

O 43 Time-resolved spectroscopy I

Time: Thursday 17:30–18:30

Room: WIL C207

O 43.1 Thu 17:30 WIL C207

Nanosopic ultrafast space-time-resolved spectroscopy — ●W. PFEIFFER¹, T. BRIXNER¹, C. SPINDLER¹, and F.J. GARCÍA DE ABAJO² — ¹Physikalisches Institut, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — ²Centro Mixto CSIC-UPV/EHU, Apartado 1072, 20080 San Sebastián, Spain

We analyze a recently proposed new scheme for ultrafast spectroscopy with nanometer spatial and femtosecond temporal resolution [1] that enables the direct spatial probing of nanoscale energy transfer or charge transfer processes. The interaction of polarization-shaped laser pulses with a nanostructure allows us to control the spatial and temporal evolution of the optical near-field. Employing a learning algorithm, the field is tailored such that pump and probe excitation occur at different positions. Both excitations can be restricted to sub-diffraction extensions and are separable on a nanometer length scale. Substantial control according to different goals is demonstrated and the limits of controllability are investigated. The dominating control mechanism is local interference of near-field modes that are excited with the two independent polarization components of the incident light pulses and hence polarization pulse shaping is essential to achieve substantial control in the optical near-field. The influence of other control mechanisms is discussed and a number of possible applications using different nanostructures such as planar and three-dimensional arrangements of nanoparticles are presented.

[1] T. Brixner, F. J. García de Abajo, J. Schneider, and W. Pfeiffer, Phys. Rev. Lett. 95, 093901 (2005).

O 43.2 Thu 17:45 WIL C207

Competing Processes at Ice-Metal Interfaces: Ultrafast Electron Transfer and Solvation Dynamics — ●JULIA STÄHLER, CORNELIUS GAHL, UWE BOVENSIEPEN, and MARTIN WOLF — Freie Universität Berlin, Fachbereich Physik, Arnimalle 14, 14195 Berlin, Germany

The ultrafast dynamics of excess electrons photo-injected into ultrathin amorphous D₂O layers on Ru(001) and Cu(111) are investigated using time-resolved two-photon photoelectron spectroscopy. On both substrates we observe electron localization and solvation in the ice films, competing with back relaxation to the metal. We identify two regimes of back transfer: (i) For $t < 300$ fs the initial back transfer rate is four times higher on Ru(001) ($\tau_1^{Ru} = 34$ fs) compared to Cu(111) ($\tau_1^{Cu} = 140$ fs). (ii) At $t > 300$ fs we observe a slower back transfer, which is comparable for both systems ($\tau_2^{Ru} = 302$ fs, $\tau_2^{Cu} = 316$ fs). By an empirical model calculation based on rate equations, which nicely reproduces our data, we show that the back transfer is initially determined by the electronic surface band structures of the two substrates, whereas the population decay at later times is dominated by tunneling through a potential barrier at the interface, which arises from the rearrangement of the surrounding polar molecules during solvation.

O 43.3 Thu 18:00 WIL C207

Time-Resolved Two-Photon Photoemission study of C₆₀ — ●ARNE ROSENFELDT, BENJAMIN GÖHLER, and HELMUT ZACHARIAS — University of Münster, 48149 Münster, Germany

Up to 200 ML of C₆₀ on Cu(111) are illuminated by tunable 365 to 415 nm (3.40 to 2.99 eV) radiation of 50 ps pulse duration. Delayed 263 nm (4.71 eV) photons have sufficient energy to probe half of the LUMO and the LUMO+1. A rate equation fitted to the electron dynamics suggests a short lifetime of about 126 ps for the LUMO, and another one longer than 5 ns, which may originate from excitons with the same energy as the LUMO. The excitation probability of these states is measured as a function of photon energy, showing a maximum at 392 nm (3.16 eV). Using four-wave mixing in Xenon radiation at 150 nm (8.27 eV) is generated to investigate low-lying states. For measurement of long lifetimes a second laser is electronically delayed. The project is financially supported by the DFG in the SPP1093 “Dynamik von Elektronentransferprozessen an Grenzflächen”

O 43.4 Thu 18:15 WIL C207

Ultrafast interfacial electron transfer probed with two-photon photoemission — ●LARS GUNDLACH, RALPH ERNSTORFER, SILKE FELBER, RAINER EICHBERGER, and FRANK WILLIG — Hahn-Meitner-Institute, Department: Dynamics of Interfacial Reactions SE4, Glienickerstrasse 100, 14109 Berlin

Two-photon photoemission (2PPE) is the method of choice for investigating ultrafast interfacial electron transfer because of its excellent sensitivity and time resolution. With this technique one can address adsorbates at coverages far below a monolayer. With the same chromophore perylene fixed with different anchor and bridge groups at the surface of rutile TiO₂(110) the corresponding 2PPE transients revealed the relevant parameters that characterize the contributing processes.

Instantaneous optical injection on one hand and slow injection over a long distance on the other hand were realized. Direct optical charge transfer was realized with the chromophore catechol that is known to form a charge transfer complex with Ti atoms on the surface of TiO₂. The slow injection cases were realized by inserting rigid molecular bridges. The same experimental systems were investigated with fs-transient absorption. Comparing the different 2PPE transients and the corresponding transient absorption transients for the identical systems revealed the physical processes and time scales that control the 2PPE transients. 2PPE measurements with different angle of incidents and polarizations of the pump pulse revealed the binding geometry of the different anchor groups on the surface.