Ultrafast dynamics of collective excitations in solids

355. Wilhelm und Else Heraeus-Seminar

September 11 – 15, 2005

Vitte, Hiddensee island, Germany
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Dear Participant!

Recent years showed an enormous advance in our understanding of laser-induced ultrafast electron-, phonon- and spin-dynamics in solid matter. An increasingly important question of interest is the interaction of these excitations with each other, the importance of screening and the principles of resulting collective phenomena.

It is the aim of the WE-Heraeus Seminar *Ultrafast Dynamics of Collective Excitations in Solids* to intensify the discussion of collective phenomena on pico- and femtosecond timescales generated by the interaction of different quasiparticles like electron-hole pairs, phonons, polarons or spin waves. Thereby, the developed understanding of various single-particle excitations will be brought together to further the insight into underlying principles of collective phenomena among the lattice-, electron- and spin-subsystems.

The workshop will bring together leading experimental and theoretical experts and young researchers from a variety of fields focusing on ultrafast collective phenomena in solids. It is one purpose of the meeting to establish links between these different subfields. In order to stimulate the scientific exchange, *round table discussions* highlighting controversial topics and promising routes for future experiments will be conducted.

The program of the workshop is meant to give an overview of our current understanding of coherent and collective processes, interactions among different quasi-particles and challenges in the respective fields. Furthermore, it is expected to serve as a starting point for discussions and future interactions of the researchers. The five sessions will cover the following topics.

- **Coherent phonons**
  The present understanding of the optical excitation mechanism, coherent phonons in confined structures and latest developments in time-resolved X-ray diffraction studies will be presented and discussed.

- **Correlated electrons**
  Here, ultrafast studies in the far infrared, visible and X-ray regime will be discussed in combination with theoretical investigations. Metal-insulator-transitions and superconductors are in the focus since they represent outstanding examples for lattice mediated electronic correlations.

- **Quasi-particle interactions**
  The built up of charge screening and coupling of excited electrons to the lattice is responsible for the quasi-particle character of e.g. charge carriers or phonons in general. The respective dynamics will be addressed for semi- and superconductors.

- **Spin dynamics**
  Although spin-relaxation times in magnetic matter are usually considered to reside in the ns-regime, optically excited spin dynamics extend well into the ultrafast regime. The observed phenomena bear the potential to identify elementary interaction pathways (e.g. magnon-phonon-coupling).
Surface dynamics
Surface and interfaces open interesting pathways to determine elementary processes by systematic structural variation of the system. Beside surface sensitive non-linear optical approaches, photoelectron spectroscopy facilitates electron-momentum resolved studies and thereby addresses scattering processes directly.

Thanks to the contributions of all of you we were able to put together what we think is a very exciting program. We are particularly grateful to the Wilhelm und Else Heraeus-Stiftung and its managing director, Dr. Ernst Dreisigacker, who made this workshop possible. We also would like to acknowledge our home institutions, the Freie Universität Berlin and the Friedrich-Schiller-Universität Jena, for their support.

We hope that all of you will enjoy the four days on Hiddensee!

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## Program Overview:

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Detailed Program

Sunday September 11

16:00 – 18:00  
**Arrival & registration**

18:00 – 18:10  
**Welcome**

**Coherent Phonons**

**Chairman: H. Zacharias**

18:10 – 18:20  
**Introduction**  
H. Zacharias (Universität Münster, Germany)

18:20 – 19:00  
**Invited Talk**  
Mechanisms for ultrafast generation of coherent phonons and spin excitations  
R. Merlin (University of Michigan, Ann Arbor, USA)

19:00 – 19:40  
**Invited Talk**  
Coherent phonons in semiconductors and semiconductor heterostructures  
T. Dekorsy (Universität Konstanz, Germany)

20:00  
**Dinner**
Monday September 12

07:30 – 09:00  Breakfast

09:00 – 09:20  Contributed Talk
Ultrafast dynamics of coherent THz phonons in transition metals
M. Hase (National Institute for Materials Science, Tsukuba, Japan)

09:20 – 10:00  Invited Talk
Constrained density functional theory calculations of photo-excited phonons
Stephen Fahy (University College Cork, Ireland)

10:00 – 10:20  Contributed Talk
Coherent lattice dynamics studied by femtosecond X-ray diffraction
Y. Hironaka (Tokyo Institute of Technology, Japan)

10:20 – 10:40  Coffee Break

10:40 – 11:00  Contributed Talk
The collapse and revival and the Fano interference with a complex asymmetry parameter for coherent phonons in semimetals
O.V. Misochko (Russian Academy of Sciences, Chernogolovka, Russia)

11:00 – 11:20  Contributed Talk
Coherent phonons in a semiconductor superlattice studied by femtosecond x-ray diffraction
M. Bargheer (Max-Born-Institute, Berlin, Germany)

11:20 – 12:00  Invited Talk
Femtosecond X-ray measurement of coherent lattice vibrations
D. von der Linde (Universität Duisburg-Essen, Germany)

12:00 – 14:00  Lunch Break

14:00 – 15:20  Roundtable discussion
Possibilities of future short-pulse accelerator based XUV and X-ray sources
Discussion leader: A. Cavalleri (University of Oxford, UK)

15:20 – 15:40  Coffee Break
Correlated electrons

Chairman: J. Demsar

15:40 – 15:50  
Introduction  
J. Demsar (J. Stefan Institute, Ljubljana, Slovenia)

15:50 – 16:30  
Invited Talk  
Probing quasiparticle dynamics in the magnetoresistive pyrochlore Tl₂Mn₂O₇ using mid-infrared spectroscopy  
R. D. Averitt (Los Alamos National Laboratory, USA)

16:30 – 17:10  
Invited Talk  
Femtosecond X-ray diffraction and absorption in solids  
A. Cavalleri (University of Oxford, UK)

17:10 – 17:30  
Contributed Talk  
Correlated many electron dynamics with the multi-configuration time-dependent Hartree-Fock (MCTDHF) method  
Mathias Nest (Universität Potsdam, Germany)

17:30 – 17:50  
Coffee Break

17:50 – 18:10  
Contributed Talk  
Importance of singlet to triplet processes versus traps in pentacene and tetracene single crystals  
V. K. Thorsmølle (École Polytechn. Féd. de Lausanne, Switzerland.)

18:10 – 18:50  
Invited Talk  
Ultrafast dynamics of photoinduced phase transitions in 1D correlated electron systems  
H. Okamoto (University of Tokyo, Japan)

18:50 – 19:30  
Invited Talk  
Relaxation of hot quasi-particles in a d-wave superconductor  
P. J. Hirschfeld (University of Florida, Gainesville, USA)

19:30 – 19:50  
Poster Introduction

20:00 – 21:00  
Dinner

21:00  
Poster Session I
**Tuesday, September 13**

07:30 – 09:00  *Breakfast*

**Quasi-particle interactions**

**Chairman: P. J. Hirschfeld**

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<td>P. J. Hirschfeld (University of Florida, Gainesville, USA)</td>
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<td>09:10 – 09:50</td>
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<td>Ultrafast quantum kinetics of elementary interactions in semiconductors</td>
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<td>P. Leitenstorfer (Universität Konstanz, Germany)</td>
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<td>09:50 – 10:10</td>
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<td>Photoexcited electron dynamics in Kondo insulators and heavy fermions</td>
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<td>J. Demsar (J. Stefan Institute, Ljubljana, Slovenia)</td>
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<td>10:10 – 10:50</td>
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<td>Ultrafast measurements of quasiparticle dynamics in cuprates and spin diffusion in quantum wells</td>
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<td>N. Gedik (California Institute of Technology, Pasadena, USA)</td>
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<td>10:50 – 11:10</td>
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<td>11:10 – 11:50</td>
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<td>Transient insulating, conducting and superconducting phases probed with ultrashort THz pulses</td>
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<td>R. A. Kaindl (University of California, Berkeley, USA)</td>
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<td>11:50 – 12:10</td>
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<td>Broadband time-domain THz spectroscopy of semi-conducting organic crystals</td>
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<td>M. Koeberg (Leiden University, The Netherlands)</td>
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<td>12:10 – 12:50</td>
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<td>The coherent coupled electron-lattice response of Si to an optical impulse</td>
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<td>H. Petek (University of Pittsburgh, USA)</td>
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<td>12:50 – 13:10</td>
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<td>Spectrally resolved coherent spin dynamics in 2DEGs</td>
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<td>A. Puzlys (University of Groningen, The Netherlands)</td>
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<td>13:10 – 14:40</td>
<td><em>Lunch Break</em></td>
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14:40 – 16:00  
**Roundtable discussion**

**Atomic spatial resolution in ultrafast science**
Discussion leader: M. Wolf (Freie Universität Berlin, Germany) & M. Bargheer (Max-Born-Inst., Berlin, Germany)

16:00 – 16:20  
**Coffee Break**

**Spin dynamics**

**Chairman: M. Dürr**

16:20 – 16:30  
**Introduction**
M. Dürr (BESSY, Berlin, Germany)

16:30 – 17:10  
**Invited Talk**

**Ultrafast spin dynamics in ferro- and antiferromagnets**
T. Rasing (University of Nijmegen, The Netherlands)

17:10 – 17:30  
**Contributed Talk**

**Ultrafast spin dynamics in ferromagnets probed by X-ray spectroscopy**
C. Stamm (BESSY, Berlin, Germany)

17:30 – 18:10  
**Invited Talk**

**Magnetization dynamics in ferromagnetic disks and squares**
C. Back (Universität Regensburg, Germany)

18:10 – 18:30  
**Coffee Break**

18:30 – 18:50  
**Contributed Talk**

**Second harmonic generation in NiO – the necessity to go beyond electric dipole approximation**
G. Lefkidis (Universität Kaiserslautern, Germany)

18:50 – 19:30  
**Invited Talk**

**Spin dynamics in metallic ferromagnets at femto- and picosecond time scales**
E. Beaurepaire (I.P.C.M.S., Strasbourg, France)

19:40 – 19:50  
**Poster Introduction**

20:00 – 21:00  
**Dinner**

21:00  
**Poster Session II**
**Wednesday, September 14**

07:30 – 09:00  *Breakfast*

09:00 – 09:20  **The Wilhelm and Else Heraeus Foundation**  
E. Dreisigacker (W. & E. Heraeus Stiftung)

09:20 – 09:40  **Contributed Talk**  
The short and dynamic life of high wave vector spin waves  
M. Etzkorn (Max-Planck-Inst. f. Mikrostrukturphysik, Halle, Germany)

09:40 – 10:20  **Invited Talk**  
The role of charge/spin correlation and electron-phonon interaction in the singles particle excitations in bulk metals and at surfaces  
E.V. Chulkov (Donostia International Physics Center, Spain)

**Surface dynamics**

Chairman: D. Menzel

10:20 – 10:30  **Introduction**  
D. Menzel (Technische Universität München, Germany)

10:30 – 11:10  **Invited Talk**  
Ultrafast electron, spin and lattice dynamics on lanthanide surfaces: Optical excitation of a coherent phonon-magnon mode at Gd(0001)  
M. Wolf (Freie Universität Berlin, Germany)

11:10 – 11:30  *Coffee Break*

11:30 – 11:50  **Contributed Talk**  
Collective and single-particle dynamics in time-resolved two- photon photoemission  
W. Pfeiffer (Universität Würzburg, Germany)

11:50 – 12:30  **Invited Talk**  
Up and down - electron dynamics at Si(100)  
M. Weinelt (Max-Born-Institut, Berlin, Germany)

12:30 – 12:50  **Contributed Talk**  
Direct observation of femto- and attosecond electron dynamics in adsorbates  
A. Föhlisch (Universität Hamburg, Germany)

12:50 – 14:00  *Lunch Break*
14:00 – 14:40 Invited Talk
Excitation and ultrafast dephasing of coherent adsorbate-substrate vibration on metal surfaces
Y. Matsumoto (Institute for Molecular Science, Okazaki, Japan)

