

7th Symposium on Ultrafast Surface Dynamics

August 22-26, 2010
Brijuni Islands, Croatia

BOOKLET OF ABSTRACTS

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Dear colleagues, participants and guests of USD7,

The Local Organizing Committee of the 7-th International Symposium on Ultrafast Surface Dynamics (USD7, <http://usd7.ifs.hr/>) has a great pleasure to welcome you to the beautiful island of Veliki Brijun off the Istrian coast in northern Adriatic, chosen for the USD7 venue in the year 2010. This conference is the seventh in the series of successful biannual meetings initiated to bring together the scientists and students from research institutions and universities throughout the world to report and discuss current developments and future directions of research in the field of ultrafast dynamics at surfaces and interfaces. Earlier meetings of the USD series were held at Ascona (Switzerland 1997), Ringberg (Germany 1999), San Sebastian (Spain, 2001), Telluride (USA 2003), Abashiri (Japan 2006) and Kloster Banz (Germany 2008).

One of the central goals of surface science is to develop microscopic understanding of interactions at surfaces and elementary steps in surface reactions. The dynamics of such elementary processes occur on the (sub)femtosecond time scale and are governed by ultrafast excitation and relaxation processes involving atomic and electronic mass and charge transport, electronic and vibrational energy transfer, and related phenomena. The objective of this symposium is to discuss recent experimental and theoretical advances in the analysis and understanding of these ultrafast surface phenomena. The symposium encompasses invited and contributed oral presentations, posters, as well as extensive discussions. Special emphasis is placed on the following research topics:

- Electronic states, electron dynamics and energy transfer at surfaces and interfaces
- Vibrational energy transfer and wave-packet dynamics at surfaces
- Ultrafast dynamics and spectroscopy of coherent surface and adsorbate excitations
- Attosecond physics and ultrafast X-ray pulses
- Laser-induced surface femtochemistry and dynamics in adsorbates and molecular films
- Spin-dependent dynamics and spin-polarized transport at surfaces and interfaces, magnetooptics and ultrafast magnetization
- Ultrafast spectroscopy and coherent control

- Advances in ultrafast surface spectroscopies
- Ultrafast dynamics in nanostructured and correlated materials.

A palette of invited lectures and contributed talks and posters selected for presentation at USD7 should provide a thorough insight into the rapid development of this exciting research field whose academic importance and technological impact have now been recognized worldwide. This makes it a challenging and promising area of research for many years ahead, with our USD community firmly established on its front.

The Local Organizing Committee and the Institute of Physics in Zagreb as the organizing institution are especially grateful to a number of research institutions that have made generous contributions to the USD7 budget, because without their financial support the organization of the symposium would not have been possible. Our special thanks go to the Fritz-Haber-Institute (Berlin), Max-Born-Institute (Berlin), Donostia International Physics Center (San Sebastian), University of Texas (Austin), University of Duisburg-Essen and University of Pittsburgh. We acknowledge the support to the symposium from the companies VG Scienta, SPECS, Femtolasers, Coherent and Wiley-VCH. Lastly, our thanks also go to the Congress Department of the Brijuni National Park for hosting the meeting and Ms. Loreta Boneta for undertaking the burden of accommodation and registration book-keeping.

Branko Gumhalter

On behalf of the organizers of USD7

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Program

Day One — Sunday, August 22nd

10:00-12:00	Registration		
12:00-14:00	Lunch and Beach		
14:00-15:40	Registration		
15:40-16:00	Welcome address		
<u>Chairman: BRANKO GUMHALTER</u>			
16:00-16:40	Christoph Lienau	Ultrafast Nano-Optics: Novel light and electron sources for surface science	16 (I1)
16:40-17:20	Christian Spielmann	Time-resolved x-ray absorption spectroscopy using high harmonic radiation	17 (I2)
17:20-18:00	Break		
18:00-18:40	A. Winkelmann, C.-T. Chiang, W. C. Lin, P. Yu, F. Bisio, H. Petek, J. Kirschner	Optical control of spin in nonlinear photoemission from metal surfaces	18 (I3)
18:40-19:20	To be decided		19 (I4)
19:20-20:40	Dinner		

Day Two — Monday, August 23rd

<u>Chairperson: MARTIN WEINELT</u>			
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09:40-10:00	J. Güdde , K. Klass, G. Mette, M. Dürr, U. Höfer	<i>Time-resolved investigation of laser-induced diffusion by SHG microscopy</i>	21 (C1)
10:00-10:20	Tsuneyuki Nakamura, Naoyuki Hirata, Atsushi Nakajima	<i>Electronic Structure of H-Terminated Silicon Surfaces [H-Si(111)] Studied by Two-Photon Photoemission</i>	22 (C2)
10:20-10:40	Thomas Fauster , Shin'ichiro Tanaka, Katsumi Tanimura	<i>One- and two-photon photoemission from silicon surfaces</i>	23 (C3)
10:40-11:20	Break		

11.20-11.40	Martin Kammner , Simone Möllenbeck, Anja Hanisch-Blicharski, Annika Kalus, Paul Schneider, Boris Krenzer, Michael Horn-von Hoegen	<i>Ultrafast Time Resolved Electron Diffraction of Adsorbate Dynamics on Silicon Surfaces</i>	24 (C4)
11.40-12.00	S. Mathias , C. La-O-Vorakiat, P. Grychtol, R. Adam, M. Siemens, J. M. Shaw, H. Nembach, M. Aeschlimann, C. M. Schneider, T. Silva, M. M. Murnane, H. C. Kapteyn	<i>Ultrafast, Element-Specific, Demagnetization Dynamics Probed using Coherent High Harmonic Beams</i>	25 (C5)
12.00-12.20	Yoichi Ogata , Nguyen Anh Tuan, Goro Mizutani	<i>Rotational anisotropy of optical second harmonic generation from Pt nanowires with boomerang-like cross-sectional shapes</i>	26 (C6)
12.20-13.00	Uwe Bovensiepen	<i>Ultrafast phenomena probed with time-resolved photoelectron spectroscopy</i>	27 (I6)
13.00-16.00	Lunch and Beach Discussion		

Chairperson: ULRICH HOEFER

16.00-16.40	Martin Aeschlimann	<i>Spin-dependent dynamics of carriers injected across interfaces with organic semiconductors</i>	28 (I7)
16.40-17.00	A.I. Shkrebtii , J.L. Cabellos, B.S. Mendoza, F. Nastos, J.E. Sipe	<i>Coherent control of injection current at surfaces and interfaces: Si(111) and GaAs(110), clean and with adsorbate</i>	29 (C7)
17.00-17.40	Break		
17.40-18.00	M. Muntwiler , Q. Yang, X.-Y. Zhu	<i>Charge-Transfer Excitons at the Interface of Organic Semiconductors</i>	30 (C8)
18.00-18.20	Oliver L.A. Monti , Mary P. Steele, Michael L. Blumenfeld	<i>Near- and Far-Field Effects on Excited States at Organic Semiconductor / Metal Interfaces</i>	31 (C9)
18.20-19.00	Julia Stähler , Michael Meyer, Uwe Bovensiepen, Martin Wolf	<i>Ultrafast electron transfer across polar molecule-metal interfaces: strong coupling vs. molecular screening</i>	32 (I8)
19.00-20.20	Dinner		
20.20-22.00	Poster Session		

Day Three — Tuesday, August 24th

Chairperson: THOMAS FAUSTER

09.00-09.40	Kunie Ishioka	<i>Coherent optical phonons on semiconductor surfaces studied with transient reflectivity</i>	33 (I9)
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09:40-10:00	Kazuya Watanabe , Ken-ichi Inoue, Ikuyo F. Nakai, Yoshiyasu Matsumoto	<i>Ultrafast vibrational dynamics of CO on Pt(111) studied by time-resolved SFG with phase-sensitive detection</i>	34 (C10)
10:00-10:20	T. Yasuike , K. Watanabe, K. Nobusada, Y. Matsumoto	<i>Two competing mechanisms for photoinduced Cs dynamics on Cu(111)</i>	35 (C11)
10:20-10:40	Hrvoje Petek	<i>Imaging the femtosecond time scale correlated electron-nuclear dynamics in surface photodesorption</i>	36 (C12)
10:40-11:20	Break		
11:20-11:40	A. Ouvrard, A. Ghalgaoui, Z. Guo, B. Bourguignon	<i>Contrasted behaviour of terrace and edge CO adsorbed on Pd nanoparticles studied by pump probe SFG</i>	37 (C13)
11:40-12:00	A. Melnikov , I. Razdolski, T. Wehling, E. Papaioannou, P. Fumagalli, O.A. Akt-sipetrov, A. Lichtenstein, U. Bovensiepen	<i>Ultrafast transport of laser-induced spin polarized carriers in Au/Fe/MgO(001)</i>	38 (C14)
12:00-12:20	K. M. Döbrich , A. Goris, I. Panzer, A. B. Schmidt, M. Pickel, M. Donath, and M. Weinelt	<i>Spin-dependent lifetimes of hot electrons in 3d ferromagnets</i>	39 (C15)
12:20-13:00	Martin Weinelt , Christian Eickhoff, Martin Teichmann	<i>Two-dimensional Fano resonances on Si(001)</i>	40 (I10)
13:00-16:00	Lunch and Beach Discussion		

Chairperson: ANATOLI CHKREBTII

16:00-16:40	Tianquan Lian	<i>Ultrafast Multi-Exciton Dissociation Dynamics in CdSe Quantum Dots</i>	41 (I11)
16:40-17:20	E.V. Chulkov	<i>Lifetimes of electron excitations on clean and nanostructured metal surfaces</i>	42 (I12)
17:20-19:40	Outing		
19:40-20:40	Dinner		
20:40-21:20	Meeting of the USD Steering Committee		

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Chairperson: MARTIN WOLF

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09:40-10:00	B. Gumhalter , P. Lazić, N. Došlić, V. M. Silkin, E. V. Chulkov	<i>Ultrafast electron dynamics in pump-probe spectroscopies of surfaces: from transient excitonic to quasi-stationary polaronic states</i>	44 (C16)

10:00-10:20	Daria Popova , Dr. Andreas Bringer, Prof. Stefan Blügel	<i>Theoretical investigation of ultrafast laser-induced magnetization dynamics in many-electron systems</i>	45 (C17)
10:20-10:40	Masahiro Shibuta, Takashi Yamada , Kenta Yamamoto, Keisuke Miyakubo, Toshiaki Munakata	<i>Resonance Effects on Two-Photon Photoemission of Lead Phthalocyanine Thin Films on Graphite</i>	46 (C18)
10:40-11:20	Break		
11:20-11:40	G. Lefkidis , W. Hübner	<i>Logic functionalization and angular momentum conservation in magnetic nanoclusters</i>	47 (C19)
11:40-12:00	I. Radu , K. Vahaplar, C. Stamm, T. Kachel, H. Dürr, T. Ostler, R. Chantrell, A. Tsukamoto, A. Itoh, A. Kirilyuk, Th. Rasing, A.V. Kimel	<i>An Element-specific View on Ultrafast Magnetization Reversal of GdFeCo</i>	48 (C20)
12:00-12:20	T. Rohwer, S. Hellmann, M. Wiesenmayer, C. Sohrt, A. Stange, M. Kalläne, S. Mathias, L. Kipp, K. Rossnagel, M. Bauer	<i>Ultrafast collapse of periodic long-range order tracked by time-resolved XUV-ARPES</i>	49 (C21)
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Chairperson: PEDRO MIGUEL ECHENIQUE

16:00-16:40	Henry M. van Driel	<i>Coherent Control and Nonlinear Optics in Carbon Nanotubes and Graphitic Layers</i>	51 (I15)
16:40-17:00	P. Pervan , I. Pletikosić, M. Kralj, M. Milun	<i>Electronic properties of graphene on Ir(111)</i>	52 (C22)
17:00-17:20	Daniel Niesner , Thomas Fauster	<i>Image-potential states of graphene on iridium</i>	53 (C23)
17:20-18:00	Break		
18:00-18:20	Michael Krüger , Markus Schenk, Peter Hommelhoff	<i>From perturbative to strong-field emission: energy-resolved ultrafast laser-induced electron emission from sharp metal tips</i>	54 (C24)
18:20-19:00	Fulvio Parmigiani , Matteo Montagnese, Stefania Pagliara, Gianluca Galimberti, Gabriele Ferrini	<i>Ultrafast electronic response and charge transfer at surfaces</i>	55 (I16)
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Day Five — Thursday, August 26th

Chairperson: MARTIN AESCHLIMANN

09:00-09:40	R. Huber , A. Leitenstorfer, A. Sell, A. A. Anappara, G. Biasiol, L. Sorba, A. Tredicucci, K. v. Volkmann, T. Kampfrath, M. Wolf	<i>Ultrafast dynamics of THz excitations: from observation to coherent control</i>	56 (I17)
09:40-10:00	Laurenz Rettig , Rocío Cortés, Setti Thirupathaiah, Uwe Bovensiepen, Martin Wolf, Hermann A. Dürr, Philipp Gegenwart, Thomas Wolf, Jörg Fink	<i>Anisotropic electron dynamics and electron-phonon coupling in Fe-pnictides</i>	57 (C25)
10:00-10:20	Ryuichi Arafune , Mayuko Q. Yamamoto, Noriaki Takagi, Maki Kawai	<i>Excitation energy dependence of photoelectron-phonon interaction on Cu surface excited by laser light</i>	58 (C26)
10:20-10:40	Masaki Hada , Kunio Okimura, Jiro Matsuo	<i>Structure dynamics of epitaxial VO₂ film on c-Al₂O₃ by using an in-air time-resolved X-ray diffraction</i>	59 (C27)
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11:20-11:40	J. E. Johns, E. A. Muller, J. M. J. Frechet, C. B. Harris	<i>Ultrafast Localization of Photo-injected Charges by Alkyl Side Chains in Organic Semiconductors</i>	60 (C28)
11:40-12:00	Ken Onda , Keiki Fukumoto, Shinya Koshihara	<i>Dynamics of Monolayer Water on a Single Crystalline TiO₂ Surface in Air by VSFG Spectroscopy</i>	61 (C29)
12:00-12:20	Kazuhiro Yabana , Yasushi Shinohara, Yosuke Kawashita, Jun-Ichi Iwata, Tomohito Otobe, George F. Bertsch	<i>Ab-initio description for laser-induced electron-phonon dynamics in dielectrics</i>	62 (C30)
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Chairperson: HRVOJE PETEK

16:00-16:40	Simon Wall , Daniele Brida, Stephen Clark, Henri Ehrke, Dieter Jaksch, Hiroshi Okamoto, Giulio Cerullo, Andrea Cavalleri	<i>Quantum Interference Effects in the Ultrafast Dynamics of a Low Dimensional Mott Insulator</i>	64 (I19)
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17.40- 18.20	Jun Kubota	<i>Dynamics of chemical reactions at surfaces under thermal excitation by laser irradiation and probed by time resolved SFG spectroscopy</i>	66 (I20)
18.20- 18.40	Takuya Harada, Keiichiro Matsuishi, Jun Oi, Yu Oishi, Fumihiko Kannari	<i>Measurement of local plasmon resonance of Au nanorods on femtosecond laser polarization using autocorrelation dark-field microscopy</i>	67 (C32)
18.40- 19.20	Jean-Yves Bigot	<i>Ultrafast transfer of angular momentum in ferromagnets</i>	68 (I21)
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20:00- 22:00	Dinner		

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Zefeng Ren , Lehua Chen, Christian Frischkorn, Martin Wolf	<i>A supersonic molecular beam doser for surface science research</i>	P13
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Abstracts of Oral Presentations

Ultrafast Nano-Optics: Novel light and electron sources for surface science

Christoph Lienau

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www.uno.uni-oldenburg.de

Functional nanomaterials are essential building blocks for a variety of modern technologies. Understanding and controlling the structure and function of nanomaterials requires novel and powerful microscopes providing ultrahigh spatial but also ultrahigh temporal resolution. Consequently a considerable amount of new, often accelerator-based ultrafast light, x-ray and electron sources are currently under development throughout the world. In this talk I want to show that also nanotechnology itself offers powerful and partially rather surprising novel microscopy tools which are based on the ability to manipulate and control light on nanometer length and femtosecond time scales. For this, I will discuss different recent developments in the field of ultrafast nano-optics, including a new optical microscope based on the localisation of propagating coherent surface plasmon polariton wavepackets, an ultrafast switchable metallic mirror and a novel approach for a spatially and temporally highly resolved electron microscope.

Time-resolved x-ray absorption spectroscopy using high harmonic radiation

Christian Spielmann

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The development of reliable femtosecond solid-state laser brought new possibilities into time-resolved spectroscopy. For the first time it became possible, in principle to monitor the nuclear motion of molecules, crystal lattices and other out-of-equilibrium structures. However, usually it is very difficult to map the experimental observations to the structural dynamics. Therefore, experimental approaches are needed that can overcome the limitation of optical studies for structural determination, while the high temporal resolution of femtosecond lasers is maintained. Structural techniques such as X-ray diffraction, X-ray absorption spectroscopy (XAS), or X-ray photoelectron spectroscopy deliver much more direct information about the structure. The key to the successful realization was the development of laser driven x-ray sources. In this contribution we report on the optimized generation of soft x-ray pulses via high harmonic generation. We will show that we are able to generate broad continuous spectra ideally suited for XAS. High-order harmonic (HH) generation is a suitable way to generate coherent, well collimated electromagnetic radiation in the extreme ultraviolet and soft-x-ray region [1]. However, the observed low conversion efficiency, especially at higher photon energies is a serious limitation for several applications. Applying non-adiabatic self-phase-matching, quasi-phase-matching [2], and x-ray parametric amplification[3] overcomes these limitations. They allowed the generation of coherent radiation up to few keV's [2] with suitable photon flux for the spectroscopic applications. The XUV signal has been intense enough in an energy range up to 3500 eV opening the way to EXAFS (extended x-ray absorption fine structure). In a first proof-of principle experiment we followed structural changes in Silicon after excitation with an intense laser pulses probed above the L and K absorption edge. The modulation in the EXAFS spectrum is in reasonable agreement with measured phonon spectra of amorphous-Si reported in the literature[4].

[1] C. Winterfeldt, C. Spielmann, G. Gerber, Rev. Mod. Phys. **80** , 117(2008).

[2] J. Seres, et al., Nature Phys. **3** , 878(2007).

[3] J. Seres, et al., Nature Phys. , **6** 455(2010).

[4] E. Seres, C. Spielmann, Appl. Phys. Lett.**91** 121919(2007).

