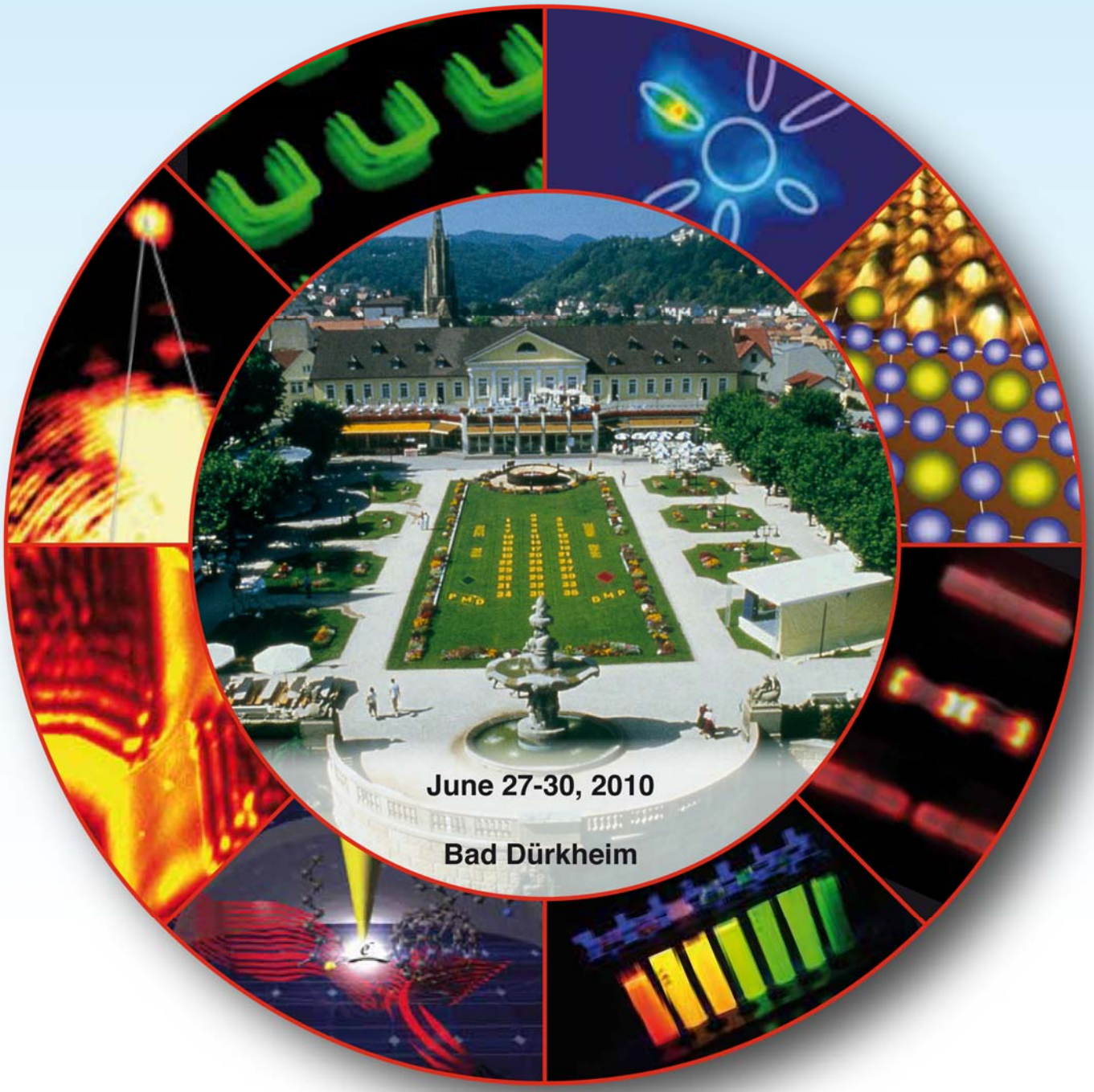
UNO-2

2nd International Workshop on Ultrafast Nanooptics

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.

This workshop is the follow up meeting of the first "Workshop on Ultrafast Nanooptics" which was organised as a "Else and Wilhelm Heraeus Seminar" in 2008 in Bad Honnef/Germany. This second workshop will give the opportunity to present and discuss the recent developments in the field. The workshop also serves as the annual meeting of the DFG SPP 1391 priority program members.

Walter Pfeiffer and Martin Aeschlimann

Program Overview

Time	Sunday 27.6.	Monday 28.6.	Tuesday 29.6.	Wednesday 30.6.
8:50		**Opening Remarks**		
9:00		**Mark I. Stockman** (Georgia State University, USA) *Ultrafast Nanoplasmonics*	**Daan Brinks** (ICFO, Barcelona, Spain) *Nanoscale Coherent Control: Single Molecules and Optical Antennas*	**Tobias Kampfrath** (AMOLF, The Netherlands) *Ultrafast Adiabatic Manipulation of Slow Light in Photonic Crystals*
9:40 *10:00*		**M. Betz** (TU Dortmund, Germany) **T.M. Bernhardt** (Universität Ulm, Germany)	**M. Agio** (ETH, Zürich, Switzerland) **K. Lindfors** (Universität Stuttgart, Germany)	**D. Chen** (University of Twente, The Netherlands) **A.W. Holleitner** (TU München, Germany)
10:20		Coffee Break	Coffee Break	Coffee Break
11:00 *11:20*		**T. Brixner** (Universität Würzburg, Germany) **M. Silies** (Universität Oldenburg, Germany)	**R. Vogelsang** (MPI, Stuttgart, Germany) **C. Strüber** (Universität Bielefeld, Germany)	**N.M.Buckanie** (Universität Duisburg-Essen,Germany) **T.Leißner** (Universität Kiel, Germany)
11:40 *12:00*		**C. David** (Instituto de Optica, CSIC, Spain) **T. Utikal** (Universität Stuttgart, Germany)	**B. Hecht** (Universität Würzburg, Germany) **B. Chichkov** (Laser Zentrum Hanover, Germany)	**Discussion** **Closing Remarks**
12:30		Lunch	Lunch	Lunch
14:00		**Cyriaque Genet** (University of Strasbourg, France) *Optical Properties of Metallic Nanostructures: Plasmons, Aperture and Molecules*	**Olivier Martin** (EPFL, Lausanne, Switzerland) *Controlling light at the nanoscale with different types of plasmonic antennas*	Departure
14:40 *15:00*		**D. Zhang** (Universität Tübingen, Germany) **S. M. Foreman** (Stanford University, USA)	**R. Bratschitsch** (Universität Konstanz, Germany) **D. Leipold** (Universität Oldenburg, Germany)	
15:20		Coffee break	Coffee break	
16:00 *16:20*		**H. Yanagisawa** (Universität Zürich, Switzerland) **C. Ropers** (Universität Göttingen, Germany)	**J. Förstner** (Universität Paderborn, Germany) **P. Vasa** (Universität Oldenburg, Germany)	
16:40		**Poster Session**	**Y. Grynko** (Universität Paderborn, Germany)	
17:00	Arrival and Registration		**T. Schumacher** (MPI, Stuttgart, Germany)	
17:20 *17:40*			**F. Schertz** (MPI, Mainz, Germany) **T. Pertsch** (Universität Jena, Germany)	
19:00	Buffet Supper	Dinner	Dinner	
21:00		**Informal discussion**	**Informal discussion**	

Workshop Program

Sunday, June 27, 2010

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

Monday, June 28, 2010

07:45	*BREAKFAST*
08:50	*W. Pfeiffer* **Opening remarks**

Session 1

9:00	*Mark. I. Stockman (Georgia State University, USA) - invited* **Ultrafast nanoplasmonics**
9:40	*C. Ruppert, S. Thunich, G. Abstreiter, A. Fontcuberta i Morral, A. W. Holleitner, M. Betz* *(TU München, TU Dortmund, EPFL Lausanne)* **Coherent control of µA currents in single GaAs nanowires**
10:00	*M.E. Vaida, K. Hinrichs, T.M. Bernhardt* *(Universität Ulm)* **Methyl bromide photodissociation on supported gold clusters: Size dependencies and perspectives for a laser-selective photochemistry**
10:20	*COFFEE BREAK*

Session 2

11:00	*A. Reiserer, J.-S. Huang, B. Hecht, T. Brixner* *(Universität Würzburg)* **Subwavelength functional elements for femtosecond plasmonic signals**

Monday, June 28, 2010

11:20 *M. Mascheck, S. Schmidt, M. Silies, P. Vasa, D. Leipold, E. Runge, K. Kitamura, T. Yatsui, M. Ohtsu, C. Lienau*
(Universität Oldenburg, TU Ilmenau, University of Tokyo)
Localizing few-cycle light pulses in space and time in random dielectric media

11:40 *C. David, F. J. García de Abajo*
(Instituto de Óptica - CSIC, Madrid, Spain)
Spatial nonlocality in the optical response of metal nanoparticles

12:00 *T. Utikal, T. Zentgraf, M. Lippitz, and H. Giessen*
(MPI and Universität Stuttgart, University of California, USA)
Group Index Enhanced Third-Harmonic Generation in Hybrid Plasmonic Systems

12:30 *LUNCH*

Session 3

14:00 *Cyriaque Genet, Th. Ebbesen (ISIS - University of Strasbourg) - invited*
Optical properties of metallic nanostructures: plasmons, apertures and molecules

14:40 *X. Wang, A. Horneber, J. Mihaljevic, K. Braun, H.-J. Egelhaaf, C.J. Brabec, D. Zhang, A.J. Meixner*
(Universität Tübingen, Konarka Technologies GmbH)
Tip-enhanced ultrafast spectroscopy and microscopy of organic solar cell blend film

15:00 Seth M. Foreman, C. Kealhofer, M.A. Kasevich
(Stanford University, USA)
Low-work function field emission tips triggered by an ultrafast laser

15:20 *COFFEE BREAK*

16:00 *H. Yanagisawa, C. Hafner, P. Dona, M. Klöckner, D. Leuenberger, T. Greber, M. Hengsberger, J. Osterwalder*
(Universität Zürich, Switzerland)
Optical control of field-emission sites by femtosecond laser pulses

Monday, June 28, 2010

16:20 *M.Gulde, R. Bormann, A. Weismann, S. Yalunin, C. Ropers (Universität Göttingen)*
Strong-field photoelectron emission from metal nanotips

