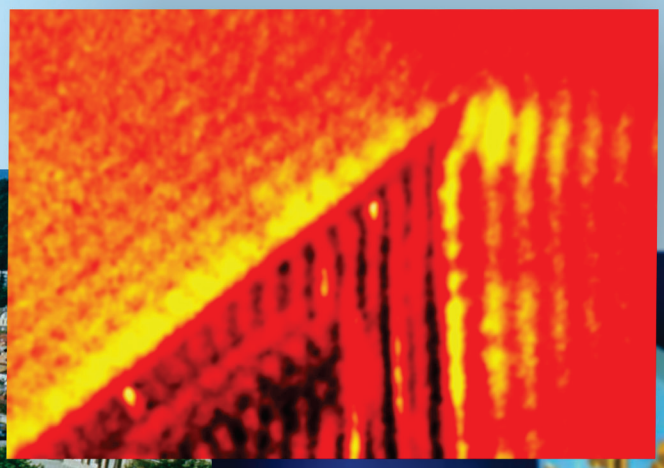
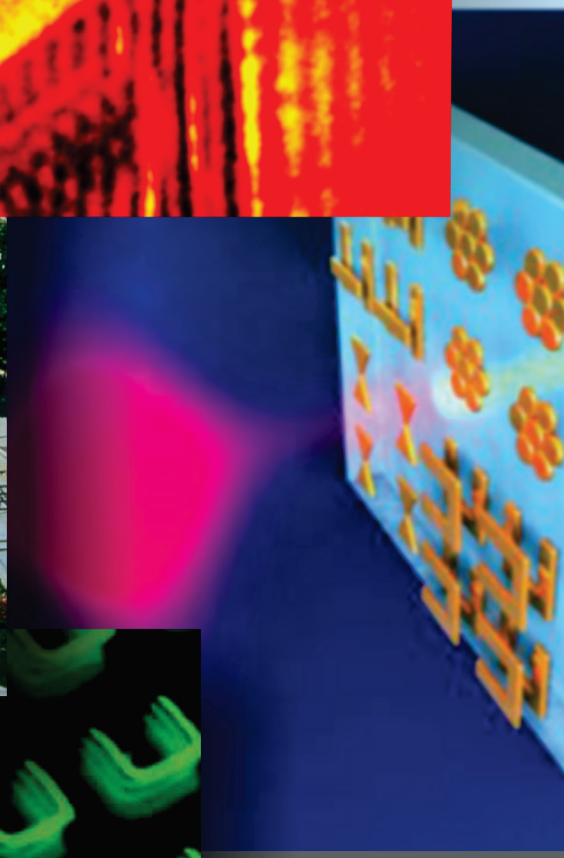
UNO-3

3rd International Workshop on Ultrafast Nanooptics

Bad Dürkheim, June 16-19, 2013

Book of Abstracts





SPP 1391

Scope and Aim

Ultrafast nanooptics, i.e. the combination of nanooptics and ultrafast laser spectroscopy, is a new and rapidly evolving field in physics and neighboring disciplines.

From June, 16-19, 2013, the "3rd International Workshop on Ultrafast Nanooptics (UNO-3)" will be held in Bad Dürkheim. This workshop continues the series started with an "Else and Wilhelm Heraeus Seminar" in 2008 in Bad Honnef/Germany and the "UNO-2" held also in Bad Dürkheim/Germany in 2010. The advances in the rapidly evolving field of ultrafast nanooptics will be presented and discussed. In addition the workshop serves as the annual meeting of the priority program members.

Walter Pfeiffer and Martin Aeschlimann

Workshop Program

Sunday, June 16, 2013

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

Monday, June 17, 2013

07:45	*BREAKFAST*
08:50	**Opening remarks**

Session 1

9:00	*F. Javier Garcia de Abajo - invited* *(ICFO Barcelona, Spain)* **Plasmons in graphene and graphene-related molecules**
9:40	*Richard Ciesielski, Matthias Handloser, Torben Winzer, Ermin Malic, Achim Hartschuh (LMU, München, TU Berlin, Germany)* **Controlling the charge carrier relaxation and photoluminescence in graphene using chirped laser pulses**
10:00	*Marten Richter, Felix Schlosser, T. Sverre Theuerholz, Alexander Carmele, Shaul Mukamel, Andreas Knorr* *(TU Berlin, Germany & University of California, USA)* **Theory of spatially resolved spectroscopy of coupled emitters and their quantum statistics**
10:20	*COFFEE BREAK*

Session 2

11:00	*Markus Raschke - invited* *(JILA, Boulder, USA)* **Ultrafast antenna coupled infrared vibrational dynamics and control**

Monday, June 17, 2013

11:40 *Nicolò Accanto, Jana Nieder, Lukasz Piatkowski, Marta Castro-Lopez, Francesco Pastorelli, Daan Brinks, Niek F. van Hulst, Niek F. van Hulst (ICFO & ICREA, Barcelona, Spain)*
Coherent control of the second harmonic emitted by a single nano-particle via femtosecond pulse shaping

12:00 *Monika Pawłowska, Christian Rewitz, Sebastian Goetz, Peter Geisler, Gary Razinskas, Enno Krauss, Bert Hecht, Tobias Brixner (Universität Würzburg, Germany)*
Coherent control of plasmon propagation in top-down fabricated optical nano-circuits

12:30 *LUNCH*

Session 3

14:00 *Hrvoje Petek – invited (University of Pittsburgh, USA)*
Ultrafast coherent multi-photon photoemission spectroscopy and microscopy

14:40 *Philip Kahl, Simon Sindermann, Christian Schneider, Alexander Fischer, Martin Aeschlimann, Frank Meyer zu Heringdorf (Universität Duisburg-Essen, TU Kaiserslautern, Germany)*
Time-resolved wave packet observation of surface plasmon polaritons provided by normal incidence photoemission electron microscopy

15:00 *Christoph Lemke, Till Leißner, Alwin Klick, Jörn Willers Radke, Jacek Fiutowski, Jakob Kjelstrup-Hansen, Horst-Günther Rubahn, Michael Bauer (Universität Kiel, Germany & University of Southern Denmark)*
Real-time imaging of the interaction of a SPP wave-packet with a localized receiver

15:20 *COFFEE BREAK*

16:00 *Martin Esmann, Simon F. Becker, Bernard B. da Cunha, Jens H. Brauer, Ralf Vogelgesang, Petra Groß, Christoph Lienau (Universität Oldenburg, Germany)*
Radiation pattern imaging of a sharp gold taper for scanning near-field optical microscopy

Monday, June 17, 2013

16:20 *Cristian González, Daniela Bayer, Elena A. Ilin, Egbert Oesterschulze, Martin Aeschlimann (TU Kaiserslautern)*
Sphere based cantilevers for SNOM

16:40 **Poster Session**

19:00 *DINNER*

21:00 **Informal Discussion**

Tuesday, June 18, 2013

07:45 *BREAKFAST*

Session 4

9:00 *Mark I. Stockman - invited*
(Georgia State University, Atlanta, USA)
Solids in ultrafast and strong optical fields: new phenomena

9:40 *K. Lindfors, M. Pfeiffer, B. Fenk, F. Phillipp, P. Atkinson, A. Rastelli, O. G. Schmidt, H. Giessen, M. Lippitz (MPI-FKF, Stuttgart, SCoPE University of Stuttgart, MI-IS Stuttgart, IFW Dresden, Germany)*
11 nm alignment accuracy in positioning plasmonic nanostructures on self-assembled GaAs quantum dots

10:00 *Steffen Michaelis de Vasconcellos, Olivier Gazzano, Clementine Symonds, Joël Bellessa, Aristide Lemaître, Pascale Senellart, Torsten Stiehm, Christian Heidrich, Rudolf Bratschitsch*
(Universität Münster, Germany, LPN-CNRS, Marcoussis, France, LPMCN, Université Lyon, TU Chemnitz, Germany)
Confined Tamm plasmon modes

10:20 *COFFEE BREAK*

Session 5

11:00 *Xiaoyang Zhu - invited*
(Columbia University, New York, USA)
Exciton fission, quantum coherence, and solar energy conversion beyond the limit

Tuesday, June 18, 2013

11:40 *David Leipold, Martin Silies, Manfred Mascheck, Christoph Lienau, Erich Runge (TU Ilmenau, Germany)*
Ultrafast dynamics of localized light modes

12:00 *M. Aeschlimann, M. Birlo, T. Brixner, D. Differt, M. Hensen, C. Kramer, F. Lükermann, P. Melchior, M. Piecuch, W. Pfeiffer, H. Stiebig, C. Strüber, P. Thielen (Universität Bielefeld, TU Kaiserslautern, Universität Würzburg, Germany)*
Localization of photonic modes in thin film solar cells