Optical control of spin in nonlinear photoemission from metal surfaces

A. Winkelmann¹, C.-T. Chiang¹, W. C. Lin¹, P. Yu¹, F. Bisio²,
H. Petek³, J. Kirschner¹

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The interaction of polarized light with electronic states that are influenced by spin-orbit coupling provides mechanisms for selective excitation of spin-polarized electrons in nonmagnetic and magnetic solids. The result of spin-orbit coupling in photoexcitation bears close analogy to the effect of a magnetic field, and it provides the means to control magnetic and other spin-dependent phenomena by optical excitation processes on time scales of the order of the applied laser pulse lengths.

We demonstrate how to optically control the spin polarization of photoelectrons emitted in nonlinear photoemission at Cu(001) surfaces under the influence of spin-orbit coupling in the copper electronic structure [1,2].

At magnetic surfaces, spin-selective excitation of photoelectrons can lead to magnetization dependent photoemission intensity changes (magnetic dichroism). As an implication of this effect in two-photon-photoemission experiments, magnetic dichroism from unoccupied quantum well states in ultrathin cobalt films grown on Cu(001) is discussed [3,4].

[1] A. Winkelmann, F. Bisio, R. Ocaña, W.-C. Lin, M. Nyvlt, H. Petek, J.Kirschner, *Phys. Rev. Lett.* **98**, 226601 (2007).

[2] A. Winkelmann, W.-C. Lin, F. Bisio, H. Petek, and J. Kirschner, *Phys. Rev. Lett.* **100**, 206601 (2008).

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To be decided

High harmonic generation for time-resolved photoelectron spectroscopy

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In this contribution we will present our approach to elucidate the evolution of the electronic structure of matter during ultrafast processes such as phase transitions and chemical reactions. We aim at mapping the valence electron rearrangements in real time as atoms are moving. Valence band photoelectron spectroscopy is probably the most direct way to map the valence electronic structure. In order to follow the electronic structure evolution on the femtosecond time scale of atomic motion ultrashort vacuum ultra violet (VUV) pulses are needed for time-resolved valence band photoelectron spectroscopy. We use high-order harmonics of a femtosecond laser as generated in a gas cell [1] or capillary [2] to produce up to 10^{10} photons/s at a photon energy of 23 eV.

With a stable and reliable source for femtosecond time-resolved spectroscopy in mind, different ways of generation, monochromatization, focussing and flux determination of femtosecond VUV pulses will be discussed. We use diffraction gratings with grazing incidence reflection mirrors [1], transmission zone plates [2] or reflection zone plates to monochromatize and focus the radiation. The flux is determined on an absolute level with calibrated detectors. Assets and drawbacks of the different approaches will be discussed in the light of their application for ultrafast surface dynamics.

As an example for the application of this approach we present the ultrafast dissociation of Br₂ molecules in the gas phase [3]. A laser pump pulse initiates dissociation and, for the first time, the entire occupied valence electronic structure is followed from the excited molecule to the free atoms.

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Time-resolved investigation of laser-induced diffusion by SHG microscopy

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We report the development of an experimental technique to investigate the fluence-dependent dynamics of laser-induced surface processes like desorption or diffusion. In our previous work we have shown that diffusion of adsorbates on vicinal metal surfaces induced by femtosecond laser pulses can be sensitively monitored by optical second harmonic generation (SHG) [1]. The strong non-linear dependence of the diffusion rate on laser fluence made it possible to apply a two-pulse correlation (2PC) technique to study the dynamics of the energy transfer between laser-excited substrate electrons to the adsorbate degrees of freedom in the time-domain on a femtosecond time scale [2]. A non-linear dependence on laser fluence is characteristic of all kinds of laser-induced surface processes. It results in a strong variation of the rate across the laser beam profile, which requires either a correction or a spatially selected detection. For this purpose we have extended our experimental setup by a SHG microscope which makes it possible to observe laser-induced diffusion or desorption for a range of laser fluences simultaneously. We illustrate its operation on the basis of a time-domain study on the diffusion of atomic oxygen on vicinal Pt(111). We observe an increase of the width of the 2PC as a function of laser fluence, a phenomenon that should occur for all substrate mediated laser-induced surface reactions [3]. The results are discussed on the basis of different variants of the electronic friction model.

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[3] M. Lawrenz *et al.*, Phys. Rev. B **80**, 075429 (2009).

Electronic Structure of H-Terminated Silicon Surfaces [H-Si(111)] Studied by Two-Photon Photoemission

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Investigating electronic states localized at the surfaces allows us to understand elementary processes of chemical reactions and the mechanisms of charge transfer at the surfaces. In particular, understanding the electronic structure of semiconductor surfaces is very important for surface carrier dynamics. As a prototype of the passivated semiconductor surface, information on the surface properties of a hydrogen-terminated Si(111), abbreviated as H-Si(111), is required from a technological as well as fundamental point of view as an unreconstructed and atomically flat surface.

In this presentation, we demonstrate the capability of two-photon photoemission (2PPE) spectroscopy to probe the surface-state transitions as well as the bulk-state transitions. To date, there has been only one report of experimental evidence for an image-potential state on semiconducting materials, which is a 2PPE experiment on Si(100). Now, we report the detection of an image-potential-state resonance on the H-Si(111). We also show the information on the resonant transitions between the image-potential state and the occupied surface state of the H-Si(111) from the photon-energy dependence [1].

The surface and bulk electronic states of H-Si(111) were studied by 2PPE. With the polarization and emission-angle dependence of the 2PPE spectra, five series of electronic structures for H-Si(111) can be identified. For $h\nu < 4.6$ eV, peak A arises from a surface resonance at ~ 1.01 eV from E_F , and at the photon energy higher than 4.6 eV, peak A indicates photoemission from an intermediate state, corresponding to an image-potential state at 3.61 eV from E_F . Other structures, B, C, and D arise from bulk states, while structure E occurs from an electron-hole excitation process. This work has revealed the surface resonance and the image-potential state on H-Si(111) for the first time.

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One- and two-photon photoemission from silicon surfaces

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One- and two-photon photoelectron spectra with the same total photon energy should give information on the same initial and final states. Differences show the influence of intermediate states in two-photon photoemission (2PPE). Of particular interest are also bulk bands where the conservation of perpendicular momentum leads to peak shifts, resonances and peak narrowing [1,2].

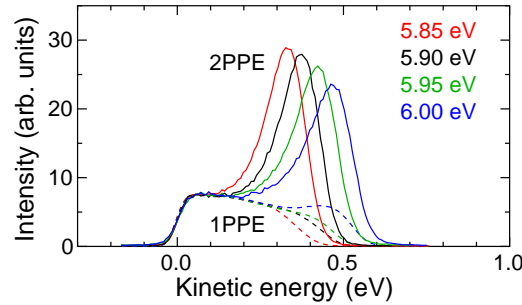


Figure 1. One- and two-photon photoemission spectra in normal emission from Si(100)(2×1) for the same total photon energy and p-polarized light.

We have measured 1PPE and 2PPE from Si(100)(2×1) using the 4th and 2nd harmonic from a Ti:sapphire laser (see Fig. 1). From the photon energy dependence we assign the 2PPE peak to the occupied dangling bond state D_{down} . However, the peak is observed weakly in 1PPE for 6 eV photon energy only. Obviously, 2PPE spectra from D_{down} are strongly influenced by intermediate states which could be related to unoccupied bridge bonds [3].

Bulk transitions observed in 1PPE are seen in 2PPE on some Si surfaces and not on others. Based on these observations we try to develop a consistent picture of the relationship between one- and two-photon photoemission.

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Ultrafast Time Resolved Electron Diffraction of Adsorbate Dynamics on Silicon Surfaces

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Ultrafast time resolved electron diffraction is an excellent technique to study dynamic processes of surfaces like phase transitions and vibrational mode coupling on a picosecond timescale after excitation by a fs laser pulse. The laser energy will excite the electron system and heat the topmost atomic layers by electron-phonon coupling. In our experiment surface sensitivity is obtained by a RHEED (reflection high energy electron diffraction)-geometry [1].

The transient dynamics of the metallic $(\sqrt{3}\times\sqrt{3})$ and the $(\sqrt{3}\times\sqrt{7})$ -Pb reconstructions on Si(111) has been studied in a laser-pump and electron-probe setup. The de-excitation and energy dissipation into the substrate has been measured through the transient surface temperature using the Debye Waller effect on the diffraction pattern. We observe two different time constants of 100 ps and 2800 ps for the de-excitation of the vibrational modes for both systems. These long time constants can be explained by the huge difference in mass of Si and Pb atoms which prevents effective coupling of the Pb vibrational modes to the phonon bath in the Si substrate.

In order to study the dynamics of strongly driven phase transitions at surfaces far away from thermal equilibrium we performed time resolved diffraction on the Peierls like phase transition from a (8×2) to a (4×1) reconstruction of an Indium terminated Si(111) surface upon laser excitation [2]. This phase transition is observed by preparing the (8×2) well below 90 K and excitation of the surface with a fs-laserpulse. After excitation the (8×2) -diffraction spots instantaneously disappear, while the intensity of the (4×1) spots increases. This increase of the (4×1) spot intensity excludes an explanation by the Debye-Waller-Effect and is evidence for a true structural phase transition at a surface.

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Ultrafast, Element-Specific, Demagnetization Dynamics Probed using Coherent High Harmonic Beams

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Element-specific magnetization dynamics on nanometer length scales and femtosecond timescales is a topic of intense current interest. Questions on, e.g., femtosecond magnetization processes, ultrafast dynamics of nanodomains, or ultrafast interaction in magnetic multilayer systems have motivated a variety of studies using magneto-optic effects probed by ultrafast light pulses. Magneto-optical dynamics studies currently make use of either ultrafast lasers or x-rays from large-scale synchrotrons. Ultrafast pulses from femtosecond lasers (~ 20 fs) make high time resolution easily accessible, however, these studies lack element selectivity. X-rays from synchrotrons, on the other hand, enable element-specific probes using photons with energy near the absorption edges of ferromagnetic materials. Moreover, nanometer imaging capabilities are also possible using x-rays. However, the relatively poor time resolution available from x-ray facilities (~ 100 fs using femtosecond slicing) to date has been too slow to resolve the fastest magnetization dynamics.

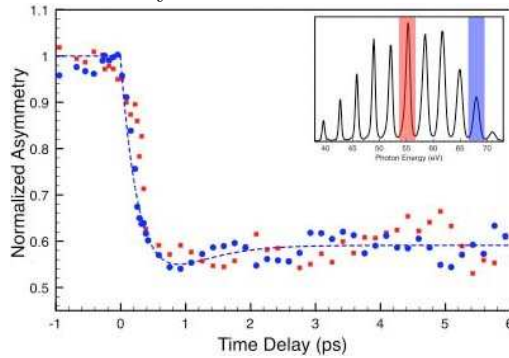


Figure 1: Time-resolved measurement of ultrafast demagnetization in a Permalloy film probed using few-fs high-harmonic beams around the Ni (blue circle) and Fe (red square) M-edge.

Ultrafast, coherent, table-top, x-ray sources based on high-harmonic upconversion of femtosecond lasers promise to overcome these limitations by providing a new tool to study how magnets work at the shortest time and length scales, with element specificity. In this work, we use few-femtosecond extreme ultraviolet (EUV) pulses from high harmonic generation (HHG) to extract demagnetization dynamics and hysteresis loops of a compound material for the first time [La-O-Vorakiat *et al.*, PRL 103, 257402 (2009)]. We measure the fastest, elementally-specific, demagnetization dynamics of any approach to date, with a time resolution of 55 fs.

Rotational anisotropy of optical second harmonic generation from Pt nanowires with boomerang-like cross-sectional shapes

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The nonlinear optics of metallic nanowires is a subject of increasing interest because of the strong anisotropy due to the electronic confinement. Nonlinear optical phenomenon such as second-harmonic generation depends strongly on the symmetry of the structure of the medium. Previously, we measured SH intensity from a C_s -symmetric Pt nanowire array on a faceted MgO(110) substrate. Their cross-sectional shapes were observed by transmission electron microscope (TEM). Then the investigation of SH response from nanowires of other cross-sectional shapes have become desirable. In this research we have observed C_{2v} -symmetric Pt nanowires with the cross sections of boomerangs by TEM and investigated the SH signal from them.

Pt nanowires on the faceted MgO(110) substrates were fabricated by a shadow deposition technique in UHV of 9.5×10^{-7} Pa. From both hand sides of a faceted MgO template, oblique deposition of Pt (2nm) was performed. After fabrication of the Pt nanowires, a very thin (80 μ m) part of a flake with MgO(001) faces and the nanowire cross-sections was observed by TEM. The probe-light pulses with a fundamental photon energy of 2.33eV was generated by a mode-locked Nd:YAG laser. The fundamental light was focused onto the sample with the incidence angle of 45° . The SH signal from the sample was observed.

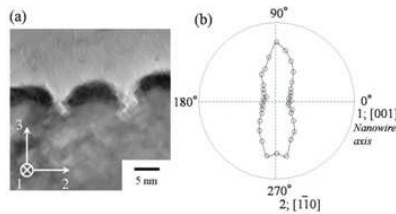


Figure 1. (a) TEM image of Pt NWs, (b) SH intensity in p -in/ p -out polarization configuration.

Fig. 1(a) shows a TEM image of the cross sections of Pt nanowires on the MgO(110) faceted template. According to the TEM image, the shapes of Pt were like boomerangs. The SH intensity in p -in/ p -out polarization configuration depends strongly on the rotation angle φ as seen in Fig.1(b). The SH pattern in s -in/ s -out polarization configuration was very weak. This is because the shapes of the metal cross sections have local mirror symmetry in direction 2.

Ultrafast phenomena probed with time-resolved photoelectron spectroscopy

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Femtosecond time- and angle-resolved photoemission using ultrashort laser pulses is well established for the analysis of excited states and their relaxation in the time domain. This approach has been very successfully applied to metal and semiconductor surfaces, as well as interfaces between adatoms or molecular layers on metal substrates. In order to investigate electron dynamics in the bulk of materials and to profit from spectral and momentum resolution of photoelectron techniques transport effects of the optically excited charge carriers have to be considered. In this talk exemplary approaches using time-resolved photoelectron spectroscopy that aim at bulk dynamics are presented and the current status of results will be presented. (i) Epitaxial growth of metallic layers on semiconductor substrates of 1 to 20 monolayer thickness facilitate within the substrate's band gap a spectral window to analyze electron dynamics in the metal layer with suppressed transport effects. (ii) Layered materials like cuprates or tritellurides can be cleaved in vacuum and the obtained surfaces can give good estimates for bulk dynamics. Such materials furthermore present various cooperative phenomena like superconductivity and charge density wave formation, which become thereby accessible to photoelectrons in time-resolved experiments.

The experimental results reported here have been obtained in close collaboration with R. Cortes, P. S. Kirchmann, L. Rettig, M. Wolf at the Freie Universität Berlin, which is gratefully acknowledged.

Spin-dependent dynamics of carriers injected across interfaces with organic semiconductors

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Organic semiconductors (OSC) have considerably attracted the interest of the scientific community due to the possibility of implementing very low-cost and versatile electronic devices based on organic thin-film materials. Recently, different pioneering experiments suggested that OSC represent as well a major opportunity for application in the growing field of spintronics.

Two fundamental requirements for engineering organic spintronics devices are (i) the efficient injection of spin-polarized carriers at a hybrid inorganic-organic interface (where the inorganic material acts as source of spin polarized carriers), and (ii) a suitable spin diffusion length in the OSC themselves. Since OSC are generally characterized by moderate spin diffusion lengths but very long spin lifetimes, the most straightforward and promising route for application of OSC in spintronics is to exploit the high spin injection efficiency achievable in OSC. In this context, the ability to control and tune the spin functionality of hybrid inorganic-organic (HOI) interfaces is a central issue.

In this contribution, we will show how the spin- and time-resolved two-photon photoemission technique can be employed to quantify the spin properties of the considered HOI interface in terms of a set of microscopic parameters that fully characterize its spin functionality in the accessible energy range, the so-called pure spin-injection region [1,2]. Furthermore, we developed a microscopic interface model [3] including the following spin-dependent events for an electron crossing the interface: (i) transmission without energy loss and without spin-flip; (ii) transmission without energy loss but with a spin-flip; (iii) reflection or transmission with energy loss. Applying this model allowed us, for example, to identify the presence of a spin polarized hybrid state at the interface between a thin cobalt film and the OSC copper phthalocyanine.

[1] M. Cinchetti et al., *Nature Materials* **8**, 115-119 (2009).

[2] H. Ding et al., *Phys. Rev. B* **78**, 075311 (2008).

[3] M. Cinchetti et al., *Phys. Rev. Lett.*, in print.

Coherent control of injection current at surfaces and interfaces: Si(111) and GaAs(110), clean and with adsorbate

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Various coherent control experiments are based on the use of ultrafast optics. We demonstrate theoretically that the so-called one-beam coherent control effect (see, *e.g.* [1]) can be used as a new probe of surfaces and interfaces in cubic semiconductors with or without bulk inversion symmetry. The motivation is that surface sensitive non-invasive optical techniques as, *e.g.*, reflectance anisotropy spectroscopy (RAS) or second harmonic generation (SHG), crucial for understanding surfaces and interfaces, are not sensitive or cannot be applied to many important systems: RAS is not sensitive to, *e.g.*, optically isotropic Si(111), while SHG signal for III-V or II-VI cubic compounds is influenced by nonzero bulk contribution. The coherent control effect, which we propose, vanishes in the bulk of both homopolar and compound cubic semiconductors, but is allowed for most surfaces or interfaces due to their reduced symmetry. We calculated the injection current from the first principles for prototypical Si and GaAs surfaces, namely hydrogenated and clean Si(111)(2 × 1), and the clean and Sb-covered GaAs(110)(1 × 1). To get even more detailed access to the surface/interface properties, we applied a microscopic layer-by-layer formalism that separates the optical response from each subsurface atomic layer. The effect is shown to be sensitive to the surface or interface and its response can be explicitly interpreted in terms of the surface electronic structure. Calculated effect magnitudes indicate that the coherent control current should be easily observable experimentally and demonstrates distinctive photon energy dependence for all of the surfaces considered. This can be a very efficient surface sensitive technique since (i) the injection current originates only from the surface or interface of cubic systems, (ii) it can be easily extracted from any spurious photovoltaic backgrounds due to its dependence on the phase difference between two orthogonally polarized beams; (iii) the coherent control current response is essentially dependent on the microscopic atomic structure, surface symmetry, type of the bonding and adsorbate. Last but not least, this effect can be detected with terahertz radiation.

