O 11: Time-Resolved Spectroscopy I

Time: Monday 13:30–15:00 Location: MA 043

O 11.1 Mon 13:30 MA 043

Two-photon photoemission of image-potential resonances in front of the Si(100) surface — Jens Kopprasch[1], •Christian Eickhoff[1], Irina Ostapenko[1], Cornelius Gahl[1], and Martin Weinelt[1,2] — [1]Max-Born-Institut, Max-Born-Straße 2A, 12489 Berlin — [2]Freie Universität Berlin, Arnimallee 14, 14195 Berlin

We have investigated the dangling-bond states and image-potential resonances on the Si(100) 2×1 surface by means of bichromatic two-photon photoemission. Optical parametric amplification generates 70 fs ultraviolet pulses with tunable photon energies between 4.5 and 5.5 eV. These allow us to populate unoccupied states up to the Si(100) vacuum level, probed by the IR fundamental at a fixed wavelength of 795 nm. Besides the occupied dangling-bond state D_{up}, we resolve the first two image-potential states with binding energies of $E_1 = 0.62$ eV and $E_2 = 0.18$ eV with respect to the vacuum level. Using these energies we obtain a surface dielectric-constant of $\epsilon = 11.2$ which is close to the silicon bulk-value of $\epsilon = 11.9$.

Tuning the photon energy of the pump pulse across the D_{up} to $n = 1$ and D_{up} to $n = 2$ transitions we find a significant variation of both the 2PPE peak positions and the intensities. Before resonance we observe the D_{up} initial state with the kinetic energy increasing with the pump-pulse photon-energy. Above resonance the D_{up} intensity is significantly reduced and shifted to the respective image-potential resonances at constant kinetic energy. These intensity variations indicate interference between the transition to the discrete image-potential resonance and transitions to the continuum of unoccupied bulk states.

O 11.2 Mon 13:45 MA 043

Combining density functional and density matrix theory: Optical excitation and electron relaxation at the Si(001) 2×1 surface — •Norbert Bücking[1,3], Peter Kratzer[2], Matthias Scheffler[3], and Andreas Knorr[1] — [1]Institut für Theoretische Physik, Technische Universität Berlin, 10623 Berlin, Germany — [2]Fachbereich Physik, 47048 Duisburg, Germany — [3]Fritz-Haber-Institut der MPG, 14195 Berlin, Germany

A theoretical two-step approach to investigate the optical excitation and subsequent phonon-assisted relaxation dynamics at semiconductor surfaces is presented and applied to the Si (001) 2×1-surface: In the first step, the electronic band structure and the Kohn-Sham wave functions are calculated by density-functional-theory (DFT) within the LDA. In the second step, dynamical equations are derived from density-matrix theory (DMT), whereby an optical field is considered via $\mathbf{A} \cdot \mathbf{p}$-coupling and phonon induced relaxation by a deformation potential coupling term. Into these equations, the numerical results of the DFT calculation (Kohn-Sham eigenvalues and wave functions) enter as coupling matrix elements. By numerically solving the dynamical equations, the time-resolved population of the excited states can be evaluated. The results for the Si (001) surface correspond to the findings of recent experiments, in particular a short (intra-surface-band scattering) and a long (bulk-surface band scattering) timescale are dominating the relaxation process. The value of the experimental short timescale is reproduced by our calculations, whereas the long timescale cannot be accurately described by our theory.

O 11.3 Mon 14:00 MA 043

The Atomistic-Continuum Modeling of Short Pulse Laser Interaction with Semiconductors — •Dmitriy Ivanov and Baerbel Rethfeld — Physics Department, Technical University of Kaiserslautern, Kaiserslautern, Germany

The understanding of fundamental mechanisms behind the sub-wave length surface modification on semiconductors is of a great importance for Information Technologies. However, strong laser-induced phase perturbations, occurring under conditions of nonequilibrium between free laser-generated carriers and phonons, make the experimental and theoretical study of short pulse laser nanostructuring on semiconductors difficult. Previously, the atomistic-continuum approach for modeling of short-pulse laser interactions with metals have been proven as an efficient tool when studying processes of laser melting, ablation, and nanostructuring on metals. In present work, a computational technique that combines the advantages of different approaches into the atomistic-continuum model for semiconductors is developed on the example of Si. In the combined model, 1) the kinetics of fast non equilibrium phase transformations is treated at atomic level with Molecular Dynamics method, and 2) the description of laser light absorption by free carriers, their transport dynamics, and strong laser-induced non equilibrium between free carriers and phonons are accounted for in the continuum part by means of free carrier dynamics model.