12:30 *LUNCH*

Session 6

14:00 *Dmitry A.Yarotski, Mark Hagmann, Min Ah Seo, Rohit P. Prasankumar, Antoinette Taylor – invited (Los Alamos Nat. Lab., USA)*
Ultrafast characterization and imaging: from the nanoscale to the mesoscale

14:40 *C. Ruppert, F. Förster, A. Zrenner, J. B. Kinzel, A. Wixforth, H. J. Krenner, M. Betz (TU Dortmund, Universität Paderborn, Universität Augsburg & CeNS LMU München, Germany)*
Active plasmonics with surface acoustic waves

15:00 *Thorsten Schumacher, Daniela Ullrich, Mario Hentschel, Harald Giessen, Markus Lippitz (MPI-FKF, Stuttgart, Universität Stuttgart, Universität Bayreuth, Germany)*
Optical antennas for ultrafast spectroscopy of single CdSe nanoobjects

15:20 *COFFEE BREAK*

16:00 *Pascal Engelke, Slawa Schmidt, Petra Groß, Christoph Lienau (Universität Oldenburg, Germany)*
Adiabatic nanofocusing of few-cycle laser pulses using a deformable mirror for wave-front adaptation

Tuesday, June 18, 2013

16:20 *Jing Qi, Thomas Kaiser, Thomas Pertsch, Carsten Rockstuhl (Universität Jena, IAP & ACP, Jena, Germany)*
Highly resonant and directional optical nanoantennas

16:40 *Stefan Linden (Universität Bonn)*
Ultrafast nonlinear optical response of metallic nanostructures: Collective effects and hybrid materials

17:00 *Lara Wimmer, Georg Herink, Katharina Echternkamp, Daniel Solli, Sergey Yalunin, and Claus Ropers (Universität Göttingen, Germany)*
Controlling the Photoemission from Nanostructures with Ultrashort THz-Pulses

19:00 *DINNER*

21:00 **Informal Discussion**

Wednesday, June 19, 2013

07:45 *BREAKFAST*

Session 7

9:00 *Ilya Akimov – invited*
(TU Dortmund, Germany)
Active control of plasmon-polaritons in periodic metal structures

9:40 *Bernd Metzger, Mario Hentschel, Thorsten Schumacher,Gelon Albrecht, Markus Lippitz, Harald Giessen (SCoPE University Stuttgart, MPI-FKF, Stuttgart, Germany)*
Nonlinear optical spectroscopy of complex plasmonic nanoantennas

10:00 *Shakeeb Bin Hasan, Christoph Etrich, Robert Filter, Carsten Rockstuhl, Falk Lederer*
(ACP Jena, Germany)
Enhancing the second harmonic generation of plasmonic nanowire antennas by tailoring their terminations

10:20 *COFFEE BREAK*

Wednesday, June 19, 2013

Session 8

11:00 *S.H. Chew, A. Gliserin, F. Schertz, S. Nobis, P. Geissler, M. Kübel, J. Schmidt, K. Pearce, B. Hecht, P. Hommelhoff, H.-J. Elmers, M.F. Kling, U. Kleineberg*
(LMU, München, Germany, MPQ-MPG, Universität Würzburg, Germany)
Toward coherent control of nanoplasmonic fields measured by carrier-envelop phase-tagged photoelectron emission microscopy

11:20 *Lennart Seiffert, Sergey Zherebtsov, Frederik Süßmann, Mathias Arbeiter, Johannes Stierle, Jürgen Plenge, Valerie Mondes, Philipp Rupp, Christina Graf, Eckart Rühl, Matthias Kling, Thomas Fennel*
(Universität Rostock, MPI-QO, Garching, FU Berlinf, Germany)
Dielectric nanospheres under strong few-cycle laser pulses: probing near-fields via phase-controlled electron recollision

11:40 *Matthias Hensen, Detlef Diesing, Dominik Differt, Ingo Heesemann, Christian Strüber, Adelheid Godt, Walter Pfeiffer*
(Universität Bielefeld, Universität Duisburg-Essen, Germany)
Single nanoparticle assisted few-cycle laser pulse induced local multiphoton internal photoemission in metal-insulator-metal junctions

12:00 *Jan Vogelsang, Björn Piglosiewicz, Slawa Schmidt, Doojae Park, Petra Groß, Christoph Lienau*
(Universität Oldenbug, Germany)
Electron motion in few-cycle laser fields around metallic nanostructures

12:20 **Discussion and Closing Remarks**

12:30 *LUNCH*

END OF SEMINAR / DEPARTURE

Abstracts of oral Presentations

(in chronological order)

Plasmons in graphene and graphene-related molecules

F. Javier Garcia de Abajo

ICFO – Institut de Ciéncies Fotóniques, Mediterranean Technology Park, 08860 Castelldefels (Barcelona), Spain

We will review recent results on the study of collective electron excitations (plasmons) in systems of low dimensionality, including small metallic nanoparticles, graphene, and simple molecules. In particular, we will show that plasmons in graphene can be used to achieve electrical modulation of light in a robust, solid state environment. Plasmons in polycyclic aromatic hydrocarbons, which can be regarded as small versions of graphene, will be also shown to exhibit remarkable tunability and strong plasmonic response, thus revealing their interest in the new field of molecular plasmonics. Quantum effects in these systems, ranging from nonlocality to the discreteness of the electronic transitions involved, will be shown to lead to exciting new physics and a wealth of potential applications

Controlling the charge carrier relaxation and photoluminescence in graphene using chirped laser pulses

Richard Ciesielski[1], Matthias Handloser[1], Torben Winzer[2], Ermin Malic[2], Achim Hartschuh[1]

[1]*Department Chemie und CeNS, Ludwig Maximilians Universität München, Butenandtstr. 5-13, 81377 Munich, Germany*

[2]*Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Technische Universität Berlin, Hardenbergstr.36, 10623 Berlin, Germany*

Charge carrier dynamics in graphene happen on femtosecond timescales. Experimentally, we show how the non-linear photoluminescence (NL-PL) depends on the chirp of a broadband laser pulse [1] (60nm, centred at 800nm) by employing a pulse shaping setup that enabled us to freely change the spectral phase. We use femtosecond laser sources, a spatial light modulator, and a standard confocal microscope with an objective of high numerical aperture (NA=1.4). Pulse compression to the (time domain) Fourier limit in the focus is achieved by grating compressors and an iterative phase optimization process (MIIPS [2]). With this setup, we control the NL-PL intensity and polarization of graphene in agreement with theoretical modelling [3].

[1] M. Handloser, T. Winzer, G. Piredda, N. Hartmann, A. Lombardo, A. Guggenmos, R. Ciesielski, A. Comin, U. Kleineberg, A.C. Ferrari, E. Malic, and A. Hartschuh1,2013, Controlling the charge carrier relaxation and photoluminescence in graphene using chirped laser pulses (to be submitted).

[2] Lozovoy, V.V., Pastirk, I., Dantus, M., 2004. Multiphoton intrapulse interference.IV. Ultrashort laserpulse spectral phase characterization and compensation. Opt. Lett. 29, 775–777.

[3] Malic, E., Winzer, T., Bobkin, E., Knorr, A., 2011. Microscopic theory of absorption and ultrafast many-particle kinetics in graphene. Phys. Rev. B 84, 205406.

Theory of spatially resolved spectroscopy of coupled emitters and their quantum statistics

Marten Richter[1], Felix Schlosser[1], T. Sverre Theuerholz[1], Alexander Carmele[1], Shaul Mukamel[2], Andreas Knorr[1]

[1]*Technische Universität Berlin, Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Hardenbergstr. 36 EW 7-1, 10623 Berlin, Germany*

[2]*Department of Chemistry, University of California, Irvine, CA 92697-2025, USA*

Field localization by metal nanoparticles and the use of spatio-temporal shaped pulses allows the selective excitation of individual nanstructures. In this contribution we combine the spatial control of the light field with coherent spectrosopy of coupled quantum dots close to metal nanostructures. It is shown that the information about the delocalization of the exciton wavefunction over a coupled QD-system can be obtained from localized coherent spectroscopy. Beyond using metal nanoparticles for controlling the optical field on the nanoscale, hybrid nanostructures can be constructed in a way that the metal plasmon excitations are close to resonance with the QD excitons. The coupling between the plasmons of the metal nanoparticle and the excitons of the quantum dots forms new hybridized plasmon-polariton states. Based on these new quantum states we analyse the quantum statistics of the plasmon/photon system using the second order photon correlation function. We show that under cw-excitation the Förster interaction between the quantum dots has a great impact on the quantum statistics of the plasmon-photon system.

