O 13: Symposium: Ultrafast Nanooptics II

Time: Monday 14:15–18:00 Location: H38

Invited Talk O 13.1 Mon 14:15 H38

Nanoscale optical spectroscopy based on local field enhancement — •Achim Hartschuh[1], Huihong Qian[1], Tobias Gokus[1], Neil Anderson[2], and Lukas Novotny[2] — [1]Department Chemie und Biochemie and CeNS, LMU Muenchen, Germany — [2]The Institute of Optics, University of Rochester, USA

Spectroscopic techniques with nanoscale spatial resolution are essential for the detection and analysis of individual nanoobjects and nanostructured materials. We study and apply a near-field optical technique that is based on local field enhancement at the apex of a laser-illuminated metal tip. In this scheme, locally enhanced excitation and radiation fields in close proximity to the near-field probe are used to amplify the optical response of the sample. At present, using monochromatic cw-excitation, we achieve a spatial resolution of about 10 nm for both photoluminescence and Raman scattering of carbon nanotubes [1, 2]. Different contributions to the signal enhancement of photoluminescence and Raman scattering are evaluated based on experimental data achieved simultaneously for the same nanotube. The high-resolution capability of the technique is accompanied by an enhanced detection sensitivity making it ideally suited for nanoscale surface and subsurface imaging [3]. We discuss strategies for the advancement of the technique and its combination with ultrafast laser spectroscopy.

[1] A. Hartschuh et. al, Phys. Rev. Lett. 90, 095503 (2003). [2] A. Hartschuh et. al, Nano Lett. 5, 2310 (2005). [3] N. Anderson, Nano Lett. 6, 744 (2006).

Invited Talk O 13.2 Mon 14:45 H38

Coherent, Nonlinear, and Ultrafast Nanoplasmonics — •Mark Stockman — Department of Physics and Astronomy, Georgia State University, Atlanta, USA

This talk introduces and reviews recent new ideas and progress in coherent, nonlinear and ultrafast nanoplasmonics. It includes a brief Introduction to the topic and forefront, focus areas based partially on original contributions, including ultrafast, coherent, nonlinear, and stimulated phenomena. Spaser will be one of the focus points of the talk as an ultrafast generator of local optical field on nanoscale. We also consider dynamic, controllable, ultrafast localization of optical energy on the nanoscale and nonlinear photoelectron emission coherently controlled by the phases of the ultrashort excitation pulses. Finally, we consider a recent development based on the combination of the adiabatic concentration of optical energy on nanoscale and spatio-temporal modulation of surface plasmon polaritons.

Invited Talk O 13.3 Mon 15:15 H38

Attosecond resolved photoemission from metal surfaces — •Ulrich Heinzmann — University of Bielefeld, Faculty of Physics

The combination of the comb of high harmonics of a phase-stabilized ultrashort laser pulse focussed into a rare gas atomic beam[1] with an optimized multilayer mirror system as soft x-ray monochromator[2] yields isolated soft x-ray pulses of 250 and less attoseconds duration[3]. Thus photoelectron and Auger electron emission processes in atoms have been studied time resolved with a sub-fs time resolution[4]. The report discusses the efforts to apply these techniques in attosecond time resolved photoemission of condensed matter. It presents very first results of attosecond resolved photoelectron emission spectroscopy from a single crystal[5] and discusses what can be learned about the electronic dynamics in the bands of the surface and of the few atomic bulk layers the photoelectrons come from.

[1] R. Kienberger et al. Nature **427**, 817 (2004)
[2] A. Wonisch et al. Applied Optics **45**, 4147 (2006)
[3] E. Goulielmakis et al. Science **305**, 1267 (2004)
[4] M. Drescher et al. Nature **419**, 803 (2002)
[5] A. Cavalieri et al. to be published (2007)

30 min Break

O 13.4 Mon 16:15 H38

Ultrafast Spectroscopy of Single Gold Nanoparticles — •Markus Lippitz[1,2,3], Meindert van Dijk[1], and Michel Orrit[1] — [1]Huygens Laboratory, University of Leiden, The Netherlands — [2]4. Physics Institute, University of Stuttgart — [3]Max Planck Institute for Solid State Research, Stuttgart

Metal nanoparticles in the size of 1-100 nm have properties different from bulk metal. The particle size distribution present in even the best chemical preparation methods blurs the picture one can get. Optical spectroscopy of a single particle at a time removes the sample inhomogeneity and gives direct access to the individual particle's properties. We present nonlinear optical experiments on single gold nanoparticles.

