Final report

DinL: Leibniz Graduate-School

Dynamics in new Light

Young Investigators Workshop on Ultrafast Dynamics 2014 – Winklmoos Alm, Germany

Leibniz-Institute: Max Born Institute **for Nonlinear Optics und Short Pulse Spectroscopy – Berlin**

Reference number: SAW-2011-MBI-4 48

Project period: November 2011 – May 2015

Contact partner: **Prof. Dr. Marc Vrakking and Prof Dr. Martin Weinelt**

Content

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Technische Universität Berlin (TUB)	*Stefan Eisebitt, Andreas Knorr, Thomas Möller*
Universität Potsdam (UP)	*Peter Saalfrank, Matias Bargheer*
Fritz-Haber-Institut der Max-Planck-Gesellschaft (FHI)	*Ralph Ernstorfer, Angel Rubio, Julia Stähler, Martin Wolf*
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1. Executive Summary

The physical and chemical properties of matter are ruled by elementary processes that take place within a few femtoseconds (1 fs = 10^{-15} s). Ultrashort optical pulses and in particular the invention of the Ti:sapphire laser in 1982 made these timescales experimentally accessible. In recent years the wavelengths of ultrashort pulses have been pushed to the vacuum ultraviolet and X-ray regime. This has opened a plethora of new research areas at the frontier of ultrafast science.

Based on these new developments and the concomitant scientific challenges and opportunities the Max Born Institute (MBI) established the graduate school *Dynamics in New Light* (DinL), which was funded from November 2011 to May 2015. The mission of DinL was to improve the education in the field of ultrafast X-ray science for graduate students with a degree in physics or chemistry. To this end, we brought together research groups of the MBI, the Berlin Universities, the Helmholtz-Zentrum-Berlin and the Fritz-Haber-Institut Berlin. Thereby research in XUV/X-ray high-harmonic generation and plasma science was combined with experimental efforts at synchrotron and Free Electron Laser sources. This created an effective channel for knowledge transfer between the laser and the FEL communities in both experiment and theory.

In order to provide a broad basis for scientific exchange and education among students and supervisors and thereby increase the attractiveness of the graduate school we decided to open the school to associated members. Thereby we were able to significantly increase the number of permanent graduate students to 18, with up to 10 regularly funded by the Leibniz society. Thesis work covers the fields of structural dynamics, ultrafast phase transitions, dynamics in nanostructures and molecular systems. The individual thesis projects are described in short abstracts in Chapter 2. Lists of DinL publications and student talks at international conferences may be found in Appendices A and B.

All PhD students profited from scientific and general job-skill training programs (Chapt. 3) and obtained travel support for their contributions on national and international workshops and conferences (Appendix B). In addition DinL covered a few experimental running costs. Besides series of scientific seminars, DinL PhD students organized a Spring School in 2012, a Young Investigators workshop on Ultrafast Dynamics in 2014, and a Workshop in 2015 (Appendices C, D, and E). Furthermore, the PhD students of DinL joint courses and workshops of our Berlin partners, the Helmholtz Virtual Institute *Dynamic Pathways in Multidimensional Landscapes* and the International Max-Planck Research-School *Complex Surfaces in Materials Science*.

Altogether DinL students published up to now 97 articles, 40 as first authors, with 23 % in high-ranking journals (11 Physical Review Letters, 1 Journal of the American Chemical Society, and 10 Nature Journals). Moreover, the PhD students of DinL presented their scientific results in more than 100 contributions on national and international conferences.

Mostly starting their Phd work in 2012, to date most of the PhD students have graduated (15 of 18), and 33 % with summa cum laude. Based on their publication records we expect all graduate students to finish within 2016.