Ultrafast antenna coupled infrared vibrational dynamics and control

Markus B. Raschke

Department of Physics, Department of Chemistry, and JILA, University of Colorado, Boulder, CO 80303, USA

The extension of ultrafast spectroscopy to the nanoscale to access the underlying spatial inhomogeneities of far-field ensemble averaging spectroscopy has been a long-standing challenge [1,2]. Its application to infrared vibrational molecular spectroscopy is particularly underdeveloped yet highly desirable for its chemical sensitivity and access to chemical dynamics. We combine scattering scanning near-field optical microscopy (*s*-SNOM) with femtosecond infrared spectroscopy for ultrafast molecular vibrational nano-spectroscopy in the mid-infrared. The metallic scanning probe tip provides a nanoscale field localization and near-field coupling via evanescent modes with the induced coherent vibrational polarization. Via its enhanced coupling to the far-field electromagnetic density of states this gives rise to tip scattered *radiative* emission of the vibrational molecular free-induction decay (FID) in competition against the rapid intra- and inter-molecular nonradiative decay in the condensed phase [3]. With increasing degree of near-field coupling between molecular excitation and metallic antenna tip we can control the rate of mode transfer and thus controllably *enhance* the vibrational relaxation. We demonstrate a drastic tip-enhanced increase in *sensitivity* of ultrafast IR spectroscopy by $>10^4$ probing ensembles as low as 100 vibrational oscillators reaching the realm of single molecule IR spectroscopy [4].

We take advantage of the optical antenna properties and field localization of the metallic scanning probe tip. The antenna mediates the mode mismatch between the near-field molecular coherent polarization and far-field density of states succeeding in the competition against the otherwise dominant non-radiative decoherence. I will discuss the generalization of this approach to any form of linear and nonlinear wavemixing techniques including multi-dimensional spectroscopy which will allow for vibrational quantum coherent control and full spatio-temporal imaging of vibrational dynamics on the nanoscale [5].

[1] A. Kubo, N. Pontius, and H. Petek, Nano Letters **7**, 470 (2007).

[2] M. Aeschlimann, M. Bauer, D. Bayer, T. Brixner, F. J. García de Abajo, W. Pfeiffer, M. Rohmer, C. Spindler, and F. Steeb, Nature, **446**, 301 (2007).

[3] X. G. Xu and M. B. Raschke, Nano Lett. **13**, 1588 (2013).

[4] X. G. Xu, M. Rang, I. M. Craig, and M. B. Raschke, J. Phys. Chem. Lett. **3**, 1836 (2012).

[5] R. L. Olmon and M. B. Raschke, Nanotechnology **23**, 444001 (2012).

Coherent control of the second harmonic emitted by a single nanoparticle via femtosecond pulse shaping

Nicolò Accanto[1], Jana Nieder[1], Lukasz Piatkowski[1], Marta Castro-Lopez[1], Francesco Pastorelli[1], Daan Brinks[1], Niek F. van Hulst[1], Niek F. van Hulst[2]

[1]ICFO Institut de Ciencies Fotoniques, Mediterranean Technology Park, 08860 Castelldefels (Barcelona), Spain

[2]ICREA Institució Catalana de Recerca i Estudis Avançats, 08015 Barcelona, Spain

Ultrafast pulse shaping in individual nano-particles (NPs) requires full femtosecond pulse control on a nanometer spatial scale. In a typical pulse shaping experiment on the nano-scale one monitors the variation in the optical response of one NP induced by a change in the phase-time properties of the excitation laser pulse. Finely tailor the pulse in its phase-time domain with femtosecond accuracy and on a nanometer spatial scale is therefore a crucial part of such experiments. The second harmonic (SH) emitted by a single NP, being a coherent phenomenon, is sensitive to the spectral phase (i.e. time profile) of a laser pulse, which makes these NPs very good test-systems for femtosecond/nanometer pulse shaping experiments given they small (100nm) dimensions. In this work we investigated the coherent SH response of single NPs (e.g. BaTiO3, Fe(IO3)3, Au) subject to a femtosecond laser pulse whose spectral phase could be controlled by means of a pulse shaper. First, using the SH generated by a single 150nm NP we were able to compress a distorted laser pulse in the time domain down to its Fourier limit (17fs) performing multiphoton intrapulse interference phase scans (MIIPS). In a second experiment we showed that, applying a known spectral phase onto the laser pulse, it is possible to deterministically control the SH spectrum from individual NPs as small as 100nm. These two experiments together demonstrate full femtosecond pulse control on a nanometer spatial scale, which is a fundamental requirement for pulse shaping experiments and coherent control on individual NPs.

Coherent control of plasmon propagation in top-down fabricated optical nano-circuits

Monika Pawłowska[1], Christian Rewitz[1], Sebastian Goetz[1], Peter Geisler[2], Gary Razinskas[2], Enno Krauss[2], Bert Hecht[3], Tobias Brixner[1]

[1]*Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany*

[2]*Nano-Optics and Biophotonics Group, Experimentelle Physik 5, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany*

[3]*Röntgen Center for Complex Material Systems (RCCM), Am Hubland, 97074 Würzburg, Germany*

Numerical simulations and an analytic approach based on transmission line theory are used to design splitters for nano-plasmonic signal processing that allow to arbitrarily adjust the ratio of transmission from an input into two different output arms. By adjusting the geometrical parameters of the structure, either a high bandwidth or a sharp transmission resonance is obtained. Switching between the two arms can be achieved by modulating the effective refractive index of the waveguide. Employing the instantaneous Kerr effect, switching rates in the THz regime are potentially feasible. The suggested devices are of interest for future applications in nanoplasmonic information processing.

Ultrafast Coherent Multi-Photon Photoemission Spectroscopy and Microscopy

Hrvoje Petek

University of Pittsburgh, Pittsburgh, PA 12560 USA

We study surface plasmon polariton (SPP) generation, propagation, diffraction, interference, focusing, and decay by femtosecond time-resolved photoemission electron microscopy (PEEM) and electromagnetic simulations. Equal pump-probe pulses with interferometrically defined delay excite two-photon photoemission from Ag surfaces. The imaging of the spatial distribution of photoemitted electrons by PEEM reveals a nonlinear map of the total surface electromagnetic fields impressed on the sample. On a nanostructured surface the images reveal coherent polarization gratings formed by superposition of the incoming excitation pulses and propagating SPP wave packets that are generated at nanofabricated coupling structures. By changing the delay between the pump and probe plusses in ~330 as steps we record movies of the evolving coherent polarization at the Ag/interface, which reflect the evolution of the surface electromagnetic fields. We report the latest experimental and simulation studies on representative plasmonic coupling structures.

Time-resolved wave packet observation of surface plasmon polaritons provided by normal incidence photoemission electron microscopy

Philip Kahl[1], Simon Sindermann[1], Christian Schneider[2], Alexander Fischer[2], Martin Aeschlimann[2], Frank Meyer zu Heringdorf[1]

[1]*Faculty of Physics and Center for Nanointegration Duisburg-Essen (CeNIDE), Universität Duisburg-Essen, 47057 Duisburg*

[2]*Department of Physics and Research Center OPTIMAS, University of Kaiserslautern, 67663 Kaiserslautern*

In order to observe surface plasmon polaritons (SPPs) in a photoemission electron microscopy (PEEM) experiment, ultrashort (15 fs) laserpulses of 800nm wavelength are directed onto a surface with plasmonic Ag or Au nanostructures. In the past, non-linear photoemission under grazing incidence was used to obtain a Moiré type contrast of propagating SPPs waves. If the light impinges perpendicular onto the surface (normal incidence) a direct imaging of the plasmon is possible, as the observed fringes resemble the SPP wavelength. In time-resolved experiments under normal incidence conditions, the direct observation of isolated SPP wave packets is expected.

Real-time imaging of the interaction of a SPP wave-packet with a localized receiver

Christoph Lemke[1], Till Leißner[1], Alwin Klick[1], Jörn Willers Radke[1], Jacek Fiutowski[2], Jakob Kjelstrup-Hansen[2], Horst-Günther Rubahn[2], Michael Bauer[1]

[1]*Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, Kiel, Germany*

[2]*Mads Clausen Institute, NanoSYD, University of Southern Denmark, Sønderborg, Denmark*

Using interferometric time-resolved photoemission electron microscopy (ITR-PEEM) operated in a counter-propagation detection mode [1] we studied in real-time the interaction of an ultrashort SPP wave packet with a gold nanodisk acting as localized plasmonic receiver. SPP wave packets are excited via 12 fs laser pulses at an excitation ridge located at a distance of 10 µm from the receiver. At impact of the SPP a clear and transient enhancement of the photoemission signal from the receiver is observed accompanied by a dipolar emission characteristic. Complementary data recorded under direct laser-excitation of the nanodisk in combination with Huygens-based simulations [2] enable us to interpret the response of the receiver in terms of a predominant coupling of the SPP to a dipolar mode of the nanodisk. The instantaneous response of the receiver to the plasmonic excitation hints to a strong damping of the dipolar mode which most likely arises from radiation damping and an efficient coupling to the substrate.

[1] C. Lemke, T. Leißner, S. Jauernik, A. Klick, J. Fiutowski, J. Kjelstrup-Hansen, H.-G. Rubahn, and M. Bauer Opt. Express., 20,12877, (2012).

