O 72: Symposium: Beyond Optical Wavelengths: Time-Resolved Spectroscopy of Surface Dynamics with EUV and XUV Radiation II (Invited Speakers: Wilfried Wurth, Hermann Dürr, Shik Shin)

Time: Thursday 14:00–17:00 Location: HE 101

Invited Talk O 72.1 Thu 14:00 HE 101

First Experiments at FLASH and the Core-Hole Clock — •Wilfried Wurth — Institut für Experimentalphysik, Universität Hamburg, Luruper Chaussee 149

Time-resolved soft x-ray spectroscopy has the potential to provide unique element specific and chemical state selective information on the complex wave packet evolution at surfaces and interfaces.

With the core-hole-clock method this potential has been utilized to determine ultrafast electron delocalization at specific atomic sites. However, here, dynamic information is only accessible in the time range given by the fixed decay time of core-excited states. In order to really follow the temporal evolution at selected atomic sites, we have to develop atom specific and chemically selective X-ray spectroscopy further towards a femtosecond time resolving technique.

With the Free-Electron Laser in Hamburg (FLASH) at DESY a unique source for femtosecond XUV-pulses with unprecedented brillance is operational since 2005. In the talk I will present some ideas how femtosecond X-ray pulses from Free Electron Lasers can be used to study surface and interface dynamics. First time-resolved experiments from FLASH will be presented and implications for future experiments will be discussed.

This work is supported by the BMBF in the framework of the Forschungschwerpunkt 301 FLASH: Matter in the light of ultrafast and extremely intense X-ray pulses.

O 72.2 Thu 14:30 HE 101

Excitation of electrons in Silicon with an ultrashort XUV free electron laser pulse: a Monte-Carlo study — •Nikita Medvedev and Baerbel Rethfeld — Technische Universitaet Kaiserslautern, 67653, Germany

The new light source FLASH at DESY in Hamburg provides ultrashort high intensity XUV pulses. We study the interaction of this new kind of irradiation with condensed matter theoretically. In this contribution we present first simulations, applying Classical Trajectory Monte Carlo simulation (CTMC) to describe the dynamics of electronic excitation and ionization within a solid silicon target, irradiated with femtosecond XUV laser pulse (25 fs, $\hbar\omega = 38$ eV). The CTMC-method was extended to take into account the electronic band structure and Pauli's principle for electrons excited into the conduction band. Secondary excitation and ionization processes were included as well. We calculate the temporal distribution of the density of exited and ionized electrons, the energy of these electrons and the energy distribution function. The influence of the band structure on the redistribution of free electrons on subpicosecond time-scale is studied. It is demonstrated that the final kinetic energy of free electrons is much less than the total energy provided by the laser pulse, because some part of energy is spended to overcome ionization potentials and is kept by holes. It was found that the total number of free electrons is significantly less than estimated by $n_e = \hbar\omega/E_{\mathrm{gap}}$. We introduce the concept of an "effective band gap", which can be applied to estimate the free electron density for collective electronic excitation with a high-intensity XUV laser pulse.

O 72.3 Thu 14:45 HE 101

Single-shot femtosecond EUV-pump/visible-probe cross-correlation on GaAs — •Theophilos Maltezopoulos[1], Stefan Cunovic[2], Marek Wieland[3], Martin Beye[3], Armin Azima[1], Harald Redlin[1], Maria Krikunova[3], Roland Kalms[3], Ulrike Fruehling[1], Filip Budzyn[3], Wilfried Wurth[3], Alexander Foehlisch[3], and Markus Drescher[3] — [1]HASYLAB at DESY, Notkestrasse 85, 22607 Hamburg, Germany — [2]Department of Physics, Bielefeld University, Universitaetsstrasse 25, 33615 Bielefeld, Germany — [3]Institut fuer Experimentalphysik, University of Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany

