Dynamic Pathways in Multidimensional Landscapes

Report of Contributions

https://events.hifis.net/e/pathways2013

Contribution ID: **1** Type: **not specified**

Nonmagnetic linear dichroism in fluorescence spectra of cubic solids

The determination of the absorption coefficient via transmission experiments is not always feasible. Common alternative methods for X-ray absorption spectroscopy (XAS) are the detection of electron and fluorescence yield (FY), implying that these are proportional to the absorption coefficient. This assumption is generally justified for the electron yield. The FY is influenced by saturation effects which can be reduced by choice of the experimental geometry. Furthermore the fluorescence process involves a twofold dipole transition, which implies that the FY is not only determined by the probability to absorb a photon, but also by the probability to emit a photon in the direction of the detector. The emission varies in direction and polarization, depending on the symmetry of the core hole and the states the core hole is filled from. In consequence even a saturation-free FY spectrum is generally not reflecting the X-ray absorption cross section. On the other hand, FY in a particular experimental geometry allows to probe the contribution of states of different symmetry selectively. Even in high-symmetry crystals, like cubic transition metal oxides, which exhibit no linear dichroism in XAS we observed dichroic effects in the fluorescence yield. In addition to paramagnetic MnO and CoO we examined antiferromagnetically ordered NiO in form of a bulk crystal as well as a crystal thin film. In the NiO thin film magnetic linear dichroism occurs in XAS originated by a preferred spin orientation due to a distortion of the cubic symmetry. We could distinguish the dichroic effects caused by the fluorescence process from magnetic linear dichroism in XAS. To explore the angle and polarization dependent fluorescence yield spectroscopy, we combined an experimental and theoretical investigation.

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Session Classification: Quantum materials

Track Classification: 1

Contribution ID: **2** Type: **not specified**

Phase separation in complex oxides: RTiO3

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Complex oxides display an unparalleled richness of physical phenomena arising from the coupling of their charge, spin and orbital degrees of freedom, with cuprate high Tc superconductors and colossal magnetoresistive (CMR) manganites as flagship materials systems. For the CMR systems, phase separation is believed to play a crucial role in creating the hypersensitivity to external stimuli such as external field [1]. In this contribution I will report our experiments on perovskite titanate systems, which are a t2g materials analogy to the CMR systems with which they share much underlying physics.
In particular, I will deal with calcium-doped rare earth titanium oxides, which exhibit charge and orbital ordering (CO/OO) during a temperature-driven metal-insulator transition (T-driven MIT). These systems are hypersensitive to the tuning of the hole-doping level, whereby the electrical transport then differs by several orders of magnitude [2], as occurs with external field in the CMR manganites. In this talk, I will present recently recorded data aimed at the investigation of the phase separation dynamics during T-driven MIT in titanates at LCLS using X-ray photon correlation spectroscopy techniques [3]. This is the first time that the single crystal coherent x-ray diffraction patterns have been recorded at 120Hz in the time domain.

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Session Classification: Quantum materials

Track Classification: 2

Contribution ID: **3** Type: **not specified**

Ab initio simulations of X-rays probing ultra-fast dynamics

We employ quantum chemical calculations and molecular dynamics simulations in the study of ultra-fast solution dynamics. The theoretical tools are crucial for the interpretation of X-ray and photo-electron spectra of aqueous solutions.

Simulations of photo-induced chemical processes require a computational framework, in which we can calculate spectra in excited states and of molecules undergoing dissociative dynamics. Hence, we have chosen to work with multi-configurational (restricted active space SCF) quantum chemical calculations, which can give a balanced description of many electronic states simultaneously. This is essential both for the description of the dynamical processes and for the spectrum simulations in particular for resonant inelastic X-ray scattering process.
The theoretical tools also allow for the simulation of dynamical effects inherent in the spectroscopies, such as core-hole induces proton-transfer.

Applications to aqueous electrolyte are presented

- the effects of hydrogen bonding around tri-iodide
- the pH dependence in the x-ray emission of simple amino acids
- L-edge x-ray absorption of transition metal complexes

showing the importance of combining experimental and theoretical data for reaching insights into the electronic structure and ultra-fast dynamics.

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Session Classification: Molecular dynamics 2

Track Classification: 1

Contribution ID: **4** Type: **not specified**

The MID station at the European X-ray Free-Electron Laser facility

The Materials Imaging and Dynamics (MID) station at the forthcoming European X-ray Free-Electron Laser facility (XFEL.EU) will allow investigations of materials using wide- and small-angle X-ray scattering techniques. The special features of the radiation from XFEL.EU allow novel experiments to be conducted, for instance exploiting the temporal structure and coherence properties of the X-rays. Of particular interest are the investigations of fast and ultra-fast dynamics using X-ray speckle correlation techniques (XPCS) and coherent X-ray diffractive imaging (CXDI).
The MID station will also provide new possibilities for time-resolved scattering in general and the use of very high energies (up to the 100 keV range) is also foreseen for certain materials science experiments. The instrument will also provide an optical laser system allowing optical pump / X-ray probe measurements at highest repetition rates (up to 4.5 MHz). The MID station features appropriate X-ray optics to tailor the beam for the aforementioned experiments with variable spot sizes between 100 & 1 μm and smaller.
The instrumentation foreseen comprises a multi-circle diffractometer as well as an in-vacuum sample handling system. Highly specialized and optimized 2D pixel detectors are currently being developed to be able to fulfill the scientific goals of the MID station.
Additionally, an X-ray split & delay will be implemented allowing X-ray pump / X-ray probe, two-color and stereo imaging experiments, ultrafast X-ray tomography, and speckle visibility spectroscopy on summed images.
At the end of the instrument, a diagnostic endstation will provide essential information for data analysis, e.g. the intensity and spectrum of the individual X-ray pulses.

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Session Classification: Hot topics and posters

Track Classification: 1

Contribution ID: **5** Type: **not specified**

Correlated non-adiabatic proton-hole motion after photoionization

The protonated water dimer irradiated with the free-electron laser FLASH at DESY (90 eV) undergoes Coulomb-explosion after losing one of its valence electrons via photoionization [1]. Because of the close-lying outer valence orbitals and various conical intersections, non-adiabatic effects are presumably important for the overall dynamics. Ab initio potential energy surfaces for the three lowest di-cationic states have been computed at the CASSCF level. A regularized diabatization scheme is used to obtain a diabatic representation of the vibronic Hamiltonian [2]. The quantum dynamics of the photo-fragmentation has been investigated using the multi-configuration time-dependent Hartree (MCTDH) method [3]. We find that the correlation between the motion of the shared proton and the newly created hole builds up in less than five femtoseconds. The build up of the proton-hole correlation is a consequence of strong non-adiabatic effects [4,5].