[2] C. Lemke, C. Schneider, T. Leißner, D. Bayer, J. W. Radke, A. Fischer, P. Melchior, A. B. Evlyukhin, B. N. Chichkov, C. Reinhardt, M. Bauer, and M. Aeschlimann, Nano Letters 13, 1053 (2013).

Radiation Pattern Imaging of a Sharp Gold Taper for Scanning Near-Field Optical Microscopy

Martin Esmann, Simon F. Becker, Bernard B. da Cunha, Jens H. Brauer, Ralf Vogelgesang, Petra Groß, Christoph Lienau

Institut für Physik, Carl von Ossietzky Universität, Carl-von-Ossietzky Str. 9-11, 26129 Oldenburg

Adiabatic nanofocusing of surface plasmon polaritons (SPP) on tapered metallic waveguides bears great potential as a novel method for apertureless scanning near-field optical microscopy [1,2]. Plasmon polariton wavepackets are launched on a grating-coupler and ideally come to a complete halt at the taper apex where a single point-dipole like light source is formed. We have incorporated such tapers obtained from electrochemical etching followed by focused ion beam milling of the grating coupler in a scanning probe setup [2]. An obstacle still present in our current setup is the fact that not all tapers, although they are obtained in the same process, yield the same amount of field enhancement at the apex. We believe that one reason is the fact that a pure dipole at the apex of the taper is only formed by the lowest rotationally symmetric eigenmode of the taper. Higher modes that are also excited on the coupler may disturb the imaging process as they radiate into the far field before reaching the taper end. Thus, they only contribute to background signals. We have therefore developed and implemented a k-space imaging technique to analyze the different eigenmodes emitted from tapered metallic nanowaveguides [3]. The analysis of k-space patterns suggests that only those SPP fields that are coupled to the lowest (rotationally symmetric) and first higher eigenmode (dipole symmetry) of the taper propagate as bound modes into close proximity of the taper apex. The decay length of the near-field contribution of the lowest eigenmode is on the order of 14 nm. Our approach allows us to use the spatial symmetry of the re-radiated light as an indicator for the tip-sample coupling. By applying k-space filtering near-fields can be extracted easily. This represents an important step forward towards coherent spectroscopy with ultrahigh spatial resolution.

[1] M. I. Stockman, PRL 93, 137404 (2004)

[2] S. Schmidt et al., ACS Nano 6, 6040 (2012)

[3] M. Esmann et al., arXiv:1304.6060

Sphere based cantilevers for SNOM

Cristian González, Daniela Bayer, Elena A. Ilin, Egbert Oesterschulze, Martin Aeschlimann

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We present the development and first measurements with a new kind of cantilever sensors, which allow an enhanced transmission of light keeping the possibility to perform SNOM (Scanning Near Field Microscope) measurements in contact to the sample. The characterization performed by measuring Au nano-rings show a clear polarization dependence effect as well as the enhancement of the transmission. These new cantilevers are a modification of the traditional aperture cantilevers for aperture based SNOM and consist, basically, of a glass micro-sphere right at the apex of the tip of the sensor.

Solids in ultrafast and strong optical fields: new phenomena

Mark I. Stockman

Ludwig Maximillian University, Munich, Germany

Max Planck Institute for Quantum Optics, Garching, Germany

Department of Physics and Astronomy, Georgia State University, Atlanta, GA 30303, USA

We report theory and experimental results for a new class of phenomena in condensed matter optics when strong optical fields ~ 1 - 3 V $Å^{-1}$ reversibly change the solid within an optical period [1-5]. Such fields, if adiabatic, cause phenomena such as the Wannier-Stark localization and anticrossings of adiabatic levels. During a single-oscillation strong optical pulse, a dielectric undergoes a reversible transition to a semimetal, which follows the instantaneous optical field during time intervals on order of hundred attoseconds. Such a pulse drives Ampere-scale currents in dielectrics and controls their properties, including optical absorption and reflection, and extreme UV absorption in a non-perturbative manner. Applied to a metal, such a pulse causes an instantaneous and reversible loss of the metallic properties. These are fastest phenomena in optics. They offer potential for petahertz-bandwidth signal processing.

1. M. Durach, A. Rusina, M. F. Kling, and M. I. Stockman, Metallization of Nanofilms in Strong Adiabatic Electric Fields, Phys. Rev. Lett. 105, 086803-1-4 (2010).
2. M. Durach, A. Rusina, M. F. Kling, and M. I. Stockman, Predicted Ultrafast Dynamic Metallization of Dielectric Nanofilms by Strong Single-Cycle Optical Fields, Phys. Rev. Lett. 107, 0866021-5 (2011).
3. A. Schiffrin, T. Paasch-Colberg, N. Karpowicz, V. Apalkov, D. Gerster, S. Muhlbrandt, M. Korbman, J. Reichert, M. Schultze, S. Holzner, J. V. Barth, R. Kienberger, R. Ernstorfer, V. S. Yakovlev, M. I. Stockman, and F. Krausz, Optical-Field-Induced Current in Dielectrics, Nature 493, 70-74 (2013).
4. M. Schultze, E. M. Bothschafter, A. Sommer, S. Holzner, W. Schweinberger, M. Fiess, M. Hofstetter, R. Kienberger, V. Apalkov, V. S. Yakovlev, M. I. Stockman, and F. Krausz, Controlling Dielectrics with the Electric Field of Light, Nature 493, 75-78 (2013).
5. V. Apalkov and M. I. Stockman, Metal Nanofilm in Strong Ultrafast Optical Fields, arXiv:1209.2245 [cond-mat.mes-hall], 1-5 (2012).

11 nm alignment accuracy in positioning plasmonic nanostructures on self-assembled GaAs quantum dots

K. Lindfors[1], M. Pfeiffer1, B. Fenk[1], F. Phillipp[3], P. Atkinson[4], A. Rastelli[4], O. G. Schmidt[4], H. Giessen[2], M. Lippitz[1]

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Embedding a single quantum emitter in an artificial nanostructure allows engineering the optical properties of the emitter. Plasmon resonant structures are particularly interesting for this purpose, as they allow confining electromagnetic fields to the nanoscale. This results in highly modified light-matter interaction. The extreme field localization requires the plasmonic structure to be positioned and oriented with respect to the quantum system with nanometer scale accuracy. Here we describe a technique which allows us to fabricate nanostructures positioned with respect to single GaAs quantum dots with an accuracy of 11 nm. The quantum emitters used in our work are self-assembled GaAs/AlGaAs quantum dots buried approximately 20 nm beneath the semiconductor surface with exciton emission at 760 nm wavelength. To position nanostructures with respect to single quantum dots we first determine the position of single emitters by imaging a characteristic surface topography feature located above the quantum dots using scanning electron microscopy. In this way we obtain the coordinates of the emitter in a coordinate system defined by a grid of markers. This allows us to use nanofabrication methods, e.g., electron beam lithography, to fabricate the desired nanostructure aligned to selected target quantum dots. As a first example of our positioning method we have fabricated gold nanorod antennas aligned to single quantum dots using electron beam lithography. By varying the aspect ratio of the nanorod antenna, the spectral position of the transverse and longitudinal plasmon resonances can be tuned. By tuning one of the resonances to coincide with the exciton transition in the quantum dot we have managed to engineer the polarization anisotropy of the emission. The developed positioning method is a promising route to realizing more advanced devices, e.g., nanoscale optical circuits making use of single photon emitters.

Confined Tamm plasmon modes

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Three dimensional spatial confinement of the optical field is crucial for many future photonic devices like nano-lasers, photodetectors and quantum light sources. We present a new type of microcavity based on the optical Tamm plasmon state, which is formed at the interface between a distributed Bragg reflector (DBR) and the metal. The Tamm plasmon can be easily confined latterly by simply shaping the metal layer. Strong three-dimensional confinement of Tamm plasmon modes is achieved by a thin gold microdisk on top of a planar GaAs/AlGaAs DBR. Discrete optical modes are evidenced both experimentally and theoretically, with quality factors up to 1200 for the fundamental mode. These modes exhibit a zero in-plane wave vector, allowing for an excellent coupling to quantum dot excitons and the vertical emission of photons. With a deterministic lithography method we couple single self-assembled InGaAs quantum dots (QD) to the confined Tamm plasmon mode. The excitonic transitions of the QD are shown to experience an acceleration of their radiative lifetime if the exciton transition is spectrally resonance with the mode, while a remarkably strong inhibition of the spontaneous emission (by a factor of up to 40) is measured if the exciton transition is off-resonance. Our study shows that this structure allows the fabrication of efficient single photon sources with a high brightness.