14:40 – 15:20 Invited Talk
Time-resolved coherent photoelectron spectroscopy of image-potential states
Ulrich Höfer (Universität Marburg, Germany)

15:20 – 15:40 Concluding remarks

15:40 Conference outing to Inselblick

20:00 Barbecue

Thursday, September 15

07:30 – 09:00 Breakfast

09:00 Departure
Abstracts

Coherent phonons: 15 – 28
Correlated electrons: 29 – 39
Quasi-particle dynamics: 41 – 53
Spin dynamics: 55 – 63
Surface dynamics: 65 – 76
Poster I (Monday): 77 – 88
Poster II (Tuesday): 89 – 100
Coherent Phonons
Mechanisms for ultrafast generation of coherent phonons and spin excitations

R. Merlin

FOCUS Center and Department of Physics, University of Michigan, Ann Arbor, MI 48109-1120, USA
e-mail: merlin@umich.edu, http://www-personal.umich.edu/~merlin/

Recent work on the generation of coherent low-lying excitations by ultrafast laser pulses will be re-viewed, emphasizing the microscopic mechanisms of light-matter interaction. The topics covered include long-lived phonons in ZnO [1], squeezed magnons [2], spin- and charge-density fluctuations [3] and cyclotron resonance [4] in GaAs quantum wells. In addition, unpublished results on surface-avoiding phonons in GaAs-AlAs superlattices and magnons in ferromagnetic Ga1−xMnxAs will be discussed. It will also be shown that frequencies can be measured using pump-probe techniques with a precision comparable to that of Brillouin scattering [1]. It is now widely accepted that stimulated Raman scattering (SRS) is (often but not always) the mechanism responsible for the coherent coupling [5]. Results will be presented showing that SRS is described by two separate tensors, one of which accounts for the excitation-induced modulation of the susceptibility, and the other one for the dependence of the amplitude of the oscillation on the light intensity [6]. These tensors have the same real component, associated with impulsive coherent generation, but different imaginary parts. If the imaginary term dominates, that is, for strongly absorbing substances, the mechanism for two-band processes becomes displacive in nature, as in the DECP (displacive excitation of coherent phonons) model. It will be argued that DECP is not a separate mechanism, but a particular case of SRS. In the final part of the talk, an attempt will be made to identify emerging areas of research on coherent excitations and coherent control, relevant to condensed matter systems, that could benefit from ultrafast electron and x-ray diffraction studies.

The impulsive excitation of coherent lattice vibrations with ultrahot laser pulses in a wide variety of materials allows to gain insight into nonlinear light-electron-phonon interaction on a femtosecond time scale. I will discuss the generation of coherent optical and acoustic phonons in semiconductors and semiconductor heterostructures with respect to different driving forces and how these can be distinguished. Especially in semiconductor superlattice zone folded acoustic phonons can be selectively excited by multiple pulses via an impulsive stimulated Raman process. In the same system optical phonons are coherently excited via coupling to coherent electronic wavepackets (Bloch oscillations).

Ultrafast dynamics of coherent THz phonons in transition metals

M. Hase\(^1\), J. Demsar\(^2\), K. Ishioka\(^1\), and M. Kitajima\(^1\)

\(^1\)National Institute for Materials Science, Japan,
\(^2\)J. Stefan Institute, Jamova 39, Ljubljana, Slovenia.

e-mail: hase.muneaki@nims.go.jp, http://www.nims.go.jp/ldynamics/index.html

Under the irradiation of solids with femtosecond laser pulses, coherent oscillations of Raman active phonons can be excited through electronic excitation via real or virtual intermediate states, which are characterized by displacive and impulsive excitations, respectively. There have been extensive studies of generation of coherent phonons,[1,2] control of coherent phonons,[3] and of dephasing dynamics of coherent phonons,[4] in semimetals, semiconductors, and superconductors in the past decade or so. Investigation of the coherent phonons has recently opened a possibility of controlling phase transitions and electron-phonon scattering phenomena,[5] and thus it has been the focus of much attention because of its wide applications as well as its fundamental interest.

Very recently, the surface coherent optical phonon (2.9 THz) have been observed in Gd metal using second harmonic generation (SHG) technique,[6] followed by the observation of bulk coherent optical phonons in Gd using the transient reflectivity (TR) technique.[7] Coherent surface vibrations on Cs/Pt(111) system have also been detected using the SHG technique.[8] The SHG technique has mainly enabled studying surface coherent optical phonons on metallic surfaces. However, investigations of bulk coherent optical phonons in metals using conventional TR pump-probe technique are still few, mainly due to very short optical penetration depth in metals stemming from the absorption by free electrons.

Here, we report on the observation and dephasing dynamics of bulk coherent optical phonon in Zn and Cd using a pump-probe reflectivity technique. The samples used were single crystals of Zn and Cd with cut and polished (0001) surface. The pump-probe measurements were carried out in a temperature range between 7 and 295 K. The source of femtosecond optical pulses used was a mode-locked Ti:sapphire laser (30 fs, 87 MHz) with the pump and probe powers fixed to 60 and 5 mW, respectively (spot size was ~70 microns). We used a lock-in detection with the pump-beam chopped at 2 kHz. The penetration depth of the laser light of 800 nm was estimated to be 13 nm based on the absorption coefficient of 7.6 x 10^5 cm^-1. Therefore the contribution to the signal from the surface oxide layer is negligible.

Figure 1 shows $\Delta R/R$ signal observed in Zn and Cd at 7 K as a function of the time delay after the excitation pulse. Initial transient non-oscillatory response is due to the excitation and relaxation of nonequilibrium electrons. Since the interband electronic transition near the L-point occurs at around 800 nm, this transition dominates the generation of nonequilibrium electrons in Zn, while both the interband and intraband electronic transitions would contribute to the generation of nonequilibrium electrons in Cd. The second component in the response is the oscillatory signal due to the coherent excited phonon mode. The period of the coherent phonons are 430 fs (=2.32 THz) in Zn, and 760 fs (1.32 THz) in Cd, respectively, both of which correspond to the bulk E\(_{2g}\) optical phonon. It is to be noted that in addition to Zn and Cd, we tried to measure coherent phonons also in Mg (0001), however we observed only weak electronic response and no coherent phonons in the TR signal (not shown). The amplitude of the coherent optical phonon is significantly larger in Zn than that in Cd. This suggests that the interband electronic transition, which contributes to the imaginary part of the dielectric function Im(\(\epsilon\)) as shown in the inset of Fig. 1, governs the excitation of the coherent optical phonon. In fact, at the
laser energy of 1.55 eV, \( \text{Im}(\varepsilon) \) shows a dominant peak for Zn, while it shows a weak shoulder for Cd. For Mg, however, \( \text{Im}(\varepsilon) \) is almost zero, supporting the suggestion that this mechanism is indeed the main driving force.

A most striking difference in the coherent phonon characters in metals from those in semimetals is a dramatic temperature dependence of the coherent phonon amplitude; the amplitude decreases with the temperature. We present a model based on the photo-absorption of electrons to account for the observed temperature dependence. As the results, the model fits our data very well, indicating that the coherent phonon response follows the density of photoexcited electrons above the Fermi energy.

The damping rate of the coherent \( E_{2g} \) mode in Zn increases upon increasing the temperature. This behavior is well accounted for by the anharmonic decay model, in which the optical phonon decays into the two acoustic phonons. Therefore, the damping of the coherent \( E_{2g} \) mode in Zn is governed by the anharmonic phonon-phonon coupling.[9]

Fig. 1. Transient reflectivity change observed in Zn and Cd at 7 K. The inset shows the imaginary part of the dielectric functions.

Constrained Density Functional Theory Calculations of Photo-excited Phonons

S. Fahy
Tyndall National Institute and Department of Physics, University College Cork, Ireland and FOCUS Centre, Department of Physics, University of Michigan, Ann Arbor, MI 48109-1120, USA., e-mail: s.fahy@ucc.ie e, http://www.physics.ucc.ie/staff/sfahy.html

We use recently developed density functional calculations of the electronic energies in solids, constraining the occupation of valence and conduction bands, to examine displacive excitation of coherent phonons by ultra-fast optical excitation of carriers. By understanding the phonon dynamics and their effects on the reflectivity modulation, we can obtain greater understanding of the complex dynamics of electron-hole diffusion and recombination, lattice heating, electronic softening and lattice anharmonicity. We look specifically at the contributions of electronic softening and lattice anharmonicity in tellurium and bismuth, using constrained density functional theory. In each case, the contribution of electronic softening is much larger than that of lattice anharmonicity for electron-hole plasma densities less than about 1.5% of the valence electrons, which is in the range of excitation usually obtained without sample damage.

Coherent lattice dynamics studied by femtosecond X-ray diffraction

Y. Hironaka¹, J. Irisawa¹, J. Saitoh¹, K. Kondo¹, K. Ishioka, K. Kitajima², and K. G. Nakamura¹,³
¹Materials and Structures Laboratory, Tokyo Ins. of Techn., Nagatsuta, 4259 Midori, Yokohama 226-8503, Japan
²National Institute for Materials Science, Tsukuba 305-0047, Japan
³Institute of Molecular Science, Okazaki, 444-8585, Japan
e-mail: hironaka@msl.titech.ac.jp

Coherent atomic motion so called “Coherent Phonon”, can be generated in solids by irradiation of ultra short laser pulses having a pulse duration much shorter than the period of the atomic vibration. It has been optically observed by a pump and probe method in various materials.

As the collective atomic motion of optical phonon mode does not change the barycentric position of lattice, it was considered to be impossible to measure the optical phonon mode by the conventional X-ray diffraction technique (by the angular shift of diffracted intensity). However, the couple of presence of coherent phonon and ultrafast time-resolved X-ray diffraction technique enable to detect the optical phonon motion in the crystal. Recent development of ultra short high-power laser system enables to realize the X-ray measurement in sub-picosecond time domain. Thus, the femtosecond X-ray diffraction technique enables to analyze not only acoustic phonons but also optical phonons excited in the crystal. K. Sokolowski, et al.[1] first measured the coherent optical phonon mode of Bi using the femtosecond X-ray diffraction technique. In the application of modern concepts of phase transition and chemical reaction involve dynamical changes of the atomic arrangements, and the understanding of temporal atomic motion is essential. In this point of view, time-resolved X-ray diffraction technique is the most powerful tools to detect not only the crystal structure but also the dynamics of transient phenomena such as phonon induced ultra-fast phase transition. In this experiment, we concentrated longitudinal phonon mode in CdTe [111] direction which was laser excited coherent LO phonon near the Brillouin zone center.

In the time-resolved X-ray diffraction, the information of coherent optical phonon will appear as the temporal modulation of X-ray diffracted intensity. According to the X-ray diffraction method, the parallel components of atomic deviation to the reciprocal lattice vector resolves amplitude of structure factor, which is well known as Debye factor for the case of incoherent phonons. In the case of coherent atomic motion the diffracted intensity modulates as the function of phonon frequency. Transverse atomic motion which is perpendicular to the reciprocal lattice vector will be excluded. Ultra-short characteristic X-ray pulse was generated by focusing the femtosecond laser (Ti:Spphire laser running at a repetition rate of 10 Hz delivered pulse of 400 mJ in 50 fs at a center wavelength of 800 nm with a contrast ratio of 10⁻⁶.) to the Cu tape target (=30 µm) in vacuumed chamber, and diffracted X-ray was detected by the X-ray CCD camera at the symmetrical Bragg diffraction geometry. The synchronized femtosecond laser pulse (=50 fs) with probe X-ray pulse was irradiated on the CdTe(111) surface with the power density of 161 GW/cm². The delay time between X-ray pulse and laser pulse were controlled by the optical delay line. X-ray penetration depth in CdTe for Cu Kα is approximately 0.7µm at the Bragg angle of 11.88 degrees and it is somewhere in the region of optical absorption depth (=0.5 µm). Then, most X-rays were diffracted from the laser excited region.