17:00 **Poster Session**

19:00 *DINNER*

21:00 **Informal Discussion**

Tuesday, June 29, 2010

07:45 *BREAKFAST*

Session 4

9:00 *Daan Brinks, M. C. Lopez, R. Hildner, F.D. Stefani, T.H. Taminiau, A.G. Curto, F. Kulzer[3], N. F. van Hulst (ICFO and ICREA Barcelona, Universidad de Buenos Aires, Université Lyon) - invited*
Nanoscale coherent control: single molecules and optical antennas

9:40 *X.-W. Chen, A.B. Ghasemi, V. Sandoghdar, M. Agio (ETH Zurich, Shahid Beheshti University Theheran)*
Coherent excitation of single emitters with optical antennas and ultrashort laser pulses

10:00 *M. Pfeiffer, K. Lindfors, C. Wolpert, H. Giessen, M. Lippitz, P. Atkinson, A. Rastelli, M. Benyoucef, O. G. Schmidt (Universität Stuttgart, MPI Stuttgart, IFW Dresden)*
Enhancing photoluminescence of single self-assembled GaAs quantum dots using plasmonic nanoparticles

10:20 *COFFEE BREAK*

Session 5

11:00 *R. Vogelgesang, J. Dorfmüller, K. Kern, C. Rockstuhl, C. Etrich (MPI Stuttgart, Universität Jena)*
Linear Plasmonic Nano-Antennas: Experiment, Simulation, and Theory

Tuesday, June 29, 2010

11:20 *M. Aeschlimann, M. Bauer, D. Bayer, T. Brixner, S. Cunovic, A. Fischer, P. Melchior, W. Pfeiffer, M. Rohmer, C. Schneider, C. Strüber, P. Tuchscherer, D.V. Voronine*
(TU Kaiserslautern, Universität Kiel, Universität Würzburg, Universität Bielefeld)
Deterministic control of nanooptical excitations in plasmonic nanostructures

11:40 J.-S. Huang, P. Geisler, C. Brüning, J. Kern, J.C. Prangsma, P. Biagioni, B. Hecht *(Universität Würzburg)*
Atomically flat single-crystalline gold nanostructures for plasmonic nanocircuitry

12:00 *B. Chichkov, C. Reinhardt, A. Seidel*
(Laser Zentrum Hannover)
Recent progress in two-photon-polymerization-based ultra-fast nanooptics applications

12:30 *LUNCH*

Session 6

14:00 *Olivier J.F. Martin (EPFL - Lausanne, Switzerland) - invited*
Controlling light at the nanoscale with different types of plasmonic antennas

14:40 *R. Bratschitsch, T. Hanke, G. Krauss, D. Träutlein, A. Leitenstorfer*
(Universität Konstanz)
Few-Cycle Nonlinear Optics with Single Plasmonic Nanoantennas

15:00 *D. Leipold, M.Maschek, S. Schmidt, M. Silies, T. Yatsui, K. Kitamura, M. Ohtsu, E. Runge, C. Lienau*
(TU Ilmenau, Universität Oldenburg, University of Tokyo)
Photon modes and second harmonic generation in ZnO nano-needle arrays

15:20 *COFFEE BREAK*

16:00 *M. Wand, T. Meier, J. Förstner*
(Universität Paderborn)
Ultrafast nonlinear optical response of metal surfaces

Tuesday, June 29, 2010

16:20 *P. Vasa, R. Pomraenke, W. Wang, G. Cirmi, E. De Re, S. Schwieger, D. Leipold, E. Runge, G. Cerullo, C. Lienau*
(Universität Oldenburg, TU Ilmenau, Politecnico di Milano)
Ultrafast control of the large Rabi splitting in metal-J-aggregate hybrid structures

16:40 *Grynko, J. Förstner, T. Meier*
(Universität Paderborn)
Simulation of the nonlinear response of split ring resonators with the Discontinuous Galerkin Time Domain method

17:00 *T. Schumacher, K. Kratzer, M. Lippitz*
(MPI Stuttgart, Universität Stuttgart)
Plasmon Hybridization Enhances the Nonlinear Response of Single Metal Nanoparticles

17:20 *F. Schertz, M. Schmelzeisen, H.J. Elmers, G. Schönhense, H.J. Butt, M. Kreiter*
(MPI Mainz, Universität Mainz)
Near-field measurements on nanoscopic sphere-on-plane systems by means of PEEM

17:40 *S.B. Hasan, J. Reinhold, T. Paul, C. Helgert, A. Chipouline, C. Rockstuhl, T. Pertsch*
(Universität Jena)
Intrinsic and Extrinsic Nonlinearities of Nanooptical Systems *Y.*

19:00 *DINNER*

21:00 **Informal Discussion**

Wednesday, June 30, 2010

07:45 *BREAKFAST*

Session 7

9:00 *Tobias Kampfrath, D.M. Beggs, T. F. Krauss, L. (Kobus) Kuipers*
(AMOLF Amsterdam, FHI Berlin, University of St Andrews) - invited
Ultrafast adiabatic manipulation of slow light in photonic crystals

Wednesday, June 30, 2010

9:40 *D. Chen, P. van der Walle, F. Segerink, L. Kuipers, J.L. Herek*
(University of Twente, FOM Amsterdam)
Second-harmonic on hole array generated in a long-lived resonance

10:00 *L. Prechtel, L. Song, S. Manus, D. Schuh, W. Wegscheider, A. W. Holleitner*
(Walter Schottky Institut and TU München, LMU München, ETH Zürich)
Time-resolved ultrafast photocurrent spectroscopy using THz stripline circuits on carbon nanotubes and graphene

10:20 *COFFEE BREAK*

Session 8

11:00 *N.M. Buckanie, P. Kirschbaum, C. Wirtz, S. Sindermann, F.Meyer zu Heringdorf*
(Universität Duisburg-Essen)
Surface Plasmon Coupling and Manipulation using Nonlinear PEEM

11:20 *T. Leißner, J. Kjelstrup-Hansen, K. Thilsing-Hansen, M. Bauer, H.-G. Rubahn*
(Universität Kiel, University of Southern Denmark)
Interaction of para-hexaphenylene based nanofibers with plasmonic substrates studied by phase resolved photoemission electron microcopy

11:40 **Discussion and Closing Remarks**

12:30 *LUNCH*

END OF SEMINAR / DEPARTURE

Abstracts of oral Presentations *(in chronological order)*

Ultrafast Nanoplasmonics

Mark I. Stockman

Department of Physics and Astronomy, Georgia State University, Atlanta, GA 30302, USA

Nanoplasmonics deals with collective electron dynamics on the surface of metal nanostructures, which arises as a result of excitations called surface plasmons. The surface plasmons localize and concentrate optical energy in nanoscopic regions creating highly enhanced local optical fields. They undergo ultrafast dynamics with timescales as short as a few hundred attoseconds. From the latest developments and original work in nanoplasmonics, we will consider SPASER (quantum nanoscale optical generator and ultrafast amplifier), attosecond nanoplasmonic field microscope, ultrafast coherent control on the nanoscale, and SPIDER (surface-plasmon-induced drag-effect rectification) that leads to generation of nanoscale THz fields by femtosecond polaritonic pulses in metal nanowires.

Coherent Control of µA Currents in Single GaAs Nanowires

C. Ruppert[1], S. Thunich[2], G. Abstreiter[3], A. Fontcuberta i Morral[4], A. W. Holleitner[3], M. Betz[2]

[1]*Physik-Department E11, TU München*
[2]*Experimentelle Physik 2, TU Dortmund*
[3]*Walter-Schottky-Institut, TU München*
[4]*Laboratoire des Materiaux Semiconducteur, EPFL Lausanne*

Coherent control of photocurrents is known from bulk semiconductors. To transfer the concept to single nanowires, we use a phase-stable superposition of 1550 nm femtosecond pulses from a compact Er:fiber source and their 775 nm second harmonic. The generated current is characterized by analyzing the potential difference at the contacted wire ends. From bulk material it is known that the time derivative of the current density injection rate scales sinusoidally with the relative phase of the pulse pair. For the nanowire, we observe a phase independent offset and a phase dependent photocurrent amplitude that is indicative of a coherently controlled current. From the ~50 pA average current in the external circuit, it is straightforward to estimate a peak current in the wire. The duration of the ultrafast current burst is limited by the momentum relaxation time in GaAs (200 fs) but has to be at least as long as the 100 fs duration of the excitation pulse. Taken together with the 90 MHz repetition rate of the laser system, we conclude a magnitude of the peak current as large as several µA. Further experiments confirm the coherently controlled current as arising from a third order optical nonlinearity as expected for a coherently controlled current. In addition, the analysis of current injection for various polarization configurations of the fundamental/second harmonic pulse gives interesting insight into the wire's optical anisotropy. These techniques intrinsically represent the fastest way to generate charge currents in nanostructures.

Methyl bromide photodissociation on supported gold clusters: Size dependencies and perspectives for a laser-selective photochemistry

Mihai E. Vaida, Kira Hinrichs, Thorsten M. Bernhardt

Institute of Surface Chemistry and Catalysis, University of Ulm, Albert-Einstein-Allee 47, 89069 Ulm, Germany

Supported metal clusters are of utmost importance in chemical catalysis. Size-selected metal clusters on single crystal substrates and well-defined ultra-thin oxide films represent versatile model systems for the investigation of the physical and chemical properties of such nano-systems.

We employ femtosecond laser pump-probe mass spectrometry with resonance enhanced multi-photon ionization to reveal the time-dependent photodissociation dynamics of methyl bromide molecules adsorbed on supported noble metal clusters. This experiment reveals striking size-dependencies in the transient methyl desorption signal from gold clusters which are interpreted in terms of the cluster dimensionality on a magnesia substrate.[1-3]

In an effort to identify the fate of the photodissociation products on the surface, scanning tunneling microscopy is applied to analyze the supported clusters after the photoreaction. The combination of methyl product mass spectrometry and local surface analysis opens new possibilities to optimize laser pulse shapes for enhanced photoreaction and selective field localization on the surface.

Femtosecond time resolved pump-probe mass signal of methyl cations detected from a bare MgO/Mo(100) substrate (circles) and from the same surface covered by gold clusters (triangles). The inset shows the quadratic pump power dependence recorded for the excitation of CH3Br on the bare MgO/Mo(100) substrate.[3]

[1] M.E. Vaida, P.E. Hindelang, T.M. Bernhardt, J. Chem. Phys. 129 (2008) 011105.

[2] M.E. Vaida, T.M. Bernhardt, ChemPhysChem 11 (2010) 804.