Exciton fission, quantum coherence, and solar energy conversion beyond the limit

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The absorption of one photon by a semiconductor material usually creates one electron-hole pair, but this general rule breaks down in a few organic semiconductors, such as pentacene and tetracene, where one photon absorption may result in two electron-hole pairs in a process called singlet fission. Recent measurements in our group by time-resolved two-photon photoemission spectroscopy on crystalline pentacene and tetracene provided the first spectroscopic signatures in singlet fission of a critical intermediate known as the multiexciton state. More importantly, population of the multiexciton state is found to rise concurrently with that of the singlet state on the ultrafast time scale upon photo excitation. This observation provides an experimental foundation for a quantum coherent mechanism in which the electronic coupling creates a quantum superposition of the singlet and the multiexciton state immediately following optical excitation. However, direct electronic coupling between singlet and multiexciton states is too weak to explain the experimental observation. Indirect coupling via charge transfer intermediate states is two orders of magnitude stronger and dominates the dynamics for ultrafast multiexciton formation. Density matrix calculation for the crystalline tetracene lattice qualitatively accounts for the experimental observations and reveals the critical roles of the charge transfer states and the high dephasing rates in ensuring the ultrafast formation of multiexciton states.

Ultrafast dynamics of localized light modes

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Localization of light by multiple coherent scattering received considerable renewed interest recently[1]. Novel ultrafast near-field experiments revealed hallmarks of localization, e.g., of plasmon waves on corrugated silver surfaces [2] and visible light in forests of randomly distributed ZnO nano-needles [3]. In both cases, the relevant modes are strongly damped by radiative and Ohmic losses. In general, loss processes oppose localization by multiple coherent scattering, simply because they reduce the number of scattering events. Here, we consider theoretically a model system with open boundaries in order to study localization in lossy systems and how the individual modes evolve in time. We find modes showing typical features of both, localized and extended character in the same energy range. The long-living modes resemble in many aspects those observed in [3].

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Localization of photonic modes in thin film solar cells

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The absorption of thin-film solar cells can be enhanced by increasing the effective light path in the absorptive layer via light trapping. We investigate amorphous thin-film Si solar cells with nanotextured internal interfaces by means of optical spectral interferometry (SI) and coherent two-dimensional (2D) nanoscopy [1] providing complementary information about localized photonic modes. SI probes the electric field of the ultrashort near-infrared laser pulses scattered in the sample. Spatially resolved SI reveals the formation of localized photonic modes in the absorptive amorphous Si layer with coherence lifetimes up to about 100fs. Coherent 2D nanocoscopy combining coherent 2D spectroscopy with photoemission electron microscopy allows investigating these photonic modes with even higher spatial resolution of about 50 nm. In our experiments we observe hot-spot electron emission confirming strongly localized electric field distributions in the absorptive Si layer. Besides the high spatial resolution coherent 2D nanoscopy offers spectroscopic information about the hot spot emission. For this the measured 2D nanospectra are fitted with a damped Lorentzian oscillator model yielding spatially-resolved information about lifetimes and spectral shifts of localized photonic modes with sub-diffraction resolution. The quantitative match of lifetime distributions extracted from SI and 2D nanoscopy indicate that the highly localized photonic modes have the same origin as the long coherence lifetimes seen in the scattered fields. We conclude that effective light trapping in amorphous thin film solar cells by nanotextured interfaces involves the generation of strongly localized photonic modes via multiple scattering in the sample.

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Ultrafast Characterization and Imaging: From the Nanoscale to the Mesoscale

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We present two ultrafast characterization techniques spanning the nanoscale to the mesoscale. First we describe our novel technique for microwave frequency-comb generation in a tunneling junction by nonlinear intermode mixing of 15fs pulses. This microwave frequency comb scanning tunneling microscope (MFC-STM) has produced 200 harmonics of the fundamental pulse frequency. Applications of the MFC-STM include nanoscale characterization of dynamic electronic and magnetic response of materials. Next, we describe our new imaging method of ultrafast wide field microscopy which enables the characterization of time-dependent carrier dynamics with high sensitivity, femtosecond time resolution and sub-micron spatial resolution over range of tens of microns. Applications to carrier dynamics in semiconductor and nanowires will be presented.

Active Plasmonics with Surface Acoustic Waves

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We analyze the impact of electrically driven surface acoustic waves (SAWs) on plasmonic devices. In a first step, we demonstrate a SAW driven converter of light into surface plasmon polaritons (SPPs). An otherwise unstructured metal thin film is deformed by travelling acoustic waves on a piezoelectric substrate underneath. This spatially periodic corrugation enables to bridge the momentum gap between free-space radiation and surface-bound modes. This concept is realized with unstructured gold films on a LiNbO3 wafer such that 500 MHz surface acoustic waves induce 1 nm surface ripples on the metal. For near-infrared light we observe a 0.01% efficiency for launching SPPs. As a next step, we explore the impact of such radio-frequency SAWs travelling across a commensurable, static gold grating, i.e., a paradigm for a pre-defined SPP injector. In essence, the electro-mechanically induced, dynamic surface deformation deliberately modulates the launcher's coupling characteristics on sub-nanosecond timescales as required for device applications. The modulation of the SPP injection efficiency is as large as 2% and is monitored in real time by a stroboscopic technique utilizing SAWs synchronized to an optical pulse train.

Optical antennas for ultrafast spectroscopy of single CdSe nanoobjects

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Ultrafast nonlinear spectroscopy investigates the deviations from linear light-matter interaction on very short timescales. The already weak nonlinear signals are reduced further when single nanoobjects such as quantum dots, molecules, or nanoparticles are the systems of interest. We present a method to resolve ultrafast carrier dynamics of excitonic states in an individual CdSe nanowire. We observe various fast and long living effects in the transient absorption spectra that we attribute to a highly excited electron-hole plasma and excitonic state bleaching. To enhance this nonlinear response we developed a method to place gold nanoantennas in close vicinity to the nanowires. Here we present first experimental results of these hybrid systems.

Adiabatic nanofocusing of few-cycle laser pulses using a deformable mirror for wave-front adaptation

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By adiabatic nanofocusing, light can be transported along conical metal tapers in the form of a surface plasmon polariton (SPP) [1]. Experimentally a highly confined light source with few nanometer spatial and few femtoseconds time resolution has been realized by grating-coupling a spectrally broad-band light source onto a gold taper [2]. Scattering and reflective losses of the SPP between grating coupling and the taper apex are reduced greatly by the use of ultra-smooth single-crystalline gold tapers, enabling propagation distances of tens of microns. The over-all coupling efficiencies that are reached with these gold tapers, however, typically remain below 1%. We have identified a wave-front mismatch between the illuminating laser pulses and the conical surface of the tip as a possible cause for the restricted coupling efficiency. Here we demonstrate shaping of the wave front to match the surface curvature of the gold taper using a deformable mirror. The wave front curvature was adapted to the tip surface using an evolutionary algorithm. We found that the coupling efficiency of the light to surface SPP wavepackets was improved considerably, resulting in an enhancement by a factor 8 of the light scattered from the tip apex. Interferometric frequency resolved autocorrelation techniques confirmed that the time structure of the 6-fs pulses was preserved. We anticipate that the demonstrated technique offers considerable improvement for scanning near field optical microscopy techniques.

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Highly resonant and directional optical nanoantennas

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The unique ability of optical nanoantennas to manipulate light at the subwavelength scale has intrigued intensive investigations on their versatile functionalities. But one of the mostly encountered limitations of the nanoantennas is the insufficient low quality factor. To mitigate this problem, we propose a novel nanoantenna design that has a significantly enhanced quality factor. In our antenna design, we support the electric dipolar nanoantenna with plasmonic gratings. This design simultaneously combines the functionalities of plasmonic gratings as incidence coupler, SPPs reflector and radiation collimator into one nanoantenna system. When a Bragg condition is met, the reflection coefficient tends to be significantly enhanced and the radiative losses of the nanoantenna will be largely reduced. This leads to an extraordinary enhancement of the resonance intensity and quality factor. Nevertheless, the field distributions of the resonant mode remain largely undisturbed. The radiation pattern of our nanoantenna still possesses two main lobes comparable to an ordinary dipole antenna but with much higher directivity. With the extraordinary enhancement of the resonance intensity, our optical nanoantenna design is highly beneficial for the applications in the ultrafast nonlinear optics, e.g. increasing the conversion efficiency of higher harmonic generation. The simple geometry of our nanoantenna allows the feasibility for the experimental realization and the design theory can be easily extended to other geometries.

Ultrafast nonlinear optical response of metallic nanostructures: Collective effects and hybrid materials

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Particle plasmons govern the optical properties of metallic nanostructures in the visible and in the near-infrared spectral region. Excitation of these particle plasmons with ultrashort light pulses can give rise to a spatiotemporal concentration of the electromagnetic field in the vicinity of the metallic nanostructure. The intensity in these "hot spots" can exceed the incident intensity by orders of magnitude.