The Free Electron Laser in Hamburg (FLASH) is currently the most intense femtosecond light source in the extreme-ultraviolet (EUV) range, which facilitates efficient pumping of inner shells in solid targets. For pump-probe experiments, a synchronized optical fs laser system is available at the facility. In our set-up, the EUV pulse (wavelength 28 nm, pulse length 20-30 fs) and the visible pulse (400 nm, 130 fs) are non-collinearly overlapped in space and time on the surface of a GaAs crystal. Along its path, the EUV pump pulse changes the reflectivity of the GaAs sample for the visible probe pulse, the latter being imaged onto a CCD array. The spatial position of the reflectivity change captures the dynamics of the process in a single exposure. The technique can be utilized to determine individual EUV-visible delays with a precision of about 40 fs.

15 min. break

Invited Talk O 72.4 Thu 15:15 HE 101

Ultrafast Magnetization Dynamics Probed by Femtosecond X-Ray Spectroscopy — •Hermann A. Dürr — BESSY GmbH, Albert-Einstein-Str. 15, 12489 Berlin, Germany.

When the electronic system of a solid is rapidly heated by absorbing a femtosecond optical laser pulse it takes time to re-establish thermal equilibrium. This timescale is ultimately determined by energy transfer from the electronic system to the lattice. For ferromagnets this process can also lead to an ultrafast quenching of the ferromagnetic order. Angular momentum conservation dictates that an exchange of spin angular momentum with a reservoir such as the lattice should occur. So far real time studies of these processes were limited to the use of femtosecond laser pump-probe spectroscopy [1]. We show that an efficient novel channel for angular momentum dissipation to the lattice can be opened by fs laser excitation of a ferromagnet [2]. The quenching of spin angular momentum and its transfer to the lattice with a time constant of 120 fs is determined unambiguously with x-ray magnetic circular dichroism by separating spin and orbital contributions to the magnetization. Fs time-resolved x-ray absorption spectroscopy shows an unexpected increase in valence electron localization during the first 120 fs possibly providing the driving force behind fs spin-lattice relaxation.

[1] E. Beaurepaire, et al., Phys. Rev. Lett. 76, 4250 (1996).
[2] C. Stamm, et al., Nature Materials 6, 740 (2007).

O 72.5 Thu 15:45 HE 101

Non-equilibrium spin-dynamics of Gd(0001) studied by magnetic linear dichroism in 4f core-level photoemission — •Alexey Melnikov[1], Helena Prima-Garcia[2], Martin Lisowski[1], Tanja Giessel[2], Ramona Weber[2], Roland Schmidt[2], Cornelius Gahl[2], Nadezhda Bulgakova[3], Uwe Bovensiepen[1], and Martin Weinelt[1,2] — [1]Freie Universität Berlin, Fachb. Physik, Arnimallee 14, 14195 Berlin, Germany — [2]Max-Born-Institut, Max-Born-Straße 2 A, 12489 Berlin, Germany — [3]Institute of Thermophysics SB RAS, pr. Lavrentiev 1, 630090 Novosibirsk, Russia

The magnetic linear dichroism of the Gd 4f core-level is studied in a time-resolved photoemission (PE) experiment at BESSY II employing 1.55eV, 100fs laser pump and 60eV, 50ps synchrotron-radiation probe-pulses. We define the dichroic contrast $\Delta M(T)$ at temperature T averaging the difference of PE spectra for opposite magnetization direction. $\Delta M(T)$ follows the spontaneous magnetization, vanishes at the Curie temperature T_C, and can therefore be used to map the order of the 4f spin-system. The latter is reduced upon optical excitation of the 5d6s valence electrons. Remarkably, $\Delta M(T)$ remains at 80% of the equilibrium value while the lattice temperature increases up to T_C. A simulation of $\Delta M(T(\tau))$ by a quasi-equilibrium model predicts an at least two times larger reduction of $\Delta M(T)$ for delays τ below 100 ps. We conclude that at these early times equilibration between lattice and 4f spin subsystems has not been established. This reveals principal differences to the spin dynamics in itinerant ferromagnets, where the valence electrons excited by the laser also carry the magnetic moment.