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Charge-Transfer Excitons at the Interface of Organic Semiconductors

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The concept of an organic photovoltaic cell relies crucially on efficient charge separation at the interface of two different organic materials [1]. Ideally, strongly bound and localized excitons which were produced by light absorption would break up to form free carriers at the interface. Due to poor dielectric screening of charge in organic materials, however, the Coulomb attraction is long-ranged and relatively strong. As a consequence, the electron and hole – even though located on opposite sides of the interface – may be trapped in a localized charge-transfer (CT) exciton state.

This presentation will discuss the observation of a Rydberg-like series of CT exciton states at the surfaces of pentacene and tetracene thin films using femtosecond time-resolved two-photon photoelectron spectroscopy (TR-2PPE). The surface of an organic thin film serves as a model system for a donor-acceptor interface where the role of the electron acceptor is taken by vacuum. The energetics of the CT excitons can be understood qualitatively in a conceptually simple Coulomb picture where the electron (in vacuum) is attracted by the image potential at the surface and the Coulomb potential of the hole [2]. Depending on the pump photon wavelength, different states from the lowest state with a binding energy around 0.4 eV up to the image potential state, representing the two-dimensional continuum, are excited. The lifetimes of the CT exciton states is of the order of 50-100 fs. Higher lying states decay rapidly into lower states [3].

[1] X.-Y. Zhu, Q. Yang, M. Muntwiler, *Acc. Chem. Res.* **42**, 1779 (2009)

[2] M. Muntwiler, Q. Yang, W. A. Tisdale, X.-Y. Zhu, *Phys. Rev. Lett.* **101**, 196403 (2008)

[3] Q. Yang, M. Muntwiler, X.-Y. Zhu, *Phys. Rev. B* **80**, 115214 (2009)

Near- and Far-Field Effects on Excited States at Organic Semiconductor / Metal Interfaces

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We present an investigation of the evolution of highly excited states at the interface of the dipolar organic semiconductor vanadyl naphthalocyanine on highly oriented pyrolytic graphite. Using twophoton photoemission we observe several image states at sub-monolayer to few-monolayer coverages: The first image state evolves seamlessly out of the graphite image state, with increased binding energy between zero and one monolayer, while the second image state appears at coverages of just below one monolayer and is pinned to the vacuum level. These findings can be understood in the context of a simple electrostatic model [1] that considers how molecular levels and vacuum level are influenced differentially by the strong electrostatic fields present at the interface with dipolar molecules: While the vacuum level rises continuously with coverage, the molecular states are significantly depolarized as a function of electric fields in the near-field regime. This depolarization, while originally generating a hybrid molecular anion / graphite image state whose binding energy increases with coverage, ultimately diminishes the coupling of the image state to the molecular affinity level by driving the affinity level out of resonance with the image state. Interfacial electrostatic fields may therefore be used to manipulate in a concrete fashion interfacial charge transfer processes such as indirect photoinduced interfacial electron transfer.

[1] Michael L. Blumenfeld, Mary P. Steele, Oliver L.A. Monti, J. Phys. Chem. Let. **1**, 145 (2010).

Ultrafast electron transfer across polar molecule-metal interfaces: strong coupling vs. molecular screening

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Heterogeneous charge transfer plays a crucial role for the development of molecular electronics such as, for example, OLEDs. Depending on the side conditions, the respective charge transfer dynamics vary significantly. In particular, in the presence of (polar) molecules, the electron transfer (ET) reaction may be strongly influenced by the screening properties of the involved molecules, as they determine the timescales on which electron transfer occurs.

We adsorb D₂O and NH₃ on Cu(111) and Ru(001) surfaces as model systems for systematic investigation of the ultrafast ET dynamics at polar molecule-metal interfaces by means of time-resolved two-photon photoelectron (2PPE) spectroscopy. With a first laser pulse, electrons are excited in the metal and injected into the adsorbate layer (solvent) where they localize at favorable sites. The subsequent electron (back) transfer to the substrate is monitored with a second, time-delayed laser pulse. Our study unveils that, in the case of amorphous adsorbate structures, (i) molecular screening is observed for $\tau > 200$ fs after electron injection, (ii) before, ET is dominated by the strong coupling to the substrate states. Moreover, we vary the coupling strength of the excess electrons to the respective substrate by variation of the structure, morphology, and layer thickness of the adsorbate. In particular the structural and morphological transition from amorphous layers to 3D crystallites results in a considerable increase of excess electron residence times from ultrafast timescales up to several minutes.

Coherent optical phonons on semiconductor surfaces studied with transient reflectivity

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Coherent optical phonons are the lattice atoms vibrating in phase to each other over a macroscopic spatial region. They are excited either by electric field of the optical pulses through non-linear susceptibility (*impulsive stimulated Raman scattering*) or as a result of photoexcitation into the excited electronic states (*transient depletion field screening* and *displacive excitation of coherent phonons*). Pulse-trains and pulse-shaping techniques have enabled mode-selective excitation of the coherent phonons and have achieved vibrational states far from thermal equilibrium. Our experiments with shaped pulses have revealed that, for a given photon energy, the amplitude of the coherent phonons is determined by the spectral component at the phonon frequency. Pump-probe reflectivity/transmissivity measurements, second harmonic generation (SHG), and X-ray diffraction (XRD) are the three major detection techniques of the coherent phonons. While SHG and XRD monitor atomic layers and bulk crystals, respectively, the reflectivity measurements on opaque semiconductors can tune the probing depth by tuning the wavelength of the probe light. We demonstrate that near-UV pulses monitor exclusively the depletion layer of doped GaAs, in which ultrafast dynamics of coherent phonons and plasmon-coupled modes are in striking contrast with their counterpart in the bulk.

Ultrafast vibrational dynamics of CO on Pt(111) studied by time-resolved SFG with phase-sensitive detection

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Time-resolved sum-frequency generation (SFG) has been known as a powerful method to elucidate vibrational dynamics at surfaces. In this paper, we report on our recent achievement to combine a phase-sensitive (heterodyne) detection with the femtosecond IR-visible SFG spectroscopy for carbon monoxide adsorbed on Pt(111) under an UHV condition. We show that the novel technique enables us to obtain separately the real and imaginary parts of the transient changes in $\chi^{(2)}$, and that the $\text{Im}[\chi^{(2)}]$ of C-O stretching region shows marked transient asymmetry upon excitation with 400 nm, 150 fs pulses, which is ascribed to Fano interference between C-O stretching mode and substrate electronic excitations. We have found that the excitation of frustrated modes enhances the nonadiabatic coupling of the C-O mode with substrate electronic excitation and leads to the spectral asymmetry.

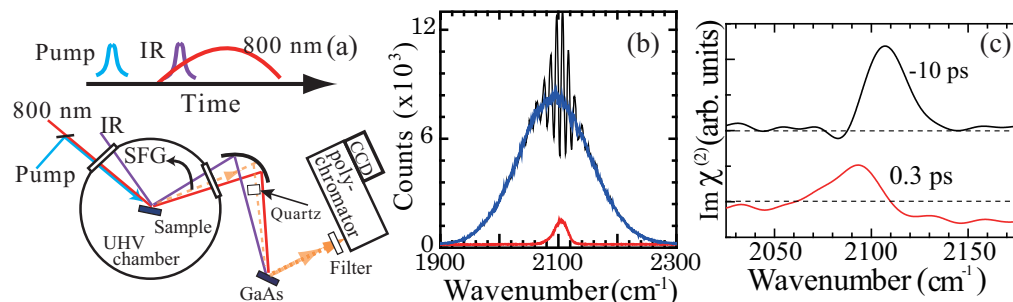


Figure 1. (a) A schematic diagram of setup for time-resolved SFG with heterodyne detection. (b) SFG spectra of on-top CO on Pt(111) observed with heterodyne (black) and homodyne (red) in addition to a spectrum of local oscillator (blue). (c) Time-resolved $\text{Im}[\chi^{(2)}]$ spectra of C-O stretching band. Pump-IR delay time is indicated in the figure.

Two competing mechanisms for photoinduced Cs dynamics on Cu(111)

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Femtosecond pump-probe spectroscopy has clarified the generation mechanism of photoinduced coherent nuclear motions for various systems ranging from isolated molecules to bulk solids. However, the mechanism for adsorbates on metals is less explored and the most fundamental question remains to be answered: What type of electronic transitions is responsible for kicking the adsorbates? In the present paper, we report our experimental and theoretical studies on this question in the photoinduced coherent nuclear motions of Cs adsorbed on Cu(111). Contrary to our previous reports on the other alkalis, where substrate-mediated excitations were inferred exclusively, we show signatures of surface-localized processes whose contribution depends on excitation photon energy. Time-resolved SHG measurement with ultra-high vacuum has been done in the manner previously reported [1]. We have found that the initial phase of the coherent vibration of the Cs-Cu stretching mode shows marked excitation wavelength (λ_{ex}) dependence: the oscillation shows sin-like behavior for $\lambda_{\text{ex}}=800$ nm, while cos-like oscillations were observed for $\lambda_{\text{ex}}<560$ nm. In addition, the power dependence of the initial amplitude shows a strong saturation behavior at $\lambda_{\text{ex}}=800$ nm, which indicates the excitation mechanism depends on the pump photon energy. The adsorbate-metal surface systems generally have a few metastable surface-localized excited states embedded in the metal continuum states. We consider a simplified three-state model [2] consisting of the ground, surface-localized excited, and background continuum states. Under the resonant excitation, the nuclear wavepacket simulation actually gives the sin-like oscillation and the saturation tendency. The experiment with $\lambda_{\text{ex}}=800$ nm is the first experimental realization of the transient-adsorbate mediation mechanism recently proposed in [2]. For $\lambda_{\text{ex}}<560$ nm, the substrate excitation channels open. The substrate-mediated hot electrons scatter on the surface-localized state. The corresponding electronic state gives the cos-like oscillation. We propose that these two mechanisms are fundamental for adsorbates on metals. In both mechanisms, the surface-localized state plays essential roles to displace the adsorbate position.

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Imaging the femtosecond time scale correlated electron-nuclear dynamics in surface photodesorption

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We employ time-resolved two-photon photoemission to characterize electronic structure and photoinduced dynamics of chemisorbed alkali atoms on noble metal surfaces. Photoinduced charge transfer excitation of the lowest energy sigma resonance of Cs on Cu or Ag surfaces turns on repulsive forces between atom and surface initiating nuclear wave packet motion on a dissociative potential energy surface. Energy, momentum, and time resolved measurements of photoemission from desorbing atoms provide information on the nuclear wave packet motion and the concomitant changes in the surface electronic structure. In particular, we use time-dependent momentum imaging of photoemission from the excited state to explore the correlation between the electron and nuclear motions.

Contrasted behaviour of terrace and edge CO adsorbed on Pd nanoparticles studied by pump probe SFG

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Nanoparticles are the subject of many studies due to their applications in various fields, like plasmon enhanced optical detection of biological molecules, or nanocatalysis. Using linear absorption femtosecond laser techniques, the sensitivity has reached single nanoparticle detection even in biological environments. The dynamics of nanoparticles following femtosecond laser excitation has been studied by linear absorption pump-probe experiments. It was shown the increasing importance of surface scattering of the electrons as the particle size decreases, and the ability to excite vibrational modes of the nanoparticle.

To obtain more specific information on processes occurring at the nanoparticle surface following a femtosecond laser pulse, a technique able to detect specifically adsorbed molecules is required. Pump probe sum-frequency generation (SFG) with femtosecond pulses is a unique tool to monitor photodesorption [1], surface diffusion [2], and energy relaxation on single crystals [1,3]. In this work, we report such experiments on CO adsorbed on a model catalyst, namely Pd nanoparticles epitaxially grown on a MgO film grown on Ag(111), of size a few nm. SFG spectra show CO at terrace bridge adsorption site and at several sites located at nanoparticle edges, in agreement with a previous SFG study done with picosecond lasers [4]. Vibrational frequencies are observed to vary with coverage and particle size. Pump probe experiments show broadening and red shift of vibrational bands similar to the case of single crystals, and photodesorption is observed. The most striking feature is that terrace sites are much more sensitive to electronic excitation and photodesorption than edge sites, showing that photoelectrons are weakly coupled to CO at edge sites.

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Ultrafast transport of laser-induced spin polarized carriers in Au/Fe/MgO(001)

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The ultrafast magnetization dynamics induced by a transport of spin polarized carriers is a hot topic due to the fundamental interest in spin excitations, their coupling to electron and lattice sub-systems and applications in spintronics and data storage. To understand the underlying elementary processes, we have developed a time domain approach to probe the hot electron-induced spin dynamics. Here we report on experiments in epitaxial Au/Fe/MgO(001) structures, where the Au layer thickness is much larger than the optical penetration depth and comparable to the electron mean free path. Hot carriers are excited in the magnetized Fe layer by 800 nm 35 fs 30 nJ pump pulses of Ti:sapphire oscillator focused from the side of MgO substrate. Owing to the exchange-split Fe band structure, minority and majority carriers are excited at different energies and concentrations. These *hot*, or *non-equilibrium*, spin-polarized carriers propagate across the Fe/Au *interface* towards the Au *surface* forming a spin-polarized current with the charge component screened by a displacement of *equilibrium* carriers in Au. The transient spin polarization (SP) of the Au *surface* is detected by (*surface sensitive!*) magneto-induced second harmonic generation from the probe pulse focused from the side of Au layer of thickness d_{Au} . After a 40 fs pump-probe delay similar to $d_{Au}=50$ nm divided by the Fermi velocity, a detectable magneto-optical signal builds up, changes its sign at a 300 fs time scale and then vanishes at a 1 ps time scale. Increasing d_{Au} increases the delay of this build-up and stretches the time profile. According to *ab initio* band structure calculations, the observed non-monotonicity can be attributed to different life times of hot carriers with different SP. If the negative SP is transported predominantly in the ballistic regime, the transport of positive SP is essentially diffusive and thus has a considerably smaller velocity. Under certain experimental conditions it can be suppressed almost completely, which results in a single-polarity 50-100 fs pulse of spin current corresponding to spin polarized carrier transport on the order of 10^{10} A/cm². The polarity is defined by the magnetization of Fe layer and thus can be easily varied in experiments on the spin transfer torque-induced magnetization dynamics.

Spin-dependent lifetimes of hot electrons in 3d ferromagnets

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The spin-dependent lifetimes of electrons excited to energies of a few hundred meV above the Fermi energy E_F —so-called hot electrons—of the 3d ferromagnets Fe, Co and Ni have been investigated experimentally and theoretically for more than a decade. They are of high technological relevance through the associated giant magneto-resistance effect and are of fundamental interest—some of the underlying microscopic physical processes are not yet fully understood. The first and up to now only spin-resolved two-photon photoemission (SR-2PPE) measurements have been performed in Refs. [1,2], where thin metal films were grown on Cu(100) and the surfaces were covered with Cs to lower the work functions.

We present bichromatic infrared pump and ultraviolet probe SR-2PPE data that give experimental access to smaller energies above E_F and thereby extend the experimental data range, without the need of Cs adsorption. We find good agreement with the previously measured data [1,2] in the overlapping energy range and discuss discrepancies with recent *ab-initio* calculations. A possible reason for the discrepancies are spin-flip exchange-scattering processes that we could observe on the clean, i.e. Cs-free, Co surface.

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Two-dimensional Fano resonances on Si(001)

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The interference between different indistinguishable excitation paths is a fundamental quantum mechanic phenomenon in cooperative systems. The coupling between discrete and continuum states can significantly modify its interaction with light resulting in Fano resonances. These interference phenomena have enabled the understanding of many systems in atomic and nuclear physics and artificial structures have been built to explore the nature of Fano resonances in the solid state.

We present a two-dimensional extension of Fano's theory for the description of two discrete states both degenerate with the continuum. As a model system we have studied transitions between the dangling-bond and image-potential states at the (100) surface of silicon, which are degenerated with the bulk continuum of valence and conduction bands. Tuning the photon energy reveals Fano line-shapes of the intensities in both the initial- and intermediate-state signals. These double state - double continuum interference phenomena allow us to deduce coupling strengths between surface and bulk states in silicon, and to estimate the lifetime of single hole excitations in an inhomogeneously broadened system.

Ultrafast Multi-Exciton Dissociation Dynamics in CdSe Quantum Dots

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Charge transfer to and from quantum dots (QDs) is of intense interest because of its important roles in QD-based devices, such as solar cells and light emitting diodes. Recent reports of multiple exciton generation (MEG) by one absorbed photon in some QDs offer an exciting new approach to improve the efficiency of QD-based solar cells and to design novel multi-electron/hole photocatalysts. However, two main challenges remain. First, the efficiency of MEG process remains controversial and may need to be significantly improved for practical applications. Second, the utilization of the MEG process requires ultrafast exciton dissociation prior to the exciton-exciton annihilation process, which occurs on the 10s to 100s ps time scale. In this presentation we report a series of studies of exciton dissociation dynamics in quantum dots by electron transfer to adsorbed electron acceptors. We show that excitons in CdSe can be dissociated on the a few picosecond timescale to various adsorbates. As a proof of principle, we demonstrate that multiple excitons per QD (generated by multiple photons) can be dissociated by electron transfer to adsorbed acceptors (J. Am. Chem. Soc. 2010, 132, 4858-4864). We will discuss the dependence of these rates on the size and the nature of the quantum dots and possible approaches to optimize the multiple exciton dissociation efficiency.

Lifetimes of electron excitations on clean and nanostructured metal surfaces

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Interaction between lattice and electron subsystems as well as interaction within each of these subsystems is crucial to understand mechanisms of single-particle excitation dynamics, i.e. lifetime of excitations. The lifetime sets the duration of excitation and in combination with the velocity determines the mean free path, a measure of influence of the excitation. In this presentation I discuss recent theoretical results on the decay of excited electrons and holes on clean metal surfaces, at single adatoms, at islands as well as in overlayers and free standing thin films. Different decay mechanisms and different kinds of interactions - elastic and inelastic electron- electron (e-e) interaction as well as electron-phonon (e-ph) interaction and the role of spin (spin-orbit and exchange interaction) - are analysed. E-ph decay channel is shown to be important for all systems considered. In the e-e decay channel the electron (hole) decay can be realized via creation of electron-hole pairs or plasmon excitation. Dimensionality effects in the lifetime of electrons and holes on metal surfaces and the role of screening and intra- (inter-) band transitions are also discussed.