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Session Classification: Molecular dynamics 2

Track Classification: 2

Contribution ID: **6** Type: **not specified**

Ultrafast Energy Transfer to Liquid Water by Short and Intense THz Pulses

Liquid water is the single most important medium in which chemical and biological processes take place. Rather than acting as passive environment, the dynamics of water during chemical and biological processes play a fundamental role in the solvation and stabilization of reaction intermediate. The possibility to generate sub-ps and very intense THz pulses at free-electron lasers in full synchronization with the X-rays (XFEL) opens the possibility to time-resolved investigations of transient state of water and of molecular species dissolved in it. Liquid water has been the subject of time-resolved X-Ray spectroscopy studies at XFELs in the past. It's response to intense pulse in the infrared domain, in which mostly intramolecular vibrations are excited, and the subsequent energy dissipation processes, have been extensively studied in the past. At low frequency, THz light couples to low energy collective modes of the liquid.

Here, we investigated the response of liquid water to one-cycle,200fs long THz pulses spectrally centered at about 100/cm (~3THz). The THz pulse does not target any particular mode of the liquid. At an intensity of about 10^(10) W/cm^(-2), the pulse transfers energy mostly to translational modes of the water monomers along the polarization axis of the electric field. In a time-scale of 500fs to 1ps the energy redistributes to hindered rotational modes first, and to intramolecular vibrations last. This implies that the energy supplied by the THz can potentially activate chemical processes long before the large amount of energy supplied leads to volume increase and vaporization of the medium. Radial Distribution function and X-Ray diffraction pattern of water at certain time intervals are showing that water looses tetrahedral hydrogen bond structure with time due to pulse. Water reaches to a quasi-equilibrium state which is gas like very hot liquid. In this study, we have used CP2K package for Ab-Initio Molecular Dynamics.

We are also investigating the energy gain by Phenol molecule in an environment of liquid water by THz pulse.

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Session Classification: Molecular dynamics 2

Track Classification: 3

Contribution ID: **7** Type: **not specified**

Investigating the role of spin-lattice coupling in the ultrafast demagnetization of GdTb alloys

In ultrafast demagnetization of ferromagnetic metals after femtosecond laser excitation, a central question is how spin angular momentum can be transferred under the optically excited nonequilibrium conditions. One would expect this to happen on the timescale of spin-lattice relaxation, which has however been found to be on the order of 100 ps for Gd [1], much slower than ultrafast demagnetization.

We aim to clarify the role of spin-lattice coupling in femtosecond laser-induced demagnetization by investigating Gd, Tb and their alloys as a model system. By alloying Gd and Tb, the strength of spin-lattice interaction can be tuned from the weak 4f-lattice coupling of pure Gd, which is mediated by the 5d conduction electrons, to the strong direct spin-lattice coupling of pure Tb. Pure Gd and Tb show laser-induced demagnetization in two steps, with the time constant of the second, slower step depending on the strength of coupling of the 4f magnetic moments to the lattice [2].

In time-resolved magneto-optical Kerr effect measurements on GdTb alloys, we see a decrease of the time constant of the second demagnetization step from 33 ps to 9 ps with the Tb content increasing from 0 to 70%, which we assign to the continuous increase of spin-lattice coupling due to the growing Tb fraction. In time- and element-resolved x-ray magnetic circular dichroism measurements at the BESSYII Femtoslicing concomitant dynamics of the Gd and Tb 4f magnetic moments is observed. Compared to pure Gd, the Gd magnetic moments in the alloy experience a coupling to the lattice which is increasing with the Tb content. Consequently, the second step of demagnetization of Gd in the alloy is accelerated compared to the pure material. In contrast, the time constant of the first demagnetization step, which is found to vary between 1.8 ps and 1.2 ps, is not correlated to the amount of Tb in the alloy. These results are in good agreement with our previous conclusion [2] that the faster demagnetization time constant is determined by relaxation of 5d electrons, while the slower timescale is set by the efficiency of spin-lattice coupling and the transfer of the angular momentum change to the lattice.

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Session Classification: Quantum materials

Track Classification: 3

Contribution ID: **8** Type: **not specified**

Magnetization Compensation Point and All-Optical Switching

Understanding ultrafast all-optical magnetization switching (AOS), i.e. the permanent reversal of the magnetization by the sole action of a femtosecond laser pulse in the absence of any applied magnetic field, is a challenging issue which could have tremendous impact for the magnetic recording industry. While a qualitative agreement between spin atomistic simulations and experiments exists[1], the exact role played by the magnetization compensation point TM in the ultrafast demagnetization[2] and switching[3] in ferrimagnetic rare-earth transition-metal alloys is still not completely clear. By combining femtosecond X-ray transmission measurements with picosecond time-resolved photo-emission electron microscopy (PEEM), both using X-ray magnetic circular dichroism, we report on new insights into the AOS mechanism in GdFe based ferrimagnetic alloys. In agreement with previous experiments and theoretical predictions, AOS is seen below and above TM, and in particular against a 0.18 T magnetic field. However, at temperatures far from TM, no AOS is observable. Collapse of the reversed domain could be ruled out using time-resolved XMCD PEEM imaging. Static imaging of the magnetic domain configuration after AOS reveals that no domain wall (DW) motion occurs within the 100 nm spatial resolution, ruling out a nucleation and growth switching mechanism favored by the DW velocity divergence at TM. Furthermore, investigation of the formation speed of the transient ferromagnetic-like state as a function of TM shows very pronounced variations. These results provide evidence that the TM is somehow a more important condition for the formation of the transient ferromagnetic-like state[4] and the occurrence of AOS than initially thought.

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Session Classification: Molecular dynamics 2

Contribution ID: **9** Type: **not specified**

Non-linear effects in x-ray emission of liquid water with ultra-high fluence at LCLS

The oxygen K-edge x-ray emission (XE) spectrum of liquid water at ultra-high x-ray fluences was studied at the SXR Instrument of the x-ray free electron laser (FEL) LCLS. XE spectra were measured from a liquid micro-jet in vacuum for a wide range of x-ray fluences by varying x-ray spot size, flux and pulse duration. The threshold fluence for reproducing reference data measured at BESSY II could be determined to be approximately 0.1 J/cm^2 for 100 fs long LCLS x-ray pulses. For higher x-ray fluences, strong deviations from the reference spectrum were discovered. A non-linear behavior of the emission intensity with increasing incident x-ray fluence and a distortion of the spectral shape were observed in particular. These can be modeled by taking into account the reabsorption of x-ray emission by valence-excited states. We propose a mechanism by which x-ray emission photons from core-hole decay are reabsorbed in the sample by water molecules in valence-excited states formed via auger decay. This novel effect of valence-excited state absorption competes with other non-linear effects such as stimulated x-ray emission. Due to the much longer valence-hole lifetime compared to the core-hole lifetime it becomes important already for comparably low fluences.
The data and the model will be presented and implications as well as possible applications for future XE studies at FELs will be discussed. More than that, experimental parameters and geometries for minimizing valence-excited state absorption and maximizing stimulated x-ray emission will be presented.

