O 20: Nano-optics of metallic and semiconducting nanostructures (time resolved)

Time: Tuesday 10:30–13:00 Location: SCH A216

O 20.1 Tue 10:30 SCH A216

Analytic solution to optimal control of plasmon propagation in nanostructures — •Philip Tuchscherer[1], Christian Rewitz[1], Dmitri V. Voronine[1], F. Javier García de Abajo[2], Walter Pfeiffer[3], and Tobias Brixner[1] — [1]Institut für Physikalische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [2]Instituto de Optica, CSIC, Serrano 121, 28006 Madrid, Spain — [3]Fakultät für Physik, Universität Bielefeld, Universitätsstr. 25, 33516 Bielefeld, Germany

Localization and adaptive control of optical near-fields using polarization-shaped laser pulses have previously been demonstrated theoretically and experimentally. Spatial localization and ultrafast switching of near-fields has a wide range of potential applications such as space-time resolved 'nano-femto' spectroscopy, quantum information processing, and nanoplasmonic devices.

We have now found an analytic solution to the problem of optimal control of plasmon propagation direction. The contrast between energy deposition at two particular locations within a nanostructure array can be manipulated with amplitude and polarization-shaped laser pulses.

Optimal fields from this deterministic approach are found to agree well with those from evolutionary optimization. Apart from the contrast of local energy deposition, nonlinear signals are also controlled by compressing the near fields at the desired site.

The analytic approach gives insight into the mechanisms involved and presents a way to simplify and improve experiments.

O 20.2 Tue 10:45 SCH A216

Dephasing of Optical Magnetism in Magnetic Photonic Crystals — •Michael Geiselmann[1,2], Tobias Utikal[1,2], Markus Lippitz[1,2], and Harald Giessen[1] — [1]4. Physikalisches Institut, Universität Stuttgart, Germany — [2]Max Planck Institut für Festkörperforschung, Stuttgart, Germany

We investigate the time dynamics of optical magnetism in a magnetic photonic crystal. The structure consists of gold cut-wire pairs with a magnesium fluoride spacer on top of a HfO2 slab waveguide. In the cut-wire pair structure, a symmetric and an antisymmetric plasmonic mode can be optically excited. The antisymmetric mode is regarded as a magnetic resonance. With the correct grating period the magnetic resonance in the wires can be coupled to the photonic mode, which is excited in the slab waveguide. In the strong coupling regime, a polaritonic system is formed, termed a magnetic plasmon polariton. In the experiment we study its femtosecond time dynamics using a nonlinear third-harmonic autocorrelation technique. We find that due to the coupling to the waveguide mode the dephasing of the system can be drastically prolonged up to 60 fs. To the best of our knowledge, this is the first experimental demonstration of temporally resolved magnetism in a magnetic photonic crystal.

O 20.3 Tue 11:00 SCH A216

Ultrafast dephasing of the surface plasmon polaritons in metallic nanoparticles: Influence of the chemical environment — •Martin Wortmann, Nils Borg, Frank Hubenthal, and Frank Träger — Institut für Physik and Center for Interdisciplinary Nanostructure Science and Technology – CINSaT, Universität Kassel, Heinrich-Plett-Straße 40, D-34132 Kassel

It is well known that intrinsic size effects for metal nanoparticles have a significant influence on the electron dynamics and lead to a reduction of the dephasing time of the surface plasmon resonance. To take this effect into account, the parameter A has been introduced, which quantifies the influence of all independent damping contributions A_i, with $A = \sum A_i$ (Matthiessen law). Although theoretically well understood, an exact knowledge of A as well as a microscopic understanding of the involved processes, in particular of the chemical surrounding, are still lacking.

In this contribution we present first results on the influence of an inhomogeneous chemical environment on the A-parameter. We have investigated silver nanoparticles supported on sapphire and quartz substrates. The obtained A-parameters amount to $A_{\mathrm{sap}} = 0,20$ nm/fs and to $A_{\mathrm{qua}} = 0,54$ nm/fs, respectively. The latter value is higher, because quartz induces chemical damping. A subsequent coverage of the samples with SO_2 opens a second damping channel and the A-parameters increase. Interestingly, for both SO_2 covered samples the A-parameter is the same and amounts to $A = 1,76$ nm/fs. This leads to the conclusion, that the two different damping channels are not independent.

O 20.4 Tue 11:15 SCH A216

Enhancing optical near-fields in nanoantennas via shaped ultrashort laser pulses — •Christian Rewitz[1], Philip Tuchscherer[1], Dmitri V. Voronine[1], Andreas Reiserer[1], Jer-Shing Huang[2], Bert Hecht[2], and Tobias Brixner[1] — [1]Institut für Physikalische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [2]Nano-Optics and Biophotonics Group, Department of Experimental Physics 5, University of Würzburg, Am Hubland, 97074 Würzburg, Germany

Femtosecond laser pulse shaping has been developed into an efficient tool for the manipulation of interference phenomena on the nanoscale. We show that this technique can be applied to tailor the ultrafast temporal response of the strongly confined and enhanced optical near-fields in the feed gap of resonant optical antennas.