2. Graduate Students and Thesis Work

Structural Dynamics

1	Tobias Tyborski (MBI)	*Ultrafast mid-infrared responses of NaBH4, KDP in aqueous solution, and bulk plasmons in highly doped ZnO*
2	André Bojahr (UP)	*Nonlinear Phononics on the Nanoscale*
3	Michele Puppin (FHI)	*High-Harmonic generation-based XUV light source for time- and angle- resolved photoelectron spectroscopy at high repetition rates*
4	Melanie Müller (FHI)	*Probing currents and atomic structure in nanomaterials by femtosecond low-energy electrons*
5	Lutz Waldecker (FHI)	*Electron-lattice interactions in photoexcited solids*

Phase Transformations

6	Michael Schneider (TUB)	*Nonlinear intensity scaling of the magnetic small-angle scattering signal under high-intensity, femtosecond X-ray pulses.*
7	Björn Frietsch (MBI and FUB)	*Magnetization Dynamics of Itinerant and Localized Electrons in Lanthanide Metals*
8	Dr. Daniel Wegkamp (FHI)	*Ultrafast electron dynamics and the role of screening*
9	Johannes Flick (FHI)	*Exact and approximate correlated Electron-Boson Dynamics*
10	Tanja Dimitrov (FHI)	*Exact Functionals on a Lattice*

Nanostructures

11	T. Sverre Theuerholz (TUB)	*Theoretical description of hybrid systems*
12	Christopher Raschpichler (FUB)	*X-ray scattering and photoemission experiments of free nanoparticles*
13	Maria Müller (TUB)	*Ultrafast nanoplasma dynamics in clusters investigated with highly intense FEL pulses via ion spectroscopy and scattering techniques*

Molecular Systems

14	Dr. Sascha Birkner (MBI)	*Strong Field Ionization of Atoms and Molecules: Electron-Ion Coincidence Measurements at High Repetition Rate*
15	Lisa Torlina (MBI)	*The analytical R-matrix approach to strong field dynamics*
16	Dr. Shiladitya Banerjee (UP)	*Time-dependent approach to molecular photophysics*
	Dr. Gernot Füchsel (UP)	*Femtosecond-laser induced chemistry at metal surfaces*
17	Dr. Kristjan Kunnus (HZB)	*Probing dynamic pathways and electronic structure of coordination complexes with soft x-ray spectroscopy*
18	Dr. Simon Schreck (HZB)	*Potential Energy Surfaces, Femtosecond Dynamics and Nonlinear X-Ray-Matter Interactions from Resonant Inelastic Soft X-Ray Scattering*

Structural Dynamics

1. Tobias Tyborski – Max-Born-Institut Berlin / Humboldt-Universität zu Berlin
Supervisor / Mentor: Thomas Elsässer (MBI) / Andreas Knorr (TUB)

Ultrafast mid-infrared responses of $NaBH_4$, KDP in aqueous solution, and bulk plasmons in highly doped ZnO

In this work, the vibrational dynamics of BH_4^-- and phosphate ions were studied with nonlinear infrared spectroscopy. A further topic was the non-equilibrium dynamic of bulk plasmons in n-doped ZnO layer systems. Pump-probe techniques and 2D spectroscopy were applied. BH_4^- ions are a key unit for the mobile storage of hydrogen and B-H vibrations essential to understand the uptake and release of hydrogen. A cascaded energy dissipation into the BH_4^- environment from high- to low- frequency B-H vibrations was measured on a ps-timescale. Phosphates are the major hydration sites of biomolecules for which $H_2PO_4^-$ ions represent a reference system. $H_2PO_4^-$ (phosphate) vibrations decayed on a sub-ps-timescale and the distinct fluctuation dynamics of water led to homogeneously broadened line shapes. Furthermore, a strong nonlinear response of a bulk plasmon in a Ga-doped ZnO layer system was measured. It was caused by a sub-ps modification of the ensemble-averaged electron mass.