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Session Classification: X-Ray interactive with matter 1

Track Classification: 1

Contribution ID: **10** Type: **not specified**

Femtosecond RIXS of Fe(CO)5 in ethanol –resolving ultrafast excited state and ligand substitution dynamics in solution

Using ultrashort X-ray pulses from the Linac Coherent Light Source we carried out a pump-probe resonant inelastic x-ray scattering (RIXS) experiment on Fe(CO)5 photodissociation in ethanol. This allowed us to follow the evolution of the valence electronic structure of the iron carbonyl species in an element- and chemical-selective way with a 300 fs (FWHM) time resolution. Supported by novel ab initio electronic structure calculations, we were able to identify the relevant reaction steps all the way from excited-state relaxation involving changes in the spin state to subsequent complexation with solvent molecules of the first coordination sphere.

Upon charge transfer excitation with 266 nm laser light the Fe(CO)5 molecule relaxes to a low-lying excited ligand field state which, due to the Jahn-Teller effect, leads to dissociation of one CO ligand. We identified that such relaxation is very fast (<100 fs) and results in the creation of electronically excited Fe(CO)4 as a primary photoproduct. The electronically excited molecule could be detected via a characteristic anti-Stokes RIXS peak at a negative energy transfer of -2 eV. This primary photoproduct then relaxes in only 150 fs to the triplet ground state of Fe(CO)4. Although in the electronic ground state, triplet Fe(CO)4 is highly reactive. Due to the missing CO ligand it is sterically unsaturated and lacking electrons in the valence shell. Reactive collisions with the hydroxyl groups of the ethanol molecules in the first solvation shell therefore lead to rapid complexation. As a result of the large amount of vibrational excess energy, a dynamical quasi-equilibrium between Fe(CO)4 and Fe(CO)4EtOH is established in less than 1 ps. Subsequent cooling of the system, which is expected to take 10-20 ps, leads to formation of final "cooled" Fe(CO)4EtOH product.

In addition to the well-known high chemical sensitivity of RIXS, we have experimentally demonstrated here the potential of femtosecond RIXS as a tool to probe excited state dynamics. This follows from the fact that RIXS probes directly the spectrum of valence states referenced to the energy of the RIXS initial state. Therefore RIXS from electronically excited states leads to appearance of characteristic anti-Stokes peaks well separated from the parent molecule signal. We believe that femtosecond RIXS can become a very valuable technique in resolving complex excited-state dynamics.

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Session Classification: X-Ray interactive with matter 1

Track Classification: 2

Contribution ID: **11** Type: **not specified**

Photoinduced charge transfer dynamics in coordination compounds

Coordination compounds are appealing targets for the development of light-harvesting devices. Currently Ru(II)-complexes are widely used but Cu(I)-compounds, in particular Cu(I)-phenanthrolines, are considered as promising alternative. In comparison Cu is more cost effective and environmentally friendly [1]. The metal-to-ligand-charge-transfer (MLCT) excited state of these Cu(I)-phenanthroline complexes is known to undergo structural reorganization, with the pseudotetrahedral D2d symmetry in the ground state changing to a flattened D2 symmetry in the MLCT state. This promotes ligation with solvent molecules, forming a so-called exciplex intermediate [2], dramatically shortening the lifetime of their excited state. Therefore, sterically active ligands could potentially be used to control the MLCT excited state properties of these compounds [3]. Using fs transient optical absorption spectroscopy, we have studied the ultra-fast kinetics, structural and electronic dynamics of new Cu(I)-phenanthroline complexes with varying degrees of steric hindrance in solution to identify structural origins of transient spectroscopic changes after their photoinduced MLCT excitation. We have demonstrated that for complexes with bulkier ligands a subpicosecond rising component is absent, which suggests that the Jahn–Teller distortion observed in other Cu(I)-phenanthroline complexes is effectively blocked by these functional groups. This implies that the pseudotetrahedral D2d symmetry is maintained and the solvent accessibility is reduced. These results together with our recent static X-ray emission spectroscopy experiments provide guidance for future X-ray structural studies in the ultrafast time regime, as well as for synthesis facilitating its applications in solar energy devices.

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Session Classification: Molecular dynamics 1

Contribution ID: **12** Type: **not specified**

Photoinduced Lattice Dynamics in BiFeO3 monitored by Femtosecond X-ray Diffraction

BiFeO3 had a deep impact in the field of multiferroics, since it is magnetic and ferroelectric at room temperature, opening a wide field of applications, e.g. for spintronics and memory devices which can be addressed both magnetically and electrically. Furthermore, it is highly desirable to photo-control the polarization and magnetization in BiFeO3 directly by ultrafast optical excitation.
Here we use femtosecond laser pulses with a photon-energy of 3.1eV (λ = 400nm) to excite a 40nm BiFeO3 thin film above its band gap of 2.8eV. Ultrafast X-ray diffraction (UXRD) at a laser-driven Plasma X-Ray Source (PXS) is applied to follow the photoinduced lattice dynamics on a sub-picosecond timescale. We observe a sound velocity-limited evolution of the strains in the excited BiFeO3 within 10ps indicating an instantaneous stress, which further decays on a ns time scale. From the considerable Bragg peak broadening we can conclude that the photovoltaic origin, driving the ultrafast lattice dynamics, has an inhomogeneous spatial distribution for early (fs) as well as late (ns) time scales after excitation, which is a direct evidence for trapped charges in the thin film.

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Session Classification: Quantum Materials 2

Track Classification: 1

Contribution ID: **13** Type: **not specified**

Following Strain-Induced Mosaicity Changes of Ferroelectric Thin Films by Ultrafast Reciprocal Space Mapping With a Convergent Beam

We investigate coherent phonon propagation in a thin film of ferroelectric Pb(Zr0.2Ti0.8)O3 (PZT) by ultrafast X-ray diffraction (UXRD) experiments, which are analyzed as time-resolved reciprocal space mapping (RSM) in order to observe the in- and out-of-plane structural dynamics simultaneously.[1] The mosaic structure of the PZT leads to a coupling of the excited out-of-plane expansion to in-plane lattice dynamics on a picosecond timescale, which is not observed for out-of-plane compression. In our opinion the ultrafast reciprocal space mapping (URSM) technique is a next key part to fully understand the coupling between physical and structural dynamics in PZT and other domain forming materials.
Furthermore, we present a detailed characterization of the utilized diffractometer setup. We show that the resolution function of the diffractometer is determined by the convergence and energy bandwidth of the incoming X-rays. For quasi-perfect crystalline samples with insignificant in-plane Bragg peak broadening we can correct the measured reciprocal space maps for the known resolution function of the diffractometer in order to achieve high resolution rocking curves with improved data quality. For this case, the resolution of the diffractometer is not limited by the convergence of the incoming X-ray beam but is solely determined by its energy bandwidth.