The both of diffracted signals from the laser perturbed and unperturbed surface of CdTe were detected at the same time, and normalized intensity was used for the reflectivity analysis. Time
zero, when the X-ray probe pulse and the laser pump pulse arrived at the surface of CdTe cointstantaneously, was determined by the sudden fall of the X-ray diffracted intensity. As the power density of 161 GW/cm² was not enough to generate the ultrafast disordering, a few picosecond lag time to the absolute zero point would be included. The observed diffracted intensity showed temporal modulation. Fourier power spectrum of the time-dependent intensity indicated single spike peak and residual small broad peaks. The single peak at 5.07 THz was obtained and it is equivalent to the LO phonon frequency at the Brillion zone center [2]. The coherent LO phonon was also obtained at 5.02 THz by using optical reflecting type pump and probe experiments on the same sample. Gabor transform, which is fourier transform with moving gaussian window, expressed the relaxation of the LO phonon and suggested phonon-phonon dynamics in CdTe.

The collapse and revival and the Fano interference with a complex asymmetry parameter for coherent phonons in semimetals

O.V. Misochko₁, M. Hase², K. Ishioka², and M. Kitajima²

₁Institute of Solid State Physics, Russian Academy of Sciences, 142432 Chernogolovka, Moscow region, Russia
₂Materials Engineering Laboratory, National Institute for Materials Science, 305-0047 Tsukuba, Japan

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One naturally expects that the most interesting physics in time-domain can be accessed at low enough temperature and high-fluence excitation. The low temperatures are needed for the creation of coherent (in Glauber' sense) phonon wave packet, with fluctuation properties identical to those of vacuum state, whereas the high fluence pulses provide the possibility to achieve such large amplitude displacements for atoms under which the anharmonism of lattice potential be essential. To demonstrate that these expectations are based on a solid ground, we have carried out the pump-probe experiments on semimetals (Bi and Sb) at helium temperature making use of high-fluence pulses. In both crystals, we have observed simultaneous excitation of the coherent $A_{1g}$ and $E_g$ phonons. The doubly degenerate $E_g$ phonons occur for strongly excited lattice at low temperature only, but nearly vanish at higher temperature. The initial phases of the coherent $A_{1g}$ and $E_g$ phonons are $\phi/2$ shifted, so the $A_{1g}$ phonons are excited displacively, while the $E_g$ phonons – impulsively.

We have also observed that in the non-linear regime, where the phonons’ oscillation parameters depend on fluence, both phonons are chirped in frequency. Furthermore, at sufficiently low temperature of the lattice and above threshold fluence there appears a collapse and revival for both phonon amplitudes. That is, at fluence greater than a threshold value, the oscillations die out up only to a characteristic (collapse) time as they did below the threshold, whereas at some time later the oscillations revive [1]. The collapse is due merely to the relative dephasing of the various elements of the phonon field, but the revival is entirely due to the grainy nature of the phonon field. The collapse and revival phenomenon is satisfactorily explained by the dynamics of a wave packet in an anharmonic potential. During the collapse-and revival regime, the phonon wave packet periodically breaks up and reconstitutes in its original form, thus giving convincing evidence that the phonons are in a quantum state, with no classical analog. A peculiar spectrum renormalization (emergence of a long-lived phonon mode and of higher phonon harmonics) along with an estimate of the phonon density during the collapse and revival phenomenon give certain hints on the exact nature of the transient quantum state [2], which we tentatively ascribed to Fröhlich condensation.

In Fourier transformed spectra, both phonons display an asymmetric (varying in time) line shape with more spectral weight at low frequencies testifying to a quantum (Fano) interference between the phononic and electronic degrees of freedom. However, the attempts to fit their line shapes with the Fano formulas revealed that the asymmetry parameter $q$ is time dependent and, furthermore, it can be positive or negative for different time delays.
A detailed study making use of a sliding-window Fourier transform performed for the $E_g$ phonon in bismuth exposed that the asymmetry parameter $q$ is a complex number oscillating between the positive and negative values with the frequency of the difference ($A_{1g} - E_g$) mode. This complex asymmetry parameter $q$ gives evidence on broken time-reversal symmetry in our experiments. Possible reasons for the broken time-reversal symmetry are discussed.

Coherent phonons in a semiconductor superlattice studied by femtosecond x-ray diffraction

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Reversible structural changes of a nanostructure were measured non-destructively with sub-picometer spatial and sub-picosecond time resolution via X-ray diffraction (XRD). The spatially periodic femtosecond excitation of a GaAs/AlGaAs superlattice results in coherent lattice motions with a 3.5 ps period, directly monitored by femtosecond X-ray pulses at a kilohertz repetition rate. Small changes $\Delta R/R=0.01$ of weak Bragg reflexes $R=0.005$ were detected. The phase and amplitude of the oscillatory XRD signal around a new equilibrium demonstrates the displacive excitation of the zone folded acoustic phonons as the dominant mechanism for strong excitation.

X-ray diffraction combined with the time resolution of femtosecond pump-probe experiments can directly monitor the ultrafast dynamics of nuclei triggered, e.g., by electronic excitation with sub-picometer and sub-picosecond accuracy. This method yields direct and quantitative information on the coherent lattice motion in contrast to all optical techniques which measure the phonon amplitude indirectly via the modulation of the susceptibility. This, together with the prospect of enabling "molecular movies" of chemical reactions, drives considerable efforts to improve time resolved optical pump – x-ray probe experiments, using both synchrotron facilities and table top x-ray plasma sources.

We report on nondestructive femtosecond XRD measurements of minute reversible structural changes in a nanostructured solid using a tabletop femtosecond x-ray source [1]. These changes conserve the crystal volume and occur in the femtosecond time domain. As a prototype sample representative for a larger class of inorganic and organic nanostructures, we chose a GaAs/AlGaAs superlattice. A femtosecond laser pulse impulsively excites electron-hole pairs in the lowest subband of the GaAs quantum wells (QWs), thus weakening the interatomic bonds by transferring valence band electrons (bonding orbitals) into conduction band states (anti-bonding orbitals). The crystal lattice responds to such excitation with an expansion of the wells and a concomitant compression of the AlGaAs barriers, and vice versa in the next half period, thus triggering a coherent acoustic standing wave. The amplitude of this motion is a fraction of only 0.00015 of the lattice constant, and as can be inferred directly from the measured intensity, which is directly proportional to the lattice compression. The signal (Fig. 1) was taken as the intensity difference between the pumped and the un-pumped region on the SL sample around the (002) reflex of bulk AlAs. The pump-probe data in Fig. 1 (symbols) display a periodic intensity modulation of the –1st order peak of the 002 reflex as a function of the time delay. The oscillation period of 3.5 ps corresponds to a frequency 0.29 THz. In contrast, the angular position of the –1st order peak is constant with negligible fluctuations, demonstrating that the volume of the crystal remains unchanged in the measured time range. The intensity modulations show a reflectivity change of 0.03 for a peak with the reflectivity 0.005, a sensitivity much higher than in earlier subpicosecond diffraction experiments.

This kind of displacive excitation of coherent phonons (DECP) [2] is a generic example of phase-coherent atomic motion, initiated in each unit cell of a (molecular) crystal. DECP is the solid state analog of the Franck-Condon principle for electronic excitation of molecules. Similar to the
In the case of bulk optical phonons there are controversial claims on the phonon excitation mechanism in superlattices, i.e., Raman-excitation vs. displacive excitation [3,4]. While the DECP mechanism yields oscillations in the excited electronic state around a displaced equilibrium position, the Raman process excites phonons in the electronic ground state with unchanged equilibrium. Our experiment provides direct evidence for the displacive excitation mechanism for ZFLAPs in semiconductor SLs in the high excitation regime. It clearly shows that the oscillations of the nuclei occur around a displaced equilibrium, i.e. in the excited electronic state. [5]

Femtosecond X-ray measurement of coherent lattice vibrations

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There are various ways of using ultrashort laser pulses to generate and detect coherent lattice vibrations. One possibility is to use a pump probe scheme in which a first laser pulse excites lattice vibrations and a second, delayed laser pulse probes the excited system. Lattice vibrations produce weak modulations of the optical properties, for example, changes in the optical reflectivity, which can be detected and measured by a suitable probe pulse. The duration of vibrational cycles of high frequency lattice vibrations is typically 50 to 100 fs. Currently laser pulses of just a few femtoseconds can be generated. Thus available time resolution is high enough to temporally map out even the highest frequency lattice vibrations. However, the spatial resolution in optical experiments is determined by the wavelength of light, typically several hundred nanometers, which is far too large for also resolving the position of the individual vibrating atoms.

Recently, ultrashort X-ray pulses have become available with wavelengths of only a few Angstroms, and rapid progress can be expected in the development of femtosecond X-ray sources. Thus it is within reach to perform optical pump/X-ray probe experiments, where the x-ray probe pulse provides both femtosecond temporal and Angstrom spatial resolution. The combination of atomic scale temporal and spatial resolution will eventually provide snapshots of atomic structures including coherent and incoherent motion of the atoms associated with lattice vibrations.

Experiments will be described in which femtosecond X-ray pulses obtained from a laser-produced plasma are used to perform time-resolved X-ray diffraction experiments in Bismuth. Strong coherent optical phonon modes can be excited in Bi by means of displacive excitation. X-ray diffraction reveals the changes in the position of atoms thus providing a direct picture of the changes in the atomic positions associated with a coherent vibrational mode of the lattice.

Correlated Electrons
Probing quasiparticle dynamics in the magnetoresistive pyrochlore $\text{Tl}_2\text{Mn}_2\text{O}_7$ using mid-infrared spectroscopy

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Ultrafast optical techniques are gaining increasing importance in the investigation of materials where strongly coupled degrees of freedom (i.e. charge, spin, lattice, orbital) determine functionality. A common goal of such investigations is to obtain insight into the ground state properties by perturbing the material and observing the subsequent return to equilibrium. Important information is obtained in such experiments since a subpicosecond probe pulse can temporally resolve dynamics at the fundamental timescales of electronic and atomic motion. Correlated electron materials including the doped transition metal oxides (TMO) and their undoped parent compounds are of particular interest for the investigation of photoinduced phenomena. There have been a number of ultrafast optical studies on TMOs including high-$T_c$ superconductors and colossal magnetoresistive manganites [1,2]. Our work has primarily used optical pump-terahertz probe and all optical pump-probe spectroscopy to investigate quasiparticle dynamics in correlated electron materials [2].

In this study, we utilize mid-infrared probe pulses to investigate quasiparticle dynamics in the magnetoresistive pyrochlore $\text{Tl}_2\text{Mn}_2\text{O}_7$. Using infrared pulses enables tracking of the Drude plasma edge shift following photoexcitation, providing direct insight into photoexcited carrier lifetimes. Our results reveal the dominance of spin fluctuations in determining the fate of photoexcited carriers in this compound.

At $T_c = 120$ K, $\text{Tl}_2\text{Mn}_2\text{O}_7$ becomes a half-metallic ferromagnetic metal [3]. However, in comparison to the widely studied manganites such as $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$, the CMR mechanism is expected to be very different since double exchange and Jahn-Teller effects are negligible due to the low carrier density and absence of the Jahn-Teller ion $\text{Mn}^{3+}$ [3]. Thus, time-resolved measurements on $\text{Tl}_2\text{Mn}_2\text{O}_7$ present an interesting point of comparison with previous work on the manganite perovskites [2].

$\text{Tl}_2\text{Mn}_2\text{O}_7$ was synthesized by a high pressure solid state reaction [3]. The measured ferromagnetic Curie temperature is 120 K. The optical pump, mid-IR probe system is a 1 kHz regenerative amplifier producing 2 mJ, 60 fs pulses at 800 nm. The amplifier output is split into two beams to excite the sample and pump an optical parametric amplifier (OPA). The signal and idler beams from the OPA are mixed in a nonlinear crystal to generate the tunable 3-20 µm mid-IR probe through difference frequency generation. A pump wavelength of 800 nm was used to excite the sample and all measurements were done in reflection with the pump and probe s polarized. The use of an 800 nm (1.55 eV) pump is important since 1.55 eV photons excite electrons into the minority spin manifold. Thus, we selectively photoexcite electrons into the band which is responsible for conduction in $\text{Tl}_2\text{Mn}_2\text{O}_7$, allowing us to probe the free carrier response.
Figure 1 depicts measurements at 5 μm for various temperatures; the pump fluence was 740 μJ/cm², exciting an initial carrier density of $4.2 \times 10^{20}$ cm$^{-3}$. At all temperatures, the traces show a rapid change in reflectivity at a pump-probe delay $\tau=0$, followed by a fast relaxation with a time scale of a few picoseconds and a slower component with a time scale of tens to hundreds of picoseconds.