2. André Bojahr – Universität Potsdam
Supervisor / Mentor: Matias Bargheer (UP) / Martin Wolf (FHI)

Nonlinear Phononics on the Nanoscale

Phonons are the quanta of lattice vibrations and the main heat carrying particles in semiconductors. The coupling of different phonon-modes in the high GHz until THz frequency range governs the thermal transport in such materials.
In my thesis I invented experimental techniques to generate GHz narrow-band acoustic phonon wave packets with very high amplitudes. With these nanometric phonon wave packets I investigated the nonlinear phonon wave mixing. Using ultrafast x-ray diffraction and time-resolved Brillouin scattering I could demonstrate the second harmonic generation of such high frequency phonons as a first example of the new field “Nonlinear Phononics” as an analogue of nonlinear optics which summarizes a kaleidoscope of different nonlinear optical effects.

3. Michele Puppin – Fritz-Haber-Institut der MPG / Freie Universität Berlin
Supervisor / Mentor: Ralpg Ernstorfer (FHI) / Martin Weinelt (FU)

High-Harmonic generation-based XUV light source for time- and angle- resolved photoelectron spectroscopy at high repetition rates

The aim of this thesis is the development of an efficient femtosecond source with 500 kHz repetition rate, with a photon energy exceeding 20 eV. Such light pulses are used for time- and angle-resolved photoemission experiments (tr-ARPES) capable of accessing the full Brillouin zone of most materials. The key component of the light source is an optical parametric chirped-pulse amplifier (OPCPA) based on a hybrid fiber-slab Ytterbium-based laser system. The laser provides 30 µJ pulses with sub-20 fs duration, capable of High Harmonic Generation at 22 eV and 500 kHz repetition rate. The high-repetition rate enables superior counting statistics as only few photoelectrons per pulse can be detected before observing spectral distortions due to space charge effects. The system has been applied for mapping the unoccupied conduction band of WSe_2 in the whole Brillouin zone of the material.

4. Melanie Müller – Fritz-Haber-Institut der MPG / Freie Universität Berlin
Supervisor / Mentor: Ralph Ernstorfer (FHI) / Martin Weinelt (FU)

Probing currents and atomic structure in nanomaterials by femtosecond low-energy electrons

A novel concept for femtosecond point-projection microscopy (fsPPM) and low-energy electron diffraction (fsLEED) was developed utilizing laser-triggered nanotips as pulsed electron source, delivering either divergent or collimated electron wave packets in the sub-kV energy range. Due to the large scattering cross-section of low-energy electrons and their high sensitivity to electric fields, this approach allows for probing ultrafast currents and electric fields in nanoobjects as well as atomic structure in low-dimensional materials with femtosecond time resolution. As a proof-of-concept, we investigated photocurrents in axially p-i-n doped InP nanowires with 100 fs temporal and few 10 nm spatial resolution. As a major step towards increased spatio-temporal resolution, we further realized an ultrashort electron point source driven non-locally by surface plasmon polaritons launched 20 µm away from the tip apex triggering ultrafast electron emission from the apex upon broadband nanofocusing.

5. Lutz Waldecker – Fritz-Haber-Institut der MPG / Freie Universität Berlin
Supervisor / Mentor: Ralph Ernstorfer (FHI) / Ludger Woeste (FU)

Electron-lattice interactions in photoexcited solids

In this project, a setup for femtosecond electron diffraction (FED) was developed and implemented. The technique was applied to study the energy transfer between electrons and the crystal lattice in photoexcited materials. In Aluminium, it is found that the widely applied two-temperature model, albeit being able to reproduce the shape of the data, is insufficient to microscopically describe the energy-flow. An advanced model was introduced (non-thermal lattice model), which captures the microscopic processes much better. In addition, the setup for FED is applied, together with optical pump – optical probe experiments, to study the photoinduced phase transition from the crystalline to the amorphous state in the phase change material GST. The data reveals that photoexcitation does not change the potential energy surface of the material, but has a drastic effect on the optical properties. The phase transition occurs thermally, i.e. by thermal melting of the material and subsequent interactions with the substrate.

Phase Transformations

6. Michael Schneider – Technische Universität Berlin

Supervisor / Mentor: Stefan Eisebitt (TUB) / Martin Weinelt (FU)

Nonlinear intensity scaling of the magnetic small-angle scattering signal under high-intensity, femtosecond X-ray pulses.