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Session Classification: Molecular dynamics 1

Track Classification: 1

Contribution ID: **14** Type: **not specified**

Ultrafast dynamics in antiferromagnetic materials

Antiferromagnetic (AFM) materials have been predicted to show ultrafast magnetic dynamics since no macroscopic magnetization exists and dynamics might not be restricted by conservation of angular momentum. Out of recent studies investigating ferromagnetic materials like pure 4f elements (Gd,Tb,..) or a combination of 3d and 4f elements with (GdFeCo,..) it turned out that that the fast demagnetization time of the pure 4f elements Tb and Gd is around 750fs and in the combination with 3d's it is around 430fs. [1,2].

We addressed antiferromagnetic dynamics in the two 4f systems Ho and EuTe. Holmium is a helical antiferromagnetic metal while EuTe is a semiconductor with collinear antiferromagnetic order. Our optical pump, time-resolved resonant soft x-ray magnetic diffraction experiments have been performed at the FEMTOSPEX facility at BESSY II and at the SXR-beamline at LCLS, Stanford. The dynamics in Ho was mapped using the strong resonant x-ray scattering signal of the helical magnetic (00τ) superstructure reflection. It is generated by FM-planes whereby there is a turning angle of around 45° between each layer along the c-axis. The dynamics in EuTe are measured on the (½, ½, ½) superstructure reflection which comes from a doubled unit cell along the (111) as every Eu layer is antiferromagnetically ordered and spaced by a not magnetic Te layer.

We found, that independent of its individually magnetic moment Ho and EuTe can loose their magnetic order in a sub ps regime, 200fs for Ho and down to 450fs for EuTe. For both materials this is surprisingly fast but is in the first order directly explainable due to the fact that there is not net-orbital momentum which has to be transferred in contrast to the FM-materials.

By recording time resolved q-scans (mainly in EuTe) we find that the thin film magnetic profile is modified in a completely different way than for elevated temperatures in thermal equilibrium. This gives insight that also the structure itself is involved in the disordering process. We addressed this point with the measurements at the LCLS looking on a structural Bragg reflection of EuTe.

In summary we present ultrafast demagnetization dynamics in two antiferromagnets at low temperatures with surprising insights in this relatively new field looking on the delay and spatially resolved scans.

Supported by the BMBF through contract 05K10PK2.

Literature:

[1]M.Wietstruk etal,PRL106,127401(2011)

[2]I.Radu etal, Nature205,472(2011)

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Session Classification: Quantum materials

Track Classification: 4

Contribution ID: **15** Type: **not specified**

Engineering Ultrafast Magnetism

Controlling magnetic states of matter on ultrashort timescales is crucial to engineering the next-generation magnetic devices combining ultrafast data processing with ultrahigh-density data storage.

Here, we report on femtosecond laser-driven dynamics of multi-sublattice magnetic materials, with both ferromagnetic and antiferromagnetic coupling between sublattices, investigated using element-specific, femtosecond time-resolved XMCD. These measurements [1], fully supported by phenomenological and atomistic spin simulations, provide evidence for a demagnetization time that scales with the elemental magnetic moment and varies with the sign of the exchange interaction. As such, one can control the speed of magnetization processes in multi-sublattices materials, being either switching or demagnetization, by properly choosing the magnitude of the constituent magnetic moments and the sign of the exchange interaction that couples them, as exemplified for the case of a synthetic ferrimagnet.
[1] I. Radu et al., (submitted)

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Presenter: RADU, Ilie (Helmholtz-Zentrum Berlin für Materialien und Energie)

Session Classification: Quantum Materials 2

Track Classification: 4

Contribution ID: **16** Type: **not specified**

2PPE measurement on Sb2Te2S with an angle-resolving time-of-flight spectrometer

We present two-photon photoemission measurements on the p-doped topological insulator Sb2Te2S. The Dirac cone of this system lies above the Fermi energy and is therefore only accessible using two-photon photoemission.

We detect the photoemitted electrons with an angle-resolving time-of-flight spectrometer. This instrument allows us to measure the kinetic energy E as a function of both k directions without rotating the sample. Our laser system with two OPAs gives us the opportunity to tune the photon energy of the infrared and ultraviolet pulse independently. This allows us to create a resonant population and efficient probing of the Dirac cone.

Sb2Te2S shows a Dirac cone with direction-dependent linear dispersion close above the Fermi level and hexagonal warping at higher energies. Polarization-dependent measurements manifest the spin texture of the Dirac cone.

The population dynamics of Sb2Te2S have been analyzed. The femtosecond lifetimes of the first and second image-potential states show an n^3 dependence. The Dirac cone is populated by indirect filling via the conduction band. Therelaxation of the Dirac cone happens on a picosecond timescale.

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Session Classification: Molecular dynamics 1

Track Classification: 2

Contribution ID: **17** Type: **not specified**

Ultrafast Magnetization Dynamics of Gadolinium: Towards a Complete Picture

We designed and performed a time- and angle- resolved photoemission experiment to investigate the electronic structure of a ferromagnetic Gadolinium film during ultrafast laser-induced demagnetization and subsequent thermal recovery.
To get a complete picture of the magnetization dynamics we track the binding energy of the exchange-split valence band, which mediates the exchange interaction between the 4f orbitals for adjacent atoms and investigate in addition in the same experiment the polarization of the 4f system by magnetic linear dichroism. This "complete"picture of the magnetic system allows us to evaluate current theories of magnetization dynamics. Comarison with published MOKE and XMCD data give insight to the magnetic properties measured by these methods in the non-equilibrium regime during the first picoseconds.

Methods: We use a 10kHz Ti:sapphire laser system (1.5 mJ/pulse) to create high harmonic radiation in an argon gas. For the presented data we used the 23rd harmonic at 36eV photon energy to perform time- and angle- resolved photoemission. The desired harmonic is selected by an XUV monochromator, giving an ultimate energy resolution of approximately 100 meV.
The 10nm thick Gadolinium film was grown epitaxially on W(110) by e-beam evaporation. In plane magnetization of the sample is done without changing the position of the sample by a free standing coil.