We have been able to show that the induced change in reflectivity $\Delta R/R$ is primarily determined by the change in carrier density ($\Delta R/R(t) = 1/R(\partial R/\partial n)\Delta n(t)$ where $\Delta n(t)$ is the time dependent carrier density). There is a relatively temperature independent fast (~10ps) initial relaxation in $\Delta R/R$ due to carrier trapping, leaving fewer photoexcited carriers to contribute to the Drude-like response. Nearly all the carriers follow this relaxation pathway for $T \ll T_c$, similar to other ferromagnetic metals [2]. However, upon approaching $T_c$, the magnitude of the transient reflectivity signal increases significantly, both at $\tau=0$ and at longer time delays ($\tau\geq50$ ps), after the carriers have reached a thermal distribution. A large long time offset appears in $\Delta R/R$ near and above $T_c$, indicating the establishment of a long-lived photoexcited carrier population.

A detailed analysis of the data in Figure 1 reveals that the photoexcited carrier density is strongly influenced by spin disorder throughout the measured temperature range (10-300K), with static spin disorder dominating the dynamics for $T \leq 0.75T_c$, dynamic spin fluctuations dominating for $T \geq 1.4T_c$, and exceptionally long lifetimes (>1.5 nanoseconds) measured for $0.75T_c \leq T \leq 1.4T_c$. In particular, spin disorder opens a channel for the efficient conversion of electron-hole singlets to triplets without spin flip processes thereby reducing the probability for recombination. Our results are consistent with a scenario whereby carriers are localized at the scale of the magnetic correlation length $\xi$. This suggests that Tl$_2$Mn$_2$O$_7$ may be a particularly simple example (i.e. low carrier density and no JT phonons) where the transport properties are determined by intrinsic nanoscale inhomogeneities that occur at a second order phase transition.

Femtosecond X-ray diffraction and absorption in solids

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I will summarize some of our recent experimental work, aimed at the study of dynamic phenomena in correlated oxides and in ferroelectrics. We combine several ultrafast optical and x-ray techniques, which range from femtosecond spectroscopy in the mid-IR to femtosecond x-ray diffraction and absorption. I will discuss experiments that seek to clarify the dynamics of phase transitions in strongly correlated Vanadium Dioxide and in the CMR-manganite PrCaMnO\textsubscript{3}. I will also cover recent femtosecond x-ray diffraction experiments in Ferroelectric LiTaO\textsubscript{3}.

Correlated many electron dynamics with the Multi-configuration time-dependent Hartree-Fock (MCTDHF) method

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We present the Multi-Configuration Time-Dependent Hartree-Fock (MCTDHF) method for correlated quantum dynamics of many electron systems. The correlated motion of electrons plays an essential role in many branches of physics and chemistry, like photoelectron spectroscopy, scanning tunneling microscopy, etc. This is even more so, since laser pulses on the attosecond time scale allow to study the time-resolved dynamics of electron systems out of equilibrium. The MCTDHF method is applied to various problems, like excitation/ionization by laser pulses, inelastic electron scattering, inverse photoemission, and electron impact ionization. We discuss the properties of the method in the framework of one-dimensional model systems. Familiar concepts from nuclear quantum dynamics, e.g. Complex Absorbing Potentials, propagation in imaginary time and the wave packet approach to spectroscopy are applied to correlated many electron dynamics, and the differences and new aspects are discussed. Some comparisons with another method for many electron dynamics, the time-dependent configuration interaction singles (TD-CIS) method, are made.

Importance of singlet to triplet processes versus traps in pentacene and tetracene single crystals


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The strong potential for use of organic semiconductors in technological applications has, in recent years, spurred a tremendous research effort in these materials [1]. In particular the polycrystalline pentacene (with one the highest mobilities in this class of materials) is a leading candidate for applications in organic electronics. Advances in ultrafast femtosecond spectroscopic techniques have facilitated investigations of excited state processes including the dynamics of excitons, polarons, traps, and photoinduced electron transfer. This will lead to a better understanding of the optical and electronic properties and the underlying photophysics of these materials.

Here we present optical pump-probe measurements of photoinduced (PI) changes in the reflectivity $\Delta R/R$ and transmissivity $\Delta T/T$ of tetracene and pentacene single crystals and films. Both tetracene and pentacene belong to the polycrystalline series of organic crystals which, in order of increasing molecular size includes naphthalene, anthracene, tetracene, and pentacene. The energy level of the first triplet exciton $E(T_1)$ decreases faster than the first singlet exciton energy $E(S_1)$ with increasing molecular size. Thus, the energy difference $E(S_1)-2E(T_1)$ is -1.3 eV in naphthalene, -0.55 eV in anthracene, -0.21 eV in tetracene, and 0.11 eV in pentacene [2]. Therefore, in pentacene, the excitonic fission process from the lowest singlet exciton to a pair of lowest triplet excitons $S_1\rightarrow 2T_1$ is energetically allowed, while in tetracene this same process is only possible by thermally activated fission (see energy diagrams in Figure 1). This process is strongly suppressed in naphthalene and anthracene.

In these experiments we studied the photoexcited carrier relaxation dynamics in tetracene and pentacene as a function of probe photon energy aiming to elucidate the electronic structure and carrier dynamics in the singlet and triplet manifolds. From our results it is clear that both excitons as well as traps are important, and we focus on unraveling the nature of the singlet-triplet fission processes and the role of traps. Both pentacene and tetracene crystals display a very strong and long-lived PI absorption peak centered at 1.7 eV (tetracene), and 1.4 eV (pentacene), which originates from triplet state absorption following singlet fission. In pentacene this triplet production occurs on a 1 ps time scale, while in tetracene it is much slower by a factor of about 50 in agreement with the physics of the excitonic fission process. In contrast to pentacene, the $S_1\rightarrow 2T_1$ fission process in tetracene is found to be temperature dependent (see Figure 1). We infer from this data that the triplet production is not solely due to the $S_1\rightarrow 2T_1$ process [2,3], but that the $T_1$ level is additionally populated by fission processes from higher lying states, $S_n\rightarrow 2T_1$. We present the temperature dependence of the triplet production, as well as the branching ratios of the $S_1\rightarrow 2T_1$ and $S_n\rightarrow 2T_1$ fission processes, and make a comparison to the processes and timescales in pentacene.
With a good understanding of the exciton dynamics we then investigate the role of traps. In pentacene thin films, disorder and traps become more important in contrast to crystals where the trap density is much smaller. When introducing impurities such as C\textsubscript{60} (or other impurities which serve as electron traps) in pentacene thin films we find that the triplet production is suppressed by more than 95% and the dynamics are largely dominated by traps, unlike in purer crystals where exciton dynamics dominate. This is attributed to competing processes (e.g. photoinduced charge-transfer or trapping) on a faster time scale which inhibits the fission processes. Furthermore, in contrast to crystals, both C\textsubscript{60}-doped or undoped pentacene films exhibit strong temperature dependence which suggests thermal activation out of traps.

Ultrafast dynamics of photoinduced phase transitions in 1D correlated electron systems

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Control of phase transitions and related macroscopic properties by photo-irradiation is now attracting much attention. This phenomenon is called photo-induced phase transition (PIPT), and is important not only as a new phenomenon in the fields of physics and chemistry, but also as a useful mechanism applicable to future optical switching devices. When one considers applications of these phenomena to switching devices, it is essentially important to control electronic structures and physical properties in picosecond (ps) or sub-ps time scale. A key strategy toward realizing such ultrafast PIPTs is the exploration of correlated electron systems. In those systems, PIPTs are expected to be driven in ultrafast time scale, since photocarriers can trigger large changes of electronic orderings through the strong interaction between photocarriers and surrounding electron(spin) systems. In this talk, we will report the ultrafast PIPTs in one-dimensional (1D) correlated electron systems of transition metal compounds and organic charge-transfer (CT) complexes.

(1) Photoinduced insulator-metal transition in 1D Mott insulators
Ultrafast optical switching from a Mott insulator to a metal in a halogen-bridged Ni-chain compound was demonstrated by a femtosecond pump-probe reflection spectroscopy. Upon the irradiation of a 100-femtosecond laser pulse, the spectral weight of the gap transition is transferred to the inner-gap region. When the photoexcitation density exceeds 0.1 photon/Ni site, a Drude-like high-reflection band appears in the infrared region, signaling the formation of a metallic state. The photogeneration of the metallic state and the subsequent recovery to the original gapful state occur within a few picoseconds.

(2) Photoinduced inverse spin-Peierls transition in 1D spin systems
Ultrafast photoinduced melting of a spin-Peierls (SP) phase was investigated in a 1D spin system of K-tetracyanoquinodimethane (K-TCNQ). Photocarrier generations destabilized the SP phase, resulting in a decrease of molecular dimerizations within 400 fs of over 20 TCNQ sites per photon. Such a melting of the SP phase drove three kinds of coherent oscillations. By comparing the oscillations with the Raman bands activated by the dimerization, we demonstrated that the oscillation of 20 cm\(^{-1}\) is due to an LO phonon mode and it plays an important role for the stabilization of the SP phase.

(3) Photoinduced neutral-ionic transition and coherent control of charge and lattice dynamics in molecular crystals
Dynamics of the photoinduced neutral (N) to ionic (I) transition in a mixed-stacked CT compound, tetrathiafulvalene-p-chloranil (TTF-CA) was investigated by a femtosecond pump-probe reflection spectroscopy. 1D microscopic I domains were produced just after the photoirradiation in the N phase and decay within 20 ps. In addition to the photoinduced N to I conversion, a coherent oscillation with the period of 0.6 ps (the frequency of 54 cm\(^{-1}\)) was observed on the photoinduced reflectivity change for the intramolecular transition band sensitive to the degree of the charge-transfer between a donor of TTF and an acceptor of CA. This oscillation was reasonably assigned to the charge-lattice coupled oscillation associated with the dynamical dimeric displacements of molecules, which were driven by the spin-Peierls-like instability.
Moreover, coherent control of the charge-lattice coupled oscillation was successfully made with use of double femtosecond laser pulses. By changing the interval of the two pump pulses, the amplitude of the oscillation on the reflectivity change was periodically changed. With increase of the pump intensity, the oscillation amplitude was dramatically enhanced. Such an enhancement will be discussed in terms of the cooperative nature of the NI transition.

Quasiparticle relaxation in d-wave superconductors

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Measurements of relaxation processes of Bogoliubov quasiparticles, the elementary excitations in superconductors, can provide information on electron-phonon and electron-electron interactions, including the residual interactions leading to the pairing instability. Very recent equilibrium measurements by microwave cavity techniques and by angle-resolved photoemission have allowed one to finally obtain a coherent picture of the relaxation of quasiparticles near the d-wave gap node in the cuprates which dominate low-frequency transport. The effect of disorder is important, and it is found that a realistic model of different disorder components is required to explain experiments. I will outline the theoretical understanding of these experiments at and near optimal doping in terms of rather conventional BCS theory. I speculate about in influence of order parameter inhomogeneity on these results.