In the first subproject, we investigate collective nonlinear optical effects in periodic metal nanoparticle arrays and complex metallic nanostructures containing several tightly packed particles. A recent experiment [1] has shown that the interaction of the nanoparticles can result in a surprising non-monotonic dependence of the conversion efficiency on the lattice constant.

The second subproject is devoted to hybrid nonlinear optical materials composed of a metallic nanostructure and a dielectric nonlinear optical nanoparticle. We report on the recent progress on the fabrication of these hybrid nanostructures.

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Controlling the Photoemission from Nanostructures with Ultrashort THz-Pulses

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High localization and strong enhancement of electric fields in the vicinity of nanostructures enable field-driven electron dynamics. If the quiver amplitude of the photoelectrons exceeds the decay length of the enhanced field, the electrons escape the field in a fraction of the optical cycle, resulting in unique electron-light interactions. We show the first implementation of a field-driven streaking experiment with femtosecond NIR and single-cycle THz pulses at a nanotip to directly measure the near field at the tip apex. The photocurrent and the electron kinetic energy follow the electric field of the THz transient, observed in streaking spectra recorded as a function of delay between THz- and NIR-pulse. The tip geometry determines both field enhancement and decay length, which influence the electron trajectories. In the case of very intense NIR-pulses, hot electrons leave the metal nanotip due to THz-induced tunneling. Varying the time delay visualizes the relaxation process of the hot electron gas. This is a variation of time-resolved photoemission spectroscopy, here probed by tunneling. This experiment is a promising new method to control and compress photoemitted electron pulses and their energy spectra.

Active control of plasmon-polaritons in periodic metal structures

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The key object of plasmonics is the surface plasmon polariton (SPP) - the coupled oscillation of the electromagnetic field and the electron plasma in metals. The excitation of a SPP leads to significant electromagnetic energy localization near the metal surface and thereby enhances nonlinear effects and light-matter interaction. Current state-of-the-art criteria in telecommunication require plasmonics to be active, i.e. a possibility for control by means of an external stimulus must be provided in the timescale of several nanoseconds or shorter. In my talk I will concentrate on plasmonic crystals, which are formed by patterning the metal film with a period comparable to the SPP wavelength. In addition I will consider a hybrid magneto-plasmonic crystal where the metal is patterned at the top of the ferromagnetic film. I will describe three types of experiments where the propagation constant and ultrafast dynamics of SPP are controlled by: (i) external magnetic field [1]; (ii) femtosecond optical pulse [2]; (iii) and coherent THz acoustic phonons [3].

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Nonlinear optical spectroscopy of complex plasmonic nanoantennas

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We peform third harmonic (TH) spectroscopy of complex gold nanoantenna arrays using widely tunable sub-30 fs laser pulses [1]. The TH efficiency of rod-type antennas is found to peak slightly red-shifted with respect to the linear extinction spectrum of the antennas. This red-shift can be attributed to the shift between near-field and far-field peak intensities [2] and can be understood and modeled with a classical anharmonic oscillator [3,4]. Furhtermore we investigate the nonlinear response of dolmen-type plasmonic nanoantennas, which exhibit EIT-like Fano resonances in their linear extincton spectrum. A Fano resonance is due to the subradiant linewidth and the strongly localized near-fields highly attractive for optical nonlinearities. We find the intense TH response of these structures to peak at the lower energy mode of the coupled system. Even more, we investigate the nonlinear response of plasmonic nanoantennas embedded in different highly nonlinear materials, which allow for resonant tuning of nanoantenna enhanced nonlinear light generation.

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Enhancing the second harmonic generation of plasmonic nanowire antennas by tailoring their terminations

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Sub-wavelength light focusing of plasmonic antennas is frequently employed to enhance the nonlinear response of the system. In most cases, however, the system is designed to resonate only at the illuminating pump. In cases when nonlinear interaction at more than single frequency is desired, it is challenging to design nanoantennas that respond resonantly to more than one or better at all interacting frequencies. Considering plasmonic nanowire antennas, we hereby demonstrate the potential to engineer their resonances at more than one frequencies involved in the nonlinear process by carefully tailoring the antenna terminations. Degenerate second harmonic generation is used to demonstrate the utility of our scheme.

Toward coherent control of nanoplasmonic fields measured by carrier-envelop phase-tagged photoelectron emission microscopy

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Recently, strong carrier-envelope phase (CEP) effects have been shown to be crucial in controlling electron motion in the solids [1]. In order to investigate and control CEP effects on tailored plasmonic nanostructures, the phase-tagged time-of-flight-photoelectron emission microscope (ToF-PEEM) has been developed. First experiments measuring the CEP dependence of gold nanoparticles on gold plane (NPOP) with a sub-nanometer gap have been performed with amplified 10 kHz few-cycle laser pulses via a multiphoton photoemission process. The spectral feature obtained from the NPOP sample is modulated with a period of π and we attribute this effect to a correlation between the CEP and the kinetic energy of the electrons coming from the whole sample area rather than from the nanoparticles alone. This CEP feature shows a correlation over 10 ms of consecutive laser pulses and a detailed investigation on this contribution is underway. To further observe a CEP effect on the photoemission process and kinetic energy of electrons from tailored plasmonic nanostructures, a CEP-sensitive nanotip [1] will be used in the next experiment as a proof of principle. Various CEP-predicted nanostructures such as V-shapes [2], single ellipsoids and bowties with ultralow surface roughness will be studied as well. The phase-tagged ToF-PEEM setup provides a versatile tool for both spatial-resolved and energy-resolved studies of attosecond control of electrons in tailored plasmonic nanostructures.

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Dielectric nanospheres under strong few-cycle laser pulses: probing near-fields via phase-controlled electron recollision

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Electron emission from atoms and molecules in intense few-cycle laser pulses can be precisely controlled (with sub-fs resolution) by the carrier-envelope-phase. Recently we demonstrated that such pulses also allow unraveling and controlling ionization processes in more complex many particle systems, such as silica nanoparticles [1]. By using a beam of isolated nanoparticles, the target is replaced after every laser shot such that we could explore the regime near, at and beyond the material damage threshold. The phase-dependent electron acceleration from laser driven dielectric nanoparticles was found to be dominated by electron re-collision, where both the optical-field enhancement and nonlinear collective near-field effects at the surface contribute to the generation of high energy electrons [2]. As incoherent contributions such as thermal evaporation can be discriminated in phase-resolved experiments, the electron spectra provide an image of the transient near-field-driven dynamics under strong fields. In order to explore these capabilities we studied the electron emission from large silica nanospheres (up to 500nm diameter) to resolve the effect of field propagation on the near-field profile. Comparison with quasi-classical trajectory-based mean-field Mie Monte-Carlo simulations shows that size-dependent modifications in the near-field resulting from propagation are imprinted and can be extracted from the electron spectra. Both, experimental as well as theoretical results will be discussed.

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Single nanoparticle assisted few-cycle laser pulse induced local multiphoton internal photoemission in metal-insulator-metal junctions

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The interaction of few-cycle laser pulses (6 fs, 80 MHz repetition rate) with metal-insulator-metal (MIM) junctions offers the possibility to control ultrafast charge transport processes in the junction leading to new optoelectronic devices. For investigating the local few-cycle laser pulse induced transport mechanisms scanning internal photoemission (IPE) microscopy is applied [1]. Here we use this method to investigate the few-cycle laser pulse induced currents in Ag-TaOx-Ta and Au-TaOx-Ta MIM junctions decorated with small citrate stabilized gold nanoparticles. These nanoparticles act as optical nanoantennas for the incident radiation and lead to strong local excitation and field enhancement in the vicinity of the nanoparticles. This dominates the locally induced multiphoton internal photoemission current. The top metal electrode thickness is 40 nm and for few-cycle pulse illumination the multiphoton excitation close to the insulation layer is too low and no internal photoemission current is observed. However, deposition of small metal nanoparticles on the surface leads to a strong modification of the local optical response and in scanning reflection and IPE microscopy small spots with reduced reflectivity appear. Match of reflectivity, IPE maps and scanning electron microscopy images shows that single Au nanoparticles locally induce strong multiphoton internal photoemission currents in the order of several 100 pA. Finite-difference time-domain (FDTD) calculations predict a more than one order of magnitude higher intensity at the metal-insulator interface in contrast to a bare MIM junction without a nanoparticle and the maximum local field strength in the insulating layer is in a range where strong field phenomena start to become relevant.

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Electron motion in few-cycle laser fields around metallic nanostructures

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Sharp metallic nanotips irradiated with few-cycle laser pulses are promising candidates as sources of highly confined electron wavepack- ets of short temporal duration and high spatial directivity. Specifically the strong-field regime, where electron emission by tunneling becomes dominant over multi-photon and above-threshold ionization, enables the generation of (sub-)femtosecond electron wavepackets. For suffi- ciently long wavelengths and short pulses, electrons are accelerated to escape the near field within one half-cycle of the laser oscillation [1,2]. Here, we study for the first time the effect of the carrier envelope phase (CEP) of few cycle laser pulses on the motion of electrons emitted from metallic nanostructures by strong-field tunneling. Sharp gold tips with an apex radius of only 5nm are illuminated with CEP-stable few-cycle near-IR pulses at 1.6 μm and the kinetic energy spectra of the emitted electrons are recorded. We observe pronounced modifications of the photoelectron spectra when varying the CEP. We use a modified three-dimensional Simpleman model to explain our findings. Our results shed new light on the light-driven motion of electrons in the near field of metallic nanostructures on sub-femtosecond time scales.