Ref.: Carley et al. Phys. Rev. Lett. 109 057401 (2012)

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Session Classification: Quantum Materials 2

Track Classification: 5

Contribution ID: **18** Type: **not specified**

Coupling of GHz phonons to ferroelastic domain walls in SrTiO3

SrTiO3 (STO) is a dielectric perovskite that shows quantum paraelectric behavior at low temperatures. It is widely used as a substrate for the growth of perovskite layers and has been intensively studied for its various structural phase transitions. The elastic behaviour of STO has been studied since the 60ies and has recently attracted attention due to the high mobility of domain walls in the antiferrodistortive phase.

We use time domain Brillouin-scattering techniques for studying the linear and nonlinear acoustic response of STO across its ferroelastic transition at Tc = 105 K. We excite a metallic transducer film on the surface in order to generate high amplitude strain pulses into the STO crystal underneath. Using ultrafast X-Ray-diffraction we are able to observe the expansion of the transducer and thus determine the induced transient strain in STO. Above Tc we observe that for a strain amplitude of 0.18% the sound velocity for compressive strain exceeds the tensile strain velocity by 3%. This behavior can be modeled in a linear chain and is explained by the anharmonicity of the interatomic potential. Below Tc we observe a giant slowing down of the sound velocity by 12% which we interpret as superelastic response due to the coupling of the GHz phonons to ferroelastic domain walls. This has so far only been observed for lower frequency phonons. We conclude that this phenomenon occurs for GHz sound only when the strain amplitude is large enough to couple to domain walls because the unit cell deformation exceeds the tetragonal distortion of the domains. We argue that the relevant time scale that makes superelastic behavior possible also for low strain amplitudes at low frequencies is determined by the time T = D/vs it takes sound to propagate through the average size D of the domains. The observed phenomena can also be interpreted in terms of a transient phase which is established under high positive and negative pressure, however, we prefer a discussion as a non-equilibrium process in a domain-forming phase.

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Session Classification: Molecular dynamics 1

Contribution ID: **19** Type: **not specified**

High amplitude coherent phonons and their nonlinear behavior

We use inelastic scattering of ultrashort visible and hard X-ray pulses as a probe of the transient occupation of phonon modes constituting large amplitude strain-wavepackets. These Brillouin scattering experiments permit the time-resolved observation of the phonon dynamics including the phonon damping and nonlinear interaction. We use a variety of excitation schemes to synthesize high amplitude phonon wavepackets. By pumping a nanometric metal layer with one or multiple pulses we can shape the imprinted phonon spectrum. Such high strain fields can lead to several anharmonic phenomena like sum and difference frequency mixing, which could be termed "nonlinear phononics"in analogy to nonlinear optics. These nonlinear phenomena depend strictly on the excited phonon spectra. For high amplitude single cycle phonon pulses we have observed the self-steepening of the pulse fronts.[1] Spectrally narrow phonon wavepackets exhibit difference- and sum-frequency generation. We quantitatively model the dynamics by coupled anharmonic oscillators. New measurements of high amplitude strain pulse propagation in SrTiO3 at the phase transition of 105K reveal a superelastic behavior which is connected to the coupling of the high pressure sound pulse to the motion of domain walls. We think our new insights in the field of nonlinear phononics could help to establish new ultrafast experiments in which the direct optical excitation of a material by an ultrashort laser pulse is replaced by a highly intense ultrashort pressure pulse which excites the material triggering the dynamics by electronic excitation.

[1] Bojahr et al, PRB 2012 86(144306)

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Session Classification: Molecular dynamics 1

Contribution ID: **20** Type: **not specified**

Ultrafast Magnetization Dynamics Viewed from the Non-equilibrium Band Structure

We present recent results from ultrafast laser-driven magnetization dynamics experiments on the rare-earth local-moment ferromagnets Gadolinium and Terbium. Our experiment combines angle-resolved photoemission spectroscopy (ARPES) with the VUV photon energies and time resolution made possible by high-order harmonic generation to scrutinize the non-equilibrium band structure following excitation of the ferromagnet by a short infrared pulse. On the timescale of a few picoseconds, transient changes of the minority and majority components of the exchange-split valence band reveal significant insight into the underlying microscopic processes. Furthermore, linear magnetic dichroism in photoemission allows us to simultaneously follow the magnetic response of the localized 4f electrons, from which the magnetic moment arises.

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Session Classification: Molecular dynamics 1

Contribution ID: **21** Type: **not specified**

Ultrafast reemergence of microscopic spin order in GdFeCo

Over the past decade, femtosecond optical laser pulses have fueled the discovery of ultrafast spin dynamics in magnetic materials that go beyond our equilibrium understanding of magnetism. Many of these phenomena are potentially suitable for future technological applications. For example, fs optical laser pulses can act as a stimulus to reverse the magnetization direction in ferrimagnetic GdFeCo, a phenomenon called all-optical switching [1]. However, much remains to be understood about these non-equilibrium spin dynamics, particularly how they manifest on nanometer lengthscales where magnetic order emerges. Further, our recent work on GdFeCo utilizing ultrafast x-ray diffraction showed that the non-equilibrium spin dynamics on the nanoscale can evolve very differently from the bulk [2]. Using the ultrabright and ultrafast x-ray pulses from the Linac Coherent Light Source (LCLS), we probed the fs magnetic response of GdFeCo with spatial resolution down to 10nm at both the Gd M5 (1190 eV) and Fe L3 (707 eV) absorption edges. Our results showed that GdFeCo displays nanoscale chemical and magnetic inhomogeneities that affect the spin dynamics. In particular, we observed Gd spin reversal in Gd-rich nanoregions within ~ 1 picosecond following optical excitation. These nanoregions of Gd 4f spin reversal result from a non-local flow of angular momentum originating from Fe 3d spins in surrounding areas. In addition, new time-resolved x-ray diffraction results from LCLS show that this nanoscale spin order not only persists against the disorder caused by the optical excitation, but that the reversed nucleated areas grow in size for the first 10ps. In this talk, I will discuss our recent work demonstrating the non-local flow of angular momentum with ultrafast x-ray diffraction measurements. I will then present our new results, which show the reemergence and growth of nanoscale spin order following optical excitation in GdFeCo.

[1] Stanciu, C. D. et al. All-Optical Magnetic Recording with Circularly Polarized Light. Phys. Rev. Lett. 99, 047601 (2007).

[2] Graves, C.E., Reid, A.H., et. al. Nanoscale spin reversal by non-local angular momentum transfer following ultrafast laser excitation in ferrimagnetic GdFeCo. Nature Mater. 12, 293 (2013).