Using finite-difference time-domain (FDTD) simulations followed by Fourier transformation, we obtain the impulse response of a nanostructure in the frequency domain, which allows obtaining its temporal response to any arbitrary pulse shape.

Additionally to this deterministic approach, an adaptive scheme involving an evolutionary algorithm can be used to obtain the optimal spectral phase that pre-compensates for the dispersion induced by the antennas. Comparing deterministic and adaptive results allows for a better understanding of plasmonic resonances in nanostructures. The presented control mechanisms will be of importance for experiments involving light-induced processes in nanometer-scale volumes.

O 20.5 Tue 11:30 SCH A216

Ultrafast Coherent Control of Third-Harmonic Generation in Plasmonic Nanostructures — •Tobias Utikal[1,3], Mark I. Stockman[1,2], Albert P. Heberle[1], Markus Lippitz[3], and Harald Giessen[1] — [1]4. Physikalisches Institut, Universität Stuttgart — [2]Department of Physics and Astronomy, Georgia State University, USA — [3]MPI für Festkörperforschung, Stuttgart

We present a new technique to coherently control ultrafast nonlinear plasmonic effects on a nanometer scale. By using a nonlinear four-photon process (third-harmonic generation) our detection is not limited to the local optical intensity in the system, but additionally supports phase information. The nanostructure under investigation consists of a 1-D gold wire grating on top of a dielectric slab waveguide. The coupling of particle plasmons, optically excited in the wires, to photonic waveguide modes leads to plasmon-polaritonic eigenstates, characterized by long dephasing times, whose dynamics can be coherently controlled on a femtosecond timescale. In the experiment one polaritonic eigenstate is excited by a first sub-8 fs laser pulse (start pulse). A subsequent second pulse (control pulse) follows the start pulse after a few tens of femtoseconds. Dependent on the exact phase delay, the control pulse either stops the polariton oscillation or re-excites it again. A third pulse (probe pulse), which is aligned at a small angle to the start and control pulse, is continuously shifted in time. Photons of the probe pulse together with photons of the start and control pulse create a nonlinear third-harmonic signal depending on the coherently controlled polarization of the polariton.

O 20.6 Tue 11:45 SCH A216

Subwavelength spatio-temporal control of the local excitation of metal nanostructures — Martin Aeschlimann[1], Michael Bauer[5], •Daniela Bayer[1], Tobias Brixner[3], Stefan Cunovic[2], Frank Dimler[3], Alexander Fischer[1], Javier García de Abajo[4], Viktor Myroshnychenko[4], Walter Pfeiffer[2], Martin Rohmer[1], Christian Schneider[1], Felix Steeb[1], Christian Strüber[2], and Dmitri V. Voronine[3] — [1]University of Kaiserslautern, Germany — [2]University of Bielefeld, Germany — [3]Ludwigs-Maximilian-Universität, Würzburg, Germany — [4]CSIC, Madrid, Spain — [5]University of Kiel, Germany

Using time-resolved two-photon photoemission electron microscopy we demonstrate simultaneous spatial and temporal control of nanooptical fields. Based on the recent demonstration of ultrafast adaptive near field optics, we now investigate directly the temporal evolution of the local excitation by spatially resolved cross correlation measurements.

Planar silver nanostructures manufactured by e-beam lithography are excited by polarization shaped pump pulses and the corresponding local excitation is probed by a time-correlated probe excitation using circularly polarized laser pulses. The time-resolved cross correlation traces for different regions of the nanostructure show a clear variation of their relative intensities. This confirms that the polarization-shaped incident laser pulse does indeed switch between two different excitation patterns within a time scale that can be controlled almost freely and is limited only by the spectral bandwidth of the used coherent light source. First spatio-temporal control experiments will be shown.

O 20.7 Tue 12:00 SCH A216

Femtosecond Surface Plasmon Characterisation on Nanostructured Surfaces — •Jose Francisco Lopez-Barbera, Brian Ashall, Fergal O'Reilly, and Dominic Zerulla — University College Dublin, School of Physics, Dublin 4, Ireland.