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Abstracts of Poster Presentations

(sorted by poster number)

#	Presenting Author	Title
1	*B. Förg*	Attosecond streaking from metal nanotips
2	*J. Förstner*	Near-field coupling and second-harmonic generation in split-ring resonator arrays
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Poster 1:

Attosecond streaking from metal nanotips

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We have completed and commissioned the attosecond beamline (AS-5) at MPQ to study nanoplasmonic fields with attosecond precision. In our studies on isolated nanosystems such as metal nanotips, we aim to reach deep insight into the collective electron dynamics in intense laser fields employing attosecond nanoplasmonic streaking, which we have also studied theoretically (1). Nanosystems can exhibit vastly different field profiles across its surface. In general, the gradients will depend on the effective radius of curvature and be more pronounced for complicated, resonant antenna geometries. A second mechanism leading to an effective inhomogeneity is the travel of the streaked electron in combination with the localized character of the plasmonic fields 1. For fields with a strong localization at the surface and fast photoelectrons, they will leave this field even before the light oscillations terminate. For spatially more extended fields and moderate electron energies, they are ponderomotively accelerated. In pioneering studies, we have explored the streaking from Au nanotips (with about 100 nm apex radius). First experimental results are compared to trajectory calculations, which indicate contributions from the tip shank and the tip apex, where the apex emission leads to higher streaking amplitudes. Both contributions are delayed with respect to each other. The experiments open the door to exciting future research into nanolocalized attosecond phenomena.

(1) Süßmann, F. & Kling, M. F. Attosecond nanoplasmonic streaking of localized fields near metal nanospheres. Phys. Rev. B 84, 121406(R) (2011)

Poster 2:

Near-field coupling and second-harmonic generation in split-ring resonator arrays

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Artificial magnetism in split-ring resonator arrays (SRR) is one of the prospective ways to create metamaterials with large second harmonic conversion rates. However, the detailed mechanism of the origin of the SHG in this case remains unclear. We use numerical simulations to calculate the linear and nonlinear optical response from split-ring resonator (SRR) arrays and to study collective effects between the constituent SRRs that determine spectral properties of the SHG. We apply the Discontinuous Galerkin Time Domain (DGTD) method utilizing its advantage in flexibility, higher order accuracy, and capability of handling non-linear problems. In a semi-classical approach, we consider non-linear electron flow confined in a metallic structure as cold plasma and evaluate the hydrodynamic Maxwell-Vlasov model including a Fermi pressure contribution. Our model is able to qualitatively reproduce and explain the non-monotonic dependence of the spectral SHG transmission measured experimentally for SRR arrays with different lattice constants.

Poster 3:

Synthesis and in situ analysis of multimode excitations in top-down fabricated plasmonic nanocircuits

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Plasmonic modes supported by noble-metal nanostructures offer strong sub-wavelength confinement and therefore promise the realization of nanometer-scale integrated optical circuits with well-defined functionality. The implementation of optical nanocircuitry requires the possibilities to fabricate arbitrary circuit designs as well as to selectively excite, propagate and re-emit specific eigenmodes within a circuit. In our work we investigate the fundamental properties of micro-fabricated optical two-wire transmission lines and branched Y-shaped optical splitters. Both structures support multiple modes that can selectively be excited by carefully optimization of an incoupling antenna and by choosing a well defined excitation position as well as laser polarization. For experimental visualization of the two fundamental modes within the two wire transmission line a mode-detector structure is used which spatially separates the farfield emission of both modes. The transmission properties of pure modes as well as superpositions of modes - which in the case of the Y-splitter structure lead to selective excitation of either of both arms - are investigated. This is achieved by controlling the relative phase between two perpendicular polarized input pulses and in turn determine the near-field intensity distribution along the transmission line. As the control of the propagation direction is achieved solely by the polarization, the spectral phase of the pulse is left as a free parameter that can be used to tailor the temporal structure of the near field.

Poster 4:

Driving Strong-Field Photoemission with mid-infrared Pulses into the sub-cycle regime

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We investigate strong-field photoemission from sharp nanotips in a sub-cycle acceleration regime, driven by ultrashort pulses at near-and mid-infrared wavelengths. Photoelectron spectra are measured using a time-of-flight spectrometer, enabling the exploration of a broad parameter range. Kinetic energies of hundreds of electronvolts are experimentally observed. Wavelength and intensity dependend photoelectron spectra show scalings unique to nanostructures. Our theoretical models reveal that these electron dynamics arise from the localization of the enhanced near-field.

Poster 5:

Circular Plasmons in cross shaped nanoantennas

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Cross shaped nanoantennas that are designed to carry angular momentum have been fabricated by focused ion beam (FIB). The nanoantennas can be resonantly excited by Ti:sapphire femtosecond laser pulses. The emitted electron intensity from individual antennas, as determined by a photoemission electron microscope, is a measure for the plasmonic field enhancement. The resonant wavelength scales with the size of the antennas. The sample is illuminated from the front side at grazing incidence or from the backside at normal incidence. In both geometries the photoemission intensity of some antennas shows an unexpectedly pronounced polarization dependence, which can be interpreted as circular plasmon mode.

Poster 6:

The complex dispersion relation of surface plasmon polaritons at gold/para-Hexaphenylene interfaces

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The propagation of surface plasmon polaritons (SPP) at gold/dielectric interfaces is characterized by the complex frequency-wave vector dispersion relation. In this article we describe an experimental approach that enables one to gain access to both, the real and imaginary part of the SPP dispersion relation, by means of two-photon photoemission electron microscopy (2P-PEEM). A comparison of experimental 2P-PEEM results obtained for a gold/vacuum interface with calculations based on optical permittivity data [1] demonstrates the ability of the presented approach. A systematic study of the dispersion relation of gold surfaces covered with thin para-Hexapheneylene films shows how effective the propagation of surface plasmon polaritons can be tuned by adjustment of the dielectric film thickness. Deviations of the experimental results from effective index calculations indicate the relevance of thin film peculiarities arising from the details of the growth process.

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Poster 7:

The role of phase cycling in coherent 2D nanoscopy

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Coherent two-dimensional (2D) nanoscopy is a femtosecond time-resolved nonlinear technique which combines the principle of coherent two-dimensional spectroscopy with photoemission electron microscopy (PEEM) [1]. Using a sequence of four ultrashort laser pulses for excitation ensures high temporal resolution and the detection of the emitted photoelectrons with a PEEM guarantees high spatial resolution on the nanometer scale [2]. As opposed to conventional 2D spectroscopy the detected signal is non-optical and incoherent, but the desired coherent information, e.g. the photon-echo signal, can nevertheless be retrieved by employing phase cycling [3, 4]. This procedure is based on the fact that every coherent signal contribution has a particular phase dependency of the input pulses. Therefore a 2D nanoscopy measurement is repeated for different phase combinations of the four-pulse sequence and the required contribution can be extracted by a suitable linear combination of the measured data. We show the implementation of phase cycling in 2D nanoscopy and explain the similarities and differences compared to phase matching. Furthermore we describe the possibility to reduce the measurement time by choosing the right phase-cycling procedure and to obtain different coherent signal contributions with only one phase-cycling scheme.

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Poster 8:

Light propagation and optical response in small metal clusters and metal cluster-biomolecule nanostructures

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We present methods of simulation of nonlinear optical response of small metal clusters and metal cluster-biomolecule nanostructures including time-resolved harmonic generation and time resolved photoelectron emission. The approach is based on our previously developed semi-classical field-induced surface hopping method for treatment of laser-induced nuclear dynamics and time-dependent density functional theory for electron dynamics. The results of application of these methods to various systems consisting of small metal clusters and biomolecules are presented. We also present a method to simulate ultrafast light propagation in ordered metal cluster arrays and demonstrate the possibility of optimal control of light propagation in such structures.

Poster 9:

Photon and Electron Emission from Free and Deposited Nanoparticles

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The discontinuous Galerkin time-domain method (DGTD) combines the highly flexible spatial discretization of finite-element methods with the possibility to investigate nanophotonic systems in the time-domain. Here, we investigate the interaction of three-dimensional metallic nanostructures with an incoming light field. In particular, we discuss the linear modeling of the dielectric function of metals via a combination of Drude and Lorentz terms. Furthermore, we present results on the second harmonic generation by metallic nanostructures modeled by a perturbative expansion of a hydrodynamical model.