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Session Classification: Quantum Materials 2

Track Classification: 6

Contribution ID: **22** Type: **not specified**

Electron Dynamics in Self-Assembled Monolayers of Azobenzene-Functionalized Alkanethiols on Gold

Charge and energy transfer processes at organic-metal interfaces are of key relevance in molecular electronics and functionalization of surfaces with molecular switches. Therefore the electronic structure and femtosecond electron dynamics of self-assembled monolayers (SAMs) of azobenzene-functionalized alkanethiols on gold have been investigated with time-resolved two-photon photoemission spectroscopy (2PPE).

Two excited electronic states at 2.4 and 3.6 eV above the Fermi level are attributed to unoccupied states in the chromophores, which are populated via charge transfer from the metal substrate. Their lifetimes are in the order of 100 fs for an alkyl chain length of 3 CH2 units. The same timescale has been deduced from resonant core-level spectroscopy for the charge transfer across such an alkyl chain from excited molecular states ~1 eV above the Fermi energy.

In the densely packed monolayer of the azobenzene derivative, photoisomerization is strongly suppressed due to interaction among the azobenzene moieties. Mixing the azobenzene derivative with an alkanethiol allows to introduce free space into the SAM and thereby to control the chromophore interactions. Accordingly the optical properties and the switching ability can be tuned.

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Session Classification: Molecular dynamics 1

Contribution ID: **23** Type: **not specified**

Verwey Transition in Magnetite: How fast does an insulator become a metal?

Magnetite (Fe3O4), is the first oxide where a relationship between electrical conductivity and fluctuating/localized charges was observed, with a drop in conductivity by two orders of magnitude at T=123K. The Verwey transition is accompanied by a structural change from monoclinic to cubic symmetry. Despite decades of research and indications that charge and orbital ordering play an important role, the mechanism behind the Verwey transition is yet unclear. Recently, three-Fe-site lattice distortions called trimerons have been identified as the true microscopic face of electronic order in low temperature insulating phase. We studied the real time response of insulating magnetite to optical excitation with ultrafast soft X-ray scattering. We discover this to be a two-step process. After an initial femtosecond destruction of individual trimerons in the corresponding lattice, we observe a phase separation into residual insulating trimeron and cubic metallic phases on a 1.5 ± 0.2 picosecond timescale.

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Session Classification: Quantum Materials 2

Contribution ID: **24** Type: **not specified**

Electron Dynamics in photo-excited Sodium Iodide in the gas phase

Following the pioneering femtosecond spectroscopy experiments by A. Zewail and coworkers, the coherent electronic and nuclear wavepacket dynamics in photo-excited NaI molecules are revisited by means of sub-100 fs resolution pump-probe photoelectron spectroscopy. Time, energy and angular resolved photoelectron distributions of NaI photo-excited with several pump wavelengths were been measured. The dynamic pathways in the energy-time landscape represent a full picture of the molecular wavepacket evolution upon pumping and directly reveal features such coherent superposition of and transfer of wavepacket population between different molecular states. Furthermore, the dispersion of the evolving wavepacket and the dependence of this dispersion on the initial pump energy can be seen in the spectra. The measurements represent a stringent test for current models and theories and will be compared to state of the art molecular quantum dynamics simulations.

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Session Classification: Molecular dynamics 1

Track Classification: 3

Contribution ID: **25** Type: **not specified**

Investigating Nanoscale Structure via Coherent Resonant X-Ray Scattering

Correlated materials such as transition-metal oxides exhibit nanoscale structures resulting from e.g. magnetic, orbital or charge order. While resonant scattering of soft x-rays in transmission geometry provides unique sensitivity to such ordering and can also enable high resolution imaging, most samples have to be grown epitaxially on single crystalline substrates and are not penetrable by soft x-rays in this configuration.

In this contribution we report on different approaches to make such standard samples suitable for transmission imaging experiments, having in particular resonant x-ray holography in mind. The goal is to thin the substrate to a few hundreds of nanometers over a sufficiently large transverse region. We focus on thin films of magnetite (Fe3O4) on MgO (100) as a prototypical sample system of interest. Two different thinning approaches are investigated: Mechanical thinning similar to TEM sample preparation as well as etching. Given that the transmission depends exponentially on the thickness and that we want to achieve “imaging quality”with respect to lateral thickness variations, we characterize the resulting substrate profiles using different microscopies. Furthermore, first soft x-ray SAXS results on MBE-grown magnetite films prepared in this way are presented.

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Presenter: Prof. EISEBITT, Stefan (Max-Born-Institute)

Session Classification: Molecular dynamics 1

Contribution ID: **26** Type: **not specified**

Time-resolved Imaging of the Gyrotropic Motion of Magnetic Bubbles

Magnetic bubbles are skyrmionic spin structures stabilized by dipolar interactions. Recent theoretical investigations predict a GHz gyrotropic motion of these topological configurations after excitation in a restoring potential, analogous to vortex gyration. However, in contrast to vortices, bubbles are predicted to exhibit inertial effects. Here we demonstrate the presence of an inertial mass in a magnetic bubble by imaging its gyrotropic trajectory using pump-probe x-ray holography. We find that the inertial mass is very large compared to other magnetic systems, which we attribute to the non-local energy reservoir of the bubble's breathing mode. The breathing mode is a unique feature of the geometrically confined skyrmionic spin structure, which is a direct consequence of its topology, and thus lends itself to describe the inertia of Skyrmions in terms of a topological mass.

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Session Classification: Molecular dynamics 1

Contribution ID: **27** Type: **not specified**

Investigating Nanoscale Structure via Coherent Resonant X-Ray Scattering

Correlated materials such as transition-metal oxides exhibit nanoscale structures resulting from e.g. magnetic, orbital or charge order. While resonant scattering of soft x-rays in transmission geometry provides unique sensitivity to such ordering and can also enable high resolution imaging, most samples have to be grown epitaxially on single crystalline substrates and are not penetrable by soft x-rays in this configuration.

In this contribution we report on different approaches to make such standard samples suitable for transmission imaging experiments, having in particular resonant x-ray holography in mind. The goal is to thin the substrate to a few hundreds of nanometers over a sufficiently large transverse region. We focus on thin films of magnetite (Fe3O4) on MgO (100) as a prototypical sample system of interest. Two different thinning approaches are investigated: Mechanical thinning similar to TEM sample preparation as well as etching. Given that the transmission depends exponentially on the thickness and that we want to achieve "imaging quality"with respect to lateral thickness variations, we characterize the resulting substrate profiles using different microscopies. Furthermore, first soft x-ray SAXS results on MBE-grown magnetite films prepared in this way are presented.