[3] M.E. Vaida, T. Gleitsmann, R. Tchitnga, T.M. Bernhardt, Phys. Status Solidi B 247 (2010) 1001.

Subwavelength Functional Elements for Femtosecond Plasmonic Signals

Andreas Reiserer[1], Jer-Shing Huang[2], Bert Hecht[2], Tobias Brixner[1]

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

[2]*Physikalisches Institut, Universität Würzburg, 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.

Localizing few-cycle light pulses in space and time in random dielectric media

M. Mascheck[1], S. Schmidt[1], M. Silies[1], P. Vasa[1], D. Leipold[2], E. Runge[2], K. Kitamura[3], T. Yatsui[3], M. Ohtsu[3], C. Lienau[1]

[1]*Institut für Physik, Carl von Ossietzky Universität, 26111 Oldenburg, Germany*

[2]*Institut für Mikro-und Nanotechnologien, Technische Universität Ilmenau, 98684 Ilmenau, Germany*

[3]*School of Engineering, University of Tokyo, 113-8656, Japan*

The localization of light due to multiple scattering in disordered nanoscale dielectric media is a particularly fascinating aspect of wave physics [1], offering a possibility to partially trap light and therefore to enhance its interaction with the medium. It is of particular importance in nano-optics, where the localization of surface plasmon polaritons in metallic nanostructures enables one to localize visible light to dimensions of 10 nm or even less and therefore to generate greatly enhanced local electromagnetic fields [2]. This combination of field localization and enhancement is currently finding a rapidly increasing number of applications such as locally-enhanced Raman spectroscopy, nano- and biosensing or ultrahigh resolution optical and electron microscopy.

So far, light localization due to wave interference has mostly been studied by macroscopic experimental techniques such as, e.g., coherent backscattering [1]. More direct studies are experimentally challenging since spatial light localization occurs on a (sub-) wavelength scale and the scattering dynamics happen on ultrashort, femtosecond time scales.

Here, we directly visualize, for the first time, the weak localization of light in both space and time in a disordered array of ZnO nanoneedles using a novel diffraction-limited second-harmonic microscope with few-cycle time resolution. Key hallmarks of weak localization, in particular the existence of strong spatially localized electromagnetic fields with finite coherence times, are experimentally resolved. This gives new and direct insight into the evolution of the weak (Anderson) localization of light in disordered dielectrics. Such nanoneedle arrays therefore present a highly interesting template for exploring SPP localization in disordered metallic nanostructures as well as random lasing.

[1] D.S. Wiersma, P. Bartolini, A. Lagendijk, and R. Righini, Nature **390**, 671-673 (1997)

[2] M.I. Stockman, Phys. Rev. Lett. 84, 1011-1014 (2000)

Spatial Nonlocality in the Optical Response of Metal Nanoparticles

Christin David[1], F. Javier García de Abajo[1]

Instituto de Óptica "Daza de Valdés" - Consejo Superior de Investigaciones Científcas
Serrano 121, 28006 Madrid, Spain

Spatial nonlocality is known to play an important role at distances of a few nanometers, but few efforts have been made to theoretically investigate nonlocal effects in a rigorous way. We present two different approaches to account for nonlocality in metal nanoparticles: (a) the non-retarded specular reflection model (SRM) and (b) the retarded hydrodynamical model. Comparison with available experiments results in excellent agreement with our parameter-free modeling of nonlocal effects, which produce dramatic changes with respect to the customary local theory. We show that nonlocal effects in both models produce sizable plasmon blue shift and broadening in single metal nanoparticles as well as in dimers and nanoshells. Analysis of the plasmon resonance dependence for dimers on the inter-particle spacing and nanoparticle size allows us to separate nonlocal and retardation effects within the hydrodynamical model. This study is particularly relevant for broad, active areas involving applications of local field enhancement to biosensing and nonlinear optics.

Group Index Enhanced Third-Harmonic Generation in Hybrid Plasmonic Systems

T. Utikal[1,2], T. Zentgraf[3], M. Lippitz[1,2], and H. Giessen[1]

[1] *4. Physikalisches Institut, Universität Stuttgart, Germany*

[2] *MPI für Festkörperforschung, Stuttgart, Germany*

[3] *NFS Nano-Scale Science and Engineering Center, University of California, Berkeley, USA*

In this work we perform third-harmonic generation (THG) spectroscopy in a hybrid plasmonic system consisting of a gold nanowire grating buried in a dielectric slab waveguide.

In these structures particle plasmon polaritons, which are optically excited in the wires, can be hybridized with photonic waveguide modes in the dielectric slab. By optimizing the structure geometry the spectrally broad particle plasmon resonance can exhibit an ultra-narrow and pronounced extinction dip.

We excite this hybrid plasmonic system with 150 fs laser pulses which can be spectrally tuned over the modulated plasmonic resonance and measure the generated third-harmonic light. The experiments show that it is insufficient to deduce the shape of the THG spectrum from the linear extinction. It is rather essential to consider the full information of the linear response, i.e., amplitude and phase. We find indications that the increased group index and the associated slow light around the extinction dip leads to an increase in the THG signal.

Optical properties of metallic nanostructures: plasmons, apertures and molecules

Cyriaque Genet, Thomas Ebbesen

ISIS - University of Strasbourg and CNRS, 8 allée Monge, 67000 Strasbourg, France

Nanostructures milled through metal films have revealed this last decade unique and surprising optical properties. On such structures, a key point is the possibility to excite surface plasmons. Tailoring these excitations through controlled design of metallic surfaces is an efficient way to induce original optical signatures.

Tip-enhanced ultrafast spectroscopy and microscopy of organic solar cell blend film

Xiao Wang[1], Anke Horneber[1], Josip Mihaljevic[1], Kai Braun[1], Hans-Joachim Egelhaaf[2], Christoph J. Brabec[2], Dai Zhang[1], Alfred J. Meixner[1]

[1]*Institute of Physical and Theoretical Chemistry, Auf der Morgenstelle 8, University of Tübingen, 72076 Tübingen, Germany*

[2]*Konarka Technologies GmbH, Landgrabenstr. 94, 90443 Nürnberg, Germany*

Poly(3-hexylthiophene) (P3HT) and [6, 6]-penyl-C61 butyric acid methyl ester (PCBM) are widely employed in the field of organic solar cells as the electronic donors and acceptors. The exciton creation, dissociation and separated charge transportation in the P3HT and PCBM blends are critical for achieving a high performance of the solar cells. To investigate the excition diffusion and dissociation, both high spatial resolution and ultrafast optical techniques are required. We studied the local chemical composition and photo physics of the blends on a length of a few nanometers using tip-enhanced spectroscopic mapping with the continuous wave laser [1, 2]. We discuss the correlations among the blend film morphology, the local P3HT:PCBM molecular distribution and the P3HT photoluminescence quenching efficiency based on the simultaneously recorded morphology and spectroscopic information. We will report about our progress in combining our parabolic mirror assisted tip-enhanced near-field optical microscope with an ultrafast laser system and report about nonlinear excitation behavior of inverted tip antennae. In addition, we will show new results on finite-difference time-domain (FDTD) simulations for different tip-sample distances in order to understand the interaction of the tip antenna with the sample materials.

[1] X. Wang et al, Advanced Functional Materials 20, 492 (2010).

[2] D. Zhang et al, Physical Review Letters 104, 056601 (2010).

Low-work function field emission tips triggered by an ultrafast laser

Seth M. Foreman[1], Catherine Kealhofer[1], Mark A. Kasevich[1]

Physics Department, Stanford University, 382 Via Pueblo Mall, Stanford, CA 94305, USA

We present the results of our experiments with triggering 100-nm radius Hafnium-Carbide field emission tips by a 150-MHz train of 10-fs, near-infrared, nano-Joule optical pulses. Due to the low work function of 310-oriented HfC (~3.4 eV) and a small DC field applied to the tip, two-photon over-the-barrier emission is possible for nearly all wavelengths contributing to the laser's spectrum. Such an emission process is expected to generate electron pulses of duration similar to that of the triggering optical pulses.

Two emission regimes are observed: for a wide range of low DC biases or low laser intensities, electron pulses leave the tip with a total current scaling as the optical intensity squared, consistent with two-photon over-the-barrier emission. For larger DC biases at higher laser intensities, the current contains no periodic RF frequency content and its nonlinearity becomes extreme with respect to the optical intensity. We postulate the tip is excessively heating due to its thermal time constant being much longer than the time between optical pulses. The thermal field emission current then dominates any pulsed emission. We present an experimental technique for inferring the average temperature of the tip by measuring the fraction of the total current that is pulsed, as a function of DC bias. The crossover operating point (DC bias and pulse power) where total thermal emission begins to exceed total pulsed emission is extended to higher pulsed currents by reducing the spot size of the focused laser.

We will also present preliminary work toward generating a point source of x-rays in both space and time using our optically-triggered ultrafast electron point source.

Optical Control of Field-Emission Sites by Femtosecond Laser Pulses

Hirofumi Yanagisawa, Christian Hafner, Patrik Dona, Martin Klöckner, Dominik Leuenberger, Thomas Greber, Matthias Hengsberger, Jürg Osterwalder

[1]*Physik Institut, Universität Zürich, Winterthurerstrasse 190, CH-8057 Zürich, Switzerland*

[2]*Laboratory for Electromagnetic Fields and Microwave Electronics, Swiss Federal Institute of Technology, CH-8092 Zürich, Switzerland*

We have investigated electron emission patterns induced by femtosecond laser pulses from a clean tungsten tip apex which is oriented along the [011] crystal direction, and compared them with those of field-emitted electrons without laser excitation. The laser light was focused to 4 mm onto the tip apex by a lens, and emitted electrons were detected by a position sensitive detector. We observed a striking difference in intensity distributions of the two patterns. Without laser, we observed the typical field emission pattern of a clean W tip; the intensity distributions reflect the local work functions on individual crystal facets. With laser, emission sites were the same as those without laser, but the intensity distributions become strongly asymmetric with respect to the shadow and exposed sides of the tip to the laser pulse. Strongly asymmetric modulations of the field emission intensity distributions are also observed depending on the polarization of the light and the laser incidence direction relative to the azimuthal orientation of tip apex [1, 2]. In effect, we have realized an ultrafast pulsed field-emission source with site selectivity on the scale of a few tens of nanometers. Simulations of local fields on the tip apex and of electron emission patterns based on photo-excited nonequilibrium electron distributions explain our observations quantitatively [1, 2].