We performed small-angle X-ray scattering experiments on magnetic thin-film samples using highly intense, ultrashort free-electron laser (FEL) radiation. For this type of experiment, we developed a novel sample design with a monolithically integrated fluence monitor. Using this, we carried out the first systematic study on the nonlinear scaling of the resonant magnetic scattering cross-section, covering a fluence range of 0.1mJ/cm² to 10mJ/cm² per sub-100fs FEL pulse. Our study revealed a strongly nonlinear behavior of the scattering cross-section. The high quality of our dataset allows for quantitative comparison with different proposed mechanisms including ultrafast demagnetization and increased forward scattering by stimulated emission.

7. Björn Frietsch – Freie Universität Berlin

Supervisor / Mentor: Martin Weinelt (FUB) / Karsten Horn (FHI)

Magnetization Dynamics of Itinerant and Localized Electrons in Lanthanide Metals

My thesis focuses on the ultrafast dynamics of electronic excitations in solids and how they are influenced by the screening of the Coulomb interaction between charged particles. The manifold impact of screening on electron dynamics, ranging from modifications of electron-electron scattering rates over trapping of excess charges to massive renormalization of electronic band structures is investigated via ultrafast time-resolved spectroscopy techniques in three exemplary systems. Most prominently, a photoinduced change of screening is identified as the driving mechanism of the ultrafast insulator-metal transition in VO_2. Further studies of electron dynamics in $SrTiO_3$ and amorphous ice layers illustrate the general impact of screening of the Coulomb interaction, ranging from fundamental processes to photochemistry.

8. Daniel Wegkamp – Fritz-Haber-Institut der MPG / Freie Universität Berlin
Supervisor / Mentor: Julia Stähler and Martin Wolf (FHI) / Martin Weinelt (FUB)

Ultrafast electron dynamics and the role of screening

My thesis focuses on the ultrafast dynamics of electronic excitations in solids and how they are influenced by the screening of the Coulomb interaction between charged particles. The manifold impact of screening on electron dynamics, ranging from modifications of electron-electron scattering rates over trapping of excess charges to massive renormalization of electronic band structures is investigated via ultrafast time-resolved spectroscopy techniques in three exemplary systems. Most prominently, a photoinduced change of screening is identified as the driving mechanism of the ultrafast insulator-metal transition in VO_2. Further studies of electron dynamics in $SrTiO_3$ and amorphous ice layers illustrate the general impact of screening of the Coulomb interaction, ranging from fundamental processes to photochemistry.

9. Johannes Flick – Fritz-Haber-Institut der Max-Planck-Gesellschaft Berlin
Supervisor / Mentor: A. Rubio (FHI) / Th. Elsaesser (MBI)

Exact Nonadiabatic Many-Body Dynamics: Electron-Phonon Coupling in Photoelectron Spectroscopy and Light-Matter Interactions in Quantum Electrodynamical Density-Functional Theory

Many natural and synthetic processes triggered by the light-matter interaction are routinely explained by employing various approximations. In the first part of this work, we assess the validity of the Born-Oppenheimer approximation (BOA) in photoelectron spectra and show that spurious peaks appear for the spectral function in the BOA. This artificial effect can be attributed to the factorized nature of the BOA. In the second part of this work, we apply a generalized time-dependent density functional theory (QEDFT) to describe electron-photon systems fully quantum mechanically. We focus on the exact electron-photon exchange-correlation (xc) contribution in real space using fixed-point inversions and assess the quality of the first approximate xc potential based on an optimized effective potential approach in comparison to the semi-classical (mean-field) approximation. This work has implications on new research lines at the interface between materials science and quantum optics.