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Session Classification: Molecular dynamics 1

Contribution ID: **28** Type: **not specified**

A New Era of X-ray Science: Beyond one-photon-at-a-time

My talk will give a historic perspective of the revolutionary science that was enabled by the advent of high power sources of coherent electromagnetic radiation and the implications for future scientific opportunities with x-ray free electron lasers (X-FELs). The historical journey starts with the development of radar microwave sources in the 1940s that fueled the development of nuclear magnetic resonance (NMR) techniques which by now have led to 6 Nobel Prizes. The theoretical description of NMR as coherent processes between nuclear states by Rabi and Bloch also provided the theoretical basis for the optical laser and its applications. Over the last fifty years, many of the 10 Nobel prizes based on lasers are based on unique non-linear interactions of light with valence electrons. This unprecedented electronic control is made possible by the large photon degeneracy parameter of lasers in specific modes with many photons simultaneously participating in the interaction process. These historic developments suggest that another revolution is upon us, based on non-linear x-ray interactions induced by X-FELs. The higher photon energy of such x-ray lasers generates element and chemical state specific electronic core-to-valence transitions and the high X-FEL degeneracy parameter allows manipulating and controlling them within the femtosecond core hole lifetime through multi-photon stimulated interactions. This capability is completely new and was unavailable during the first 100+ years of x-ray science. I will discuss the concepts of describing non-linear x-ray interactions and at what x-ray fluences they become important. I will also discuss what beam parameters and instrumentation are required to advance this new area of science and mention associated with chemical reactivity and energy transfer.

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Session Classification: X-Ray interactive with matter 1

Contribution ID: **29** Type: **not specified**

HIKE: the HAXPES Facility at BESSY II

In the last decade hard x-ray high kinetic energy photoelectron spectroscopy (HAXPES or HIKE) has led to a break-through in the field of photoemission due to its non destructive way of investigating the bulk electronic properties of materials and in particular buried interfaces in layered systems. Nanolayered systems are the keystones of current and future devices. The electronic, magnetic and magneto-transport properties of these structures are determined not only by the characteristics of each layer, but also by their interfaces.
The HIKE facility at the Berliner synchrotron light source BESSY II successfully combines the bending magnet source of the KMC-1 beamline [1] with a new generation electron spectrometer optimized for high kinetic energy electrons. In the present contribution several experiments performed at the HIKE HAXPES user facility [2] will be detailed with emphasis on the performance and abilities of the technique.

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Session Classification: Molecular dynamics 1

Contribution ID: **30** Type: **not specified**

Quantification of finite-temperature effects on adsorption geometries of π-conjugated molecules

Experimentally determined adsorption geometries of molecular switches are essential both for understanding their functionalities and for benchmarking ab initio calculations. The prototypical molecular switch azobenzene is investigated on the Ag(111) surface by means of the normal incidence x-ray standing wave (NIXSW) technique and dispersion-corrected density functional theory (DFT) calculations.

We show that, besides the average positions of each chemical species, the molecular geometry can be retrieved from the coherence of the NIXSW signal using a new analysis method generally applicable to all molecular adsorbate. In this way azobenzene is found to exhibit a substantial torsion in a dense monolayer. The inclusion of non-local collective substrate response (screening) in the dispersion correction scheme improves the description of azobenzene adsorption geometry. Nevertheless, we show that for a quantitative agreement with experiment explicit consideration of the (hitherto generally neglected) effect of vibrational mode anharmonicity due to finite temperature on the molecular geometry is crucial.

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Session Classification: Molecular dynamics 1

Contribution ID: **31** Type: **not specified**

New insights into ultrafast molecular dynamics with femtosecond pulses of a higher harmonic source

The current status of project P2 "Ultrafast charge transfer and isomerization dynamics of molecules" will be presented. The goal is to resolve the early-stage dynamics of energy relaxation concomitant with structural rearrangements in Fe(CO)5 in the gas phase. Photoexcitation generates a metal-to-ligand charge transfer in Fe(CO)5 that launches a sequence of processes on time scales of 20-30 fs before the molecule dissociates after approximately 150 fs. The aim is to resolve these early steps with pump-probe valence-band photoelectron spectroscopy with femtosecond VUV pulses from high-order harmonic generation. The table-top source for this and an outline of the principles of higher harmonic generation and of the Fe(CO)5 experiments will be presented.

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Session Classification: Molecular dynamics 1

Contribution ID: **32** Type: **not specified**

Lattice Response to Femtosecond Laser Excitation Studied by Time Resolved Transmission Electron Diffraction

Lattice vibrations after femtosecond laser excitation can be launched by different processes such as the relaxation of hot electrons or the coupling of the laser pulses to the lattice via nonlinear-optical mechanisms. The pathway and time scale of energy transfer form photonic excitation to the phononic subsystem is of fundamental interest to understand various physical phenomena like e.g. laser-driven structural phase transitions. Time resolved transmission electron diffraction (TED) is an ideal tool for these investigations. The different excitation mechanisms produce distinct signatures in the time evolution of the electron diffraction pattern. The possibility of observing many diffraction spots simultaneously enables us to identify the signature the atomic motion. Here we report on experimental results of lattice excitation by time resolved electron diffraction and the analysis of disentangling disordered thermal motion and directional atomic displacement in Bismuth and Nickel.

The experiments were performed on free-standing single crystalline thin films of either 22 nm Bismuth with (001)surface orientation (hexagonal notation) or 20 nm Nickel with (001) surface orientation. The electron probe pulses had an energy of 30 keV, and the pulse duration was less than 700 fs. The samples were laser-excited using pulses of 800 nm at a repetition rate of 1 kHz with 2 or 4 mJ/cm^2 incident fluence for Bismuth or Nickel, respectively.

The diffraction patterns were recorded as function of delay time between optical pump and electron probe. The integrated intensity of all the diffraction spots decreased after laser excitation. The ultrafast lattice heating can be described by the transient Debye-Waller effect, which treats the lattice displacement as disordered thermal motion by fitting the decrease of the diffraction intensities to the Debye-Waller factor for corresponding diffraction orders. The extracted time constants for Bi (4 ps) and Ni (2 ps) indicated strong material dependence of the ultrafast lattice heating. In addition a directional displacement could be studied by analyzing the temporal evolution of proper diffraction orders under specified diffraction conditions. The directed motion as longitudinal elastic waves in direction of the surface normal was observed in Nickel and Bismuth. We discuss the diffraction patterns and the possible driving mechanisms.