Invited Talk O 72.6 Thu 16:00 HE 101

Sub-meV-resolution photoemission spectrocopy on solids using VUV laser — •shik shin — Institute for Solid State Physics, Chiba277-8581, Japan

We developed the ultrahigh-resolution photoemission system using quasi-CW VUV laser. VUV laser, photoemission analyzer, and cooling system were newly developed for ultrahigh resolution photoemission.

The total resolution of the Laser-PES system is estimated to be about 0.15 eV by using the gold Fermi edge at 1.8 K at present. This is the highest resolution of the world. We will present the angle-integrated and angle-resolved photoemission results on several superconducting materials as well as the heavy Fermion materials. We will also shows the high resolution PES on surface states of Ag.

O 72.7 Thu 16:30 HE 101

Femtosecond time- and angle-resolved photoelectron spectroscopy: A tool to study non-equilibrium states of electronic structure — •Uwe Bovensiepen — Freie Universität Berlin, Fachbereich Physik, Arnimallee 14, 14195 Berlin

During the last 20 years angle-resolved photoemission spectroscopy has turned from an established surface science technique to a powerful experimental system that maps the electronic structure of valence bands in solids. Intense optical excitation by femtosecond laser pulses drives this electronic subsystem in condensed matter into a non-equilibrium state and its energy content is characterized by electronic temperatures of several thousand Kelvin. The ion-ion interaction is determined by the screened electron-ion potential and the intense optical excitation can be expected to modify the screening contribution. We excite metallic structures by 1.5 eV laser pulse at absorbed fluences of 0.1 – 1 mJ/cm^2 and probe the transient state of the electronic structure by 6 eV UV laser pulses in time- and angle-resolved photoemission. For the $5d_{z^2}$ surface state of Gd(0001) and for the highest occupied quantum well state originating from the $6p_z$ band in epitaxial Pb films on Si(111) we observe a transient binding energy increase by 50 – 100 meV before electron-phonon scattering has lowered the excess energy in the electronic subsystem. Thus, we conclude that the optical excitation leads to a stabilization of both these systems. First experiments on RTe3 charge density wave materials show that under comparable conditions even a transition from an insulating to a metallic state can be induced optically.

O 72.8 Thu 16:45 HE 101

Ultrafast charge transfer at silicon surfaces investigated by the core hole clock method: Band structure influences — Silvano Lizzit[1], Guillermo Zampieri[1,2], Luca Petaccia[1], Rosanna Larciprete[1,3], Krassimir L. Kostov[4], Georgi Tyuliev[5], and •Dietrich Menzel[6,7] — [1]Sincrotrone Trieste, Area Science Park, 34012 Trieste, Italy — [2]Centro Atómico Bariloche, 8400 S.C. de Bariloche, Argentina — [3]CNR-Istituto dei Sistemi Complessi, 00016 Monterotondo (RM), Italy — [4]Institute of General and Inorganic Chemistry, Bulgarian Academy of Sciences, Sofia, Bulgaria — [5]Institute of Catalysis, Bulgarian Academy of Sciences, Sofia, Bulgaria — [6]Physik-Dept.E20, TU Muenchen, 85748 Garching, Germany — [7]Fritz-Haber-Institut der MPG, Faradayweg 4-6, 14195 Berlin, Germany

With the well-known core hole clock method to measure charge transfer (CT) at surfaces by resonant core excitation and decay under resonant Auger Raman conditions (bandwidth of exciting photons below the lifetime width) the lifetimes of 4s electrons on core-excited Ar adsorbed on clean and H-covered n- and p-Si(100) have been determined. CT times are in the range of 1 to 4 fs. They do not depend on doping and are about twice larger on H-covered than on clean Si. Distinct structure is found as a function of excitation energy, similarly on both surfaces. It is explained by the influence of the empty band structure of Si(100), into which the excited 4s electron is transferred. The concepts and mechanisms will be discussed. Time allowing, the relation of this method to laser pump-probe techniques will be discussed.