10. Tanja Dimitrov – Fritz-Haber-Institut der Max-Planck-Gesellschaft Berlin

Supervisor / Mentor: A. Rubio (FHI) / Th. Elsaesser (MBI)

Exact Functionals on a Lattice

We employ exact diagonalization for an one-dimensional H2 model molecule and a two-site lattice model on a real-space lattice to explicitly construct the exact density-to-potential map of density functional theory and compare it to the density-to-potential map in local density approximation (LDA). The exact maps of both, the two-site model and the one-dimensional H2 molecule, show similar qualitative features missing in the LDA maps. For the two-site model, we also construct the exact density-to-wavefunction map that underly the Hohenberg-Kohn theorem in density functional theory. Having the explicit wavefunction-to-density map at hand, we are able to construct arbitrary observables as functionals of the ground-state density. We analyze the density-to-potential map, the density-to-wavefunction map and the (arbitrary) observable-to-density map as the correlation in the system grows. This work opens the possibility to construct new functionals using a coarse-grained approach.

Nanostructures

10. T. Sverre Theuerholz – Technische Universität Berlin

Supervisor / Mentor: Andreas Knorr (TUB) / Martin Wolf (FHI)

Theoretical description of hybrid systems

In this work, we investigate hybrid materials of quantum dots/molecules coupled to metallic nanostructures. Within a fully quantized theory, the coupling between the individual constituents and their collective excitations is analyzed. Calculated observables include the coherent response and the secondary emisson after laser excitation, whereas the emission statistics is analyzed using the second order photon-photon correlation function. The developed theory is tested by a direct comparison to the temporal fluorescence dynamics of a variety of quantum dot/metal hybrids, also we find that the photon emission statistics can be strongly tuned by varying the dipole-dipole interaction between two QDs coupled to a metall structure.

11. Christopher Raschpichler – Freie Universität Berlin

Supervisor / Mentor: Eckhart Rühl and Jürgen Plenge (FUB) / Marc Vrakking (MBI)

X-ray scattering and photoemission experiments on free nanoparticles

Elastic X-ray scattering and photoemission experiments are performed on free nanoparticles in a beam in vacuum. Three novel aspects of the interaction of free nanoparticles with X-rays and femtosecond laser pulses are studied: (i) Relaxation mechanisms of SiO_2 nanoparticles (d=100 nm) are studied after excitation with femtosecond UV pulses (hv=4.6-7.7 eV). (ii) Elastic X-ray scattering in the angle range 1-80° is measured, which gives detailed insight into the electronic and geometric properties of SiO_2 nanoparticles as a function of their size, surface roughness, porosity, and functionalization. (iii) Surface segregation in mixed $NaCl/Na_2SO_4$ nanoscale aerosols (d=70 nm) is probed by soft X-ray photoelectron spectroscopy, where insight is gained into the surface properties of these atmospheric model systems.

12. Maria Müller – Technische Universität Berlin

Supervisor / Mentor: Thomas Möller (TUB) / Marc Vrakking (MBI)

Time resolved studies of ultrafast nanoplasma dynamics in clusters investigated with highly intense NIR and XUV pulses via ion spectroscopy and scattering techniques

The interaction with highly intense light pulses transfers all matter immediately into a highly ionized state and thereby initializes its destruction. To investigate its dynamics in large Xe clusters single particle spectroscopy is combined with coherent diffraction imaging in single-pulse and pump-probe setups. Single particle experiments, common in the XUV and x-ray regime, overcome the limits set by averaging over focal density and cluster size distribution. Within this thesis, this approach has been transferred for the first time to the NIR regime where it allows to retrace the theoretical concepts for hydrodynamic expansion in great detail in experimental data. Within the pump-probe setup signatures of strongly enhanced absorption during different states of expansion are found, e.g. surface plasmon effects and emerging shockwaves, if the XUV-FEL pulse precedes the NIR pulse. In contrast, if the pulse order is reversed, a high level of transparency for the FEL can be temporarily induced by the NIR pulse.