When the d-wave quasiparticle system is pumped far out of equilibrium, new kinds of relaxation dynamics can arise which differ considerably from the models used to describe hot quasiparticle relaxation in conventional superconductors. I will briefly review the rather confusing experimental situation in this regard, and then try to formulate the simplest possible model problem which captures the essential aspects of the d-wave case. In particular, I sketch calculations which consider the processes by which "hot" quasiparticles produced near the antinodes of a d-wave superconductor can relax via momentum conserving electron-electron interactions. In a large region of momentum space, processes which break Cooper pairs are forbidden by energy and momentum conservation. Equilibration then occurs by scattering with thermal quasiparticles: Umklapp scattering is exponentially suppressed at low temperatures, but small-angle scattering leads to power-law behavior. By solving the Boltzmann equation analytically, one can make detailed predictions for the temperature and intensity dependence of these processes, which will be compared with experiment.
Quasi-particle interactions
Ultrafast quantum kinetics of elementary interactions in semiconductors

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The optical and transport properties of semiconductors are strongly influenced by ultrafast interaction processes between elementary excitations. There are two fundamental coupling mechanisms for free electronic charges: Carrier-carrier interactions via the screened Coulomb potential and carrier-phonon scattering. Traditionally, non-equilibrium dynamics governed by these processes has been described via the semiclassical Boltzmann equation. This picture implies that scattering events are point-like in time and do not possess internal structure. It is clear that on ultrafast time scales this approximation does not hold and new physics might arise. Specifically, the Boltzmann model is valid only under conditions where the scattering rates are substantially slower than the oscillation frequencies of the collective excitations involved, namely plasmons and phonons. However, elementary excitations in the condensed phase often feature scattering times which are in the same order of magnitude as the inverse phonon or plasmon frequencies. This regime is governed by quantum kinetics where the wave nature of particles is relevant and quantum interference phenomena are playing a dominant role [1]. To provide insight into semiconductor quantum kinetics, three experiments from this context are featured.

Carrier-phonon scattering is studied in the low-density regime where the dynamics of free electrons is dominated by interaction with the lattice. As an example, the emission of LO phonons by photoexcited hot electrons in the conduction band of GaAs has been investigated [2]. On a time scale comparable to the phonon oscillation cycle (115 fs in GaAs), scattering events of free carriers exist which do not conserve energy. As time proceeds, destructive quantum interference causes these virtual transitions to disappear and the semiclassical limit is restored.

Carrier-carrier scattering is prominent in the high-density regime where Coulomb interactions among electrons are frequent. While the bare Coulomb potential has infinite spatial range, mutual screening of positive and negative charges causes electric forces to be relevant only over small distances. However, one might ask how fast Coulomb screening is established in a free carrier ensemble in an extremely non-stationary situation far from thermal equilibrium. Ultrabroadband terahertz spectroscopy gives insight into a regime of quasi-bare Coulomb scattering early after 10 fs photoexcitation of an electron-hole plasma in GaAs. It demonstrates how many-body correlations are established on a time scale comparable to the inverse plasma cycle [3]. The transition of a purely dielectric system into a regime with phonon-plasmon coupled modes is investigated in InP [4]. A two-time femtosecond experiment demonstrates the bifurcation topology from the single LO phonon resonance of the intrinsic material into the state after photoexcitation of an electron-hole plasma which is characterized by the L⁺ and L⁻ branches of the phonon-plasmon dispersion. In addition, the density dependence of the ultrafast buildup dynamics of phonon-plasmon screening is mapped out between $5 \times 10^{16}$ cm$^{-3}$ and $5 \times 10^{18}$ cm$^{-3}$.

Recent experiments on photoexcited electron dynamics in heavy fermions have shown that the relaxation time of the photoinduced (PI) reflectivity transient increases by more than two orders of magnitude upon cooling from 300 K to 10 K [1,2]. In contrast, the dynamics of non-magnetic analogues showed similar behavior to conventional metals like Au and Ag [3], suggesting their dynamics is governed by the electron-phonon (e-ph) thermalization process, described by the so-called two-temperature model (TTM) [3,4]. The relaxation times in magnetic and non-magnetic compounds were quite similar at high temperatures (T) - thus suggesting the TTM as a starting point for heavy fermions. Indeed, the TTM analysis of the relaxation time suggested that the relaxation dynamics in heavy fermions can be accounted for by a simple e-ph thermalization, and provided that there exists a mechanism of e-ph scattering suppression when both initial and final electronic states lie within the peak in the density of states (DOS) at the Fermi level (Ef). It was argued that the small Fermi velocity compared to the sound velocity may be the origin of this suppression, since the energy and momentum conservation law in this case suppress the e-ph scattering within the peak in the DOS at Ef [1,2]. On the other hand, there were some observations not accounted for by this model, e.g. the anomalous rise-time dynamics observed in YbAgCu4 at low T [1].

Similar T-dependence of the rise-time dynamics has also been observed in superconductors [5]. Moreover, as pointed out by Schneider et al. [6], the T-dependence of the relaxation rate observed in YbAgCu4 also shows close resemblance to the data on both conventional [5] and cuprate [6,7] superconductors. The very similar behavior of the photoexcited carrier dynamics in superconductors and heavy electron systems suggests that the physics governing the relaxation dynamics in heavy electron compounds may need to be reconsidered.

The relaxation and recombination phenomena of nonequilibrium superconductors has been one of the most intriguing problems in condensed matter physics since the 1960's [8]. It was soon realized that the presence of the superconducting gap in the single particle excitation spectrum presents a relaxation bottleneck, because there are no electronic states in the gap. The minimal model that describes the relaxation of the (photo)excited superconductor was formulated in 1967 by Rothwarf and Taylor [9]. Pointing out that the phonon channel should be considered when discussing relaxation phenomena, they described the relaxation dynamics by two coupled nonlinear-differential equations [9]. While in the low perturbation limit the equations can be linearized, it was shown recently that approximate analytical solutions can be obtained for all limiting cases [5,10]. Analytical solutions enable comparison of the experimental data with the model, and one can show that the Rothwarf-Taylor (RT) model can account for both the rise-time dynamics [5,10] as well as the superconducting state recovery dynamics as a function of excitation fluence (F) and T for conventional as well as cuprate superconductors [10].

Here we present a detailed study of the carrier relaxation dynamics in heavy electron systems with the emphasis on the Kondo insulator SmB6 and heavy fermion YbAgCu4. Utilizing a low repetition rate optical parametric amplifier we were able to measure the excitation density (F) dependence of the reflectivity transient over more than three orders of magnitude in F. In both compounds the amplitude of the transient shows a sub-linear F-dependence. Furthermore, the
relaxation rate was found to be strongly F-dependent. Both observations are consistent with the RT prediction [10] implying that the relaxation process in heavy electron compounds is governed by the presence of the hybridization gap. The T-dependence of the transient amplitude and relaxation rate further support the hybridization gap scenario, and the extracted values of the hybridization gap are in good agreement with published data – see Figure 1.

Importantly, the data suggest that the hybridization gap is to a first approximation T-independent as in the case of a periodic Anderson lattice model in the U=0 limit [11]. The dynamics can be well accounted for by the phenomenological Rothwarf-Taylor model, suggesting that the carrier relaxation and recombination dynamics in heavy electron systems is governed by the presence of the hybridization gap, which is found to be only weakly temperature dependent.

Ultrafast measurements of quasiparticle dynamics in cuprates and spin diffusion in quantum wells

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Despite nearly two decades of intense research in high temperature superconductivity, the mechanism that binds Cooper pairs is still not known. When Cooper pairs break apart, elementary excitations known as quasiparticles are generated. Understanding the dynamic properties of quasiparticles such as their rates of recombination, scattering and diffusion, is critical to uncover the nature of interaction that leads to pairing.

Nonequilibrium measurements provide unique opportunity of creating and studying quasiparticles in cuprates. We used pump probe spectroscopy to determine the recombination rate of quasiparticles. In these experiments, quasiparticles are injected into single crystal samples by short laser pulses with photon energy 1.5eV. Once injected, they change the index of refraction at the laser frequency, which allows time-resolved optical measurements of their density. Measurements of the decay of quasiparticle density after pulsed injection yields the rate at which Cooper pairs reform and enter the superfluid condensate. I will report on measurements of quasiparticle decay rate for two different high temperature superconductors (YBCO and BSCCO). I will discuss the dependence of decay rate on temperature, excitation density [1] and carrier concentration [2] in the sample.

Using transient grating spectroscopy, real space propagation of quasiparticles is measured [3]. In this technique, two coherent laser pulses are interfered at the sample creating a spatially periodic quasiparticle distribution. A third beam sent on to the sample is diffracted as well as being reflected by this quasiparticle grating. Recording both diffracted and reflected signals as a function of time yields precise measurement of the quasiparticle diffusion coefficient. To measure very small diffracted signals, we implemented heterodyne detected transient reflection grating spectroscopy using a phase mask with a novel method of phase calibration [4]. We discuss the experimental setup and implications of the measured diffusion coefficients in YBCO and BSCCO.

Motivated by the success of the transient grating method in cuprates, we used a similar technique to measure spin diffusion in the two-dimensional electron gas in GaAs/GaAlAs quantum wells (QW’s). In this technique, two coherent laser pulses with perpendicular polarization interfere at the surface of a sample to produce a sinusoidal variation of photon helicity. Because of spin-orbit coupling, the photon-helicity variation is transcribed into a periodic wave of electron spin polarization, whose amplitude can be probed by diffracting time-delayed laser pulses from the surface. The rates of spin transport and relaxation can be inferred from the evolution through time of the spin polarization grating. I will report on measurements of spin diffusion in QW samples as a function of temperature (T), carrier concentration (n), and mobility (μ). We compare spin diffusion coefficient to the charge diffusion coefficient which is obtained (using the Einstein relation) from 4-contact measurements of resistivity performed on samples from the same wafer. We find that spin diffusion coefficient is smaller than charge diffusion coefficient by an amount which agrees quantitatively with the predictions of D’Amico and Vignale [5]. This constitutes the first observation of the “spin Coulomb drag” effect, which acts selectively to oppose the relative motion of oppositely aligned spin populations because of electron-electron collisions.

*Work performed in collaboration with:

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Transient insulating, conducting and superconducting phases probed with ultrashort THz pulses

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In a many-body system, Coulomb interactions between a large number of quasiparticles can generate low-energy correlated states that result in new excitations and physical properties. Ultrafast time-resolved spectroscopy is a powerful tool to study key microscopic processes in such systems by following the temporal relaxation of nonequilibrium quasiparticles. Transient insulating, conducting, or superconducting phases can occur which exhibit a characteristic spectrum of their lowest-energy elementary excitations at terahertz (THz) frequencies. Here, we discuss experiments that employ ultrashort coherent THz pulses and direct field-resolved detection to probe time-varying correlations of charge carriers in semiconductors and superconductors.

In the past, bound electron-hole (e-h) pairs in semiconductors were extensively explored at near-bandgap frequencies, while studies of their internal transitions - several orders of magnitude below the optical bandgap - remained scarce. Recently, we carried out optical-pump THz-probe experiments that explore the dynamics of bound and unbound e-h pairs in GaAs/AlGaAs quantum wells after selective near-IR excitation [1]. Resonant generation of bound pairs (excitons) results in a new low-energy oscillator around 7 meV that arises from transitions between each exciton’s internal 1s and 2p levels. Owing to the correlated e-h motion, charge-neutral excitons form an insulating phase. Above-bandgap excitation, in contrast, induces a conducting plasma of unbound e-h pairs. The distinct responses of these phases enable us to follow in time a metal-insulator transition that occurs upon exciton formation out of a gas of unbound e-h pairs, as well as its reverse process of ionization. At high excited pair densities, the exciton THz resonance exhibits a substantial weakening of the binding energy and increased broadening [2]. Densities of excitons and unbound e-h pairs are quantitatively obtained from a two-component model of the THz dielectric response. The resonance disappears at high pump powers where the response evolves into a Drude-like conducting phase of unbound e-h pairs. Comparison with near-IR absorption reveals a significantly enhanced shift and broadening of the THz resonance.