O 11.4 Mon 14:15 MA 043

Potential Energy Surface of Laser-Excited InSb — •Jessica Walkenhorst, Eeuwe S. Zijlstra, and Martin E. Garcia — Theoretische Physik, Fachbereich Naturwissenschaften, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel

A recent experiment [A. M. Lindenberg et al., Science **308**, 392 (2005)] performed on InSb suggests that ultrafast laser-induced nonthermal melting occurs due to a flattening of interatomic potentials. This study was based on Debye-Waller theory, applied in the time-domain and for non-equilibrium processes. We analyzed the nonthermal melting of InSb by using (i) first-principles electronic structure calculations for the interatomic potentials (ii) dynamical models to find the structure factors under different nonequilibrium conditions. Our calculations show that no dramatic flattening of the potential energy surface occurs. Instead, the softening of the transverse acoustic phonons at the X point suffices to explain the measured Gaussian x-ray intensity decay.

O 11.5 Mon 14:30 MA 043

Ultrafast electron dynamics in Pb/Si(111) investigated by two-photon photoemission — •Patrick S. Kirchmann, Martin Wolf, and Uwe Bovensiepen — Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin-Dahlem

We studied the ultrafast electron relaxation dynamics of hot electrons in quantum well states (QWS) in ultrathin epitaxial Pb films on Si(111) [1] by femtosecond time-resolved two-photon photoemission spectroscopy.

Up to four unoccupied QWSs are identified, which exhibit a bi-exponential decay of the hot electron population. The slower decay is assigned to a delayed filling of the metallic QWS in the Pb adlayer by scattering from electronic states of the optically excited Si(111) substrate. The faster decay is assigned to e-e scattering within the Pb film. The overall trend of the extracted decay rates is governed by Fermi liquid theory. However, a detailed analysis reveals a well resolved dip in the decay rate which occurs precisely at the binding energy of the band bottom of the first unoccupied QWS. This local minimum of the decay rate is assigned to intra-subband scattering within the Pb film.

Thus, for a comprehensive description of the electron decay in a two-dimensional metal film not only the electron density and screening parameters as in Fermi liquid theory have to be considered. Here, we show that also the electron scattering processes in the quantized band structure have to be taken explicitly into account.

[1] P. S. Kirchmann et al., Phys. Rev. B **76**, 075406 (2007)

O 11.6 Mon 14:45 MA 043

Time-resolved Electron Diffraction studies on the ultrafast temperature response of Bi and Pb on Si(111) — •Boris Krenzer, Anja Hanisch, Simone Möllenbeck, Tobias Pelka, and Michael Horn-von Hoegen — Department of Physics, University of Duisburg-Essen, 47048 Duisburg, Germany

Recent progress in developing ultrashort electron pulses has opened the wide field of investigating structural dynamics of surfaces upon short-pulsed laser excitations. Because the diffraction pattern is related to the atomic positions in a solid and its surface, direct investigation of transient structures on a femtosecond timescale became possible. Additionally, the diffracted intensity is affected by the thermal excitation of the solid and its surface. Thus, a time-resolved electron diffraction experiment yields information on the transient structure and the transient vibrational energy contained in the system at the same time.

Here we report on the fs-excitation of ultra-thin epitaxial Bi- and Pb-films on a Si(111)-substrate. For the Bi-film the initial temperature rise upon laser excitation is rather slow with a time-constant of 20 ps, which is in accordance to the predictions of the two-temperature model (2TM). Because Pb has a three orders of magnitude larger electron-phonon coupling constant the observed initial surface temperature rise is faster than for Bi but slower than the prediction of the 2TM. This discrepancy is attributed to the limited time-resolution of the experiment. However, the Pb/Si(111) is an ideal system to quantify the experimental temporal resolution and to test methods for improving the time-resolution.