Molecular Systems

13. Sascha Birkner – Max-Born-Institut / Freie Universität Berlin

Supervisor / Mentor: Marc Vrakking (MBI) / Eckhart Rühl (FUB)

Strong Field Ionization of Atoms and Molecules: Electron-Ion Coincidence Measurements at High Repetition Rate

The combination of strong few-cycle laser pulses with a reaction microscope, a spectrometer capable of detecting the momentum-vectors of all charged particles that emerge from ionization or dissociation processes in coincidence, enables precise and detailed investigations of strong field processes that occur in atoms and small molecules. We combined a reaction microscope with a 400 kHz high repetition rate NOPA-system delivering strong few-cycle laser pulses to study multi-electron dynamics in strong field ionization experiments.
Experiments on strong field ionization of polyatomic molecules (butadiene and n-butane), revealed that different ionic states are populated during the strong field ionization and hinted towards a break-down of the single active electron approximation in polyatomic molecules.

14. Lisa Torlina – Max-Born-Institut / Technische Universität Berlin
Supervisor / Mentor: Olga Smirnova (MBI) / P. Saalfrank (UP)

The analytical R-matrix approach to strong field dynamics

In this work, we present a new theoretical framework for describing the electron dynamics induced when an atom or molecule is exposed to a strong low frequency laser field, focusing especially on strong field ionization. Once the basic formalism of our theory – known as the Analytical R-Matrix (ARM) – is established for both single- and multi-electron systems, we move on to a more detailed investigation of its predictions and physical interpretation. Focusing on circularly polarized fields, we study the effects of the ionic core on the outgoing electron and validate our theory by comparing our results with exact ab initio numerical calculations. Finally, we apply ARM to analyze the physics behind the attoclock experiment. Doing so, we are able to address the question of tunnelling time delays and – by calibrating the attoclock for the one-electron case – open the way to study multielectron dynamics.

15. Shiladitya Banerjee / Gernot Füchsel – Universität Potsdam
Supervisor / Mentor: Peter Saalfrank and T. Klamroth / Angel Rubio (FHI / MPSD)

Time-dependent approach to molecular photophysics / Femtosecond-laser induced chemistry at metal surfaces

The topic of S. Banerjee's research was to develop and apply time-dependent methods for vibronic, non-adiabatic spectra for molecules of medium size (50-100 atoms). In particular, vibrationally resolved electronic absorption and fluorescence emission spectra as well as resonance Raman (rR) spectra were to be determined for molecules such as β-carotene ($C_{40}H_{56}$, 96 atoms) or so-called diamondoids (e.g., pentamantane, $C_{26}H_{32}$, 58 atoms). Concrete reasons for our interest in the photophysics of these species were anomalies and / or special features in resonance Raman spectra as determined experimentally (for β-carotene and also for modified diamondoids, work of Hildebrandt and Maultzsch groups, TU Berlin), or their potential to serve as materials with tailor-made optoelectronic properties (in case of diamondoids, work of Möller group at TU Berlin). A numerical challenge when adopting the “usual” time-independent (Golden Rule type) methods to compute vibronic spectra for molecules with many atoms, is the exponential increase of the number of Franck-Condon factors (absorption, emission) or products of dipole matrix elements (resonance Raman), respectively. A possible workaround is using time-dependent methods instead as popularized by Heller and coworkers in the 1980's, where vibronic spectra are obtained from Fourier transforms of time-dependent auto-correlation functions (absorption, emission) and cross-correlation functions (resonance Raman), respectively. S. Banerjee has written programs in which this approach is realized within the harmonic approximation for multi-mode problems, by taking geometry and frequency shifts as well as the so-called Duschinsky rotation (mode mixing) into account. He was then able to explain a previously not understood anomaly measured in the rR spectra of β-carotene. As a second application, absorption, emission and rR spectra for a large class of pristine and also artificial, diamondoids were determined. [Quite recently, this method was also applied to functionalized diamondoids with the goal to tune optoelectronic properties of these materials (Banerjee, Stüker, Saalfrank, submitted to Phys. Chem. Chem. Phys.).]