Latest developments in ultrafast pulses have presented new opportunities for the investigation of surface plasmon polaritons (SPPs) on the sub 20 fs time scale. We have extended the characterization of the resonant photon-SPP coupling processes on smooth surfaces as presented in [1], emphasising the excitation, propagation, and temporal and spectral responses of the SPPs.
Broadening these studies, we have characterized the same processes using SPP excitation on metallically coated nanostructures of specific surface symmetry and tuneability. As expected, the nanostructured samples show different temporal characteristics which are attributed to their more complex propagation mechanisms. Furthermore, using our tuneable plasmonic structures [2] we present the influences of the tuneability of such systems on the temporal characteristics of the SPPs.

[1] S. E. Yalcin et al., Appl. Phys. Lett. 93, 101103 (2008).
[2] Stephanie Rehwald, Michael Berndt, Frank Katzenberg, Stephan Schwieger, Erich Runge, Klaus Schierbaum, and Dominic Zerulla, Phys. Rev. B 76, 085420 (2007).

O 20.8 Tue 12:15 SCH A216

Surface plasmon polariton - exciton interaction in semiconductor-metal hybrids and dye-metal systems — •Stephan Schwieger[1], Parinda Vasa[2], Christoph Lienau[2], and Erich Runge[1] — [1]Technische Universität Ilmenau, 98693 Ilmenau, Germany — [2]Carl von Ossietzky Universität Oldenburg, 26129 Oldenburg, Germany

Surface plasmons polaritons (SPPs) are optical excitations that are confined to a metal surface and coupled to far-field radiation, e.g., by a periodic nanowire array. The interaction of SPPs and excitons in semiconductor quantum wells (QW) [P. Vasa et. al., PRL, 101, 116801 (2008)] or in a dye layer is investigated. The coupling strength is estimated for different systems and band structures of the coupled modes are calculated. A band gap at the exciton resonance energy is found for the dye system, which indicates strong coupling energies. In the semiconductor system, we predict an efficient energy transfer from excitons to SPPs and a clear enhancement of the SPP life time and propagation length for optimized parameters.

O 20.9 Tue 12:30 SCH A216

Ultrafast optical nonlinearities in hybrid metal-J-aggregate nanostructures — •Parinda Vasa[1], Robert Pomraenke[1], Stephan Schwieger[2], Erich Runge[2], and Christoph Lienau[1] — [1]Carl von Ossietzky Universitaet, Institut fuer Physik, 26111 Oldenburg, Germany — [2]Technische Universitaet Ilmenau, Institut fuer Physik, 98684 Ilmenau, Germany.

We report the first measurement of an ultrafast optical nonlinearity resulting from the strong interaction between Surface Plasmon Polaritons (SPPs) excited on a gold grating and excitons in a J-aggregated cyanine dye. The hybrid metal-J-aggregate nanostructure is fabricated by spin coating the dye solution on a gold grating fabricated using a focused ion beam. The hybrid structures are characterized using far-field linear reflectivity as well as photoluminescence measurements and exhibit enhanced SPP-exciton coupling in the linear optical regime. The nonlinearity is investigated by low-temperature, angle-resolved, ultrafast pump-probe spectroscopy with 20-fs-time resolution. The strong coupling results in a significant shift in the response wavelength and changes in the response time of the third order nonlinearity of the dye exciton are observed. Specifically, we observe, under certain resonance conditions, extremely strong, more than tenfold changes of the radiative lifetime of the dye exciton due to its coupling to SPP. Such a strong ultrafast nonlinear interaction between metal and excitons will be of key importance to amplify SPP excitations in such hybrid structures.

O 20.10 Tue 12:45 SCH A216

Coherent exciton-surface plasmon polariton coupling in hybrid metal-J-aggregate — •Robert Pomraenke[1], Parinda Vasa[1], Stephan Schwieger[2], Erich Runge[2], and Christoph Lienau[1] — [1]Carl von Ossietzky Universitaet, Institut fuer Physik, 26111 Oldenburg, Germany — [2]Technische Universitaet Ilmenau, Institut fuer Physik, 98684 Ilmenau, Germany.

Understanding and manipulating the interactions between quantum emitters and Surface Plasmon Polaritons (SPPs) is the key to designing and implementing novel nano-optical devices such as nano-lasers or ultrafast optical switches. Here, we report the observation of a greatly enhanced coherent coupling between Surface Plasmon Polaritons (SPPs) excited on a metal grating and excitons in a J-aggregated cyanine dye. The hybrid metal-semiconductor nanostructure is fabricated by spin coating the dye solution on a gold grating fabricated using a focused ion beam. The structure is designed to maximize the radiative interaction between the two excitations which is probed by low-temperature, angle-resolved, far-field reflectivity and photoluminescence measurements. As a result of the strong interaction between the two resonances, coupled SPP-exciton polariton modes following hybrid dispersion relations are formed. The experimental results are explained within a phenomenological, coupled oscillator model. Such a strong interaction can be used to significantly alter the optical response of an organic semiconductor or to design novel hybrid devices.