[1] H. Yanagisawa et. al., Phys. Rev. Lett. **103**, 257603 (2009).

[2] H. Yanagisawa et. al., Phys. Rev. B **81**, 115429 (2010).

Strong-field photoelectron emission from metal nanotips

Max Gulde, Reiner Bormann, Alexander Weismann, Sergey Yalunin, Claus Ropers

Courant Research Center Nano-Spectroscopy and X-Ray Imaging, Friedrich-Hund-Platz 1, 37077 Göttingen, Germany

The generation of ultrashort, localized electron pulses is of fundamental interest for future applications in time-resolved electron imaging and diffraction. Femtosecond electron sources of great spatial coherence make use of a combination of local field enhancement at metal nanotips and nonlinear photoelectric effects. Previous studies have resulted in a controversial debate about the underlying physical processes.

Here, we present our most recent theoretical and experimental results regarding ultrafast photoelectron emission from nanometric gold tips. For the first time, we conclusively show the transition between the multiphoton and the optical field emission (i.e. tunneling) regimes. Direct evidence for this transition is found from both the power dependence of the total current and the spatial characteristics of the resulting electron beam. The results are supported by theoretical modeling.

Nanoscale coherent control: single molecules and optical antennas

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We aim for complete control of single photon emitters at the nanoscale. This entails tailoring the properties of the excitation light in far and near field; targeted state preparation in single quantum systems; and control of the temporal, spatial and polarization characteristics of the emission of a nano-system.

Coupling single molecules to nano-antennas is of particular interest: the antennas will confine excitation fields to far below the diffraction limit and offer exciting possibilities to influence the femtosecond dynamics of the near field. A single molecule on the other hand is an excellent probe for changes in the local density of states, which will be expressed in changes in its fluorescence lifetime, emission spectrum, k-spectrum and polarization. The coupled system will display its own distinct dynamics and can be described as a new quantum system.

In this talk, I will focus on two advances that our group recently made in molecule and antenna research. On the one hand, we probe the femtosecond dynamics of single molecules in dense polymer environments using femtosecond pulses and control the state in which they are prepared. On the other hand, we engineer the phase characteristics of fields confined to nanometric volumes with asymmetrc optical antennas and steer hotspots of intensity around the antenna with femtosecond time resolution.

Coherent excitation of single emitters with optical antennas and ultrashort laser pulses

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The enhancement of light emission with metal nanostructures has gained considerable attention in the recent years across a broad range of areas from biophotonics to quantum optics. Under continuous weak excitation, it is well established that the fluorescence signal is proportional to the field intensity and to the apparent quantum yield. The latter accounts for the competition between the radiative and non-radiative decay rates, which are strongly modified by the presence of an optical antenna. Extensive investigations have shown that huge field enhancements can coexist with large quantum efficiencies, making these systems appealing for quantum optical applications and single-molecule spectroscopy. On the other hand, time-resolved techniques, such as pump-probe spectroscopy, coherent control and triggering single-photon sources, to mention a few, rely on (ultrafast) pulsed excitation. The immediate question that arises is thus how an optical antenna affects the response of a molecule under laser pulses of various widths, at room and cryogenic temperatures. Here the important points of concern are the competition between decay times, dephasing and pulse width, the increased interaction strength due to field enhancements, and the dispersion occurring when the pulse duration becomes comparable with the lifetime of the antenna resonance. We have performed a detailed analysis of these phenomena and found that pulsed excitation discloses new challenges and opportunities for optical antennas. We present our findings in a context that ties ultrafast nano-optics with quantum-optics.

Enhancing photoluminescence of single self-assembled GaAs quantum dots using plasmonic nanoparticles

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The light emission properties of a single quantum system may be significantly modified by placing the emitter close to a nanostructure. This allows controlling, e.g., the excitation and emission rate as well as the emission pattern of the emitter. Plasmon resonant metal structures are a particularly interesting choice for the nanostructure as the electromagnetic field is significantly enhanced at the plasmon resonance wavelength. This offers exciting possibilities in both fundamental light-matter studies as well as in applications.

We experimentally investigate the influence of plasmon resonant nanoantennas on the photoluminescence properties of individual semiconductor quantum dots (QDs). As nanoantennas we use spherical gold nanoparticles. The quantum dots are epitaxially grown AlGaAs/GaAs QDs which are buried a few nanometers beneath the semiconductor surface and photoluminesce at approximately 760 nm wavelength. The advantage of this system is that the optical properties of the QDs are very stable and the transition dipole moments have a fixed orientation. The thin barrier layer allows efficient coupling between the quantum dot and a nanoparticle positioned above the emitter. We observe an increase in the photoluminescence of up to a factor of 6 due to the nanoantenna. Based on time resolved measurements and spectral analysis of the photoluminescence we attribute the enhancement in luminescence to an increase in the excitation rate of the quantum dot.

Linear Plasmonic Nano-Antennas: Experiment, Simulation, and Theory

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Understanding the intriguing interplay between the geometry of metallic nanoparticles and the field distribution of their localized surface plasmons eigenmodes can be achieved by theoretical modelling, simulation, or experiment. In this presentation, the focus is on the example of metallic nano-wire antennas, for which we are able to demonstrate all three approaches.

To measure their eigenmodes, we use apertureless Scanning Near-Field Optical Microscopy (aSNOM) with cross-polarized excitation and scattered radiation [1]. It allows the mapping of nearly unperturbed eigenmodes of plasmonic nanostructures [2-4]. We demonstrate how, from the combinatorial imaging of many different structures under virtually identical excitation conditions, important properties of the fabricated can be extracted, such as resonant geometry, effective propagation wavelength, or reflection phase. All our results are verified by extensive FDTD simulations. Finally we discuss our recently developed analytic model for the nano-wire antennas [5].

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[3] Nano Lett. 9, 2372–2377 (2009).

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[5] J. Dorfmüller, et al., submitted.

Deterministic control of nanooptical excitations in plasmonic nanostructures

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Theoretical investigations have demonstrated the capability of deterministic approaches to efficiently control ultrafast nanooptical light-matter interactions [1]. In this work we present experimentally deterministic control of nanooptical excitations in plasmonic nanostructures using ultrashort polarization-shaped laser pulses. Spatial distributions of near-fields in gold nanoprisms are probed by two-color two-photon photoemission microscopy. In adaptive optimizations the near-fields are localized at the corners of the nanoprisms. Deterministic control is realized by applying a pi-shift to the relative phase of two polarization components of adaptively optimized pulses resulting in a switching of the localized excitation.

[1] P. Tuchscherer *et al.*, Optics Express **17,** 14235 (2009)

Atomically flat single-crystalline gold nanostructures for plasmonic nanocircuitry

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Deep subwavelength integration of high-definition plasmonic nanostructures is of key importance for the development of future optical nanocircuitry for high-speed communication, quantum computation, and lab-on-a-chip applications. So far the experimental realization of proposed extended plasmonic networks consisting of multiple functional elements remains challenging, mainly due to the multi-crystallinity of commonly used thermally evaporated gold layers. Resulting structural imperfections in individual circuit elements will drastically reduce the yield of functional integrated nanocircuits. Here we demonstrate the use of very large (>100 μm^2) but thin (<80 nm) chemically grown single-crystalline gold flakes, which, after immobilization, serve as an ideal basis for focused-ion beam milling and other top-down nanofabrication techniques on any desired substrate. Using this methodology we obtain high-definition ultrasmooth gold nanostructures with superior optical properties and reproducible nano-sized features over micrometer length scales. Our approach overcomes the current fabrication bottleneck and opens the road for the realization of extended plasmonic circuitry.

[1] J.-S. Huang, V. Callegari, P. Geisler, C. Brüning, J. Kern, J.C. Prangsma, P. Weinmann, M. Kamp, A. Forchel, P. Biagioni, U. Sennhauser and B. Hecht, Atomically flat single-crystalline gold nanostructures for plasmonic nanocircuitry, arXiv:1004.1961 (2010).

Recent progress in two-photon-polymerization-based ultra-fast nanooptics applications

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There has been a lot of progress recently in new fabrication methods to create nanostructured media from polymers and metals, including the precise placement of nanoparticles. These methods allow the creation of new compound materials and components of increased complexity with a wide range of applications. A large part of such applications lie in the field of ultrafast nanooptics, due to a combination of miniaturization and incorporation of broad-band functionalities into nanodevices.

Controlling light at the nanoscale with different types of plasmonic antennas

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Over the last century, numerous antenna structures have been developed in electrical engineering to help the radiation of electromagnetic waves at microwave frequencies. These structures include for example dipole, bowtie and patch antennas. Each structure has its advantages and limitations. For example the dipole antenna is narrow-band, while the bowtie antenna has a broad-band response.

The recent advances in nanofabrication techniques have enabled the realization of similar structures that now operate at optical frequencies and rely on the excitation of plasmonic modes in the different components of the structure. Taking advantage of plasmon resonances in metals enables the excitation of extremely strong fields within the antenna, a phenomenon that does not have a classical equivalent at microwave frequencies. In this presentation, I will concentrate on a few types of nanoscopic antennas and show how they can be used to selectively enhance specific parts of the Raman spectrum from molecules deposited on them. I will further demonstrate experimentally that the strong field gradients created in the gap of the antenna create optical forces that can be used to trap particles as small as 10nm inside the gap. This ultimate trapping at the nanoscale may provide useful means to bring analyte in regions where the field is very strong. Finally, I will show that the combination of localized and delocalized plasmon resonances in the same structure can produce a narrowing of the spectral response of the system and decouple the different components that build up the system by reducing their crosstalks. This is of interest to realize a very strong and localized field enhancement within a compact system.

Few-Cycle Nonlinear Optics with Single Plasmonic Nanoantennas

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We present intense coherent emission of visible light from single gold nanoantennas driven with few-cycle laser pulses in the near-infrared [1]. A femtosecond Er:fiber system delivers pulses of a duration of 7.8 fs with a spectral bandwidth extending from 0.85 to 1.4 eV. Gold nanostructures are fabricated via electron-beam lithography or colloidal nanomasks [2]. We observe strong third harmonic generation (THG) together with a weak second harmonic signal (SHG) and two-photon photoluminescence (TPPL). We find a cubic and quadratic dependence of the emission intensity on excitation power for the THG and SHG, respectively. The origin of the different nonlinear emission processes lies in the band structure of bulk gold. If the excitation bandwidth is restricted to photon energies below 1.1 eV, the plasmon oscillation is resonantly driven and the third-order emission dominates. In contrast, excitation above 1.1 eV favors resonant two-photon transitions between the sp- and d-bands in gold. In this case SHG and TPPL emission occurs. The third-harmonic emission allows for precise detection of frequency-resolved interferometric autocorrelation traces of individual optical antennas. A sub-cycle plasmon dephasing time as short as 2 fs is measured directly in the time domain, highlighting the ultrastrong radiation coupling and ultrabroadband response of these efficient nanodevices. We will also present our latest results concerning spatially resolved emission patterns and measurements of shape-dependent dephasing times.