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Session Classification: Molecular dynamics 1

Contribution ID: **33** Type: **not specified**

Investigation of the Spectral Fingerprints of Phonons and Magnons in Ferrimagnetic Oxides using Fourier-transformed Infrared Spectroscopy

The low-energy excitations of magnetic solids, as for instance magnons and phonons, are ultimately responsible for dynamic magnetization phenomena like demagnetization and magnetization switching, and therefore of paramount importance for a microscopic understanding of these processes.
Here, we report on a systematic study of the spectral fingerprints of these quasiparticles on ferrimagnetic Ho3Fe5O12 garnet using the Fourier-transformed Infrared Absorption (FTIR) technique. The ultimate goal of the project is to understand the role of these quasiparticles and their coupling during the thermal- and laser-driven magnetization switching processes. The measurements have been performed in transmission geometry at the THz beamline of Helmholtz-Zentrum Berlin employing photon energies between 1.2meV to 12.4meV upon varying the temperature, magnetic field magnitude and the light polarization. This approach allowed us to identify and disentangle the magnetic and phononic excitations as well as their hybrid modes. In particular, we observe pronounced peaks in the absorption spectra centered at 27cm-1 and 28.5cm-1, which we ascribe to an antiferromagnetic resonance and to a hybrid phonon-magnon mode, respectively.

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Session Classification: Molecular dynamics 1

Contribution ID: **34** Type: **not specified**

Ultrafast surface dynamics probed with time resolved photoemission at FLASH

Time resolved photoemission from core levels (trPES) at FELs is a tool for studying surface dynamics in real-time. Basis for this is the sensitivity of core-levels to their chemical environment. By probing their spectral evolution after an ultra-short excitation one can get insight on both, electronically caused excitations (fs- up to several-ps time-scales) as well as responses of the crystal lattice sub-ps to hundreds of ps).

In the present study we monitored the evolution of the 4f core levels of clean Ir(111) after an ultra-short (<120fs) optical excitation (800nm) by means of trPES at a photon energy of 198eV. The experiments were performed at the Free Electron Laser FLASH at the plane-grating monochromator beamline PG2. The setup allowed us to resolve temporal changes with a precision down to 1,1ps.

By analyzing the recorded data on a shot-to-shot basis we are able to fully characterize and control spectral shifts and broadening induced by X-ray and 800nm induced space-charge effects.

We observe a pump induced broadening in the surface component of the 4f lines which fully relaxes after 2ps. The early-component (<1,1ps) of this spectral broadening corresponds to the appearance of sidebands whereas a remaining excitation in the post-sideband time-scales (>1,1ps) can be explained by an increased electronic temperature.

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Session Classification: Molecular dynamics 1

Contribution ID: **35** Type: **not specified**

Two color x-ray pump-probe experiment on GaAs(100)

Motivated by the x-ray induced ultra-fast transient drop of optical reflectivity of semi-conductors, we performed time-resolved photoemission (trPES) experiments with x-ray pump x-ray probe in order to understand the x-ray induced changes in the electronic structure of GaAs(100). To accomplish our measurements we were using the split-and-delay unit at PG2 beamline at the free-electron laser facility FLASH, which allows, due to the existence of high harmonics in FEL radiation, (jitter free) x-ray pump x-ray probe experiments (40eV pump (fundamental) / 120eV probe (3rd harmonic)). For our trPES measurements we were using a newly developed electron time-of-flight spectrometer (SPECS THEMIS 1000 WAL) including a novel four quadrant delayline detector, which allows high transmission and high repetition rate measurements on single shot basis (400bunches@1MHz).
Compared to prior trPES measurements done with conventional hemispherical analyzers and CCD-detectors the present data show improved space-charge analyzing capabilities caused by the high read out speed of the detector and the resulting possibility to analyze each FEL shot independent.
A first analysis of the time-resolved measurement show changes of the electronic structure on sub-ps timescale.

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Session Classification: Molecular dynamics 1

Contribution ID: **36** Type: **not specified**

Molecular dynamics in small molecules studied by wavelength-selected femtosecond XUV pulses

Since demonstration of high-order harmonic generation (HHG) by femtosecond laser pulses, time-resolved XUV spectroscopy has become an attractive tool to study molecular dynamics via detection of charged particles (electrons and ions). Information collected in these experiments is typically complicated due to broad spectral content of the HHG sources. In this contribution we present implementation of an HHG-based time-compensating XUV monochromator, which is used to spectrally filter the wavelength of the XUV radiation, while keeping the temporal pulse profile unchanged. We use this setup in combination with a velocity map imaging spectrometer to investigate time-resolved dynamics of dissociative ionization of N2 molecules close to double-ionization threshold (35-50 eV). It is demonstrated how wavelength selectivity of XUV together with charged-particle detection helps to unravel ionization pathways of the molecule and map high-lying potential energy surfaces with energy resolution comparable to accuracy of quantum chemistry methods available today. First results on dynamics in triatomic molecules, such as CO2 and NO2, will also be presented.

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Presenter: Dr KORNILOV, Oleg (Max-Born-Institute)

Session Classification: Molecular dynamics 2

Contribution ID: **37** Type: **not specified**

Molecular dynamics probed via strong-field ionization of state-selected molecules fixed in space

State-selected aligned and oriented molecular ensembles serve as ideal samples to study ultrafast molecular dynamics in the molecular frame. Possible probing mechanisms are the investigation of molecular-frame photoelectron angular distributions or the detection of structural changes via X-ray and electron diffraction.

We have developed techniques to manipulate the motion of molecules in cold supersonic beams using strong inhomogeneous electric and laser fields at 1 kHz repetition rate. State-, conformer-, and size-selection is achieved, for instance, using the electric deflector, the analogue of the Stern-Gerlach deflector and the electrostatic bender for charged particles. The state-selected molecules are aligned by strong laser fields or oriented in combination of laser fields and static electric fields. The laser pulses for alignment and orientation are provided by a kHz Ti:Sapphire laser system. The laser pulse length can be varied continuously between 50 fs and 500 ps. This allows for the manipulation of the rotational degrees of freedom of the molecules non-adiabatically (impulsively) as well as adiabatically, and the study of the intermediate regime.

Here, we will present our work on the alignment and orientation of iodobenzene and carbonyl sulphide (OCS). We will discuss our findings on the (non) adiabaticity of the alignment and orientation of OCS for various pulse lengths between impulsive and adiabatic alignment and orientation. Intruiging effects of state- and laser-intensity-specific alignment, orientation without alignment, and selective detection of molecular eigenstates with ultrashort laser pulses are observed.

These samples allow the recording of molecular-frame photoelectron angular distributions (MF-PADs) to image the electronic and nuclear structure. In pump-probe type experiments one can thus follow the dynamics of these controlled systems.

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Session Classification: Molecular dynamics 1