A topic of Gernot Füchsel's project during his membership in DinL was the quantum mechanical simulation of laser induced desorption of H_2 from a Ru(0001) surface (experiments: Wolf group, FHI Berlin). A three-dimensional, two-state wavepacket model was applied in to study the energy distribution as well as the state-resolved ensemble properties of the desorbed molecules. Here, the coupling of electronic degrees of freedom (of the metal surface) to molecular degrees of freedom is essential. In a related side-project, the role of adsorbate-metal electron couplings on the scattering and dissociation of H_2 at Ru(0001) was also studied, using a Langevin molecular dynamics approach. Finally, a six-dimensional potential energy surface based on density functional was developed for CO interacting with Ru(0001), for later use in femtosecond-laser induced dynamics.

16. Kristjan Kunnus – Helmholtz-Zentrum Berlin und Universität Potsdam

Supervisor / Mentor: Philippe Wernet (HZB) / Wolfgang Sandner (MBI)

Probing dynamic pathways and electronic structure of coordination complexes with soft x-ray spectroscopy

In this thesis static and time-resolved resonant inelastic x-ray scattering (RIXS) was utilized to study electronic structure and photo-induced dynamics of 3d transition metal coordination complexes (TM-complexes) in solution. Multidimensional RIXS spectroscopy was developed for performing time-resolved liquid phase experiments at the x-ray free electron laser light sources and novel *ab initio* calculations are applied to simulate the metal $L_{2,3}$-edge RIXS spectra. Results from two types of systems are presented in this thesis. First, the interplay between inter-atomic solute-solvent and intra-atomic multiplet interactions is investigated in TM-ion aqueous solutions. Secondly, a novel femtosecond time-resolved Fe L_3-edge RIXS technique at the LCLS x-ray free electron laser light source was utilized to investigate the photoreaction following a metal-to-ligand charge transfer excitation of a prototypical metal carbonyl compound $Fe(CO)_5$ in ethanol.

17. Simon Schreck – Helmholtz-Zentrum Berlin und Universität Potsdam
Supervisor / Mentor: Alexander Föhlisch (HZB) / Peter Saalfrank (UP)

Potential Energy Surfaces, Femtosecond Dynamics and Nonlinear X-Ray-Matter Interactions from Resonant Inelastic Soft X-Ray Scattering

In this project novel methodological concepts of resonant inelastic x-ray scattering (RIXS) in the soft x-ray regime have been investigated and developed. The intrinsic fs duration of the RIXS process provides sensitivity to dynamical processes in the studied system on the fs timescale. The sensitivity of RIXS to fs nuclear dynamics in the OH group of liquid alcohols has been investigated. Furthermore it was demonstrated how ground state potential energy surfaces around selected atomic sites can be mapped by RIXS. This is becoming possible due to advances in soft x-ray beamlines and spectrometers that enable vibrational resolution. The ultrahigh peak brilliance available at x-ray free-electron lasers opens up possibilities to transfer nonlinear spectroscopic techniques from the optical and infrared into the x-ray regime. We showed that for an x-ray pulse length comparable to the core hole lifetime stimulated x-ray scattering is possible. This can be used to greatly enhance signal levels in RIXS experiments.

DinL – Training Program

Scientific Workshops

DinL Symposium 2012 - 2013

During the terms in 2012 and 2013 we had in addition to regular talks in the group seminars of the supervisors and the mentors a series of monthly colloquia. Alternatingly, all graduate students organized at their institution these meetings. After a tour through the institution and a visit of the labs, graduate students gave talks about their PhD working-plans and first results. The colloquia were completed by an extended 90 minutes tutorial and scientific lecture from an invited speaker.

Spring school 2012

In April 2012 the DinL graduate students Maria Müller and Michael Schneider from TUB organized a four-day scientific Spring School. Students retreated at the seminar hotel Villago in Eggersdorf near Berlin. Extended lectures were given by 11 external speakers and 5 supervisors of DinL. They introduced in the topics investigated by the graduate students of DinL. Ample time was reserved for extended discussions. The program may be found in Appendix C.