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Photon modes and second harmonic generation in ZnO nano-needle arrays

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Localization of visible light is a fascinating topic in modern physics. Not only is it of fundamental interest in itself, but the resulting strong electromagnetic field enhancements concentrated to dimensions below the diffraction limit allow for novel ultrafast, non-linear, nano-optical experiments and applications.

Weak localization and strong light-matter interaction can be achieved in disordered dielectric materials. In recent experiments, some of the present authors illuminated a densely packed array of ZnO nanorods fabricated by metal-organic vapor phase epitaxy. Ultrashort (6-fs) laser pulses around 800 nm are focused onto the sample with a Cassegrain objective. Pronounced spatial fluctuations of the resulting second-harmonic emission by more than an order of magnitude are seen.

In our calculations, we algorithmically create random spatial distributions of cylinders which are very similar to those seen in SEM images of the actual sample. These distributions are then used as input to a finite difference time-domain (FDTD) Maxwell solver. In our contribution, we present field distributions of photon modes obtained from 2-D calculations (i.e., cylinders extent infinitly in their axial direction) showing strong, localized field enhancements which we compare with the experimental results. Additionally, we show first results of full 3-D calculations of our complete model system (i.e., substrate layer, nano-needles, and air) which provide detailed insight in the dynamics and spatial distribution of the second harmonic generation.

Ultrafast nonlinear optical response of metal surfaces

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Our goal is the simulation of the nonlinear optical response of complex plasmonic nanostructures as, e.g., meta materials built from split ring resonators [1]. As shown by Rudnick and Stern [2], the motion of electrons perpendicular to the surface requires a quantum mechanical treatment due to nonlocal effects. As we aim to incorporate the ultrafast material response into a Maxwell solver, it is highly desirable to have a time-domain description of the electron dynamics at the surface. This is achieved by using the time-dependent density functional theory (TDDFT), which describes the time evolution of the charge density of a quantum mechanical many body system. The charge density along the normal direction of the metal surface is resolved on an Ångström scale [3] and propagated in time on a secondary grid [4]. The current density is extracted from the TDDFT simulation and incorporated into Maxwell's equations as a nonlinear source current. We use the finite difference time domain (FDTD) method for the self consistent inclusion of the light field dynamics.

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Ultrafast control of the large Rabi splitting in metal-J-aggregate hybrid structures

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The optical properties of hybrid nanostructures comprised of active materials, e.g., semiconductors or J-aggregated molecules, and metals are currently attracting substantial attention since they may form the basis for novel optoelectronic devices e.g. surface plasmon polariton (SPP) amplifiers, ultrafast optical switches or nanolasers as well as are highly interesting from a fundamental point of view. In favorable geometries, their optical properties are governed by a new class of short-lived quasiparticles, exciton – surface plasmon polaritons [1], with hitherto essentially unexplored nonequilibrium dynamics. Their binding energies, E_b, given by the spatial overlap integral of the excitonic transition dipole moment μ and the SPP electric field E_{spp}, can reach large values of several hundred meV. This implies that the optical properties of such strongly coupled hybrid structures can be drastically altered by externally manipulating either μ or E_{spp}.

Here, we study for the first time the effects of strong optical pumping on the optical properties of a strongly coupled hybrid nanostructure. A 50-nm-thick J-aggregated cyanine dye film deposited onto a gold grating, displaying strong exciton-SPP coupling with $E_b > 50$ meV is chosen. The effects of the ultrafast pumping on the dispersion relations of the coupled modes are mapped by recording transient angle-resolved low temperature differential reflectivity spectra. Strong optical switching of the sample reflectivity by more than 60% on a 100-fs-timescale and essentially complete external control over E_b, which can be reversibly switched between 0 and 50 meV within 250 fs, is demonstrated. We anticipate that such a pronounced ultrafast control of the exciton-SPP coupling is of considerable interest for different applications such as all-optical switching or SPP amplification.

[1] P. Vasa et al, Phys. Rev. Lett. **101**, 116801 (2008).

Simulation of the nonlinear response of split ring resonators with the Discontinuous Galerkin Time Domain method

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We report on our ongoing efforts to create a simulation environment that allows calculations of the linear and non-linear response of plasmonic nanostructures. In order to resolve the electronic and electrodynamic variations on and around the surface on different spatial scales we employ the Discontinuous Galerkin Time Domain (DGTD) method [1] utilizing its advantage in flexibility, higher order accuracy, and capability of handling non-linear problems. In DGTD the system of Maxwell equations is solved in the computational domain discretized into a number of conforming elements of arbitrary shape. The fields are locally expanded in terms of interpolating polynomials and the fluxes at interfaces are determined. Time integration is realized via the 5-stage 4th order Runge-Kutta Low Storage method. A Perfectly Matching Layer is applied as boundary condition. To describe a dispersive volume material we use the Drude model and include the Lorentz force non-linear term.

As an example, a U-shaped split ring resonator is illuminated with a linearly polarized short pulse leading to two polarization components of the transmitted signal. The presence of a nonlinearity leads to the doubling of the transmitted frequency in the spectrum and a peak of the 2nd harmonic appears in the perpendicular polarization component.

Currently, our simulations become unstable for long-time simulations. Nevertheless, the method shows its potential for studying multiscale non-linear effects. We plan to modify the computational scheme implementing stabilization filtering. Our long-term goal is a detailed description of both bulk and surface nonlinearities.

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Plasmon Hybridization Enhances the Nonlinear Response of Single Metal Nanoparticles

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The optical investigation of single metal nanoparticles is limited to rather large sizes due to their weak influence on focused laser radiation. Therefore it is very difficult to detect small dielectric variations, which is crucial for modern plasmonic nanosensors. We induce small, periodic variations of a nanoparticle's plasmonic properties by a heating pump puls that triggers acoustical breathing oscillations. The particle's response is monitored by a probe pulse. An optical nanoantenna increases the influence of these single dielectric objects on the laser focus. Such an antenna can be implemented by placing a bigger nanoparticle close to the smaller one that is probed.

We will show measurements of single metal nanoparticles' acoustic breathing modes as well as their first antenna enhanced detection. A model of the antenna-effect of plasmon hybridisation is presented. At the end, it allows us to analyze the individual nanomechanical properties of tiny single metal nanoparticles and study plasmonic coupling effects, without averaging over big ensembles.

Near-field measurements on nanoscopic sphere-on-plane systems by means of PEEM

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Surface immobilized gold nano-spheres above a gold surface with a gap distance in the order of a nanometer act as sphere-on-plane (SOP) optical resonator systems showing unique optical properties. In the vicinity of a flat metal surface, a second resonance band red-shifted with respect to the isolated particle resonance occurs, understandable on the basis of multipole interactions of the sphere with its own mirror image in the gold film. Several theoretical and optical far-field studies of SOP resonators were performed, investigating the dependence of the resonance wavelength on the resonator geometry and dielectric properties of the spacer material. The optical near-field of the system under fs-laser-radiation causes highly non-linear (n=3-5) photoemission processes strongly varying depending on the particle properties. Photoelectron microscopy turned out to be a valuable tool for the investigation of the optical near-field of individual SOPs. We present the gap-resonance-induced photoemission characteristics of several SOPs (individually characterized by SEM), excited by fs-laser radiation in the range from 750nm to 850nm.

Intrinsic and Extrinsic Nonlinearities of Nanooptical Systems

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The exploitation of nonlinear effects in nanooptical systems has many perspective applications, as, e.g., in novel light sources. To enhance the interaction of light with matter in such nanooptical systems, plasmonic resonances are often exploited where they may serve two distinctive purposes. On the one hand, plasmonic elements can be integrated into the device to enhance the nonlinear conversion efficiency. On the other hand, they are part of the primary effects an application relies on and the nonlinear effect is of supportive nature, e.g., to compensate for dissipation losses in plasmonic systems. In our contribution, which combines experimental and theoretical efforts, we describe our latest achievements in investigating such nonlinear effects; predominantly for localized plasmon polaritons. We explicitly distinguish between an intrinsic nonlinearity, i.e., the metallic constituents itself cause a nonlinear response, or an extrinsic nonlinearity, i.e., an additional nonlinear dielectric material integrated into the system dominates the nonlinear response. We describe our efforts to predict observable quantities theoretically on rigorous grounds using an extension to the Fourier Modal Method. The algorithm allows analyzing various nonlinear effects as long as an explicit expression for the nonlinear polarization can be provided. We compare the algorithm to approximate methods that take into account only the linear fields at the different frequencies involved.

Furthermore we describe our efforts to characterize experimentally the nonlinear response of fabricated samples comprising optical nanoantennas. Second and third harmonic generation depending on the frequency and the angle of incidence are quantified with the aim to reveal how linear resonances of the system affect the nonlinear spectrum. Experiments are compared to simulations.

We finalize our contribution with an outline on future problems, which we wish to solve in our project within the SPP.

Ultrafast adiabatic manipulation of slow light in photonic crystals

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Photonic crystals are spatial structures with a periodic modulation of the refractive index. Tuning the crystal's unit cell allows one to tailor light states with a group velocity of $c/30$ or less. Such slow light allows one to squeeze an optical pulse in a small volume, where it can be manipulated by an external stimulus of low energy [1]. In this respect, adiabatic schemes are very attractive since they offer reversibility and high conversion efficiency.

A physical system is said to evolve adiabatically when there is no energy transfer among the eigenstates of its instantaneous Hamiltonian. For example, when a vibrating guitar string is tuned slowly enough, additional harmonics are not excited [2]. Adiabatic dynamics usually rely on the external stimulus being much slower than the inverse spacing of the eigenfrequencies. Here, we show in experiment and theory that the color of a light pulse propagating in a photonic crystal waveguide can be changed adiabatically on a femtosecond time scale, even though the eigenfrequency spacing is arbitrarily small. This unexpected behavior occurs because the external stimulus respects the spatial symmetry of the system [3]. As the frequency of a 1.3-ps light pulse is shifted by more than 0.3THz with an efficiency of 80% in a 19-µm short waveguide, our scheme might be attractive for transferring signals between different frequency channels in future optical data networks [4]. We finally discuss how our approach can be used to accelerate an optical pulse by adiabatically increasing the group velocity of the light states involved.