Attosecond spectroscopy on surfaces

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Over the past decade novel ultrafast optical technologies have pushed the duration of laser pulses close to its natural limit, to the wave cycle, which lasts somewhat longer than one femtosecond ($1 \text{ fs} = 10^{-15} \text{ s}$) in the visible spectral range. Atoms exposed to a few oscillation cycles of intense visible or near-infrared light are able to emit a single XUV photon wavepacket of sub-femtosecond duration [1]. These XUV pulses together with the few-cycle (few-femtosecond) laser pulses used for their generation have opened the way to the development of a technique for attosecond sampling of electrons ejected from atoms or molecules [2]. This is accomplished by probing electron emission with the oscillating electric field of the few-cycle laser pulse following excitation of the atom by the synchronized sub-femtosecond XUV pulse. First experiments have been carried out to measure sub-femtosecond behavior of matter. Recently, the dynamics of the photoionization process on solids has been studied [3]. Not only that attosecond metrology now enables clocking of surface dynamics, but also the individual behaviour of electrons of different type (core electrons vs. conduction band electrons vs. adlayers) can be resolved. Here, we measured a time delay of about 100 as on the emission of the aforementioned types of electrons (see Fig. 1).

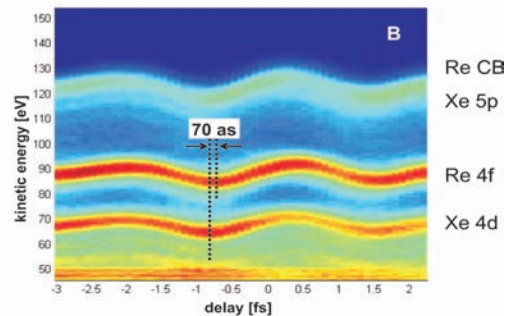


Figure 1. Spectrograms of Xe and Re electrons showing a delay in emission.

The information gained in these experiments may have influence on the development of many modern technologies including semiconductor and molecular electronics, optoelectronics, information processing, photovoltaics, electrochemical reactions, or electronically stimulated chemistry on surfaces and interfaces.

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Ultrafast electron dynamics in pump-probe spectroscopies of surfaces: from transient excitonic to quasi-stationary polaronic states

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Rapid development of pump-probe spectroscopies has greatly advanced the studies of electronic excitations in the real time domain. State- and time-resolved two-photon-photoemission (TR2PPE) measurements utilizing ultrashort laser pulses with variable delay have provided unprecedented insight into the dynamics of quasiparticles at metal surfaces. This has enabled detailed investigations of quasiparticle energetics and decay in the intermediate states of 2PPE from quasi-two-dimensional surface bands which are of fundamental interest in many aspects of surface science. In particular, occupied surface state- (SS) and unoccupied image potential-state (IS) bands constitute ideal prototype systems for studying ultrafast quasiparticle dynamics and relaxation phenomena in restricted dimensionality. So far the majority of analyses of 2PPE data from surface bands have invoked pre-existent SS- and IS-states whose many-body properties were assessed *a posteriori* by perturbation methods. However, since IS-bands arise from an interplay of the surface band gap and retarded polarization interaction, the image potential supported IS-states are fully developed only after the completion of screening of excited quasiparticle charges on the time scale of several surface plasmon cycles. Consequently, such states are mainly inaccessible in ultrafast measurements utilizing laser pulses of few femtosecond duration and delay. Instead, the evolution of an optically excited electron with energy below the vacuum level and a SS-band hole in the intermediate state of TR2PPE should be visualized as a transient exciton which with the formation of image charge evolves into a weakly correlated quasiparticle pair (IS-electron+SS-hole). The corresponding many-body scenario maps onto the problem of propagation of an electron-hole pair initially governed by excitonic interactions which in the course of time give way to polaronic interactions of each quasiparticle with the developing screening cloud and thereby to establishment of IS-band states. Temporal stages of these processes will be illustrated and quantified on the paradigmatic example of 2PPE from Cu(111) surfaces.

Theoretical investigation of ultrafast laser-induced magnetization dynamics in many-electron systems

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Ultrafast optical control of a magnetic state of a medium is presently a subject of intense research. It is of importance for the development of novel concepts for high-speed magnetic recording and information processing. Series of experiments has revealed direct optical control on magnetization via inverse Faraday effect [1]. In these experiments femtosecond circularly polarized high-intensity laser pulses are used to excite the sample. But the mechanism of the orbital momentum transfer from light to the medium, which defines the fundamental time limit on magnetic switching, is still not understood.

In order to get insight to the origin of the inverse Faraday effect we investigate the magnetization dynamics of atoms, which are components of materials used in experiments, excited by femtosecond circularly polarized Gaussian-shaped laser pulses. We study the stimulated Raman-like scattering process, which was suggested to be responsible for the magnetization reversal by light [2]. The time-dependent Schrödinger equation is solved up to the second order using the Volterra iteration method to describe the action of the laser light on the system. The time evolution of the probability of excitation from the ground state to an excited level and back to the ground state with a different magnetic state is calculated. We show, that due to this process the magnetization of the system changes after the action of the laser pulse on it.

We are thankful for the financial support of the FANTOMAS project.

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Resonance Effects on Two-Photon Photoemission of Lead Phthalocyanine Thin Films on Graphite

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Two-photon photoemission (2PPE) spectroscopy has been performed for lead phthalocyanine films on Highly Oriented Pyrolytic Graphite (HOPG) surface. Fully resolved occupied and unoccupied levels and resonant optical transitions between them are characteristics of the film surface [1]. 2PPE peaks due to photoemission from normally unoccupied levels (1ω peaks) and those due to coherent two-photon process from occupied levels (2ω peaks) show unexpected variations in intensities and widths when the pump photon energy crosses the resonances[2]. At around a resonance between molecule-derived levels, we find an intensity switching in which only the 2ω peak appears at photon energy below the resonance, and at above the resonance, the 1ω peak becomes prominent and the 2ω peak becomes very weak. The 1ω peak is broadened with increasing photon energy. These results cannot be interpreted by a simple energy level scheme, and point to further understanding of 2PPE process. The broadening of the 1ω peak can be interpreted in the framework of the hole scattering mechanism [3], and suggests that the resonantly excited state is not a molecular exciton in which the hole and electron are localized within a molecule.

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Logic functionalization and angular momentum conservation in magnetic nanoclusters

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We present an *ab initio* theory of ultrafast nanologic elements based on optical Λ -processes [1-3]. Using high-level quantum chemistry we show that in 2- and 3-magnetic-center structures both spin flips and spin transfers are possible within a hundred femtoseconds. From 3-magnetic-center clusters we are able to construct OR, XOR (CNOT), and AND gates [2]. Thus multicenter magnetic clusters allow to exploit spin dynamics for full-fledged logic functionalization.

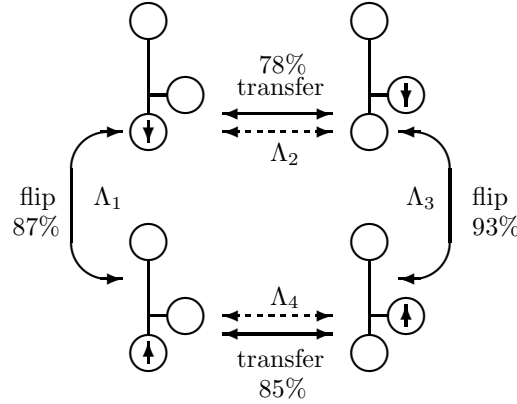


Figure 1. Possible spin-manipulation mechanisms in the Ni_3Na_2 cluster.

In order to explain the angular momentum conservation during the process we propagate in time the intragap levels of a NiO cluster under the influence of a laser pulse [3]. Using quantum optics analysis we show how the coherently induced material polarization leads to angular-momentum exchange between the light and the irradiated antiferromagnetic NiO (001) surface. We also predict a dynamic Kerr-effect, which provides a signature for monitoring spin-dynamics, by simply measuring the transient rotation and ellipticity of the reflected pump beam [1].

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An Element-specific View on Ultrafast Magnetization Reversal of GdFeCo

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Recent experiments have shown that femtosecond (fs) laser excitation is able to manipulate and even control spins in magnetic systems at unprecedented speeds [1,2]. In particular, a radical new approach in switching the magnetization using circularly polarized fs laser pulses has been recently demonstrated [2]. All these experiments have raised intriguing questions about ultrafast channels of angular momentum transfer from and to the spin system and microscopic processes governing the all-optical magnetization switching.

Here, we report on element- and time-resolved X-ray magnetic circular dichroism (TRXMCD) investigations of the fs laser-induced magnetization reversal of the ferrimagnetic GdFeCo alloy. We trigger the magnetization switching by driving the sample over its magnetization compensation temperature upon excitation with linearly polarized fs laser pulses. The subsequent dynamics of the Fe and Gd magnetic moments of the composite alloy is probed with 100 fs X-ray pulses. The TRXMCD data reveal a clearly distinct switching dynamics at the Fe and Gd sites: while Fe magnetic moment switches within 400 fs it takes around 2 ps for the Gd moment to reach the magnetization reversed state. This observation is highly intriguing since the expectation is to have identical dynamics for both sub-lattices accounting for the strong exchange interaction which governs their magnetic ordering. These results suggest a novel non-equilibrium state to be responsible for the genuine magnetization switching process where the elemental magnetic moments show a highly divergent transient behavior. The nature of the novel non-equilibrium state, its possible origins and the implications with respect to the all-optical magnetization switching mechanism will be discussed.

Funding from European Union through UltraMagnetron program and NWO is gratefully acknowledged.

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Ultrafast collapse of periodic long-range order tracked by time-resolved XUV-ARPES

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The combination of ultrafast light sources in the UV spectral regime and the technique of photoelectron spectroscopy provides a unique tool for a most direct access to ultrafast processes in condensed matter systems that couple to electronic degrees of freedom. Angular resolution enables one in this context to monitor the temporal evolution of the valence electronic band structure of a solid at selected - and possibly critical - points in momentum space [1]. In this contribution we will present time- and angle-resolved photoemission spectroscopy (trARPES) experiments using femtosecond XUV pulses generated in a high harmonic light source, to study the ultrafast dynamics of a laser-induced phase transition in 1T-TiSe₂. In response to a photoexcitation with an intense IR-pulse, we monitor the transient suppression of a phase characteristic band backfolding, indicative for a loss of long-range order in the CDW state (see Fig. 1). The characteristic response time of this process critically depends on the IR pump fluence and we observe response times as short as 20 fs. We show that dynamical screening because of the transient generation of free carriers accounts for the observed timescales, the amplitudes of suppression, and the fluence dependence.

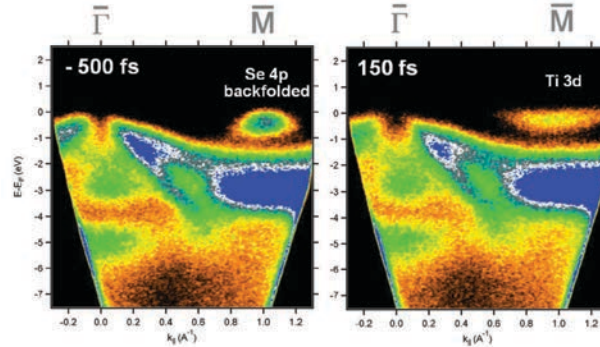


Figure 1. Time-resolved ARPES snapshots of 1T-TiSe₂ prior (left) and after (right) excitation with a 30 fs IR laser pulse; the changes at the M point arise from the transient population of the Ti 3d band and the suppression of Se 4p band backfolding due to the (2x2x2) CDW reconstruction.

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First-principles calculation of charge transfer times: analysis of the spin dependence in Cs/Fe(011)

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We have recently developed a scheme to compute resonant charge-transfer times from adsorbates on metal surfaces using first-principles density functional calculations [1]. We combine the information from surface calculations using the traditional finite slab approach with bulk calculations of the substrate material, and then compute the surface Green's function using recursive methods with arbitrary energy resolution.

We have applied this scheme to study several systems: i) alkalis on noble-metal surfaces in the low [1] and high coverage limits [2]; ii) the c(4x2)-S/Ru(0001) surface, for which we found, in agreement with core-hole-spectroscopy measurements, charge-transfer times well below the femtosecond scale [3] with a peculiar dependence on the polarization of the excitation light [4]; iii) Ar monolayers on Ru(0001) [5] with charge transfer times in the femtosecond scale.

As a new application, I will present here calculations for Cs adsorbed on Fe(011) [6]. In principle, using light with the appropriate polarization, it is possible to excite electrons with a well defined spin orientation from the spin-orbit-split $5p$ levels onto the $6s$ unoccupied resonance of Cs. At the energy of the $6s$ resonance, the calculated density of electronic state in the substrate is widely different for both spin channels. We analyze in detail if this spin-dependence of the number of available states in the substrate also translates into an appreciable spin-dependence of the elastic lifetimes.

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Coherent Control and Nonlinear Optics in Carbon Nanotubes and Graphitic Layers

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I will describe our recent experiments to examine 2nd order and 3rd order nonlinear optical processes in 1-D (Carbon nanotubes) and 2- D (graphene and graphitic films) forms of pure carbon. Results will include the study of how second harmonic response varies for the multilayer carbon systems, as one proceeds from graphene to bulk graphite samples mounted on an oxidized silicon wafer. I will also discuss recent experiments on the use of coherence control techniques to study graphite and both semiconducting and metallic nanotubes

Electronic properties of graphene on Ir(111)

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Epitaxial graphene on Ir(111) prepared in excellent structural quality [1] appears to be an attractive model system for the investigation of intrinsic electronic properties of graphene modified by weak interactions with a supporting surface[2]. Angle resolved photoemission spectroscopy (ARPES) clearly displays a Dirac cone, a hallmark of the unusual band structure of graphene[3], with the Dirac point shifted only slightly above the Fermi level which indicates marginal substrate induced doping. The Moiré structure resulting from the overlaid graphene and Ir(111) surface lattices imposes a superperiodic potential giving rise to opening of minigaps in the band structure. ARPES of the electron doped graphene reveals a whole variety of many body interactions[4]. The π^* band of potassium intercalated graphene showed noticeable renormalization within the wide energy range. Near the Fermi level the renormalization is induced by electron-phonon interactions while the change of dispersion at higher binding energies is induced by the hole-plasmon interaction.

[1] J. Coraux, A.T. N'Diaye, M. Engler, C. Busse, D. Wall, N. Buckanie, F.J.M. zu Heringdorf, R. van Gastel, B. Poelsema, and T. Michely, *New Journal of Physics* **11** 023006 (2009)

[2] I. Pletikosić, M. Kralj, P. Pervan, R. Brako, J. Coraux, A. N'Diaye, C. Busse and T. Michely, *Phys. Rev. Lett.* **102** 056808 (2009)

[3] K.S. Novoselov, A.K. Geim, S.V. Morozov, D. Jiang, M.I. Katsnelson, I.V. Grigorieva, S.V. Dubonos, and A. Firsov, *Nature* **438** 197 (2005)

[4] A. Bostwick, H. Ohta, T. Seyller, K. Horn, O. Rotenberg, *Nature Physics* **3** 35 (2007)

Image-potential states of graphene on iridium

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A single layer of graphene can be produced by decomposition of hydrocarbons on metal surfaces. The resulting adlayer is electronically almost completely decoupled from the metal as known from photoemission data [1]. A comparative study of the image-potential states in front of graphene on (111) and (100) surfaces of iridium was performed by time- and angle-resolved two-photon photoemission measurements using a display-type analyzer. The work function of a single layer of graphene (4.6 eV) is close to the value for graphite even for the Ir substrates with rather high work function >5.6 eV. Energies, dispersion, and lifetimes of the lowest two image-potential states are presented.

[1] I. Pletikosić, M. Kralj, P. Pervan, R. Brako, J. Coraux, A. T. N'Diaye, C. Busse, and T. Michely, *Phys. Rev. Lett.* **102**, 056808 (2009).

From perturbative to strong-field emission: energy-resolved ultrafast laser-induced electron emission from sharp metal tips

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We investigate electron emission from sharp metal tips induced by few-cycle femtosecond laser pulses. Laser-triggered nanoscale electron emitters should be ideal electron sources for e.g. ultrafast electron diffraction, but should also give insight into fundamental questions of light-matter interaction. Recently, a variety of experiments has been done in this field [1]. Because of the complexity of the system and the non-linear behavior involved, different experimental parameters can favor very different emission processes. Intimately linked to the processes is the question of energy width and emission duration. Here we present energy-resolved measurements that enable us to uniquely identify these processes.

In our experiment, ~ 6 fs infrared laser pulses of up to a few nJ pulse energy are focused tightly onto the apex of atomically clean tungsten tips. We observe three qualitatively different emission processes. At low laser intensity and with a small negative DC voltage applied to the tip, three- or two-photon above-barrier emission dominates (multiphoton photoemission). With large bias voltages, one-photon photo-excitation and subsequent tunneling through the barrier sets in (photoassisted field emission). At high laser intensities, peaks corresponding to the absorption of up to 9 photons are observed (above-threshold photoemission, ATP). With increasing intensity, we observe a shift of the first ATP peak towards smaller kinetic energies, reflecting the ponderomotive energy of electrons in the laser pulse. From this shift and the knowledge of the laser parameters we infer a local electric field enhancement factor of about 6 at the tungsten tip apex. Here our experiment is situated in a regime where a perturbative description of the emission process is not sufficient anymore. We present a numerical simulation that shows good qualitative agreement with the experiment and reveals that the temporal structure of the laser electric field is resolved in the emission current.

[1] Hommelhoff et al., PRL **96**, 077401 (2006), PRL **97**, 247402 (2006); Ropers et al., PRL **98**, 043907 (2007); Barwick et al., New J. Phys. **9**, 142 (2007); Tsujino et al., APL **94**, 093508 (2009); Yanagisawa et al., PRL **103**, 257603 (2009)

Ultrafast electronic response and charge transfer at surfaces

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In these last decades it has been demonstrated that the optical properties of semiconductors near the electronic bandgap are dominated by excitons and their residual interactions, producing many-body effects, such as band-gap renormalization (BGR) [1], and nonlinear optical mechanisms. In particular, a deep insight has been gained on understanding the BGR in semiconductors by means of time-resolved non-linear optical spectroscopy in the femtosecond time-domain [2]. However, in semi-metallic and gap-less systems the response to strong photoexcitations is still unclear, and the studies of the dynamics and interactions of quasiparticles have been only marginal. Graphite is an interesting and intriguing material for, at least, three reasons: i) its low-dimensional, inherently inhomogeneous character; ii) its peculiar, semimetal electronic structure, constituted of optically-active π -orbital networks, characterized by a unusual topology and vanishing density of states at the Fermi surface, combined with the presence of singular points in the density of states of empty and occupied bands. iii) It is optically active in the visible and near-UV range, allowing the photoinjection of high carrier densities in the π bands. Starting from these considerations we focus our experiments on measuring, by non-linear angle resolved photoemission, the effective mass and linewidth of the IPS in highly oriented pyrolytic graphite (HOPG) sample excited by 100 fs laser pulses. By tuning the photon energy across the π -bands saddle points, located at the M point of the Brillouin Zone (BZ), i.e. the van Hove singularity (vHs), we photo-inject a high carriers density ($2 \cdot 10^{20} \text{cm}^{-3}$) in the outermost graphene layers. Here, by showing the strong correlation among the IPS electron effective mass, the IPS total fullwidth and the IPS emission intensity versus the photon energy we reveal the electron dynamics and the band renormalization effects governing the correlated interactions of the unoccupied states in graphite. On the basis of these observations we formulate a consistent self-energy model that connects by causality, via Kramers-Kronig transforms, the IPS effective mass with the IPS total fullwidth, predicting a transient bands energy renormalization mechanism consistent with the experimental observations.