In superconductors, the THz-frequency electromagnetic response couples directly to Cooper pairs and quasiparticle excitations. We have studied the transient THz conductivity in the high-T\textsubscript{c} superconductor Bi\textsubscript{2}Sr\textsubscript{2}CaCu\textsubscript{2}O\textsubscript{8+δ} that occurs after ultrafast depletion of the superconducting condensate. The conductivity spectra and their temporal decay kinetics reveal a bimolecular kinetics of quasiparticle recombination as Cooper pairs form on a picosecond timescale. The experiments discussed here directly trace the low-energy spectra and dynamics of correlated pair states, which uncovers microscopic processes and motivates their use in future studies of collective excitations in solids.

Broadband Time-Domain THz Spectroscopy of Semi-Conducting Organic Crystals

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Charge transport in semi-conducting organic crystals has been, and is extensively studied.¹ One reason for this scientific attention is the fact that these materials provide promising building blocks for (molecular) electronic devices. However, a good understanding of the microscopic charge transport mechanism in these organic materials is currently lacking. Although several studies have indicated that inter-molecular (lattice) and intra-molecular structural reorganization, i.e. electron-phonon coupling, plays an important role in explaining some of the observed charge transport properties in these materials, no direct spectroscopic evidence of coupling between (photo-induced) mobile charges and intra-molecular vibrations exists for this class of materials.

A technique well suited for the study of the mobility of transient photo-excited carriers in semi-conductors is terahertz time domain spectroscopy (THz-TDS).² Most THz-TDS measurements have been limited to frequencies <3 THz, where the response is dominated by these mobile carriers. However, at higher frequencies (>10 THz) in organic materials also (intra-molecular) vibrational transitions can be expected, thus THz-TDS at these frequencies allows for the simultaneous measurement of the electronic and vibronic response. Because the pump induced changes in the THz probe pulse transmission through the sample are electro-optically detected in the time domain, both amplitude and phase are known, giving access to the photo-induced complex dielectric response of the sample. Owing to the electro-optical detection scheme using ultrashort pulses, in THz-TDS the dynamics of the frequency resolved spectral changes can be followed on a femtosecond timescale.

Here we use broadband THz-TDS to study both the electronic and vibronic response of a Rubrene crystal upon ultra-fast photo-excitation. The pump pulse (3.0 eV) induces an electronic transition in the Rubrene (S₀→S₁ gap 1.92-2.2 eV), generating mobile charges. It should be noted that in the THz region available in this experiment, Rubrene shows several IR active vibrational modes.

Fig. 1. The real part of the frequency resolved photo-induced THz transmission changes (ΔE/E) as a function of delay after excitation. Top: datasets for a thick (~5 µm) crystal (left) and for a thin (~2 µm) Rubrene crystal (right). Bottom: The real and imaginary parts of ΔE/E at the maximum of the signal (from 200-400 fs)
As shown in Figure 1, we observe both a broad photo-induced absorption, corresponding to mobile charges, and sharp resonances, with similar frequencies as ground state vibrational transitions. Interestingly the resonant and non-resonant signals show different dynamics, with the resonant signal disappearing after 600 fs, while the non-resonant signal persists for several picoseconds.

We have modeled our experiment using an analytical model describing the THz field transmission through a photo-excited sample with ground state absorption lines and can reproduce most of our experimental data, including the faster decay of the resonant signal compared to the non-resonant signal, assuming only a broad photo-induced absorption. Differences between some of the model input parameters and known experimental parameters, however, indicate the possible observation of phonon line broadening in the excited state.

The coherent coupled electron-lattice response of Si to an optical impulse

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The Coulomb interaction between electrons and phonons defines many of the physical properties of Si, e.g., the electrical conductivity: real and virtual scattering processes transfer carriers between different momentum states and renormalize their mass. Much of our knowledge of the electron-phonon interaction in Si is limited to transport measurements at near equilibrium conditions. Studies of the carrier relaxation in silicon by optical techniques have been restricted mainly to transient changes of index of refraction following excitation by intense laser pulses, because the indirect band gap of Si obviates more direct probes. We report on the coherent response of the coupled Si carrier-lattice system to excitation by a 10 fs, ~400 nm laser pulse. The transient anisotropic reflectivity is measured and transformed into time-frequency space revealing the dynamics of formation of the coherent longitudinal optical (LO) phonon at 15.3 THz [1]. The phonon dephasing is governed by the deformation potential interaction with the nonequilibrium electron-hole plasma.

The anisotropic transient reflectivity of n-Si(001) under ambient conditions is measured by the electro-optic (EO) sampling method. Nearly collinear, pump and probe pulses (10 fs pulse duration; 2.99-3.16 eV energy; 65-90 MHz repetition rate) are focused to a 10 µm spot on the sample. The pump pulse with 40 mW average power generates a carrier density of 5x10¹⁹ cm⁻³. After reflecting from the sample, the probe beam is analyzed into polarization components parallel and perpendicular to that of the pump, ΔR∥ and ΔR⊥, and detected with matched photodiodes. The photocurrents are subtracted, and their difference ΔR∥−ΔR⊥ recorded as a function of the pump-probe delay. The transient reflectivity is measured both with the [110] crystalline axis of Si oriented parallel and at 45° to the pump polarization to explore the Γ₂⁵ and Γ₁₂ symmetry response, respectively. Figure 1 shows the different signal components separately, as well as their difference, which corresponds to the EO response for the Γ₂⁵ symmetry. Although, the electro-optic effect is forbidden by symmetry in centrosymmetric materials such as Si in the electric dipole approximation, it can be observed in the bulk through the electric quadrupole interaction.

![Fig. 1. The parallel and perpendicular polarization components of the EO signal recorded separately and their difference for the Γ₂⁵ symmetry. The oscillations due to the coherent phonon excitation are observed only for the Γ₂⁵ geometry.](image-url)
The EO signal in Fig. 1 consists of an aperiodic component near the zero delay and 66 fs period oscillations that decay quasi-exponentially on a picosecond time scale. The initial signal is the coupled electron-phonon response to generation of an anisotropic real and reciprocal space carrier distribution by the impulsive excitation near resonant with the direct band gap of Si (E1 critical point at 3.4 eV). The phonon oscillations exhibit a frequency chirp and decay with a time-dependent dephasing rate, which represent the self-energy of the phonon due to the deformation potential interaction with the photoexcited holes. The self-energy changes with time reflecting the evolution of the nonequilibrium photogenerated plasma through carrier-carrier scattering and diffusion.

In summary, we have explored carrier-phonon interaction in Si, which is manifested in correlation effects during the quasiparticle generation, and on a longer time scale, in the carrier density-dependent self-energy of coherent phonons. Because of their practical importance and relative simplicity, the quasiparticles in Si present an inviting playground for studying quasiparticle correlation in solid-state materials.

Spectrally resolved coherent spin dynamics in 2DEGs

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The functionality of conventional semiconductor devices is based on the electronic charge. However, the electron has an additional degree of freedom: Spin, which is an intrinsic angular momentum. In comparison with conventional charge-based devices, spin-based (opto-)electronic devices have several advantages: Weaker interactions with the solid-state environment, larger coherence times, the possibility of fast optical control and higher efficiency. Recently, spin transistors, spin memories, spin batteries, spin modulated vertical-cavity surface-emitting lasers and spin quantum computers have been proposed as promising tools for future (opto-)electronics. Our research concentrates on an optical investigation of electron-spin dynamics in low-dimensional non-magnetic semiconductor nanodevices. Fundamental knowledge of the mechanisms that cause spin relaxation and spin decoherence in these low-dimensional devices, as well as the ability to manipulate spin polarization and electron-spin transport in these systems is interesting from both a fundamental and practical point of view. The two-dimensional electron gas (2DEG) formed at the interface of two semiconductors is a particularly attractive material for realizing semiconductor nanodevices1,2. At the interface, the bottom of the conduction band forms a triangular quantum well (in the direction normal to the interface plane). Only the lowest quantum state in this well (width is about 10 nm) falls below the Fermi energy, and all the conduction electrons are thereby confined to a plane. The electrons can freely move in the plane, with an extremely high mobility (mean free path in excess of a micrometer).

We used frequency and time-resolved magneto-optical Kerr effect (TRMOKE) spectroscopy for studying coherent electron-spin dynamics in an AlGaAs/GaAs 2DEG at different temperatures and in the presence of an external magnetic field. TRMOKE spectroscopy is a pump-probe technique. A circularly-polarized pump pulse generates spin-aligned charge carriers. A change in the polarization state of a later probe pulse upon reflection on the material, contains information about the transient spin dynamics of these charge carriers. Since the 2DEG is formed at the interface of two semiconductors, the response of the 2DEG is influenced by carrier dynamics in adjacent bulk layers. In order to extract the signals characteristic for electrons in the 2DEG out of the total response, we measured as a reference in the same setup on bulk n-GaAs, whose properties are well known3.

The obtained results reveal substantial differences between the coherent spin dynamics in bulk n-GaAs and the 2DEG system (see figure). We present here results from the regime where the number of photo-excited carriers is large in comparison to the equilibrium density of electrons in the 2DEG (~3·11 cm⁻²) and bulk n-GaAs3 (~1·10¹⁶ cm⁻³). In absence of an external magnetic field (top panels in the figure) the TRMOKE signal from bulk n-GaAs is characterized by an instantaneous rise, a fast initial sub-2 ps decay which can be ascribed to exciton formation, and a slower exponential sub-ns decay which speeds up with increasing temperature. The speeding of the overall decay with temperature is characteristic for the Elliot-Yafet (EY) mechanism for loss of spin coherence4. The electron spins dephase here by momentum scattering via by spin-orbit coupling. In this case the spin scattering rate is proportional to the momentum scattering rate.
In the case of the 2DEG, a slow formation (~100 ps) of the signal is observed. The slow formation, which speeds up with increasing temperature, can be understood by taking into account both spatial diffusion of the coherent spin population from adjacent bulk into the 2DEG quantum well, and kinetic-energy relaxation of photo-excited electrons in the conduction band. In contrast to the case of bulk n-GaAs, the decay of a spin-coherence in the 2DEG slows down with increasing temperature. This is characteristic for D'yakonov-Perel'-Kachorovskii (DPK) mechanism, where electron spins dephase by spin precession between momentum scattering events. The DPK mechanism is dominant in the case of high mobility samples with weak momentum scattering. In experiments with an external magnetic field applied in the plane of the sample surfaces (so, in the 2DEG plane, and normal to the optical beams), the optically oriented spins precess about the applied magnetic field at the Larmor frequency (bottom panels in the figure). From the precession frequency the electron g-factor can be calculated. For bulk n-GaAs, the g-factor is nearly constant in the measured range of magnetic fields. For the 2DEG, the g-factor is dependent on the strength of the magnetic field, temperature and the pump-photon energy. Furthermore, we observed a shift of the precession frequency in time (5% increase in 400 ps), and two precession modes (two g-factors) were evidently present in the 2DEG signals. As the g-factor depends on the electron kinetic energy, these results indicate that the signals are influenced by intra-band kinetic energy relaxation. It is not yet known whether the two g-factors result from simultaneously detecting signals from electron populations at two different levels in the quantum well, or from detecting both 2DEG and bulk-electron signals, or both exciton-bound-electron and free-electron signals.

Spin-Dynamics
Ultrafast spin dynamics in ferro- and antiferromagnets

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The manipulating of the electron spin is not only relevant for magnetic storage but may also lead to the development of novel electronic devices with new characteristics (so-called spintronics). Therefore, the investigations of the physical mechanisms underlying the manipulation of electron spin in ferromagnets, semiconductors and hybrid ferromagnet/semiconductor structures constitute at present an exciting area of research. Due to the fact that in antiferromagnets no angular momentum is associated with the order parameter, spin dynamics in these materials is intrinsically much faster than in ferromagnets, expanding the area of spin-dynamics even more [1]. Femto-second laser excitation opens the way to excite magnetic systems on a time scale much shorter than fundamental time scales such as spin-lattice relaxation or precession times. This has already lead to surprising and exciting results like changes in magnetization on a sub-picoscond time scale [2]. Fs laser pulses can also be used to generate short magnetic field pulses that allow coherent control of the magnetization dynamics [3].