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Second-harmonic on hole array generated in a long-lived resonance

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The mechanism behind second-harmonic generation from hole arrays was studied by illumination with phase shaped pulses. The measurements show that the SHG is generated in a long-lived resonance with a lifetime of 55 fs.

Time-resolved ultrafast photocurrent spectroscopy using THz stripline circuits on carbon nanotubes and graphene

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The dynamics of photo-generated charge carriers in nanoscale devices are typically detected in a time-resolved way by optical techniques such as the transient absorption technique and the time-resolved photoluminescence spectroscopy. Many questions remain concerning the separation and the transport of photo-generated charge carriers to source and drain leads. We address these questions by a novel ultrafast photocurrent spectroscopy, which is based on an optical pump-probe technique applied to co-planar striplines. The experimental setup with a picosecond time-resolution will be introduced, and first results of the time-resolved photocurrent will be shown. We will discuss polarization and charge separation effects within the carbon nanotubes and graphene as well as the influence of the contacts.

Surface Plasmon Coupling and Manipulation using Nonlinear PEEM

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Photoelectron emission microscopy (PEEM) has become an important tool to study the excitation and propagation of surface plasmon polaritons (SPPs). Surface plasmon polaritons are density oscillations of the electron gas in metals and propagate along the metal surface. The illumination of Ag-nanostructures with photons of less energy than the silver work function results in nonlinear, surface plasmon assisted photoemission.

The contrast in PEEM arises from the superposition of the electric field of laser and SPP, integrated over the detection time, i.e. the SPP can be observed in PEEM indirectly. The observed superposition pattern shows a periodicity that is directly related to the excitation geometry. The wavelength of this periodic pattern can be calculated theoretically using the moire-theory. The measurement of the k-vector of the PEEM pattern combined with the known k-vector of the laser pulses provides a possibility to determine the propagation direction of the SPP. This results in a Snell's law of refraction for the SPP excitation.

The photoemission yield decreases with increasing distance from the excitation site (here edge of the Ag-structure). Two mechanisms contribute to this behavior. During its propagation, the SPP dissipates energy through different channels such as dephasing, radiative decay and Landau-damping. The resulting decrease in the SPP wave amplitude reduces the photoemission yield. The second mechanism is the loss of the temporal overlap between the exciting light wave and the excited SPP wave as they propagate at different velocities along the surface of the Ag-structure. In this case there are less photons for the plasmon to absorb and hence the emission of electrons decreases.

Interaction of para-hexaphenylene based nanofibers with plasmonic substrates studied by phase resolved photoemission electron microcopy

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Para-hexaphenylene (p-6P) based nanofibers have been proven to support optical wave guiding down to wavelengths as short as 400 nm [1]. As a dielectric distortion on top of a metallic substrate, they have also been proposed to support the localized propagation of interface plasmon modes. P-6P nanofibers may therefore be useful as light-channeling sub-units and as highly localized plasmonic emitters in future ultrafast nanophotonic devices.

We deposited well aligned p-6P nanofibers onto microstructured gold films to investigate the interaction between optical waveguiding modes and interface plasmonic excitations. Photoemission electron microscopy was used to image the local near-field response of this plasmon-organic hybrid system excited by a femtosecond laser pulse. Located at the nanofibers, beating patterns exhibiting a high and reproducible periodicity are observed. Phase resolved interferometric measurements provide evidence that this pattern arises from localized plasmonic excitations at the fiber-gold-interface.

[1] F.Balzer et al., Appl. Phys. Lett. 82, 10 (2003)

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

#	Presenting Author	Title
1	*M. Kreiter*	Manipulating Near-Field Polarization beyond the diffraction limit
2	*C. Rewitz*	Analytic coherent control of plasmon propagation in nanostructures
3	*C. Strüber*	Nanolocalization of Ultrashort Time-Reversed Laser Pulses in Random Nanoparticle Assemblies
4	*K. Hinrichs*	Optimal spatio-temporal optical control of electron-induced processes at graphene-supported metal cluster nano-structures
5	*G. Piredda*	Near-field phase and flux controllability in the near-field
6	*E. Runge*	Surface plasmon polaritons on arrays of three-fold symmetric nanostructures
7	*C. Prohm*	Modeling the Linear and Nonlinaer Optical Response of Metallic Nanostructures in the Discontinuous Galerkin Time-Domain Method
8	*A. Schliwa*	Temperature dependent Q-factors in metallic-coated whispering-gallery microcavities
9	*A. Williamson*	Black silicon: Experimentally implemented and FDTD calculated results for photodiodes
10	*J. Shirdel*	Nonlocal, grating-coupled scattering-type near-field scanning optical microscopy of individual gold nano-particles
11	*P. Lisinetskaya*	Nonlinear optical response properties and ultrafast observables in silver nanostructures
12	*S.H. Chew*	Time- and Energy Resolved Photoelectron Emission Microscopy of Plasmonic Nanoantennas for Enhanced High Harmonic Generation
13	*A. Schmidt*	Bi-directional terahertz emission from gold coated nanogratings upon femtosecond laser irradiation
14	*V. Mondes*	Preparation of ordered metal nanostructures for high-harmonic generation by ultrafast plasmonic fields
15	*C. Schneider*	Surface plasmon observation using photoelectron emission microscopy

16	*F. Eilenberger*	Spatiotemporal characterization of ultrashort, nonlinear excitations in waveguide arrays
17	*P. Geisler*	Using PEEM to visualize the propagation of guided modes in plasmonic circuitry
18	*R. Bormann*	Strong-field photoelectron emission from metal nanotips
19	*X. Wang*	Tip-enhanced ultrafast spectroscopy and microscopy of organic solar cell blend film
20	*B. Schaaf*	Nano Antennas and their integration on a microstructured SNOM cantilever platform for Time Resolved investigations of Nanostructures
21	*J. Mihaljevic*	FDTD based calculations of the tip-induced near-field in a parabolic mirror microscope

Poster 1:

Manipulating Near-Field Polarization beyond the diffraction limit

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We introduce a new optical nanoantenna structure. The proposed nanoantenna is composed of three triangular gold nanoprisms which are assembled in a threefold symmetric configuration which share a gap. The antenna can produce a hot spot in the gap. Illumination by circularly polarized light produces a circularly polarized near field. The optical properties of the antenna are studied using 3D finite element method simulations. The length of triangles is varied to adjust the resonance frequency of the nanoantenna. The effects of gap size and shape of the triangles on the optical response of antenna are also studied. The optical response of the antenna is compared to two known antenna structures of the same type. The three-fold symmetric nanoantenna allows for full circular polarization in the gap and is significantly simpler than its four-arm counterpart as proposed by Biagoni, et. al. The simplicity of the new structure makes it more suitable for nanomanipulation of particles which have been wet-chemically synthesized.

Poster 2:

Analytic coherent control of plasmon propagation in nanostructures

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We present general analytic solutions for optical coherent control ofelectromagnetic energy propagation in plasmonic nanostructures. Tightly focused ultrashort laser pulses that are shaped in amplitude, phase, and polarization (ellipticity and orientation angle) are used to excite propagating modes on an example nanostructure consisting of a branching chain of nanospheres.Spatial localization and ultrafast switching of near-fields have a wide range of potential applications such as space-time resolved 'nano-femto' spectroscopy, quantum information processing, and nanoplasmonic devices.We decouple the interplay between two main mechanisms which are essential for the control of local near-fields. The first is a local interference of near-field modes which result from two laser pulse polarization components. Choosing the analytically computed amplitudes and phase difference of these two components, the linear flux is guided to a desired spatial position. The second control mechanism is a local pulse compression, where the temporal structure of a near-field can be manipulated at a desired spatial location. It is achieved at the target location by using the remaining free laser pulse parameter to flatten the local spectral phase.The resulting enhancement of nonlinear signals from this intuitive analytic two-step process is compared to and confirmed by the results of an iterative adaptive learning loop in which an evolutionary algorithm performs a global optimization. Thus, we gain detailed insight into why a certain complex laser pulse shape leads to a particular control target. This analytic approach may also be useful in a number of other coherent control scenarios.

Poster 3:

Nanolocalization of Ultrashort Time-Reversed Laser Pulses in Random Nanoparticle Assemblies

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The far field emission pattern of a nanoscale light emitter embedded in a nanoscale random scattering environment contains information about the localized emission. Because of the reciprocity of electromagnetic wave propagation time-reversing the outgoing wave from an emitter creates an excitation that propagates back and localizes again on a sub-diffraction limited length scale at the emitter. However, perfect nanolocalization requires the time-reversal of all outgoing wave components. We theoretically demonstrate improved nanolocalization of back-propagated waves if the emitter is embedded both in a local scattering plasmonic environment (a few tens of nanometers scale) with a strong near field coupling and in a reverberation "chamber" (a few microns scale) located in the far field of the local scattering cluster. The outgoing wave and the back-propagating plane wave are calculated in the frequency domain by self-consistently solving Maxwell's equations using a multiple scattering approach realized in the multiple elastic scattering of multipole expansions (MESME) code. To determine the degree of localization achieved by the time-reversed field we compare the spectral fluences deposited by the time-reversed field and by two separated perpendicular linearly-polarized bandwidth-limited pulses. The results demonstrate that the phase of a complex polarization-shaped laser pulse determines the localization of the radiation in a random scattering medium. Hence, the interference of different pathways of the multiply scattered incident radiation determines the local energy deposition. Furthermore, the achieved degree of localization depends on the random near-field scattering environment and the presence of a far-field reverberation chamber significantly facilitates nanolocalization.

Poster 4:

Optimal spatio-temporal optical control of electron-induced processes at graphene-supported metal cluster nano-structures

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An experimental setup is presented in which metal nano-cluster arrays consisting of graphene-supported size-selected silver clusters with extensions from a few atoms to the nanometer scale can be generated. Graphene is employed as a substrate to provide a geometric template, i.e., to define the cluster distances – its potential to also provide electronic coupling between the clusters will be a subject of investigation. Size-selected clusters will be employed to obtain perfectly monodisperse samples and to investigate the effect of the transition from discrete to plasmonic electronic cluster structure on the possibility of field localization.