[1] K.-F. Berggren and B. E. Sernelius, Phys. Rev. B **24**, 1971 (1981).

[2] D. S. Chemla and J. Shah, Nature (London) **411**, 549 (2001).

Ultrafast dynamics of THz excitations: from observation to coherent control

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Low-energy elementary excitations, such as plasmons, phonons, magnons, or excitons, play a pivotal role for the femtosecond dynamics of solids and their surfaces. Phase-locked single-cycle pulses in the terahertz (THz) spectral domain and field-sensitive detectors have been harnessed to trace these excitations on a sub-cycle time scale [1]. In addition, THz fields of more than 10 GV/m have become available [2] to study condensed matter under unprecedented bias conditions and set the quantum state of quasiparticles by coherent control. Examples are:

- (i) Resonant THz pulses promote 1s para excitons in the semiconductor Cu₂O into the 2p state, by internal Rabi cycles [3]. The results point out a promising route towards ultracold exciton gases and potential Bose-Einstein condensation.
- (ii) Intense THz transients coherently control collective magnon oscillations in antiferromagnetic NiO [4]. Being triggered by Zeeman interaction with the magnetic field of the THz pulse, this approach opens up a novel and most direct gateway to the ultrafast dynamics of electron spins in the electronic ground state.
- (iii) Finally, we explore a new limit of non-adiabatic quantum electrodynamics: Intersubband cavity polaritons in a semiconductor quantum well waveguide structure are photogenerated by 12-fs near-infrared pulses. Multi-THz transients trace the abrupt conversion of bare photons into cavity polaritons. Our structure represents the first sub-cycle switching device of ultrastrong light-matter coupling and paves the way towards non-adiabatic quantum optics [5].

[1] see e.g. R. Huber et al., *Nature* **414**, 286 (2001).

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[3] S. Leinß et al., *Phys. Rev. Lett.* **101**, 246401 (2008).

[4] T. Kampfrath et al., submitted.

[5] G. Günter et al., *Nature* **458**, 178 (2009).

Anisotropic electron dynamics and electron-phonon coupling in Fe-pnictides

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The recently discovered class of Fe-based high- T_c superconductors (SC) and their parent compounds represent an interesting correlated electron system facilitating the study of effects like intra- and interband scattering and electron-phonon coupling. A powerful method to investigate such effects is femtosecond (fs) time- and angle-resolved photoemission spectroscopy (trARPES).

Here, we present trARPES experiments of the parent compound EuFe_2As_2 and the doped $\text{BaFe}_{1.85}\text{Co}_{0.15}\text{As}_2$ high- T_c SC. We observe a strongly momentum dependent dynamics of states around the hole-pocket at the Γ -point of the Brillouin zone (BZ) where occupied states become depopulated by excited holes, whereas electrons are filling empty states within the hole-pocket. The timescales of electron and hole dynamics differ by almost an order of magnitude which cannot be explained solely by intraband scattering processes. Thus, additional interband scattering channels between the center and the boundary of the BZ have to be considered, which require momentum transfer and might lead to the excitation of zone boundary phonons.

In addition, a periodic modulation of the trARPES intensity near the Fermi level is observed. Analysis of these oscillations reveals three coherently excited phonon modes at frequencies of 5.6, 3.3 and 2.6 THz, that couple to the electronic system and modulate the chemical potential. Comparison to Raman data identifies the mode at 5.6 THz as the A_{1g} mode. The other two modes cannot be assigned to Raman active modes and might originate from zone boundary phonons.

The analysis of the excited electron distribution allows to estimate the average electron-phonon coupling parameter λ , which is an important property related to superconductivity. The small value of $\lambda < 0.5$ resulting from our experiments suggest a limited importance of e-ph coupling to superconductivity in Fe-pnictides.

Excitation energy dependence of photoelectron-phonon interaction on Cu surface excited by laser light

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In recent times, laser photoemission (LPE) spectroscopy has emerged as a powerful tool for probing surface electron dynamics. For example, two photon photoemission spectroscopy is well known as an ideal technique for monitoring the decay process of image states at the surface. We propose a novel application of this technique to measure phonon properties at a solid surface. (Here, “phonon” also refers to adsorbate vibrations.) In a previous study[1], we reported that an energy loss process due to vibrational excitation during LPE. To establish LPE spectroscopy as a novel vibrational spectroscopic tool, it is necessary to understand the mechanism of the inelastic interaction between the photoelectron and the phonon.

The present study investigates the excitation energy dependence of the cross section for this inelastic interaction on a Cu(110) surface. In the LPE spectra of the clean Cu(110) surface ($\Phi \sim 4.52$ eV), the inelastic component arising from the generated phonons appeared as the step at 15 meV [2]. The step height, which corresponds to the cross-section, rapidly decreased as the photon energy ($h\nu$) increased, for $4.5 < h\nu < 5.0$ eV. For $5.0 < h\nu < 6.0$ eV, the step height was low and the $h\nu$ dependence was weak. We also measured the $h\nu$ dependence of the O/Cu(110) surface, which has a higher work function ($\Phi \sim 5.0$ eV). A step appeared at 15 meV and the $h\nu$ dependence was weak in the energy range of $5.0 < h\nu < 6.0$ eV. These results suggest that the cross-section for this inelastic interaction was mainly determined not by the kinetic energy of photoelectron but the photon energy. Finally, we discuss a feasible scenario of the inelastic photoemission process.

This work was supported in part by the PRESTO program of the Japan Science and Technology Agency (JST).

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[2] R. Arafune *et al.*, PRB **80**, 073407 (2009).

Structure dynamics of epitaxial VO₂ film on c-Al₂O₃ by using an in-air time-resolved X-ray diffraction

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The lattice motion and displacement of atoms were characterized for the unit cell in vanadium dioxide (VO₂) grown on Al₂O₃ using static and time-resolved X-ray diffraction (XRD). The monoclinic–tetragonal phase transition of the VO₂ unit cell and the twist motion of vanadium atoms in the unit cell were observed. The time-resolved XRD measurements were performed in air with a table-top high-repetition femtosecond laser. Typical X-ray diffraction spectra are shown in Fig. 1. The results from the time-resolved XRD measurements suggested that the unit cell of low-temperature monoclinic VO₂ transformed into the high-temperature tetragonal phase very fast; however, the atoms in the unit cell fluctuated with the center of the tetragonal coordinate within about 100 ps. The propagating time scale of 100 ps corresponded well to the previously reported time scale of shear motion measured with femtosecond optical measurements or crystallography [1,2]. Thus, the time-resolved XRD measurements of the Bragg angle, intensity and width of the diffraction lines revealed the phase transition of VO₂ and the atomic motion in the unit cell simultaneously.

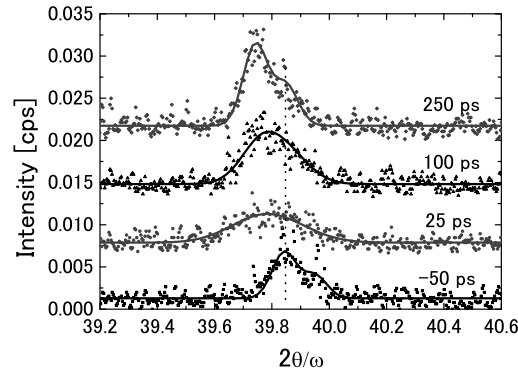


Figure 1. The typical spectra of the time-resolved XRD line from the (020) plane of VO₂. (at -50, 25, 100 and 250 ps, as marked)

[1] A. Cavalleri, Cs. Toth, C.W. Siders, J.A. Squier, F. Raksi, P. Forget, J.C. Kieffer, *Phys. Rev. Lett.* **87**, 237401 (2001).

[2] P. Baum, D.-S. Yang, A.H. Zewail, *Science* **318**, 788 (2007).

Ultrafast Localization of Photo-injected Charges by Alkyl Side Chains in Organic Semiconductors

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Organic electronics and photovoltaics suffer from low charge mobilities and an inability to disentangle morphological and molecular effects on charge transport. Specific chemical groups such as alkyl side chains are often added to enable spin casting and to improve over all power efficiency and morphologies, but their exact influence on mobility is poorly understood.

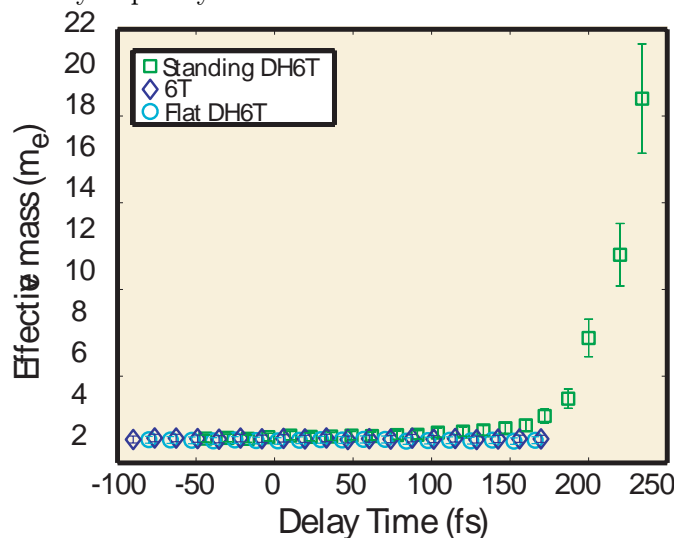


Figure 1. The effective mass of an electron is shown as a function of time in three systems. The standing layer of dihexyl-sexithiophene is found to affect a rapid increase in effective mass.

Here we use two photon photoemission spectroscopy to study the charge transport properties of the image potential state in two organic semiconductors, both with and without alkyl substituents (sexithiophene and dihexyl-sexithiophene). We show that the hydrocarbon side chains are responsible for localizing electrons within 220 fs. The localization is measured directly by following the dynamic increase in effective mass of a free electron (see Fig. 1). This implies that charge transport in alkyl containing organic semiconductors will be fundamentally limited by their side chains. We suggest that molecules which are solution processable without alkyl units may make more efficient transistors and photovoltaics.

Dynamics of Monolayer Water on a Single Crystalline TiO₂ Surface in Air by VSFG Spectroscopy

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CREST/Japan Science and Technology Agency*

Molecular dynamics on an oxide surface is a key process for various devices using oxide such as catalysts, solar cells, and electronic devices. We have been investigating the dynamics using picosecond vibrational sum-frequency generation (VSFG) spectroscopy suitable for in-situ observation. On a single crystalline TiO₂ surface, which plays important role in photo-catalytic reaction, we have succeeded in detecting monolayer water *in air* and revealing its photoinduced dynamics.

Fig. 1 shows the VSFG spectrum of a TiO₂(110) surface measured in air at room temperature. Judging from their wavenumbers, the bands at around 2800 cm⁻¹ and 3300 cm⁻¹ are assigned to CH stretching modes of hydrocarbons and OH stretching modes of bulk water. In contrast to these bands, the 1600-cm⁻¹ band is fairly unique. The intensity is hundreds times higher and the width is much narrower than the others. According to the study on water-adsorption on a TiO₂(110) surface by HREELS in vacuum [1], HOH bending mode of monolayer water is located at 1605 cm⁻¹. In addition, the band strongly depends on the rotation of the sample. Thus, we assigned the band to the monolayer water on the TiO₂(110) surface in air. These features of the band are probably caused by the resonance not only to the vibrational mode but also to the electrically excited state of surface defect site. This strong and sharp band enables us to study ultrafast dynamics of monolayer water on an oxide surface, so we carried out pump-probe measurements.

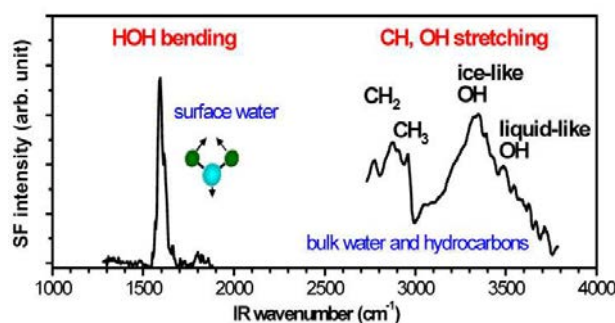


Figure 1. VSFG spectrum of TiO₂(110) in air.

[1] M.A. Henderson, Surf. Sci. **335**, 151 (1996).

Ab-initio description for laser-induced electron-phonon dynamics in dielectrics

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We have been developing a theoretical and computational approach describing electron-phonon dynamics in bulk material induced by intense and ultrashort laser pulses. Our approach relies upon the time-dependent density functional theory, a successful first-principles theory for electronic excitations. We solve the time-dependent Kohn-Sham equation in real-time and real-space. The electron dynamics can be visualized in femtosecond time scale, which helps much to get an intuitive understanding for the dynamics. The following figure shows a snapshot of electron dynamics in bulk Si induced by an ultrashort laser pulse.

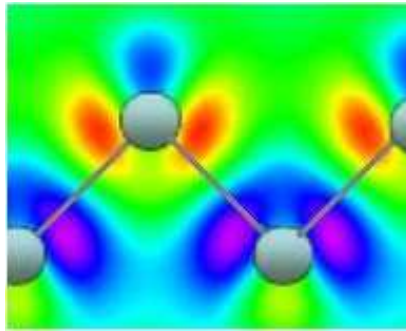


Figure 1. A snapshot of photoinduced electron dynamics in bulk Si.

We have applied our theory for several processes and have found that it provides a unified and quantitative description for the electron-phonon dynamics in femtosecond time scale. Under an intense laser pulse, it can describe optical breakdown of dielectrics induced by nonlinear ionization processes [1]. For a moderate intensity pulse, a coherent optical phonon generation can be described [2]. We have confirmed that our theory is capable of describing two distinct mechanisms, the impulsive Raman scattering and the displacive excitation, in a unified way.

[1] T. Otohe et al, Phys. Rev. B **77**, 165104 (2008); J. Phys. Cond. Matter **21**, 064224 (2009).

[2] Y. Shinohara et al, J. Phys. Cond. Matter, in press.

Coupling between 0D and 3D: hot electrons, coherent phonons, and solar energy conversion

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Semiconductor nanocrystals (NCs) are called artificial atoms because of their atom-like discrete electronic structure due to quantum confinement. They can also be assembled into artificial molecules or solids, thus, extending the toolbox for material design. We address the interaction of artificial atoms or molecules with bulk semiconductor surfaces. These interfaces are model systems for understanding the coupling between localized and delocalized electronic structures. In many perceived applications, such as nanoelectronics and solar energy conversion, interfacing semiconductor NCs to bulk materials is a key ingredient. Here, we apply the well-established theories of chemisorption and interfacial electron transfer as conceptual frameworks for understanding the coupling between NCs and bulk semiconductor surfaces. In the model system of PbSe NCs adsorbed on TiO₂(110), we show that strong interfacial electronic coupling allows hot electron transfer from photo-excited NCs. Moreover, we discovered that the transient electric field resulting from ultrafast charge separation across the PbSe-TiO₂ interface excites coherent vibrations of the TiO₂ surface atoms, whose motions can be followed in real time. These results indicate hot electron transfer from semiconductor NCs to a technologically relevant electron acceptor is possible. This effect is expected to be of general significance to other 0D/3D interfaces and may be utilized in the design of the ultimate solar cell.

Quantum Interference Effects in the Ultrafast Dynamics of a Low Dimensional Mott Insulator

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The competition between electron localization and de-localization in doped Mott insulators underpins the physics of strongly correlated electron systems. Photo-excitation, which re-distributes charge among different sites, can be used to control this cooperative many-body process on the ultrafast timescale. To date, time-resolved studies have mostly been performed in solids in which other degrees of freedom, such as lattice and spin-orbital excitations, come into play. However, the underlying dynamic physics of ‘bare’ electronic excitations has remained out of reach. To date, quantum coherent many-body dynamics have only been detected in the controlled environment of optical lattices, where the dynamics are slower and lattice excitations are absent. By using nearly-single-cycle, sub-10-fs pulses in the near-infrared, we have measured quantum-coherent excitations in the organic salt ET-F₂TCNQ, a prototypical one-dimensional Mott insulator with narrow bandwidth and weak electron-phonon coupling. After photo-doping, a new resonance appears on the low-energy side of Mott gap, which oscillates with a 25 THz frequency. Time-dependent simulations of the Hubbard Hamiltonian reproduce the oscillations at the experimentally observed frequency, showing that electronic delocalization occurs through quantum interference between bound and ionized holon-doublon pairs.

Snapshots of cooperative atomic motions in the optical becalming of charge density waves

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Femtosecond (fs) time-resolved techniques are becoming established as new tools for studying correlated electron phenomena in solids. By tracking energy relaxation pathways between various degrees of freedom (electron, spin, lattice etc.) on fs timescale, the interaction strengths amongst these subsystems can be investigated or manipulated. Charge-Density-Wave (CDW) systems, with their inherently multi-component order parameter, present a prime example of such cooperative phenomena in solids. The CDW state is characterized by the periodic modulation of the electron density and the periodic lattice distortion (PLD). In equilibrium the order parameter is given by a macroscopically occupied "frozen-in" phonon mode (static displacement of atoms away from that of the host lattice). Following perturbation with a fs optical pulse, however, the situation can be realized where on the fs timescale the electronic and lattice parts of the order parameter are uncoupled. While numerous fs time-resolved experiments have been performed on this class of materials, all were essentially probing the dynamics of the electronic subsystem and only inferred the dynamics of the PLD.

Here we report on the first study of the fs structural dynamics in a CDW system 1T-TaS₂, where dynamics of the PLD was studied directly utilizing fs electron diffraction [1]. The results reveal an ultrafast non-thermal suppression of the CDW order with a timescale of 170 fs. The high degree of correlated motions is further highlighted by the rapid electron-phonon energy transfer (350 fs) and the exceptionally fast recovery of the CDW (4 ps). These observed timescales and the degree of cooperativity are remarkable and illustrate the importance of obtaining atomic level perspectives of the dynamics directing the physics of strongly correlated systems.