Young Investigators Workshop on Ultrafast Dynamics 2014

In March 2014 we organized a five-day young investigators workshop and retreat at the Winklmoosalm (Reit im Winkl, Germany) together with the Helmholtz Virtual Institute. Besides 4 extended tutorials, every participant gave a 30 minutes presentation. Outdoor activities stimulated plenty of scientific discussion. The format of the winter school was a big success and repeated by the Virtual Institute in 2015. The program may be found in Appendix D.

DinL – Concluding Workshop 2015

A two-day concluding workshop was organized at the Max-Bon-Institute in 2015. Key lectures of the three MBI directors and of three invited external speakers framed the final reports of the DinL graduate students. The excellent talks attracted a large audience from MBI and from the participating groups. The program may be found in Appendix E.

Job-Skill Trainings

Besides the regular group seminars and teaching courses offered by the supervisors, DinL organized courses related to scientific presentation and job-skills.

Seminars by Scientists on Scientific Presentation and Publication

- Presentation techniques (Martin Weinelt, FUB)
- How to write a good abstract. Some do's and don'ts (Karsten Reuther, TU München)
- Paper writing (Nacho Pascual, NanoGUNE, San Sebastian, Spain)

Coaching of Job Skills by External Teachers

- 2-day ***Workshop on paper writing*** by Dr. Katherine Tiede (Scientific Communication Seminars and Consulting)
  Content: Introduction to scientific writing, logic and structure of papers, create a story, draw interest, distinguish between good and bad papers in your field, practical training: witing and correcting manuscripts

- 1-day ***Application seminar*** by Wolfgang Träger (Eigen.schafft Unternehmensentwicklung)

  Content: Supervision and realization of a work shop „How graduate students increase their chance to be hired in industry by an optimal application"

- 1-day ***Presentation workshop*** by Steve Weir (FU career service)

  Content: speaking without notes, non-verbal communication (body language), using communication media, dealing with unwelcome speech situations and planning of a presentation

Appendix A: List of Publications

2011

[1] **S. Schreck**, G. Gavrila, C. Weniger, P. Wernet, *A sample holder for soft x-ray absorption spectroscopy of liquids in transmission mode*, Rev. Sci. Instrum. **82**, 103101 (2011).

[2] J. Plenge, A. Wirsing, **C. Raschpichler**, B. Wassermann, and E. Rühl, *Control of Coherent Excitation of Neon in the Extreme Ultraviolet Regime*, Faraday Discuss., **153**, 361 (2011).

[3] **D. Wegkamp**, D. Brida, S. Bonora, G. Cerullo, J. Stähler, M. Wolf and S. Wall, *Phase retrieval and compression of low-power white-light pulses*, App. Phys. Lett. **99**, 101101 (2011).

2012

[4] R. Carley, K. Döbrich, **B. Frietsch**, C. Gahl, M. Teichmann, O. Schwarzkopf, P. Wernet and M. Weinelt, *Femtosecond Laser Excitation Drives Ferromagnetic Gadolinium out of Magnetic Equilibrium*, Phys. Rev. Lett. **109**, 057401 (2012).

[5] A. Paarmann, M. Gulde, **M. Müller**, S. Schäfer, S. Schweda, M. Maiti, C. Xu, T. Hohage, F. Schenk, C. Ropers, and R. Ernstorfer, *Coherent femtosecond low-energy single-electron pulses for time-resolved diffraction and imaging: A numerical study*, J. Appl. Phys. **112**, 113109 (2012).

[6] I. Josefsson, **K. Kunnus**, **S. Schreck**, A. Föhlisch, F.M.F. de Groot, P. Wernet, M. Odelius, Ab Initio Calculations of X-ray Spectra: *Atomic Multiplet and Molecular Orbital Effects in a Multiconfigurational SCF Approach to the L-Edge Spectra of Transition Metal Complexes*, J. Phys. Chem. Lett. **3**, 3565 (2012).

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Appendix B: Talks at international workshops and conferences

DinL students gave more than 30 talks on international conferences and in addition more than 50 contributions on seminars, national workshops and conferences, excluding the DinL-organized events summarized in Section DinL - Workshops.