The completely new ultra high vacuum apparatus futhermore enables the deposition of methyl halogenide molecules and the femtosecond-laser photodissociation of these adsorbates. The effect of the field localization will be probed by detecting the effectiveness of the electron-emission-induced dissociation of physisorbed adsorbate molecules (CH_3Br, CH_3Cl) on the metal clusters. Thus, the molecular fragment (CH_3) mass signal will be employed as feedback signal in the control loop to obtain the optimal electromagnetic field.

The investigation of the field localization will be performed by scanning tunneling microscopic detection of the spatial extent of the metal cluster halogenation as a function of the parameters cluster size and cluster separation.

In first experiments the cluster array formation on graphene via scanning tunnelling microscopy analysis is demonstrated. Furthermore, the photoinduced molecular dissociation of methyl bromide on supported metal clusters is investigated by femtosecond-laser pump-probe mass spectrometry.

Poster 5:

Near-field phase and flux controllability in the near-field

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Ultrafast nanooptics is an emerging field that combines the concepts and tools of ultrafast spectroscopy with those of near-field optics [1]. A basic form of coherent control in the near-field is the ability to concentrate the linear optical flux at a desired location with sub-diffraction resolution. This task requires polarization pulse shaping of the incident electric field [2].

We show, using simple examples, that the presence of quick spatial variations of the phase ("near-field phase") is an important ingredient for the controllability of linear flux in the near-field [3]. The near-field phase, that does not depend on propagation, is almost always present when light interacts with a nanostructure [4]. We compare to each other two similar nanoparticle and incident field configurations, in which near-field spatial variations of the relative phase between fields generated by orthogonal incident polarizations are present in different degrees (and are accompanied by varying degrees of spatial variations of the relative amplitude of the two interfering fields), to highlight the fact that without near-field phase variations control of flux localization is less effective.

In addition we present our first results of control of second-harmonic generation from rough gold films in a near-field optical setup.

[1] M.I. Stockman, S.V. Faleev, and D.J. Bergman; Coherent control of femtosecond energy localization in nanosystems; Phys. Rev. Lett., **88**, 067402 (2002).

[2] T. Brixner, F. J. García de Abajo, J. Schneider, C. Spindler, and W. Pfeiffer; Ultrafast adaptive optical near-field control; Phys. Rev. B, **73**, 125437 (2006).

[3] G. Piredda, C. Gollub, R. de Vivie-Riedle and A. Hartschuh; Controlling near-field optical intensities in metal nanoparticle systems by polarization pulse shaping; Applied Physics B, in press.

[4] R. Carminati; Phase properties of the optical near field; Phys. Rev. E **55**, R4901 (1997).

Poster 6:

Surface plasmon polaritons on arrays of three-fold symmetric nanostructures

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The influence of the geometry of three-fold symmetric (rotor-shaped) nanostructures on two dimensional quadratic arrays on the excitation of surface plasmon polaritons (SPPs) is studied numerically. We consider SPP related extrema of the far-field reflectivity $R(\alpha)$ as a function of the polarization angle α of the incident light. In support of recent experimental work, it is observed that these extrema shift. In particular they move away from $\alpha = 0$ and $\alpha = 9$, where one would expect them for rotationally invariant nanostructures. We also investigate the influence of details of the shape and the size of the nanostructures on the shifts. However, the polarization angle corresponding to the most efficient SPP excitation is found to be independent of the shape of the individual nanostructures. We discuss optical near- and far-field properties of the considered nanostructures.

Poster 7:

Modeling the Linear and Nonlinaer Optical Response of Metallic Nanostructures in the Discontinuous Galerkin Time-Domain Method

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DFG Forschungszentrum Center for Functional Nanostructures (CFN),
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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 8:

Temperature dependent Q-factors in metallic-coated whispering-gallery microcavities

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Metallic cavities enable light-confinement considerably smaller than the wavelength of light. The key figure of merit in this respect is the ratio of cavity quality factor, Q, to the cavity mode volume, V. For high-Q metallic-coated microresonators whispering gallery surface plasmon polaritons (SPP) achieve Q factors in the range above 1000 in experiment [1] at room temperature. One way to increase this value further, is the reduction of the high metal losses by significantly decreasing the temperature.

In this contribution we employ the Drude-Cricital-Point (DCP) model [2] to desribe the dielectric function of the metal coating. The temperature dependence is implemented by assuming a linear relationship between the damping coefficient of the Drude part of the DCP model and temperature. In this way a Kramers-Kronig consistent desription of the dielectric function is retained. The SPP and dielectric eigenmodes are calculated by solving the eigenmode problem in the frame of full vectorial finite elements analysis. High accuracy is ensured by using error-control guided adaptive finite elements up to order nine. Thus, the strongly compressed SPP modes are well resolved. The influence of disc-size, -geometry and choice of metal coating is discussed. Material dispersion and frequency of the eigenmodes are calculated self-consistently.

[1] B. Min et al. Nature 457, p455 (2009)

[2] P. Etchegoin et al. J.chem.Phys, 125, 164705 (2006)

Poster 9:

Black silicon: Experimentally implemented and FDTD calculated results for photodiodes

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Black silicon is a nano-structured silicon surface formed normally by a self-aligning, maskless process with structured surfaces often discernible by their black color and tremendous anti-reflective properties. The experimental results presented here will show directly the creation of black silicon through plasma processing and its subsequent implementation as an anti-reflective coating for photodiodes. Integrating sphere reflection measurements of physical structures are compared to FDTD calculations of modelled ideal structures and measured spectral sensitivities of black silicon photodiodes are discussed.

Poster 10:

Nonlocal, grating-coupled scattering-type near-field scanning optical microscopy of individual gold nano-particles

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Scattering-type near-field scanning optical microscopy (s-NSOM) is now routinely used for (sub-) 10-nm-resolution optical imaging of surfaces [1]. The performance is, however, often limited by a rather substantial signal background resulting from a direct optical illumination of the scattering antenna. Various ideas for a nonlocal optical excitation of the antenna apex have therefore been proposed, e.g., by grating-coupling of surface plasmon polaritons (SPP) onto adiabatic metallic tapers and three-dimensional focusing of SPP wavepackets towards the tip apex [2]. Recently, first line-scan images recorded by using such probes demonstrated 20 nm resolution and coupling of about 15% of the SPP intensity onto the tip apex [3].

Here, we use such a grating-coupled SPP microscope for the first time for s-NSOM imaging of single metallic nano-particles. We demonstrate sub-20-nm-resolution imaging of localized SPP fields and observe that more than 40% of the grating-coupled SPP field is localized at the taper apex. In order to show that such images do not simply reflect topographical artifacts, we recorded light scattering images from a larger, elliptical, gold nano-particle with 100 x 40 nm dimension. We find strong, localized intensity peaks at the edges of both the long and short axis of the nano-particle, in contrast to the regular shape of its topographical image. This indicates that the optical image essentially simply reflects the z-component of local electric field, as anticipated from the polarizability tensor of such a tip antenna. We anticipate that such a nonlocal s-NSOM will find a variety of applications in ultrahigh-resolution photoluminescence, absorption and Raman spectroscopy of nano-structures and progress along those directions will be discussed.

[1] see, e.g., D. Zhang et al, *Phys. Rev. Lett.* **104**, 056601 (2010) and references therein.

[2] C. Ropers et al. *Nano Letters* **7**, 2784 (2007).

[3] C. C. Neacsu et al, *Nano Letters* **10**, 592 (2010).

Poster 11:

Nonlinear optical response properties and ultrafast observables in silver nanostructures

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Our newly developed field-induced surface hopping method (FISH) represents a powerful tool for the simulation of dynamical observables such as time-resolved photoelectron spectra and harmonic emission signals. In the FISH approach, quantum electronic state population dynamics is combined with classical nuclear dynamics "on the fly" without precalculation of potential energy surfaces. The idea of the FISH method is to propagate independent trajectories in the manifold of adiabatic electronic states and allow them to switch the states under the influence of the laser field. Our approach allows for the direct inclusion of the laser fields in the dynamics simulations. It can be combined with the optimal control theory in order to steer dynamical processes in molecules and nanostructures by optimizing laser fields. The calculation of nonadiabatic couplings and transition dipole moments necessary for the FISH simulations has been implemented in the frame of the TDDFT method and its more efficient tight-binding version (TDDFTB).

Based on this approach, we have simulated nonlinear optical response properties and ultrafast observables in metallic nanoclusters. We show on the example of silver nanoclusters that the time-resolved photoelectron spectra and harmonic generation can be used to reveal the dynamical processes after photoexcitation. Furthermore, we demonstrate the first results on the optical properties of silver clusters and silver clusters at graphene. This represents a basis for combining the FISH method with the Maxwell Bloch approach for the simulation of light propagation in ordered arrays of metal nanoclusters at graphene. The presented methodological development will enable us to explore and control the light propagation in photonic nanostructures.

Poster 12:

Time- and Energy Resolved Photoelectron Emission Microscopy of Plasmonic Nanoantennas for Enhanced High Harmonic Generation

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Metallic bowtie nanostructures have recently been demonstrated to enhance high harmonic generation from noble gases, attributing to resonant plasmon field enhancement in the vicinity of nanostructures driven by femtosecond laser pulse excitation [1]. Two-photon photoelectron emission microscopy (2P-PEEM) is a powerful tool to image the near field enhancement and 'hot spot' photoemission from plasmonic nanostructures with a spatial resolution of ~20 nm. Besides imaging the spatial electron distribution, the kinetic energy distribution of the photoelectrons within the nanostructures can be mapped using a time-of-flight (TOF) energy analyzer utilizing a delayline detector coupled to the PEEM [2]. We will present the preliminary characterization results of the silver plasmonic nanoantennas (fabricated by electron beam lithography) by means of static 2P-TOF-PEEM using a 400 nm picosecond diode laser. However, our ultimate goal is to characterize the nanoplasmonic fields not only on a nm spatial scale but also on a ~100 as temporal scale. Therefore, the concept of an attosecond field microscope utilizing a TOF-PEEM in combination with femtosecond optical pump/attosecond XUV probe experiment will be discussed. As a first step toward this realization we also have characterized gold plasmonic nanoantennas using single attosecond XUV pulse excitation at ~90 eV photon energy for the first time.

[1] S. Kim et al., Nature 453, 575 (2008).