[1] M. Eichberger et al., submitted

Dynamics of chemical reactions at surfaces under thermal excitation by laser irradiation and probed by time resolved SFG spectroscopy

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The dynamics of surface molecules with ps time resolution gives detailed information for elemental steps of chemical reactions at surfaces such as catalytic reactions. TiO_2 surfaces have been utilized to catalysts and photocatalysts. The time resolved SFG spectroscopy with NIR pump pulses reveals the ultra-short dynamics of structural change of formate species on TiO_x . The temperature jump at TiO_x layers prepared on a Pt(111) surface was initiated by the 1064 nm pulses (35 ps pulse width) and the temperature was relaxed to the initial temperature within ns time scale. The surface species (deuterated formates) changed the structure during the temperature change, which is hard to be observed by conventional static spectroscopy. The thermal broadening of vibrational peaks and population changes between different surface species showed the unique changes of transient SFG signals [1,2].

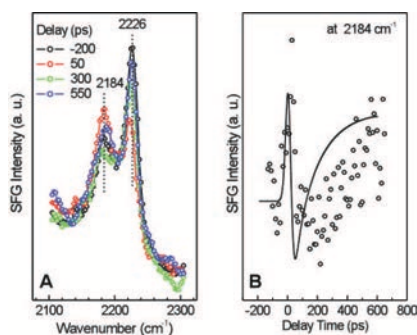


Figure 1. Transient SFG spectra (A) and signal change at 2184 cm^{-1} (B) of d-formates on $\text{TiO}_x/\text{Pt}(111)$ at 250 K under the irradiation with 1064 nm pulses [2].

Photocatalytic water splitting is one of the powerful tools to convert solar energy to solar hydrogen. We performed in-situ RAIRS experiments to understand the catalytic selectivity and the potential changes at photocatalyst surfaces [3,4]. This system is promised to be a valuable subject for the ultra-fast vibrational spectroscopy.

- [1] J. Chen, et al., J. Phys. Chem. **112**, 12477 (2008).
- [2] J. Chen, et al., J. Am. Chem. Soc. **131**, 4580 (2009).
- [3] M. Yoshida, et al., J. Phys. Chem. **113**, 10151 (2009).
- [4] M. Yoshida, et al., J. Am. Chem. Soc. **131**, 13218 (2009).

Measurement of local plasmon resonance of Au nanorods on femtosecond laser polarization using autocorrelation dark-field microscopy

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We established a new measurement technique of plasmon resonance using fringe-resolved e-field autocorrelation with dark-field microscopy. We prepared Au nanorods which have resonance wavelength around 800 nm on a SiO₂ substrate. Ultra-broadband femtosecond laser pulses are employed for excitation. The dark field microscope consists of a dark-field capacitor, object lens and a CCD. Two pulses are incident into the dark-field microscope with variable time delay. Fig. 1(a) shows the experimental result of autocorrelation measurement. The autocorrelation function and its amplitude varies by angular polarization of the femtosecond laser pulse. Fig. 1(b) shows the corresponding spectra to those in Fig. 1(b) obtained by Fourier transform. We confirmed the resonance enhancement of at 800 nm for the Au nano-rods. This new measurement scheme enables us to spatiotemporally control nanoplasmon in nanostructures using shaped femtosecond laser pulses and construct new surface reaction platforms.

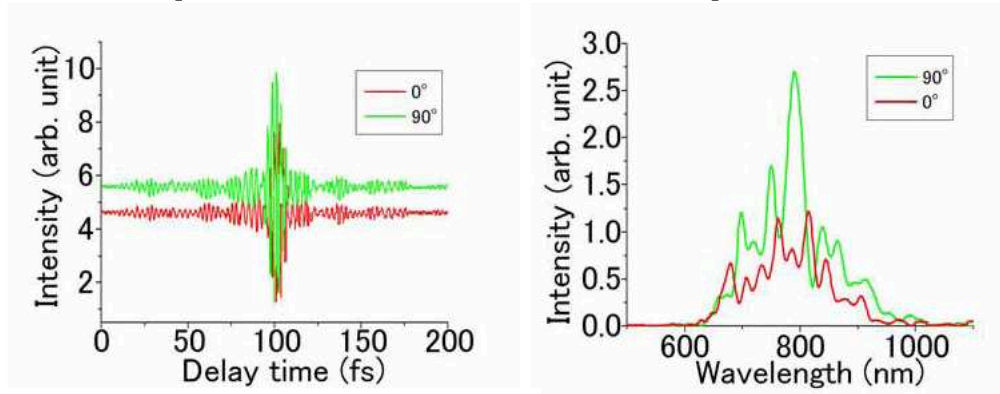


Figure 1. (a) Autocorrelation functions measured for orthogonal polarizations and (b) corresponding spectra.

Ultrafast transfer of angular momentum in ferromagnets

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Studying the ultrafast magnetization dynamics of ferromagnetic materials using femtosecond laser pulses is an interesting approach both from a fundamental point of view and for potential applications in spin-photonics. It is now well known that several steps occur after the excitation of the ferromagnet with an intense optical pulse. First the electrons and spins thermalize to a hot Fermi distribution (~ 100 fs) due to the electron-electron scattering with the appropriate many body correlations. Then they cool down by relaxing their excess energy to the lattice (~ 1 ps) by interacting with the phonons. Subsequently the energy is dissipated into the environment (a few picoseconds up to nanoseconds) which may consist of the unexcited part of the studied sample or to the substrate. The dynamics of these different subsystems can be fairly well described phenomenologically by a three temperatures model where three coupled baths are considered with temperature T_e for the electrons (charges), T_{spin} for the spins (magnetization) and T_l for the phonons (lattice dynamics). The modeling can be further improved by taking into account the non-thermal regime of the charges and spins populations which prevail until they reach their equilibrium temperatures T_e and T_{spin} . This approach is satisfying from the point of view of the energy balance.

A complementary aspect to the magnetization dynamics concerns the total angular momentum which also must be conserved. More generally, the transfer of angular momentum in a correlated spin system perturbed by a laser pulse is important to take into account as it is related to the mechanisms which induce the spin flips associated to the demagnetizing process. This is the topic that we have investigated recently and that will be reported in our presentation [1,2]. Two main aspects will be raised. First the coherent interaction between the laser and the ferromagnetic system. We will describe the corresponding dynamics of the charges and spins of Ni and CoPt films excited by 50 fs pulses. Second the dynamics of the orbital and spin moments, observed by time resolved femtosecond Xrays magnetic circular dichroism, will be described in CoPd films.

[1] J.-Y. Bigot, M. Vomir, E. Beaurepaire, *Nature Physics* **5**, 515 (2009).

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Abstracts of Posters

Influence of helium adlayers on image-potential states on Cu(111)/Ru(001)

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Nowadays image-potential states are mostly considered as a characteristic feature of metal surfaces, but in fact they have been first observed in the mid-seventies at the surface of liquid He. He films grown on metal surfaces have attracted attention due to the possibility to use image-potential states for the preparation of a dense two-dimensional electron gas. Due to the difficulties of combining cryogenic, surface science and laser technologies, however, detailed studies of these states with powerful spectroscopic techniques, such as time-resolved 2PPE have not been reported.

Here we present a time-resolved study of the decoupling of image-potential states on a Cu(111) surface by thin He adlayers using time-resolved two-photon photoemission (2PPE). The main challenge of the experiment consisted in the preparation of well defined He adlayers in combination with the laser excitation which leads to efficient laser-induced desorption. By using a specially designed lHe cryostat He adlayers up to a thickness of 2 ML could be prepared on a Cu(111) film which has been grown epitaxially on a Ru(001) substrate. By using different background pressures the thickness of the He adlayers has been varied in the range of 0-2 ML. The decoupling of the image-potential states from the metal surface results in a reduction of the binding energy of up to 310 meV for the $n = 1$ state and only of about 15 meV for the $n = 2$ state. This goes along with an increase of the lifetime from 30 fs up to 350 fs for the $n = 1$ state and from 105 fs to 360 fs for the $n = 2$ state. The decoupling, thus, is much stronger as compared to rare-gas adlayers of Xe, Kr and Ar on metals surfaces that have been studied systematically before. It is caused by the large negative electron affinity of bulk He of $EA = -1.3$ eV.

Unoccupied electronic states at atomic gold chains on vicinal Si(111) surfaces

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Atomic wires of noble metals such as gold serve as a model system for the investigation of one-dimensional electron systems. Recent experiments on Si(557)-Au have provided initial information on the unoccupied part of the electronic band structure [1,2]. We extended our research to Si(111)-(5×2)-Au and Si(553)-Au and investigated the electron dynamics of these systems.

Time-resolved two-photon photoemission experiments were carried out using femto-second laser pulses. Infrared (IR, $E_{IR} = 1.55$ eV) radiation was generated by a Ti:sapphire oscillator and subsequently frequency-tripled (UV, $E_{UV} = 4.65$ eV). It proved possible to rotate the polarizations of the IR and UV beams to align the electric field vector either perpendicular to (s-pol.) or parallel (p-pol.) to the plane of incidence. In normal emission the beams were incident on the sample at a glancing angle of 10° . Using optical dipole-selection rules, at normal emission, we evaluated the parity of the electronic states, which is either even or odd with respect to the $(1\bar{1}0)$ mirror plane orthogonal to the gold chains.

For all three surfaces we find an even, short-lived, intermediate state at about $E = E_{Vac} - 0.6$ eV below the vacuum level. We assign this state to the $n = 1$ image-potential resonance. Its short lifetime $\tau < 20$ fs can be understood in light of the absence of a band gap in the band structure of silicon for the corresponding energies.

Within the bulk band gap of silicon we observe a long-lived feature ($\tau > 600$ fs), which is pumped by the IR pulse (s-pol. or p-pol.) and probed by the p-polarized UV pulse.

We present here an analysis of our time- and energy-resolved measurements and discuss the similarities and differences between the surfaces investigated.

[1] J. A. Lipton-Duffin, J. M. MacLeod and A. B. McLean, Phys. Rev. B **73**, 245418 (2006).

[2] T. K. Rügheimer, Th. Fauster and F. J. Himpsel, Phys. Rev. B **75**, 121401(R) (2007).

Performance of a time-of-flight electron spectrometer for angle-and time-resolved 2PPE

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In recent years hemispherical electron analyzers in combination with two-dimensional detectors gained popularity in two-photon photoemission (2PPE) experiments for their ability to map the kinetic energy as well as the parallel momentum of photoelectrons in one direction within the surface plane. Many applications, however, would benefit from the time-resolved mapping of the full two-dimensional surface bandstructure. This includes the study of quasi-elastic electron scattering within electronic surface bands using time-resolved 2PPE as well as its extension for the generation of coherently excited electrical currents at surfaces.

For this purpose we have developed a position-sensitive time-of-flight spectrometer which has the capability to measure the kinetic energy as well as the full parallel momentum $\vec{k}_{||}$ of low-energy electrons photoemitted from a surface with a maximum rate of few MHz. It consists of a field-free drift tube and a delay-line detector for a position-sensitive detection in two-dimensions. The drift length can be varied in order to enhance either the acceptance angle or the energy- and momentum-resolution. With a time resolution of better than 200 ps and a spatial resolution of about 0.2 mm at a usable diameter of 37 mm, the energy resolution for low kinetic energy electrons as typical for 2PPE experiments is better than 10 meV even at the maximum acceptance angle of $\pm 25^\circ$. Furthermore, our data exhibit a dynamic range of more than three orders of magnitude.

In order to demonstrate the capabilities of this instrument we have investigated the well-characterized image-potential states of Cu(100). Optical excitation results in a populations of the parabolic bands which is initially isotropic in all directions in the surface plane. The observation of ring-shaped momentum maps for fixed kinetic energies with an isotropic intensity distributions serves as a test of the excellent imaging capabilities of the detector. Beside an isotropic depopulation due to inelastic scattering out of the excited band we observe a redistribution of the electrons within the $n = 1$ band for increasing pump-probe delay. The latter can be attributed to quasi-elastic scattering within the $n = 1$ band due to steps which break the almost cylindrical symmetry of the image-potential band.

Terahertz-laser Control of Large Amplitude Vibrational Motion in the Sub-one-cycle Pulse Limit

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Laser control is a well established research area in which many remarkable theoretical and experimental results have been achieved. Notwithstanding this success, the laser control of proton transfer reactions have yet to be reported experimentally. The complexity of this apparently simple isomerization reaction and its obstinacy to avoid control will be outlined: from the intrinsic time scale of the reaction that dictates the characteristics of the laser pulses, through the quantum nature of H-transfer, to the intrinsic multidimensionality of the reaction.

The laser control of H-transfer reactions calls for ultrashort pulses in the THz range. In the THz ultrashort pulse limit the time derivative of the pulse envelope has a significant effect on the population dynamics. Here we present a theoretical description of this phenomenon. Furthermore we show how the carrier envelope phase-sensitive dynamics arises in multi-level systems and extend optimal control theory in the THz ultrashort pulse domain.

Phonons in CdSe Quantum Dots: Influence of Surface Oxidation

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Fundamental processes in photoexcited semiconductor quantum dots (QDs) have received much attention during the last years. The optical excitation of QDs can be accompanied by the emission of phonons (exciton-phonon coupling) which are observable in resonance Raman studies as well as by fs-spectroscopy. While resonance Raman experiments show the longitudinal optical (LO) phonon, in time resolved absorption spectroscopy the LO as well as the longitudinal acoustic (LA) phonon can be seen [2-3]. Here we present time resolved pump/probe experiments on CdSe QDs. The particles were excited by ultrashort pump pulses with a central wavelength at the maximum of the lowest QD transition. The non-oscillatory electronic contribution was subtracted from the overall transient signal to obtain the purely phonon associated absorbance changes.

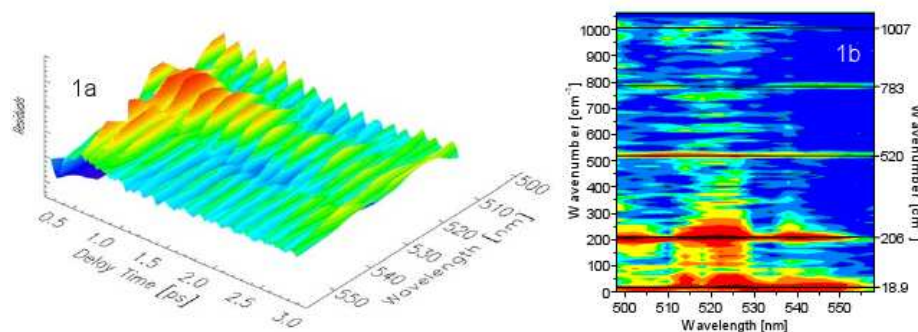


Figure 1. Transient absorbance data of CdSe QDs (a), 2D Fourier transformed spectrum (b)

The complete transient data is depicted in Fig 1a. The effect of frequency modulation with a phase shift of π at the minimum of the transient signal at 525 nm as well as a low frequency oscillation of the LA phonon and a high frequency oscillation of the LO phonon can be seen at first sight. The LA and LO phonon can be assigned to the peaks at 18,9 and 206 cm^{-1} in the 2D Fourier transformed spectrum of the residual oscillations shown in Fig. 1b. Surprisingly, the 2D Fourier transformed spectrum also has relevant signals at 520, 783 and 1007 cm^{-1} which cannot be assigned to any phonon in CdSe. In contrast, these results may be explained by a highly oxidized surface of the CdSe particles. Contrary to common quantum dot synthesis, particles used in our experiments were grown under oxygenic conditions, eventually leading to a CdO enriched surface. IR [4] and Raman [5] studies on polycrystalline CdO could determine the position of the LO phonon at 523 cm^{-1} and the second order Raman peaks, 780 and 970

cm⁻¹ for the 2LO(X) and the 2LO(L) phonon, respectively. This is in excellent agreement to the newly discovered modes for our QD sample.

- [1] R. Loudon, Proc. R. Soc. London Ser. A **275**, 218 (1963).
- [2] A. P. Alivisatos et al. J. Chem. Phys **90**, 3463 (1989).
- [3] D. M. Sagar et al. Phys. Rev. B **77**, 235321 (2008).
- [4] H. Finkenrath et al. Solid State Commun. **5**, 875 (1967).
- [5] G. Schaack et al. Solid State Commun. **19**, 315 (1976).

Electron dynamics on Si(001)

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Combining ultrafast laser excitation with energy- and angle-resolved two-photon photoemission (2PPE) the electronic properties of bulk silicon and the Si(001)-surface have been investigated. A custom-built laser- and UHV-system equipped with a 2D-electron detector gives new insight into momentum- and energy-relaxation on the femtosecond timescale.

For densities of 10^{19} cm^{-3} , carriers photo-excited to the conduction band thermalize via electron-electron scattering within 300 fs. Energy relaxation occurs on a slower 1 ps timescale via emission of optical phonons. Close to the conduction-band bottom optical phonons can not be emitted. This phonon bottleneck leads to a slow-down of electron-lattice equilibration and a raised electronic temperature long after laser excitation. Depending on the carrier density surface recombination is dominated by electron-electron scattering or phonon emission. This results in distinct momentum and density-dependent population dynamics in the unoccupied dangling bond band D_{down} .

What is more the new laser system allows us to tune the photon energy slightly above the silicon ionization potential and we thereby resolve the Rydberg-like series of image-potential states on Si(001) up to $n = 4$. Their binding energies, dispersion, and momentum-dependent decay and dephasing rates will be discussed. On silicon the image-potential resonances and the occupied dangling bond state D_{up} are degenerated with bulk states. Tuning the photon energy of the pump-laser across the resonance between D_{up} and $n = 1, 2$ we observe clear interference phenomena both in the initial and final state 2PPE signals. Simulating this double state – double continuum Fano resonances via optical Bloch equations allows us to deduce the coupling strengths between surface and bulk states. For constant coupling we obtain a rather short lifetime of about 10 fs of the photo-hole in the D_{up} dangling bond state of Si(001).