Recent progress in this area will be discussed, demonstrating in particular the use of time resolved linear and nonlinear optical methods to investigate the static and dynamic properties of magnetic hybrid structures and the possibility of direct spin manipulation with optical fields [4,5].


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Ultrafast spin dynamics in ferromagnets probed by X-ray spectroscopy

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Ferromagnetic order is one of the simplest and longest known collective phenomenon in solids. It describes the alignment of electron spins along a common direction in space and can easily be accessed by measuring the macroscopic magnetic properties of the sample. As an additional degree of freedom, the spin system stores energy and can be excited by pulsed magnetic fields or by fs laser pulses. Here we report on experiments using a laser pump and x-ray probe scheme to investigate the spin dynamics in a thin ferromagnetic film.

Recent experiments utilized fs laser pulses both as pump to excite the sample and – via magneto-optic Kerr effect – as probe. Several groups observed that the magnetization vanishes on the sub-ps time scale [1,2]. This raises new questions concerning the speed with which it is possible to transfer spin and angular momentum between electrons and the lattice.

As a complementary technique we use x-ray absorption spectroscopy to measure the dynamic response of a thin ferromagnetic film. Utilizing x-ray magnetic circular dichroism (XMCD) we observe the evolution of the magnetic moments as a function of time delay between laser pump and x-ray probe pulse. High sensitivity and the potential of applying sum rules to separately determine spin and orbital momentum of the electrons make XMCD a very powerful technique for our task [3].

Current Synchrotron-based x-ray sources have pulse durations of ~50 ps, limiting the time resolution of our pump-probe experiment. As a first improvement, we used the “low-alpha” operation of the BESSY Synchrotron, which reduces the pulse length down to 5 ps. Even shorter probe pluses can be generated by the slicing technique pioneered at the Advanced Light Source in Berkeley [4] and currently under commissioning at BESSY. First experiments are currently underway using this novel source of fs x-ray pulses.

Magnetization dynamics in ferromagnetic disks and squares

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In recent years the investigation of dynamic eigenmodes in confined magnetic structures has triggered intensive research. The response of micron and sub-micron sized to ultra short magnetic field pulses and the accurate modelling of the spin excitations following a magnetic field pulse is of central interest in advanced magnetic recording technology as switching times are further reduced and pushed well into the gyromagnetic regime. In order to understand the complex trajectories the magnetization vector may take in ultra fast switching experiments we set out with the aim of understanding the excitation spectra of micron sized magnetic elements in the linear excitation regime.

We will report on recent experiments performed using time resolved Kerr microscopy [1]. We investigate micron sized Permalloy disks, which in equilibrium are in the flux closed vortex state. Upon excitation with perpendicular or in-plane magnetic tipping field pulses a complicated dynamic response of the magnetisation evolves. Phase sensitive Fourier transformation allows the identification of several radial and azimuthal eigenmodes, which can be accounted for in a dipolar model. The azimuthal modes in this system show negative dispersion: modes with higher node number are lower in frequency, a behaviour that is reminiscent of magneto-static backward volume modes known to appear in thin magnetic films.

In addition, we analyse not only the mode spectrum of the excited modes, but also their damping behaviour. We have evidence for mode-mode coupling in the studied elements. Mode-mode coupling leads to an apparent increase of the damping parameter.

Recently, some effort has been undertaken to increase the spatial resolution available in time resolved experiments. We will summarize experiments performed at the Swiss Light Source using a time resolved soft x-ray photoemission microscope. First results demonstrate the ability to determine the modal structure of Permalloy squares in the Landau equilibrium magnetization state with high spatial and temporal resolution [2].

Second harmonic generation in NiO – the necessity to go beyond electric dipole approximation

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The discrete intragap states of both the bulk and (001) surface of NiO could be employed for a four-level ultrafast magnetic switching scenario. For this a tool is needed, both to detect the magnetic state of the sample, and to monitor the dynamics of the process. Second harmonic generation (SHG) is a very well suited tool, for it couples linearly to the antiferromagnetic order, is surface sensitive, and can detect all NiO domains.

NiO is a strongly correlated antiferromagnetic solid, which notoriously withstands both density functional approaches within the local density approximation and Hartree-Fock calculations. This material is of relevance for tunnel junctions as well as exchange bias systems and may serve as a “realistic model” system for the ultrafast switching scenario. A viable ab-initio approach to the electronic properties of NiO is based on real space quantum chemistry, the only one that can provide the discrete intragap states. NiO is modeled as a doubly embedded cluster and its intragap and lowest charge transfer energy levels of the bulk and the (001) surface are obtained with two ab-initio quantum chemistry methods. The one consists of the single excitation configuration-interaction technique with energy corrections from higher excitations [CIS(D)], and the other one from the multiconfigurational complete active space method (MC-CAS).

The symmetry analysis of the second order susceptibility tensor is made, and the various tensor elements are computed taking into account electric dipole (ED), magnetic dipole (MD) and electric quadrupole (EQ) transitions, both for the bulk and the (001) surface [1]. Comparison with experiment is performed. This implementation of nonlocality is an indispensable prerequisite for a profound judgment of whether, even in the absence of magnetism, the second harmonic signal results from the bulk (where ED transitions are frequently symmetry-forbidden) or from the surface, and, whether magnetic switching that originates from the magnetic light field is as efficient as a combination of spin-orbit coupling with the electric field under specific circumstances.

Finally, the second order susceptibility tensor is calculated in the presence of the lowest transversal optical phonon branch, within the frozen phonon approximation. Two major effects due to the temporary lowering of the symmetry are shown: the appearance of new SHG peaks even within the ED approximation, and the splitting of the existing ones.

Spin dynamics in metallic ferromagnets at femto- and picosecond time scales

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The study of magnetization dynamics is important for technological applications, since it allows one to determine the maximum working frequency of devices for magnetic storage and information processing in applications such as magnetic random access memories (M-RAM). It is also a critical issue for magnetism since the fundamental time scales involved are not yet completely identified. Spin dynamics can be excited by femtosecond laser pulses, and measured from time resolved magneto-optical effects using a pump-probe experimental set-up. Since the techniques involve optical transitions both for exciting the system and probing its magnetic response, it is always questionable as to what extend optical effects may influence the observed dynamics. Model systems will be considered here with easy magnetization axis is either in plane (nickel films) or out of plane (Co-Pt multilayers or alloys films). Different experiments supporting the conclusion that magneto-optical signals in this context predominantly reflect the spin dynamics will be presented: (i) detailed analysis of the time dependant dielectric tensor[1] , (ii) spectral variation of magneto-optic signals[2], (iii) THz emission related to the ultra-fast demagnetization[3]. Moreover, it will be shown that the absorption of femtosecond pulses induces a change of the effective field experienced by the magnetization, leading to the precession and damping at picosecond time scale[4]. This process will be discussed from real space measurements of the magnetization trajectory.

Spin waves, i.e. collective magnetic excitations, are of high interest for both fundamental as well as applied physics. We have recently shown that spin-polarized electron energy loss spectroscopy offers the unique possibility to study high energy and high wave vector spin wave excitations at surfaces [1, 2]. In our contribution, we present results of spin wave excitations measured on Co-surfaces with this technique. Our measurements show that the spin wave peaks have an intrinsic width (not determined by the spectrometer resolution) in both, the energy and the momentum space. This is expected for such high wave vector spin waves due to the strong damping by Stoner excitations. From our measurements we thus obtain information about the spin wave energy for different wave vectors as well as about the width of these excitations. The strong exchange interaction in the Co leads to high spin wave energies of up to about 400 meV. We can Fourier-transform the results of our measurements obtained in momentum/energy space into real space and time. The result for one particular measurement on an 8 ML Co-film on Cu(001) at a wave vector of 0.81 Å⁻¹ is shown in the figure. The Fourier-transformation was done with a Gaussian peak for which the parameters for energy, wave vector, and width were obtained from the experiment. The graphs show the spin wave amplitudes along the measured direction as a function of space and time. The spin wave is confined in space in about 40 Å which results from the measured width in wave vector space. As time proceeds, the spin wave amplitude decreases due to the non-zero width measured in energy. The oscillation time of the magnetic moments (given by the energy) is about 10 fs. The life time of the spin wave can be estimated to about 30 fs and thus it is only about three times larger than the oscillation time itself. In this short time the center of mass of the spin wave travels only about 15 Å.

The role of charge/spin correlation and electron-phonon interaction in the singles particle excitations in bulk metals and at surfaces

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Interaction between lattice, electron, and spin subsystems as well as interaction within each of these subsystems is crucial to understand mechanism of single-particle excitation dynamics, i.e. lifetime of excitations (LIEX). The LIEX sets the duration of excitation and in combination with the velocity determines the mean free path, a measure of influence of the excitation [1]. Interest to the study of excited particles dynamics is motivated by an important role that excited electrons and holes play in many processes, e.g. in energy, charge, and spin transport in bulk materials, at surfaces, across interfaces, and at nanosystems. In this presentation recently published and new theoretical and experimental results are discussed in terms of elastic and inelastic electron-electron (e-e) interaction for paramagnetic and ferromagnetic materials as well as in terms of electron-phonon (e-ph) interaction. E-ph decay channel is shown to be important for all systems considered being especially important for low-dimensional systems such as free standing monolayers. In the e-e decay the quasiparticle decay can be realized via creation of electron-hole pairs or plasmon excitation. In ferromagnetic systems the electron (hole) decay via the Stoner pair excitation or/and excitation of spin waves is made possible [2]. 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.


Surface Dynamics
Ultrafast electron, spin and lattice dynamics on lanthanide surfaces: Optical excitation of a coherent phonon-magnon mode at Gd(0001)

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The last few years have witnessed significant progress in the understanding of elementary scattering processes of excited electrons at surfaces and interfaces [1]. In particular the advances in femtosecond time-resolved two-photon photoemission (2PPE) spectroscopy provide direct insight into the dynamics of inter- and intraband scattering of electrons in surfaces states at metals, coherent wave packet dynamics as well as electron transfer, localization and solvation processes at adsorbate/solid interfaces. On the other hand, non-linear magneto optics (Second harmonic generation, SHG) provides access to both spin and electron dynamics at surfaces, which can be separated through the symmetry of SHG with respect to magnetization reversal on ferromagnetic samples. In this talk we discuss collective excitations at the rare earth metal surface of Gd(0001), which provides a rich dynamics based on the interplay between electron, spin and lattice and excitations.

Femtosecond electron, spin and lattice dynamics of Gd(0001) are investigated by pump-probe experiments. The electronic structure and optical absorption of the Gd(0001) surface is characterized by a spin split surface state (S₁ and S₂), which leads to resonant enhancement the second harmonic generation at this surface [2]. Upon optical excitation with 800 nm laser pulses the spin polarization of the exchange-split surface state probed by SHG is reduced by half and a coherent phonon-magnon mode at a frequency of 3 THz is observed directly in the time domain [3]. The origin of this mode is attributed to the ultrafast charge redistribution at the surface leading to displacive excitation of the lattice planes normal to the surface and periodic modulation of magnetic coupling.

Time-resolved photoemission experiments give direct access to the underlying electron dynamics and the exchange splitting of the surface state (see figure). We observe a "coherent breathing" (modulation) of the S₁ surface state binding energy mediated by...
the 3 THz lattice vibration. The pronounced drop in the spin polarization is attributed to spin-flip scattering of 5d valence electrons leading to magnon emission among the 4f moments of Gadolinium to conserve angular momentum on an ultrafast time scale. [4]. We will discuss the mechanism of the ultrafast optical induced demagnetization and collective excitation.