We reported the first observation of third-harmonic signals from individual gold particles. Excited with short pulses (100 fs) at 1600 nm, the particles generate 533-nm light, close to the plasmon resonance. Contrary to our expectation, we found the third-harmonic intensity to vary as the fourth power of the diameter of the particles, which can be explained by the nonlinear optical response of the free electrons.

In another experiment we performed time-resolved absorption measurements of single gold nanoparticles, with a sensitive common-path interferometer. The variation of the plasmon resonance after absorption of a near-infrared pump pulse is probed in the visible wavelength range. At short times, the signal of the hot electrons dominates. At longer delay times, mechanical oscillations of the whole particle can be detected by periodic shifts of the plasmon frequency.

O 13.5 Mon 16:30 H38

Electronic properties of silver nanoclusters on oxide supports: Fluorescence microscopy and femtosecond two photon photoemission — Tobias Gleitsmann, Mihai E. Vaida, and •Thorsten M. Bernhardt — Institut für Oberflächenchemie und Katalyse, Universität Ulm, 89069 Ulm, Germany

In the quest for efficiently fluorescent nano-scale materials atomic silver clusters have attracted considerable interest in recent years. Possible applications are in all optical logic devices and in advanced optical data storage media. Unexpectedly, the observed optical properties of silver clusters in the size range with up to about 20 atoms per particle indicate that these clusters cannot yet be considered fully metallic. On the contrary, the observed fluorescence requires the presence of long-lived discrete electronic levels in the cluster-support system. Employing a laser scanning microscope and femtosecond radiation it is possible to write fluorescent silver cluster nano-structures in silver oxide thin films or silver oxide containing biopolymer matrices. The information is long-time stable and can be read out with conventional low intensity light sources. In order to furthermore identify the hitherto unknown electronic structure of atomic metal clusters at surfaces and thus to elucidate the origin of the observed luminescence properties time-resolved two photon photoemission spectroscopy is applied to mass-selected silver metal clusters which have been soft-landed onto a well-defined insulating substrate.

O 13.6 Mon 16:45 H38

Influence of the interband transition on the optical properties of gold nanoparticles — •Frank Hubenthal, Nils Borg, Christian Hendrich, and Frank Träger — Universität Kassel, Institut für Physik and Center for Interdisciplinary Nanostructure Science and Technology, Heinrich-Plett-Str. 40, 34132 Kassel, Germany

We present systematic measurements of the ultrafast dephasing time T_2 of surface plasmon resonances (SPRs) in gold nanoparticles (NPs) as a function of photon energy. The objective is to investigate the influence of the interband transition on the damping of the SPR, typically described by the damping parameter A. Gold NPs were prepared by deposition of thermal atoms followed by subsequent diffusion and nucleation, i.e. Volmer-Weber-growth. Thereafter, systematic measurements of T_2 were carried out by persistent spectral hole burning. The most essential among the numerous results is the observation of an increased damping of the SPR at the onset of the interband transition. While the damping parameter A amounts to 0.20 nm/fs for photon energies below the onset of the interband transition at 1.85 eV, the damping increases to $A = 0.32$ nm/fs at 1.85 eV. For higher photon energies the contribution of the interband transition becomes the dominant damping mechanism, resulting in a reduction of A to as little as 0.15 nm/fs. The high A-Factor at 1.85 eV cannot be explained semiclassically by taking only the surface scattering of the oscillating electrons into account. In fact, we observe an increased Landau damping in Au NPs at the onset of the interband transition, which has not

been identified before for all other photon energies.

O 13.7 Mon 17:00 H38

Adaptive sub-wavelength control of nano-optical fields — Martin Aeschlimann[1], Michael Bauer[2], Daniela Bayer[1], •Tobias Brixner[3], F. Javier García de Abajo[4], Walter Pfeiffer[5], Martin Rohmer[1], Christian Spindler[3], and Felix Steeb[1] — [1]Fachbereich Physik, Technische Universität Kaiserslautern, Erwin-Schrödinger-Str. 46, 67663 Kaiserslautern, Germany — [2]Institut für Experimentelle und Angewandte Physik, Universität Kiel, Leibnizstr. 19, 24118 Kiel, Germany — [3]Physikalisches Institut, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [4]Instituto de Optica, Serrano 121, 28006 Madrid, Spain — [5]Fakultät für Physik, Universität Bielefeld, Universitätsstr. 25, 33516 Bielefeld, Germany

We combine two previously separated research fields, adaptive quantum control and nano-optics, to achieve dynamic localization of electromagnetic intensity at sub-wavelength nanoscopic spatial resolution. This is demonstrated experimentally with femtosecond polarization shaping and photoemission electron microscopy (PEEM). The electric near field in the vicinity of Ag nanostructures is controlled by employing femtosecond laser pulses with specifically modulated vectorial properties. Two-photon PEEM probes the lateral intensity distribution, and a learning algorithm optimizes the external field such that photoemission is localized in the desired places. Thus pulse shaping and quantum-control concepts are now also available with nanoscale resolution.