Angle- and time-resolved XUV photoemission from graphene/Ni(111) using High Harmonic radiation

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In surface science the structure of occupied and unoccupied electronic states is of great importance. Due to the large wave vectors towards the periphery of the Brillouin zone measurements at large $k_{||}$ require high photon energies in the VUV and XUV spectral range. Therefore synchrotron radiation sources have been advantageously employed to investigate the occupied electronic structure in photoemission experiments. To investigate the structure and especially the dynamics of unoccupied electronic states at surfaces two-photon photoemission is a widely used technique. Sufficient time resolution is achieved by High order Harmonic radiation of ultra-short Ti:sapphire laser pulses, being a table top source of XUV radiation with femtosecond pulse duration. We have developed a set-up that enables angle-resolved and time-resolved photoelectron spectroscopy. The Harmonics are generated by frequency conversion in rare gases and cover an energy range from 20 to 110 eV. ARUPS measurements at different photon energies can be performed by selecting single harmonic orders using a toroidal grating monochromator. For time-resolved measurements a Si/Mo multilayer mirror monochromator is employed which does not influence the XUV pulse duration.

The system of our interest is graphene because detailed knowledge about the unoccupied states is lacking. An interesting detail in the electronic structure of graphene is the linear band dispersion at the K point of the Brillouin zone. It leads to various new effects and the unique electronic properties of this material. Ultra-short High order Harmonics pulses provide the possibility to investigate the electron dynamics at the K point of graphene directly in the time domain.

Time- and angle-resolved photoelectron spectroscopy measurements on graphene epitaxial grown on a Ni(111) single crystal surface have been performed. Polarization dependent final state effects have been observed in the photoelectron cross section of graphene states. The band structure has been investigated. After excitation by pulses of the frequency-doubled Ti:sapphire radiation time-dependent populations in formerly unoccupied states have been observed.

Photoemission Enhancement Mechanism through the Local Surface Plasmon Resonance of Size-Selected Silver Nanoparticles

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Metal nanoparticles (NPs) have unique optical property of Local Surface Plasmon Resonance (LSPR), providing drastic enhancement of the local electric fields near the metal NPs. The metal NPs have been therefore applied widely to surface-enhanced spectroscopic measurements. LSPR of noble-metal NPs is a process of electronic excitation. Hence, photoemission from noble-metal NPs has also been found to enable enhancement by themselves [1]. However, the correlation of the photoemission processes with the LSPR or surface morphology of the deposited NPs has been still controversial to date. In this study, gas-phase synthesized Ag NPs were deposited onto a hydrogen-terminated Si(111) [H-Si(111)] surface through their size selection, and the photoemission process of the size- and coverage-defined Ag NPs was investigated by use of two-photon photoemission (2PPE) spectroscopy.

Size-selected Ag NPs were prepared by low pressure-differential mobility analyzer (LP-DMA) with a laser ablation source, and the Ag NPs (6.1 - 1.2 nm) thus produced were deposited onto an H-Si(111) substrate. In the 2PPE measurements, a laser at the photon energy ranging 2.90 - 3.23 eV (428 - 384 nm) was generated by frequency doubling of the output from a mode-locked Ti:sapphire laser.

At low coverage of Ag NPs around 0.005 monolayer equivalents (MLE), the photoemission from the H-Si(111) substrate was enhanced by 2×10^2 times. Above 0.12 MLE, in contrast, photoemission from the Ag NPs themselves was observed. From the experimental results of polarization and the photoemission angle dependence, the mechanism of the photoemission enhancement is attributed to near fields from LSPR of Ag NPs [2].

Transient spectroscopy of desorbing molecules : CO/Pt(111)

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We used time-resolved sum-frequency generation (SFG) spectroscopy with phase-sensitive detection for studying CO photo-desorption dynamics at Pt(111). After vibrational coherence of CO stretch was produced by an IR pulse, a CO-covered Pt(111) surface was irradiated by a pump pulse (400 nm, 150 fs duration) that was intense enough to desorb CO. While keeping CO coverage constant, we measured the real and imaginary parts of the nonlinear susceptibility, $\chi^{(2)}$. Time-domain plots of C-O stretching vibrational polarization were reconstructed from the amplitude and phase information of $\chi^{(2)}$. Time-resolved Fourier transform of the polarization plots reveals transient frequency shifts. Whereas the transient frequency shows redshifts within 300 fs after the pump pulse by anharmonic coupling with frustrated modes of CO, it shows blueshifts at the later time; this is indicative of a contribution of CO-Pt stretching vibrational excitation to the spectra. We discuss the vibrational dynamics of CO desorbing from the Pt(111) surface.

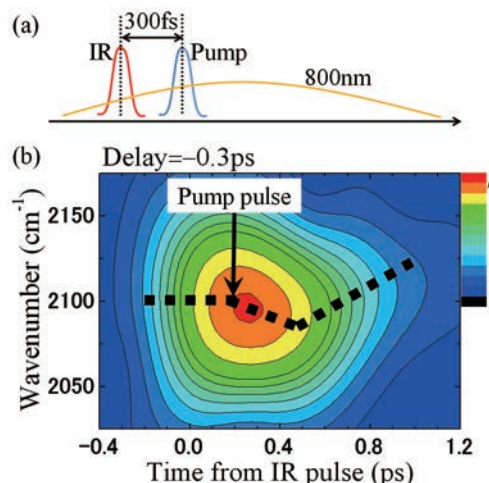


Figure 1. (a) Pulse timing of the measurement. (b) Time-frequency plots of C-O stretching vibrational polarization reconstructed from time-resolved SFG spectrum with heterodyne detection. The dotted line indicates the transient center frequency of the C-O stretching band.

Coherent Phonons of Ultrathin Bismuth Films on Si(111)-7×7

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Ultrathin bismuth (Bi) films grown on Si(111)-7×7 is known to undergo an intriguing structural transformation with increasing number of atomic layers [1]. The low-thickness phase, pseudocubic and {012}-oriented, is stable up to four atomic bilayers (BL), or 8 monolayers (ML), at room temperature. In terms of the cohesive energy, this pseudocubic phase is similar to the black phosphorus structure and shows a large spin-orbit splitting in the surface states. In the present study we performed transient reflectivity measurements [2] on the ultrathin Bi films of different thicknesses. We found that films whose thickness was between 6 and 8 ML exhibited a split in the A_{1g} phonon frequency, as shown in Fig. 1. Thicker films showed no frequency splitting but a slight blueshift, possibly due to the compressive stress at the interface. Films thinner than 6 ML gave no coherent phonon response in the reflectivity. Our observation is consistent with the lowering of the symmetry along the surface normal below 8 ML.

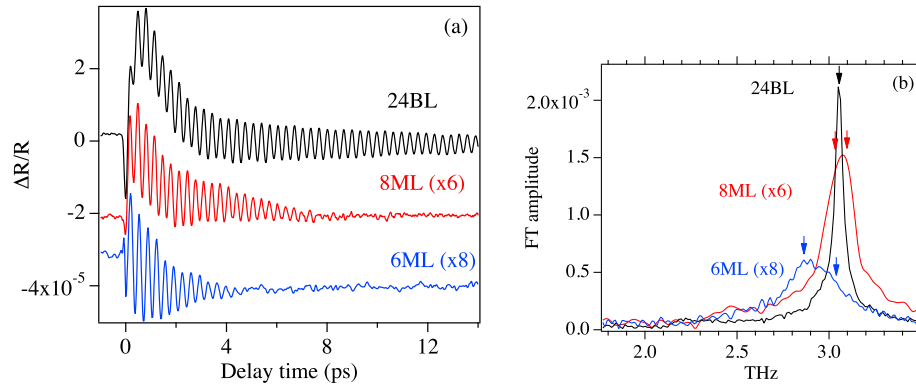


Figure 1. (a) Transient reflectivity of Bi ultrathin films of different thicknesses, and (b) its Fourier-transformed spectrum. Arrows in (b) shows the A_{1g} frequencies.

[1] T. Nagao *et al.*, Phys. Rev. Lett. **93**, 105501 (2004).

[2] Ishioka and Misochko, in *Progress in Ultrafast Intense Laser Science V*, ed. K. Yamanouchi, A. Giulietti and K. Ledingham, Chap. 2-3, Springer, Berlin (2010).

Ultrafast electron diffraction from thin (semi-)metal films

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We used ultrafast transmission electron diffraction to study the structural response of thin polycrystalline metal and epitaxially grown Bi(111) films [1] to femtosecond laser pulse excitation. In these optical pump - electron probe experiments, thin film samples were excited below the damage threshold and probed by diffracting a short (800 fs) electron bunch from the sample material.

In case of the polycrystalline metal samples (Ag, Au and Cu), the diffraction patterns consist of Debye-Scherrer like diffraction rings that prevent detailed studies of atomic motions due to the indistinguishability of crystallographic directions and low diffraction intensities. Upon optical (electronic) excitation, we observed that the lattice system is rapidly heated on the timescale of a few picoseconds, caused by electron-phonon interaction. The thermal motion of the atoms results in a decrease of the diffraction intensity that can be described by the Debye-Waller-factor. The electron-phonon energy transfer times extracted from our experimental results were found to be 4.7 ps in Au, 1.3 ps in Cu and 2.6 ps in Ag, in agreement with older all-optical measurements of the electronic temperature.

In contrast to the polycrystalline samples, we obtained discrete Bragg diffraction spots from epitaxially grown Bi(111). These samples offer the opportunity to study lattice dynamics in more detail and with higher measurement accuracy. We observed an anisotropic decrease of diffraction intensity that clearly indicates an atomic motion with preferential directions. We will discuss different effects that contribute to this anisotropy.

[1] T. Payer et al., Appl. Phys. Lett. **93**, 093102 (2008).

Numerical simulations of surface resonance spectra using 1d model potentials

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For many aspects of the energetics and dynamics of electronic states at metal surfaces and interfaces it is sufficient to use a one-dimensional description in form of a parametrized one-electron potential as the well-known Chulkov-potential which accounts for the electronic structure of the metal bulk within the two-band model of nearly free electrons as well as for the image-potential in front of the surface.

In this contribution we will discuss the application of the Numerov method in order to solve numerically the Schrdinger equation on the basis of different variants of the Chulkov-potential and show that this method provides an easy way to find discrete and continuum solutions which can describe for instance the Shockley surface state, image-potential states and resonances as well as electronic states at the interface of adsorbate covered metal surfaces. Compared to slab calculations this method allows the consideration of extended bulk and vacuum regions without the need of large computing power. Whereas it is straightforward to find bound solutions, i.e. states with energies below the vacuum energy E_{vac} and within the projected bulk band gap, the calculation of resonance spectra requires a more careful handling of the boundary conditions. Resonances, i.e. solution that are not bounded in one direction, can appear within a continuous range of energies either above E_{vac} or within a projected bulk band. The probability density of these solutions integrated within a region around the surface provide resonance spectra which can be interpreted as the effective density of intermediated states for a 2PPE process. The width of the resonances provides a measure of the elastic transfer rate of the excited electrons into the vacuum or into the bulk, respectively.

We will demonstrate the application of this method with of examples for different phenomena on clean and adsorbate covered metal surfaces, which we have studied within the last years. This includes the decoupling of image-potential states by rare-gas layers, the appearance of new quasi-bound electronic states at metal/rare-gas interfaces, the formation of a Shockley-type state at the interface between Ag(111) and PTCDA, the energies and dynamics of image-potential resonances with high quantum numbers n on Ag(111) as well as the transition between image-potential states and resonances for Ar/Cu(111).

A supersonic molecular beam doser for surface science research

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Commercial heterogeneous catalytic reactions proceed typically under high pressure condition, but not at low pressure ($<10^{-4}$ mbar). High pressure simply increases the absolute number of energetic molecules, which can surmount the reaction barrier, thereby increases absolute number of products sufficiently to be detected. Here we present a new doser for surface science research with supersonic molecular beam technique. The translational energy of molecules is easy to be tuned at a large energy scale by controlling the temperature of beam source and the carrier gas. The molecular beam source is triple differential pumped with a special designed gate valve to keep the ultrahigh vacuum condition of surface sample. The character of high translational energy will be exploited to dose some small molecules on surfaces. We expect to prepare CH_x and its isotopic radicals on Ni(111) surface by this supersonic molecular beam doser and study chemical reactions of CH_x radicals induced by either *indirectly* by hot electrons or phonons excited with ultrashort (i.e. femtosecond) visible or ultraviolet laser pulses or *directly* by resonant vibrational excitation of the CH_x radicals with ultrashort infrared lasers. We will show some test results of CO and D_2O on Ru(001).

[1] S. L. Tang, et. al., J. Chem. Phys. **84**, 6488 (1986).

[2] S. T. Ceyer, Acc. Chem. Res. **34**, 737 (2001).

Ultrafast quasi-particle dynamics in metallic quantum-wells

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We use time-resolved two-photon photoemission spectroscopy to investigate ultrafast quasi-particle dynamics in two-dimensional quantum-well systems [Ag, Pb, and Bi on Cu(111)]. This includes the study of electron-electron and electron-phonon scattering rates, the dependence of these processes on the dimensionality of the material system and the appearance of quantum-oscillations in the ultrafast dynamics. Our experimental data show that i) the electron-phonon scattering rate critically depends on the thickness of the quantum-well [1], and that ii) the electron-electron scattering rate is governed to a large extent by the dispersion of the electron quantum well bands [2]. Our findings suggest that the specific peculiarities of the electronic structure of two-dimensional quantum-wells can be used to control ultrafast dynamical properties in metals.

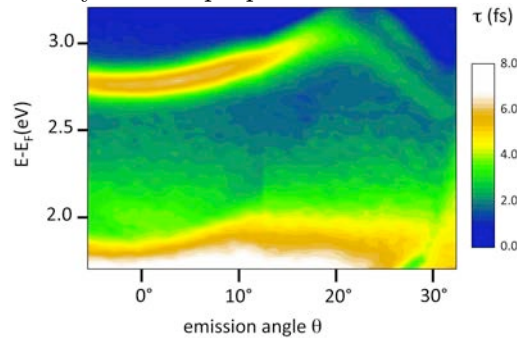


Figure 1. Inelastic lifetime map $\tau(E, k_{||})$ in a QW subband of 1 ML Pb/Cu(111) derived from an angle- and time-resolved 2PPE measurement. The lifetime τ is color-coded as a function of energy E and parallel momentum $k_{||}$. A color tone corresponds to 100 as.

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The lateral inhomogeneity of unoccupied states for PbPc and CuPc films

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Growth of organic thin film is typically inhomogeneous due to substrate-molecule and intermolecular interactions, which cause inhomogeneity of the unoccupied electronic structures at the interface. In order to reveal the inhomogeneous electronic structure, we have applied two-photon photoemission microspectroscopy (micro-2PPE) to ultrathin films (<1 ML) of lead phthalocyanine (PbPc) and copper phthalocyanine (CuPc) on graphite (HOPG). Our equipment is featured by its high-energy (30 meV) and high-lateral (0.4 μm) resolutions.

We observed PbPc derived states and Image Potential States (IPS) [1]. The surface images based on the HOMO peak became laterally uniform after annealing [2]. However, the images due to LUMO/LUMO+1 derived peaks (L/L1) are still largely inhomogeneous within $40 \times 40 \mu\text{m}^2$ area. The origin of the inhomogeneity is revealed by the coverage dependent micro-2PPE spectroscopy. The IPS peak is broadened to higher energy by 0.3 eV with deposition of PbPc (<0.8 ML). The broadening indicates that the electron of IPS is scattered by molecular clusters within the mean free path less than 10 nm. PbPc molecules are randomly distributed including nm-size clusters. The L/L1 is stabilized as the increase of the coverage. The stabilization energy for L/L1 is higher by 0.2 eV/ML than that for the HOMO peak. This indicates that L/L1 is sensitively affected by the neighboring molecules, that is, sensitive to the size of clusters. The inhomogeneity of the surface image due to the L/L1 comes from the size distribution of nm-clusters within the light spot.

In contrast to PbPc, the broadening of the IPS peak is not observed for sub-ML films of CuPc. These films are composed of two domains; islands of 1 ML coverage nucleate in the wetting layer of 0.3 ML coverage.

The high-energy resolution micro-2PPE spectroscopy is effective to resolve different growth modes of Pc molecules.

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Effects of hole scattering and Fano interference in two-photon photoemission from solid surfaces

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Two photon photoemission (2PPE) spectroscopy is one of the powerful experimental techniques for investigating electron dynamics characterizing chemical properties of material surfaces. In the conventional theories of 2PPE, it has been considered that a 2PPE spectrum is constructed by double-peak structures associated with a combination of one-photon excitation processes of electrons tentatively populating unoccupied states and direct two-photon excitation processes of electrons in occupied states. This theory is known to be successfully applicable to the cases of clean noble metal (111) surfaces, at which the well-defined surface states construct ideal systems described by simple extensions of three-level models [1,2].

However the recent experimental studies on semiconductor surfaces confirmed that the 2PPE spectra via surface resonances manifest remarkable features due to Fano interference and hence the higher energy portions of the peak structures vanish [3]. Interference effects on 2PPE processes are complicated theoretical subjects because quantum entanglement due to interference with hole scattering can be also involved [1]. In the present study, we reconstruct the microscopic theory of 2PPE from the above viewpoint of interference effects by means of the nonequilibrium Green function method [1]. We will discuss in the talk how the intrinsic physical values, *e.g.*, electron/hole lifetimes and Fano parameters, affect the 2PPE spectra.

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Disentanglement of the electronic and lattice parts of the order parameter in a Charge Density Wave system

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Recently it was shown that photoexcitation with an intense optical pulse can nonthermally drive the phase transition between the low temperature CDW and the high temperature metallic state [1,2]. Moreover, systematic studies of relaxation dynamics as a function of temperature and excitation density revealed that the melting of the modulation of carrier density can be achieved on the 100 fs timescale, and that during the process of melting and sub-ps recovery of the electronic modulation the lattice remains nearly frozen [1]. Here we report on the high resolution T-dependence studies of photoinduced reflectivity changes in the quasi 1D Charge Density Wave (CDW) compound $K_{0.3}MoO_3$, which enabled us to track the T-dependence of the coherently generated phonon modes with unprecedented sensitivity [3]. Numerous modes that appear below T_c , and show comparable softening as T_c is approached from below, are observed. By applying Time-dependent-Ginzburg-Landau (TDGL) analysis we were able to show, that these modes are a result of the linear coupling of the electronic part of the order parameter (EOP) to the $2k_F$ phonons. The EOP is found *not to* adiabatically follow the lattice modulation, explaining the longstanding dilemma why no phonon shows full softening near T_c in this class of materials. The fact that a fast electronic dynamics, whose decay time diverges near T_c , is observed in many CDW compounds [1,4,5] lead us to identify this mode as the true overdamped amplitude mode. The disentanglement of the electronic and lattice parts of the order parameter on the very short time-scale, demonstrated here for the CDW systems, could be however operational in a broader class of materials undergoing structural phase transitions.