Poster 10:

Nanolocalization of time-reversed coherent optical fields in random scattering media

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In this project, our primary goal is to localize broadband coherent optical fields in random media and random nanoantennas well below the diffraction limit via time-reversal of the measured emitted fields. A nanoscale light emitter is embedded in a nanoscale random multiply scattering environment. The underlying idea is that the far-field emission pattern of the emitter contains information about the localized emission.

The experimental realization of the time-reversal scheme requires a vector field synthesizer allowing for a complete control of the temporal evolution of the polarization state. Based on an optimization procedure using ZEMAX we have built an interferometric vector field synthesizer in an all-reflective design including an active stabilization. In order to show the functionality of the setup, we have performed first test measurements.

A succesfull demonstration of nanolocalization of time-reversed broadband coherent optical fields requires information of specific properties of the scattering nanostructures. To take this into account, simulations have been run using MESME (Multple Elastic Scattering of Multipole Expansion). In a first experimental realization we are seeking for a one-dimensional nanolocalization in nano-textured thin-film solar cells.

Poster 11:

Time-of-Flight-Photoelectron Emission Microscope for Probing and Control of Surface Plasmons at Ultrahigh Spatiotemporal Resolutions

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The attosecond plasmonic field microscope utilizing attosecond streaking spectroscopy from high harmonic generation (HHG) was developed to obtain a direct and non-invasive access to the plasmonic dynamics in nanostructured surfaces with attosecond temporal and nanometer spatial resolution. We have previously demonstrated the feasibility of time-of-flight-photoemission electron microscopy (ToF-PEEM) in combination with extreme ultraviolet attosecond pulses from a HHG source. Recently, strong carrier-envelope phase (CEP) effects were observed from a nanoscale tungsten tip when interacting with few-cycle laser pulses. Therefore, it is also very promising to investigate and control CEP effects on tailored plasmonic nanostructures. Hence, the single-shot phase-tagged ToF-PEEM has recently been developed. First experiments measuring the CEP dependence on single gold nanoparticles have been performed with 10 kHz few-cycle laser pulses via a multiphoton photoemission process. We report the first results on the observation of CEP feature from the single gold nanoparticle sample and the challenges of the experiments. We also address the possible solutions to improve the plasmonic sample quality and design for probing and control the plasmonic dynamics via CEP effect. In addition, we wish to observe the time evolution of surface plasmon polaritons (SPPs) in metallic nanostructures to explore their potential as plasmon waveguides. We characterize various silver nanostructures using 400 nm picosecond laser pulses as a first step toward achieving time-resolved two-photon photoemission (2PPE) experiments. These experiments demonstrate next steps toward the temporal characterization and CEP control of nanoscaled localized surface plasmon fields in a femtosecond optical-pump/attosecond XUV-probe scheme as well as 2PPE pump-probe experiments on SPPs in waveguides with femtosecond resolution.

Poster 12:

Determining excitation pathways in a hybrid metal-organic interface state using coherent 2D nanoscopy

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Coherent 2D nanoscopy [1] combines the principle of conventional 2D spectroscopy with photoemission electron microscopy (PEEM). While 2D spectra are well suited for studying the dynamics of quantum states as well as interactions between them, PEEM allows for the detection of the electrical near field of e.g. nano-structured samples with a resolution well below the optical diffraction limit. The lateral resolution enables us to address small ensembles of quantum systems rather than large-scale averaging.

We use coherent 2D nanoscopy to study the hybrid electronic state forming at the interface between cobalt and the organometallic complex tris(8-hydroxyquinolinato)aluminium (Alq3). We observe the excitation paths at such a hybrid metal-organic interface and monitor the subsequent electron dynamics. To extract the lifetime and energy shift of the involved electronic states we develop a model to simulate 2D photoemission spectra based on the Newns-Anderson model for chemisorption.

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Poster 13:

Optical antennas and plasmon dynamics

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Many applications of surface plasmon polaritons (SPPs) that rely on propagation and field enhancement in linear and nonlinear processes are limited by damping. However, despite being a key parameter in plasmonics, many issues related to the different relaxation pathways and their determining parameter have remained the subject of intense discussions. Despite the obvious correlation of damping with the Drude relaxation time of the supporting metal, which describes the effective momentum scattering and thus dephasing of a free electron, there are many inconsistencies in both theory and experiment. The investigation of the electrodynamics in metals is often challenging due to the high relaxation rates (on few femtoseconds time scales) associated with the intrinsic large carrier densities in metals. With the general experimental difficulty on separating the relative contributions of radiative and non-radiative channels, we resort to both theoretical and numerical methods to study the radiative and non-radiative decay of SPPs. We also compared the result to general response time of radio-frequency antennas, and reconciled the mismatch between femtosecond decay time of particle plasmons and nanosecond response time of RF antennas.

Poster 14:

Coupling mediated coherent control of localized plasmonic resonances

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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 explore the possibilities of coherently controlling the light-matter interaction in plasmonic meta-atoms with the goal of modifying the light propagation and nonlinear light emission. Our approach focuses on the strong near-field interaction within a plasmonic meta-atoms unit-cell. Recently it was shown that utilizing strong near-field coupling effects a so-called plasmon induced transparency can appear leading to a strong reduction in the scattering loss of the meta-atom. Similar to atomic physics where the electromagnetic induced transparency can lead to an enhancement of the nonlinear properties due to large dispersion we expect that the plasmon induced transparency in plasmonic meta-atoms will lead to similar effects like light storage and strong field enhancement. However, the short life times and the high energy dissipation in the plasmonic systems together with extremely small structures sizes are tremendous challenges in a practical realization. Furthermore, we investigate if the near-field coupling effects are strong enough to lead to a complete control of the optical properties. Meeting these challenges will yield a fundamental and more complete understanding of the linear and nonlinear optical properties of collective excitations in strongly coupled metallic meta-atoms and metamaterials.

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Program Overview

Time	Sunday 16.6.	Monday 17.6.	Tuesday 18.6.	Wednesday 19.6.
8:50		**Opening Remarks**		
9:00		**F. Javier Garcia de Abajo** (ICFO Barcelona, Spain) *Plasmons in graphene and graphene-related molecules*	**Mark I. Stockman** (Georgia State University, USA) *Solids in ultrafast and strong optical fields: new phenomena*	**Ilya Akimov** (Universität Dortmund, Germany) *Active control of plasmon-polaritons in periodic metal structures*
9:40 *10:00*		**R. Ciesielski** (LMU, München, Germany) **M. Richter** (TU Berlin, Germany)	**K. Lindfors** (MPI-FKF, Stuttgart, Germany) **S. Michaelis de Vasconcellos** (Universität Münster, Germany)	**B. Metzger** (Universität Stuttgart, Germany) **S. Bin Hasan** (Abbe Center of Photonics, Jena, Germany)
10:20		Coffee Break	Coffee Break	Coffee Break
11:00 *11:20*		**Markus Raschke** (JILA, Boulder, USA) *Ultrafast antenna coupled infrared vibrational dynamics and control*	**Xiaoyang Zhu** (Columbia University, USA) *Exciton fission, quantum coherence, and solar energy conversion beyond the limit*	**U. Kleineberg** (LMU, München, Germany) **Th. Fennel** (Universität Rostock, Germany)
11:40 *12:00*		**N. Accanto** (ICFO, Barcelona, Spain) **M. Pawłowska** (Universität Würzburg, Germany)	**D. Leipold** (TU Ilmenau, Germany) **C. Strüber** (Universität Bielefeld, Germany)	**M. Hensen** (Universität Bielefeld, Germany) **J. Vogelsang** (Universität Oldenburg, Germany)
12:30		Lunch	Lunch	Lunch
14:00		**Hrvoje Petek** (University of Pittsburgh, USA) Ultrafast coherent multi-photon photoemission spectroscopy and microscopy	**Dmitry A. Yarotski** (Los Alamos Nat. Lab., USA) *Ultrafast characterization and imaging: from the nanoscale to the mesoscale*	Departure
14:40 *15:00*		**F. Meyer zu Heringdorf** (U. Duisburg-Essen, Germany) **M. Bauer** (Universität Kiel, Germany)	**M. Betz** (Universität Dortmund, Germany) **M. Lippitz** (MPI-FKF, Stuttgart, Germany)	
15:20		Coffee break	Coffee break	
16:00 *16:20*		**S. F. Becker** (Universität Oldenburg, Germany) **C. González** (TU Kaiserslautern, Germany)	**P. Groß** (Universität Oldenburg, Germany) **J. Qi** (Universität Jena, Germany)	
16:40 *17:00*		**Poster Session**	**S. Linden** (Universität Bonn, Germany) **L. Wimmer** (Universität Göttingen, Germany)	
17:20	Arrival and Registration			
19:00	Buffet Supper	Dinner	Dinner	
21:00		**Informal discussion**	**Informal discussion**	