[2] J. Lin et al., J. Phys.: Condens. Matter 21, 314005 (2009)

Poster 13:

Bi-directional terahertz emission from gold coated nanogratings upon femtosecond laser irradiation

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We report on terahertz (THz) emission upon femtosecond laser irradiation (785 nm, 150 fs, 1 kHz, ≤1 mJ/pulse) of gold coated nanogratings in fused silica (500 nm period, 40 nm gold film). Previous experiments showed THz emission only in the case of rear side irradiation (through substrate) of the nanograting in the direction of laser propagation [1]. In contrast, we observed THz emission from this nanoemitter both in the direction of laser propagation and reverse in either case of rear side and front side irradiation (through air). A highly sensitive and fast superconducting transition edge sensor (TES) was used as THz detector.

The current model of THz generation is based on field enhancement due to laser excited surface plasmons. Electrons escaping the metal layer upon 3-4 photon absorption processes, experience ponderomotive forces in the exponentially decaying electric field of the surface plasmons and thus emit electromagnetic radiation [1]. Our experimental findings however, appear to be inconsistent with this model of THz emission.

Therefore, we propose another model, which explains the THz emission without the assumption of free electrons: Propagating surface plasmon polaritons constitute a transient current, which may directly emit THz radiation. Because of the small gold film thickness, plasmon polaritons on both surfaces strongly couple, resulting in simultaneous emissions from both sides of the grating. The lifetime of such surface plasmon polaritons can be calculated to about several picoseconds [2], which corresponds to the reported length of the THz pulse [1].

[1] G.H. Welsh, N.T. Hunt, and K. Wynne. Phys. Rev. Lett., 98:026803, 2007.

[2] F. Garwe, U. Bauerschäfer, A. Csaki, A. Steinbrück, K. Ritter, A. Bochmann, J. Bergmann, A. Weise, D. Akimov, G. Maubach, K. König, G. Hüttmann, W. Paa, J. Popp, and W. Fritzsche. *Nanotechnology*, 19:055207, 2008.

Poster 14:

Preparation of ordered metal nanostructures for high-harmonic generation by ultrafast plasmonic fields

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In high-harmonic generation (HHG) an atomic medium is exposed to a femtosecond laser pulse of sufficient intensity. The nonlinear HHG process leads to the formation of a comb of coherent XUV pulses of femtosecond to sub-femtosecond duration. Recently, it has been shown that the requirement of using high intensity laser pulses for the HHG process can be lowered by exploiting the local field enhancement induced by resonant plasmons within metallic nanostructures. Strong plasmonic enhancement effects are expected from ordered pattern of highly anisotropic noble metal nanoparticles. Though lithographic methods yield well-defined nanostructures of various morphologies in the micrometer range, the preparation of large arrays of ordered metallic structures by self-organization of nanoparticles appears to be favorable. This is because the nanoparticle approach yields structures with dimensions well below 100 nm on areas reaching up to square centimeters, which appears to be promising for the efficient generation of high-harmonics. Gold nanoparticles of different size and shape are prepared by colloidal chemistry. These are arranged in arrays of ordered structures by self-organization on sapphire substrates and characterized by scanning electron microscopy. The controlled distance between the nanoparticles is adjusted by variable size organic ligands or the application colloidal masks. Optimization of these structures is required in a subsequent step for an efficient enhancement of high-harmonics. The formation of XUV pulses using ordered nanostructures by plasmonic field enhancement required the construction of an XUV-monochromator for probing high harmonics in the XUV-regime. First experiments were performed using this setup, indicating that high-harmonics are formed in rare gases. Future experiments will focus on exploring systematically nanoparticle based arrays for the efficient enhancement of the HHG process in combination with short pulse laser radiation.

Poster 15:

Surface plasmon observation using photoelectron emission microscopy

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Photoelectron emission microscopy (PEEM) is a very powerful tool to map optical near fields with subwavelength spatial resolution. In combination with a stabilized interferometric setup, it is possible to study electron dynamics with attosecond temporal resolution.

The emerging research field of plasmonics combines the advantages of photonics and electronics. The study of collective electronic oscillations in metal nanoparticles (localized surface plasmons, LSP) or on metal/dielectric interfaces (surface plasmon polaritons, SPP) have the capacity to transport coherently energy on subwavelength length scales. As these structures show a huge nearfield enhancement, PEEM is a perfect instrument to study plasmonic behaviour simultaneously in time and space.

Gold stripes being excited with ultrashort laser pulses at 800 nm wavelength which exhibit a surface plasmon resonance will be presented on our poster. The interference between the propagating surface plasmon wave and the exciting laser pulse leads to a beating pattern visible in the PEEM image. We developed a fast and effective algorithm which is based on Huygen‘s principle to simulate this interference and to reconstruct the surface plasmon wave.

In addition, we performed interferometric measurements on these structures. This enables us to study the propagation of the SPPs along the gold surface.

Poster 16:

Spatiotemporal characterization of ultrashort, nonlinear excitations in waveguide arrays

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Optical fibers arrays are ideal systems to study the interplay of discreteness, periodicity, and nonlinearity [1]. One of the consequences of this interplay is the existence of Light-Bullets (LBs), [2] solitary wavepackets, which are nonlinearly self-confined in space and time, simultaneously.

We demonstrate, for the first time, conclusive experimental evidence of LBs in a fibre array, with an unpreceeded degree of regularity [3]. The demonstration is based on high resolution mapping of the ultrashort wavepackets by means of a 3D spatio-temporal cross-correlation imaging technique [4] and thorough numerical calculations.

Based on the agreement between numerical and experimental data we developed a model for exitation and evolution of a LB under the influence of higher order effects, such as the wavelength dependence of the discrete diffration, higher order dispersion and the Ramam-induced red-shift. We find that the interplay of these effects leads to an adiabatic evolution of the LB, where the LB threshold energy eventually grows higher than the LB energy itself, leading to decay.

We argue that this scenario is of very general nature for LBs, thus imposing a fundamental limit on their lifetime.

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Poster 17:

Using PEEM to visualize the propagation of guided modes in plasmonic circuitry

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Subwavelength nano-optical circuitry based on two-wire transmission lines is of great interest for the realization of ultrafast optical communication and data processing. This is because two-wire transmission lines offer a large degree of intensity confinement between the two wires which may be exploited to efficiently induce nonlinear effects. Furthermore, concepts of radiowave technology can be directly transferred to optical frequencies.

A drawback of the large confinement is the decreased propagation length, which makes it difficult to directly observe the propagation of optical signals in such waveguides. A direct observation of propagating fields and/or standing waves would allow characterizing the quality of the waveguides as well as attached components, such as optical antennas based on their impedance mismatch. In this contribution we show first results of PEEM measurements applied to nano-optical circuits based on two-wire transmission lines.

Poster 18:

Strong-field photoelectron emission from metal nanotips

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The generation of ultrashort, localized electron pulses is of fundamental interest for future applications in time-resolved electron imaging and diffraction. Femtosecond electron sources of great spatial coherence make use of a combination of local field enhancement at metal nanotips and nonlinear photoelectric effects. Previous studies have resulted in a controversial debate about the underlying physical processes.

Here, we present our most recent theoretical and experimental results regarding ultrafast photoelectron emission from nanometric gold tips. For the first time, we conclusively show the transition between the multiphoton and the optical field emission (i.e. tunneling) regimes. Direct evidence for this transition is found from both the power dependence of the total current and the spatial characteristics of the resulting electron beam. The results are supported by theoretical modeling.

Poster 19:

Tip-enhanced ultrafast spectroscopy and microscopy of organic solar cell blend film

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Poly(3-hexylthiophene) (P3HT) and [6, 6]-penyl-C61 butyric acid methyl ester (PCBM) are widely employed in the field of organic solar cells as the electronic donors and acceptors. The exciton creation, dissociation and separated charge transportation in the P3HT and PCBM blends are critical for achieving a high performance of the solar cells. To investigate the excition diffusion and dissociation, both high spatial resolution and ultrafast optical techniques are required. We studied the local chemical composition and photo physics of the blends on a length of a few nanometers using tip-enhanced spectroscopic mapping with the continuous wave laser [1, 2]. We discuss the correlations among the blend film morphology, the local P3HT:PCBM molecular distribution and the P3HT photoluminescence quenching efficiency based on the simultaneously recorded morphology and spectroscopic information. We will report about our progress in combining our parabolic mirror assisted tip-enhanced near-field optical microscope with an ultrafast laser system and report about nonlinear excitation behavior of inverted tip antennae. In addition, we will show new results on finite-difference time-domain (FDTD) simulations for different tip-sample distances in order to understand the interaction of the tip antenna with the sample materials.

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Poster 20:

Nano Antennas and their integration on a microstructured SNOM cantilever platform for Time Resolved investigations of Nanostructures

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Scanning Nearfield Optical Microscopy (SNOM) probes are presented. The polarization dependence and spectral resonance behavior of c-shape antennas in the far-field are reported and compared to simulations of the field distribution. Bowtie antennas on ITO/glass substrates were structured by e-beam lithography and their spectral resonance in the far-field was measured. As a platform for nano antennas, SNOM cantilevers were flattened by ion beam etching. The integration onto the flattened apex necessitates a homogenous PMMA layer applied by spray-coating. The probes aim at the investigation of, e.g., collective electron oscillations like localized surface plasmons (LSPs).

Poster 21:

FDTD based calculations of the tip-induced near-field in a parabolic mirror microscope

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The optical properties of molecules can be dramatically altered when they are in a close proximity of an excited metal antenna. In order to get insight into how the antenna generated near-field influences the optical properties of low quantum yield systems, we carried out FDTD simulations of a sharp laser-illuminated Au tip approaching to different substrates, such as semiconductor thin film, gold, silicon and glass. The source field was modelled according to the focal field which is achieved by a parabolic mirror as the focusing element in our experimental setup. The time-averaged field distributions in the gap between the different materials and the tip antenna were calculated regarding to different distances.

Our calculation for the diindenopyrelene (DIP)-film demonstrated that the coupling between the localized plasmon at the tip apex and the semiconductor polariton in the thin film can be achieved building up a distance-dependent high field enhancement. In this context, such a high field strength enhances not only the excitation process of DIP thin film by a factor of 10^4, but alters the ratio of radiative to non-radiative decay rate giving approx. 15 times stronger photoluminescence emission.

Our current aim is to model the P3HT/PCBM solar cell system corresponding to our experimental framework. The simulation results would assist us in interpreting and understanding of the possible interactions between the scanning tip and solar cell hybrid blend film.

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