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THEMIS: An angular resolving time-of-flight spectrometer

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The THEMIS time-of-flight spectrometer series complements the SPECS photoemission spectrometer line with an instrument ideally suited for pulsed photon sources. The design of this state-of-the-art instrument is based on the renowned PHOIBOS 100/150/225 spectrometer series. The extensively optimized electron lens with the proven 3D-DLD detector technology enables most flexible operation, with experiments ranging from extreme resolution spectroscopy to highest transmission snapshots.

Key performance features of THEMIS are parallel angle and energy detection with up to $\pm 13^\circ$ and 70 eV windows, spatial resolving modes with up to 10x magnification, accelerating and retarding operation with retard ratios from 0.05 - 1000, and a 3D-DLD detector with 3 MHz maximum count rate. In addition there is a high energy option for kinetic energies up to 15 keV.

The angular cone and the length of the energy spectrum that can be acquired in parallel depends on the lens mode. Since the THEMIS lens is optimized for minimum chromatic aberrations, a very long energy window can be acquired in parallel. By choosing the lens parameters, the performance can be varied from extremely high transmission (full spectrum per shot) up to extreme energy and angular resolution (selected small energy window per shot). The raw data from the detector (position and flight time for each electron) can be precisely transformed into emission angle and energy coordinates using a transformation matrix. Due to the high imaging quality of the THEMIS lens, the spectral and angular features of the experiment are immediately visible even in the non-transformed raw data.

Electronic States and Dynamics at Alkanethiol Self-assembled Monolayer on Au(111)

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Electronic properties of molecular nanoclusters have a great deal of attention among scientists for developing new electronic devices or catalysts. One of essential issues is that such functional clusters should be supported on a surface with keeping geometric/electronic configuration of the clusters in a gas phase. Moreover, the ordering of the clusters on the surface is also important to control and to design the electronic properties. From these points of view, formations of molecular clusters adsorbed or soft-landed on the surface have been studied so far. We have reported the self-assembled monolayer (SAM)/Au(111) surface works as a favorable environment to support molecular clusters without destruction in/on assembled monolayer consisting of its alkyl chain [1].

On the other hand, there are few examples of spectroscopic and dynamical information on the electronic states of SAM/Au(111) itself, which should provide fundamental knowledge to understand the mechanism of carrier transformation microscopically on the systems.

Here, we present electronic properties of SAM/Au(111) by using two-photon photoemission (2PPE) spectroscopy. Firstly, the unoccupied electronic states above Fermi level of dodecanethiolate- (C12-) SAM/Au(111) prepared within a liquid phase is clarified, comparing with it prepared by vapor deposition in a vacuum which have been already resolved by Zhu et al. [2]. Secondly, time- and angle- resolved 2PPE measurements are also discussed for the system to know band dispersion of the observed unoccupied levels and the decay of electronically excited states. These will be fundamental information to apply the SAM/Au(111) system as a cluster supporting layer.

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Theoretical investigation for generation of coherent phonon in bulk Si

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We have been developing a first-principles simulation method for quantum dynamics of electrons and ions induced by ultrashort laser pulses in infinite periodic systems. In our method, electron dynamics is described by the time-dependent density functional theory (TDDFT), while the ionic motion is described by classical mechanics with the force evaluated by the Ehrenfest theorem.

We have applied our method to investigate a mechanism of coherent phonon generation, taking bulk Si as an example. Our calculation confirmed that two distinct mechanisms are present in the TDDFT. The left figure shows the calculated force for ions for three frequencies across the direct band gap (2.4 eV). Below the gap, the force only exerts during the irradiation of the laser pulse, indicating impulsive stimulated Raman scattering (ISRS) mechanism. Above the gap, real electron-hole excitation induces the force after the laser pulse ended. This is the displacive excitation (DECP) mechanism. The right figure shows the calculated phase of the coherent phonon as a function of the laser frequency. The abrupt change from $\pi/2$ to 0 across the gap indicates the change of mechanism from ISRS to DECP. A change of phase from 0 to π around 5 eV region is also predicted.

These results confirm that the TDDFT is a powerful approach for the first-principles description of ultrashort electron-ion dynamics in bulk dielectrics.

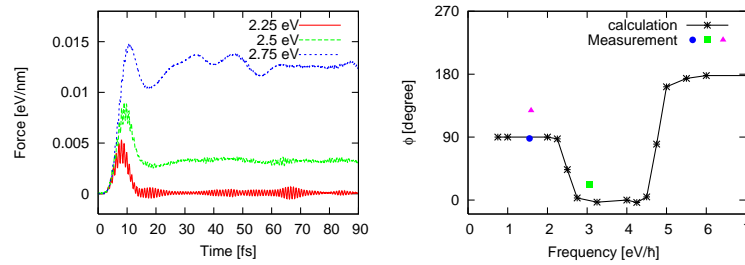


Figure 1. (Left) Calculated force as a function of time for laser pulses with three frequencies across the bandgap. (Right) Calculated phase of coherent phonon (line with crosses) and measurements (filled symbols).

Nonlinear optical characterization of hydrogenated two-dimensional honeycomb carbon (graphene), silicon (silicene) and germanium layers

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Since 2004 we have witnessed a spectacular growth of a research on graphene, a new class of 2D nanomaterial [1]. Very recently, a silicon analogue of graphene (silicene) has been produced experimentally [2] while the germanium-based monolayer film analogue has been predicted theoretically [3]. Although graphene (and its Si or Ge analogues) demonstrates a lot of exciting properties, its zero-gap band structure limits graphene application in 2D microelectronics. As was demonstrated very recently, however, hydrogenation of graphene produces a stable semiconducting material, graphane [4]. For possible device application of graphene/graphane single layer or bilayer of the material can be formed, e.g., graphene on top of SiC (0001) [5], or silicene on top of Ag(110) [3]. For such sandwiched system, experimental geometries may not provide conditions for complete hydrogenation of the 2D sheet(s). Therefore, it is important to have experimental techniques that allow characterizing the system non-destructively. We prove theoretically that linear and especially nonlinear optical techniques can provide detailed access to the properties of 2D monolayers of graphene, silicene and Ge-based nanostructures including the degree hydrogenation, symmetry, band gap and many others. To reach this goal we carry out combined simulation of structural, electronic and optical properties for hydrogenated graphene, silicene and germanium based 2D nano-sheets at different H coverages. Optical techniques considered are: second harmonic generation (SHG), both can be detected with terahertz radiation, recently proposed coherent control of injection current, optical spin injection and linear optical response.

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Time- and angle-resolved photoelectron spectroscopy at FLASH

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The Free-Electron Laser in Hamburg (FLASH) generates highly brilliant, ultrashort pulsed radiation with pulse durations down to 50 fs and photon energies up to 1000 eV. This enables a broad spectrum of novel and unique experiments. One such example is time-resolved photoelectron spectroscopy, for which various challenges concerning space-charge effects [1] and synchronization of pump and probe pulses [2] have recently been solved, as demonstrated by a proof-of-principle experiment on the layered charge-density-wave/Mott insulator 1T-TaS₂ [3].

However, to establish photoelectron spectroscopy as a completely viable technique at free-electron lasers, one has to develop a detection scheme with the highest possible efficiency. This is because the pulse repetition rates of free-electron lasers are notoriously low and the available beam time is extremely scarce.

Our proposed solution is a photoelectron spectroscopy experiment based on a novel angle-resolved time-of-flight spectrometer. This instrument will in particular feature a wide acceptance angle and a delayline detector with multihit capability. Compared to traditional detection schemes it is thus expected to enhance the detection efficiency by a factor of about 200 and, in addition, the temporal resolution by a factor of seven to about 100 fs via single-pulse detection, which avoids averaging over temporal fluctuations within pulse trains. Employing the wide photon energy range available at FLASH, the novel instrument will enable unique photoemission measurements. It will particularly become possible to perform time-resolved ARPES (angle-resolved photoelectron spectroscopy) and time-resolved XPS (X-ray photoelectron spectroscopy) in a single experiment so that, for example, atomic site-specific core-level dynamics can directly be correlated with the dynamics of Fermi surfaces. Here, we present the design of the proposed new end station at FLASH.

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Ultrafast dynamics of coherent phonons during the insulator-metal transition of VO₂

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VO₂ exhibits a first-order phase transition (PT) from an insulating, monoclinic to a metallic, rutile phase at 67 °C. Though studied in detail in the past decades by a large number of experimental and theoretical approaches, uncertainty remains about whether lattice deformation (Peirls) or strong electron correlation (Mott) dominate the transition. Femto- and picosecond time-resolved studies of the *photoinduced* transition are a promising tool to achieve better understanding of the underlying processes, as they may provide temporal separation of the lattice and charge carrier dynamics of the PT.

In the present work, we investigate the ultrafast response of a 200 nm thin VO₂ film to optical excitation (pump) in a wide range of fluences up to 30 mJ/cm². The resulting non-equilibrium ultrafast dynamics are probed by means of optical spectroscopy (800 nm, < 40 fs). At low excitation densities, the transient reflectivity response of hot carriers, which decays with a time constant of 400 fs, is superimposed with four coherent phonon modes of monoclinic VO₂ that are observed up to several ps. These modes, however, persist even when the pump fluence is increased above 10 mJ/cm², an excitation density that is commonly assumed to drive the transition to the rutile metallic phase. Only at even larger fluences (> 20 mJ/cm²), these coherent phonons, which are characteristic for the *monoclinic* VO₂ structure, are ultimately suppressed, indicating completion of the structural transition. Providing insight into the lattice dynamics during the insulator-metal transition of VO₂, these results will be related to previous time-resolved studies and discussed regarding potential disentanglement of Mott and Peirls contributions to the PT.

The Kiel-setup for time- and angle-resolved photoemission using high-harmonic light pulses

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Angle resolved photoelectron spectroscopy (ARPES) has proved to be a leading technique in identifying static key properties of complex electron systems. In a pump-probe scheme using femtosecond light pulses this technique can be extended to monitor ultrafast changes in the electronic valence structure in response to an intense optical excitation [1]. Here we present an experimental setup for time-resolved ARPES using femtosecond XUV probe pulses (see Fig. 1). We will focus on the relevant details and specifications of our system such as time and energy resolution, XUV photon flux and harmonic selection by a multilayer mirror monochromator. We show that the application of XUV pulses is particularly advantageous in recording valence structure transients at large momentum values in the vicinity or even beyond the border of the first Brillouin zone.

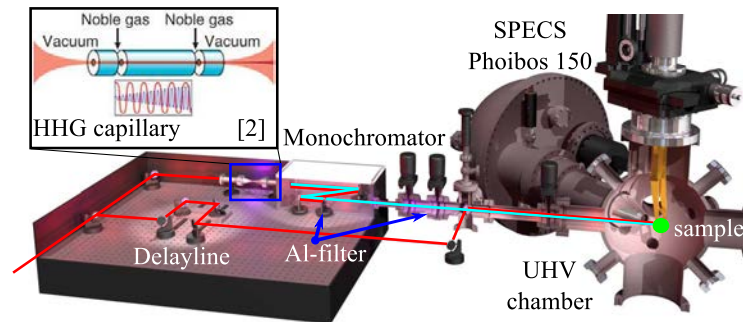


Figure 1. Scheme of the experimental setup.

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Correlation Between Geometric and Electronic Structures of Naphthalene on Cu(111)

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We focus on the naphthalene adsorption on Cu(111) as a prototype system[1,2] to clarify the correlation between geometric and electronic structures. We perform STM, LEED and 2PPE experiments under the same experimental conditions.

Figure 1 shows a summary of the coverage dependent 2PPE results. The break point of the linear change of the work function corresponds to the completion of the $(5\sqrt{3}\times 5\sqrt{3})R30^\circ$ superstructure. The first image potential states (IPS($n=1$)) are generally observed on the clean and adsorbed surfaces pinned at about -0.85eV with respect to the vacuum level (V_{vac}). However the IPS is stabilized to about -1.1 eV in this system. The stabilization was related to the resonance with the first electron affinity level (b_{2g})[2] known for the multilayer film on Ag(111)[3], but the stabilization effect is lifted for films thicker than 1 ML. This indicates that the IPS stabilization correlates with the formation of the unoccupied states specific to the $(5\sqrt{3}\times 5\sqrt{3})R30^\circ$ phase. The lifting is associated with the formation of the $(2\sqrt{3}\times 3)-1C_{10}H_8$ phase.

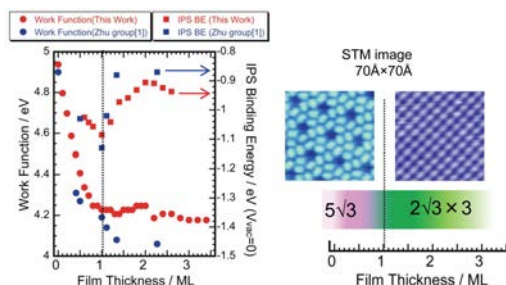


Figure 1. Coverage dependent change of the work function and IPS($n=1$) state(left) shown with the corresponding geometric structures (right).

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2PPE characterization of image-potential states of graphene/Ir(111)

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Double Rydberg series of image potential states (IPS) has been predicted for a free standing graphene [1]. However, experimental characterization and mapping of IPS's in graphene is a great challenge. For samples of limited quality and/or non-uniform thickness, only local probes such as scanning tunneling spectroscopy can be applied [2]. By epitaxial growth of graphene on Ir(111), weakly bonded large-area high-quality graphene of uniform thickness can be obtained [3]. This enabled us to use area-integrating method, i.e. two-photon photoemission (2PPE) to study first double series of IPS's (1+ and 1-), and reveal possible effects of the electronic interaction with the underlying substrate. The measured work functions of graphene and iridium are 4.9 eV and 5.65 eV, respectively. For full graphene coverage, we mapped 1+ and 1- dispersions and the binding energies of the two states which are compatible with the theoretical predictions for a free-standing graphene [1]. In order to get more insight into possible differences of a potential well at the graphene/Ir interface and on the vacuum side, we used the second harmonic of a tunable optical parametric amplifier, to extract coverage dependence of the work function and the binding energies of the image states from 0 to 1 ML of graphene.

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Separation of magnetic and non-magnetic contributions in the transient MOKE signal on Gd

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Laser induced ultrafast magnetization dynamics remains a controversial issue of magnetism after two decades of the first experiment by Vaterlaus et al. on Gd [1]. One potential reason is the presence of some effects which are not related to magnetization of the sample in linear magneto-optical signals widely used for analysis [2]. Gd with magnetic moments localized at the electronic 4f shell and indirect exchange coupling through the 5d6s valence electrons shows a transient magnetization behavior described by (i) an ultrafast and (ii) a slower quasi-equilibrium demagnetization [3] which are both slower than in 3d itinerant magnets [4]. We report here time-resolved studies of the rotation and ellipticity in the magneto-optical Kerr effect (MOKE) after fs laser pulse excitation as a function of equilibrium temperature. Experiments have been carried out in situ on Gd films of 20 nm thickness grown epitaxially on W(110) for temperatures between 40 and 200 K. We find that MOKE ellipticity and rotation show a similar but in detail different transient behavior which is attributed to the presence of the spectroscopic factor at ultrafast time scales. Considering the dependence of the spectroscopic factor on the electronic temperature we extracted transient demagnetization curves. We observe that the demagnetization time increases by order of 20 % if the temperature is increased from 40 to 200 K. Different scenarios will be discussed for an explanation of the effect including the role of surface contributions to the magneto-optical signal.

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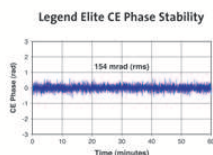
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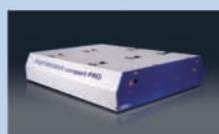
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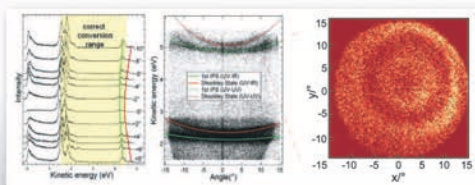
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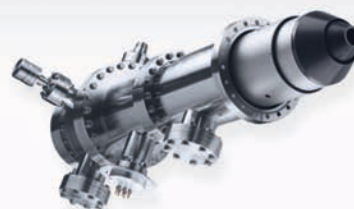
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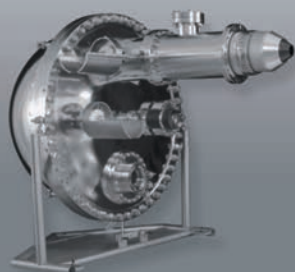
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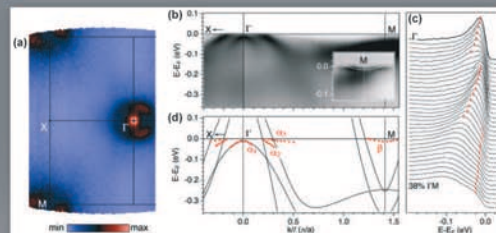
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a) Fermi surface map of $\text{FeSe}_{0.5}\text{Te}_{0.5}$ measured with 21.2eV photon energy and p-polarization at $T=1.2\text{K}$. b,c) Band dispersion along $X\Gamma M$. d) DFT band structure calculation for FeSe.

(A.Tamai et al., Phys.Rev.Lett. 104 (2010) 097002)

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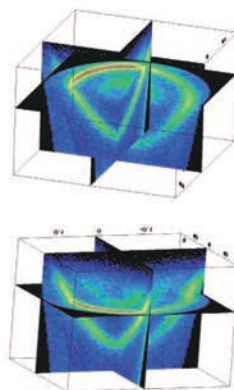
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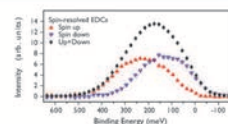


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Dynamics at Solid State Surfaces and Interfaces

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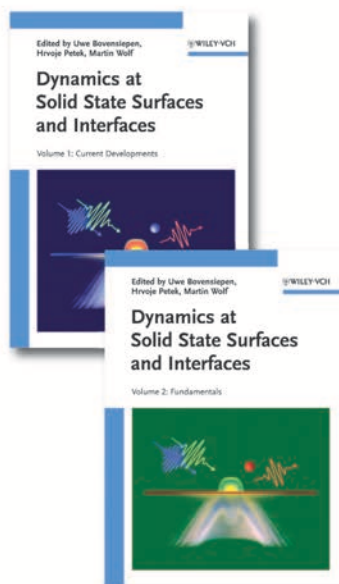
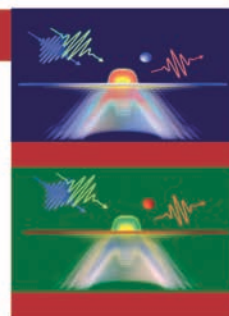
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