O 13.8 Mon 17:15 H38

Ultrafast polariton dynamics in metallic photonic crystal superlattices — •Tobias Utikal[1], Thomas Zentgraf[1,2], Jürgen Kuhl[2], Sergei Tikhodeev[3], and Harald Giessen[1] — [1]4. Physikalisches Institut, Universität Stuttgart, Germany — [2]MPI für Festkörperforschung, Stuttgart, Germany — [3]General Physics Institute, Moscow, Russia

We present time- and frequency-resolved nonlinear autocorrelation measurements of gold nanowire photonic crystal superlattices on a dielectric waveguide layer. In these structures an external light field can excite electronic and photonic modes at the same time. A strong coupling of the two resonances leads to the formation of a polaritonic state which can further be modified by introducing a periodically structured unit-cell into the lattice. The frequency resolved measurements show a rather complex behaviour of the third harmonic signal. It consists of several spectral components, which differ in intensity and time dynamics. To describe the nonlinear processes we use a simple model, in which each resonance in the fundamental extinction spectrum is considered as an eigenmode of the polaritonic system and can be described as a Lorentzian oscillator with a certain amplitude and lifetime. The resonance frequencies of these eigenmodes can mix in the nonlinear regime, leading to third harmonic and sum frequency generation. Within this model it is possible to relate the spectral components in the higher harmonic signal to the different third harmonic and sum frequency processes. We find dephasing times of the different polariton modes which range from 10 up to 60 fs.

O 13.9 Mon 17:30 H38

Plasmon Enhanced Photoemission Microscopy — •Frank Meyer zu Heringdorf and Liviu Chelaru — Universität Duisburg-Essen, Lotharstrasse 1, 47048 Duisburg

Excitation and propagation of light in metallic nanostructures by means of plasmons is one of the most promising techniques for scaling down photonic devices to their ultimate limit. This perspective has created a tremendous interest in the interaction of light with metallic particles. Two Photon Photoemission Microscopy with femtosecond laser pulses is particularly well suited to study plasmons in small metallic nanostructures, as the existence of a plasmon enhances the nonlinear photoemission yield and provides for direct visualization of the plasmon. We use the self-organization of Ag islands and wires on various Si surfaces to form structures that act as waveguides for surface plasmon polariton waves. Such waves travel along the surface of the wires and form an interference pattern with the exciting light. Pump-Probe Microscopy allows observation of the time-dependent beating pattern of the plasmon wave that is excited by a first laser pulse and probed by a second laser pulse.

O 13.10 Mon 17:45 H38

Quasi Particles as Quantum Phenomena at Surfaces — •Dominic Zerulla, Brian Ashall, and Michael Berndt — UCD Dublin, School of Physics, Dublin 4, Ireland

Surface Plasmon Polaritons (SPP's) are mixed states between photons and electron density waves propagating along the surface of a conductor. While the principle excitation of such phenomena is well understood the focus of this talk is on the detailed properties of these quasi-particles, such as localization, lifetime, causality and re-emission characteristics. The localization of surface plasmons can be used to tailor the accompanying near- and far-fields to exhibit, for example, a strongly focused character. Furthermore we will show that SPP's are ultra fast phenomena which range typically from ps to fs time scales, while the propagation lengths in the visible are of the order of 80 micrometers. Finally, we will demonstrate the stunning coherence of the phenomenon via discussing the re-emission characteristics of SPP's which nicely underline the mixed state nature of the phenomenon.

[1] G. Isfort, K.-D. Schierbaum, D. Zerulla; Causality of Surface Plasmon Polariton Emission Processes, Phys. Rev. B, 73, 033408 (2006)

[2] G. Isfort, K. Schierbaum, D. Zerulla; Polarization Dependence of Surface Plasmon Polariton Emissions, Phys Rev. B, 74, 033404 (2006)