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Collective and single-particle dynamics in time-resolved two-photon photoemission

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Time-resolved two-photon photoemission (TR 2PPES) has contributed to a better understanding of ultrafast electronic relaxation processes in metals, surface states and adsorbates. It is based on the pump-probe principle: A pump pulse excites electrons into an intermediate state and a time-delayed probe pulse then maps its decaying population. Energy resolved detection of the photoelectrons reveals the single-particle dynamics, i.e. the lifetime of distinct intermediate states in the unoccupied band structure. This is in contrast to purely optical methods like transient absorption and reflection measurements. These techniques probe directly the collective response of the electrons. On the first view, one might assume that 2PPES and optical spectroscopy yield complementary information, i.e. single-particle dynamics are determined in 2PPES experiments whereas the collective response is seen in optical spectroscopy. This is implicitly assumed in the common interpretation of TR 2PES results. However, as it is demonstrated here, the TR 2PPE signal is also influenced by the collective response of the electrons. The relationship between collective and single-particle dynamics in 2PES is therefore more complex and can alter the quantitative information drawn from 2PPE experiments.

The separation of collective and single-particle dynamics as it is presented here is based i) on a model for the 2PPE process that accounts for collective and single-particles dynamics and ii) on time-resolved 2PPE spectroscopy with interferometric resolution. The interferometric time-resolved 2PPE signal contains information on the local excitation field that is modified by the collective electron response. Hence, our model for the 2PPE process comprises the collective as well as the single particle response of the electrons. The incident light field $E_{\text{ext}}$ interacts with an ensemble of electrons and induces a collective response. The resulting internal field $E_{\text{int}}$ acts on the electrons as a time-dependent perturbation and is therefore responsible for the photoemission. The internal field is described in the frequency domain by the product of a complex response function $G(\nu)$ and the spectral distribution of the incident field. The internal field $E_{\text{int}}$ is responsible for the 2PPE process. This process is phenomenologically modeled using the density matrix propagation for a three-level system. For the excitation via a continuum of intermediate states the TR 2PPE signal is only governed by the inelastic lifetime $T(E)$ of the electrons in the intermediate state.

We use Ag nanoparticles supported on graphite to demonstrate the influence of the collective response on 2PPE and the separation of collective and single particle dynamics in TR 2PPE experiments [1]. The supported metal nanoparticles exhibit a strong collective resonance, i.e. the Mie resonance. The incident pulse spectrum at a photon energy of 3.2 eV is located in the wing of this resonance. The frequency-doubled output of an amplified Ti:sapphire laser system (pulse duration 45 fs, intensity $< 10^9$ W cm$^{-2}$) illuminates the sample. The normalized interferometric two-pulse-correlation for an intermediate state energy of 1.6 eV above $E_F$ is shown in Fig. 1b. The signal exhibits the expected 1:8 signal enhancement that is typical for a two-photon process. The low pass filtered signals for two different intermediate states shown in Fig. 1a exhibit different width and therefore reflect that the lifetime of the intermediate state at 2.8 eV above $E_F$ is smaller than for the state 1.6 eV above $E_F$. To determine the lifetime our model for the 2PPE is implemented into a fitting routine. Nonlinear optimization then yields the parameters for $G(\nu)$ and the intermediate state lifetime $T(E)$. 
The agreement of data and fit is illustrated in the inset in Fig. 1b. Neglecting the collective response leads to a significant increase of the residual (compare Fig. 1d). The comparison of the residual for both cases shown in Fig. 1b and d demonstrates that the internal field correction is essential to reproduce the experimental data. In addition our model contains the lifetime $T(E)$ of an excited electron. This lifetime causes the broadening of the TR 2PPE signal shown in Fig. 1a. A fit of our model to the experimental data yields $T(E)$. The comparison of the electron lifetimes determined with s- and p-polarized excitation confirms the presented strategy to disentangle collective and single particle dynamics in TR 2PPE experiments.

Summarizing, a general model for two-photon photoemission is presented that implements both collective and single particle dynamics. In combination with interferometric time-resolved two-photon photoemission, this model allows to determine the shape of the collective response function across the excitation spectrum and the single particle lifetime of excited electrons. Supported Ag nanoparticles were used as a model system to demonstrate the influence of the collective response in time-resolved 2PPE.

Up and down - electron dynamics at Si(100)

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Carrier dynamics in silicon is of both fundamental and technological importance. Equally relevant as the bulk electronic properties are the dynamics at device interfaces (for an overview see Ref. 1). In regard of continuing miniaturization it is evident that hot electrons will rule device performance and coupling of bulk electrons to surface and interface states will dominate recombination rates (see e.g. [2]).

At semiconductor surfaces electron scattering, trapping and recombination occur on a (sub)-picosecond timescale [1]. Here time-resolved photoemission employing ultrashort laser pulses allows to study carrier dynamics in considerable detail. A first photon excites an electron into an unoccupied intermediate state. The second photon leads to photoemission of the excited electron. Typically, to suppress direct photoemission the photon energies of pump- and probe pulses are chosen below the ionization energy of the semiconductor surface. By controlling the time delay between the two pulses the electron dynamics of the intermediate state can be studied. As in normal photoelectron spectroscopy the state is selected by measuring energy and angle of the emitted electrons. Including the time resolution in two-photon photoemission (2PPE) one obtains momentum- and energy-dependent scattering rates. As the kinetic energy of the emitted photoelectrons is low (~ 1 eV) we can only probe a small fraction of the surface Brillouin zone (SBZ) of ± 0.2 Å.

By now there is general agreement that the lower-coordinated atoms at the Si(100) surface rebond to buckled dimers. The ground state predicted by theory is a c(4 x 2) stacking of dimers ([3] and references therein). This structure is observed below 200 K in low-energy electron diffraction and predominates scanning tunneling topographs [4]. Concomitant to the dimer tilt, the two remaining half-filled dangling-bond states split to form semiconducting surface bands. The 2PPE spectra of Si(100) are dominated by interband transitions between these occupied and unoccupied surface states and emission out of transiently and permanently charged surface defects. At 90 K the occupied D\textsuperscript{up} dangling bond state is located 150 ± 50 meV below the valence band maximum VBM at the center of the surface Brillouin zone. Here the unoccupied D\textsuperscript{Down} band has a local minimum at 650 ± 50 meV above the VBM and shows strong dispersion along the dimer rows of the c(4 x 2) reconstructed surface. At 300 K the D\textsuperscript{Down} position shifts comparable to the Si conduction band minimum but the dispersion of the dangling-bond states is independent of temperature. The surface band bending for p-doped silicon is less than 30 meV, while acceptor type defects cause significant and preparation-dependent band bending on n-doped samples. Note, that these are excellent prerequisites to study carrier dynamics on p-doped Si(100).

In collaboration with Michael Rohlfing (IU Bremen) we have combined this experimental results with many-body perturbation theory to elucidate carrier relaxation and exciton formation in the dangling-bond surface conduction band [5]. Including electron-hole interaction in the many-body calculations of the quasi-particle band structure leads us to assign a dangling bond split-off state to a quasi-one-dimensional surface exciton with a binding energy of 130–meV.
Electrons resonantly excited to the unoccupied D_{Down} dangling-bond band with an excess energy of about 350 meV need 1.5 ± 0.2 ps to scatter via phonon emission to the band bottom and relax within 5 ps with an excited hole in the occupied surface band to form the exciton living for nanoseconds. At room temperature the 2PPE intensity of the surface exciton diminishes indicating faster surface recombination, i.e. phonon mediated scattering of excited electrons and holes into bulk states.

Direct observation of femto- and attosecond electron dynamics in adsorbates

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Ultrafast electron dynamics play a decisive role in processes like photo- and electrochemistry as well as in molecular electronics and single electron devices. To access these ultrafast dynamic processes experimentally, we use the core-hole-clock method employing soft-X-rays from synchrotron radiation sources.

The basis to the core hole clock method is that in polyatomic systems on the timescale of the finite core hole lifetime, different autoionization final states can be reached. In autoionization of a single, free atom, autoionization will only have a single core excited intermediate state. In contrast, in a polyatomic system the excited state created by X-ray absorption can lead due to electronic and/or vibrational coupling to relaxation in the core excited state. Based on the branching ratio of the different autoionization final states due to the dynamic processes in the core excited state, a survival probability of the initially excited state is given as a rate equation. Thus, for an atomic adsorbate with weak next neighbour interaction, two autoionization final states can be observed: a) The adsorbate core excited state remains atomically localized. b) The adsorbate core excited state couples via resonant charge transfer to delocalized (itinerant) substrate states, leading to a charge transfer final state. Hence, the branching ratio of these two autoionization channels directly relates the core hole lifetime to the survival of the atomically localized adsorbate core excited state. Likewise the charge transfer time.

For adsorbed atoms and molecules on surfaces electron transfer processes depend in fine detail on the overlap between adsorbate and substrate states, giving for example sensitivity to the surface projected band structure of the substrate. Furthermore even electron transfer rates between an adsorbed atom and a metal surface in the attosecond range are accessible.

To apply core hole clock spectroscopy to a molecular adsorbate, we need to consider two questions: a) What is the nature of the core excited state, as the symmetry properties of the molecular unit allows molecularly delocalized symmetry adapted core hole excited states, b) What are the possible electronic (CT) and the additional vibrational relaxation processes in the core excited state.

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Excitation and ultrafast dephasing of coherent adsorbate-substrate vibration on metal surfaces

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A detailed understanding of vibrational wavepacket dynamics is vital to design and perform the coherent control of reactions by tailored laser pulses. In comparison with isolated molecules in the gas phase, the wavepacket dynamics of adsorbate on metal surfaces have been rarely explored so far. Recently we have found that fs-laser pulses irradiated on Pt(111) and Cu(111) surfaces covered with submonolayer of alkali metal atoms (Na, K and Cs) induce the coherent motions of adsorbate-substrate stretching vibration [1-3]. Here, we discuss mainly how vibrational wavepackets are created and evolve.

The coherent motions of vibrational wavepackets are monitored by time-resolved second harmonic generation (TRSHG). The clean metal surface at 110 K was dosed with alkali atoms from a well-degassed alkali dispenser (SAES getters). The light sources for fs pulses are a Ti:sapphire regenerative amplifier (800 nm, 150 fs) or a home-built noncollinear optical parametric amplifier (580 nm, 25 fs). We performed pump and probe measurements at a fixed center wavelength of pump pulses. The p-polarized pump and probe beams were focused on the sample surface. The p-polarized component of the SH of the probe pulse was detected. An optical chopper was inserted in the optical path of the pump beam, and the intensity modulation in the probe SH induced by the pump pulse was detected by a lock-in amplifier at various pump-probe delays. The sample temperature was kept at 110 K during the measurements.

Typical TRSHG traces from the Cs-covered Pt(111) surface show clear oscillation with the frequency of 2.2~2.4 THz due to coherent vibrational motions excited by pump pulses at 580 nm. We found that there are two oscillating components in the TRSHG traces, which are attributed to Cs-Pt stretching vibration and surface phonon of Pt. The damping of the oscillations in the TRSHG traces is mainly due to dephasing of the coherent motions. We found that the damping is accelerated as the pump fluence increases. This is attributed to the detuning by anharmonic coupling with lateral vibrational modes excited by substrate hot electron scattering. [2]

The intensity of SHG signals is significantly increased by alkali atom adsorption and shows some "resonance" features at a fixed wavelength of pump pulses when the SH intensity is plotted as a function of alkali-metal coverage. The "resonance" features are attributed to the resonant transitions between electronic states involved in SHG. The oscillation amplitude does not necessarily follow the intensity alteration of SHG, but appears only in a limited coverage range. In addition, the initial phase for the Cs-Pt stretching mode falls in the range from -170 to -190 degrees. These facts indicate that not all electronic excitations induce the coherent vibrational motions, but some resonant electronic excitations that displace the position of the alkali atom are responsible.

In order to explore what electronic excitation is responsible for the creation of the coherent vibrations, we measured the photon energy dependence of TRSHG traces from a Na-covered Cu(111) surface, since the electronic structure has been well characterized in this adsorption system by two photon photoemission.[4] The photon energy of the pump pulse were varied from 2.0 to 2.5 eV, while the photon energy of the probe pulse was kept constant at 2.1 eV. Although the modulation amplitude of the adsorption system was much smaller than those observed in the alkali-covered Pt(111) surfaces, they show a clear excitation photon energy dependence. At 2.0 eV, no modulations were observed in the TRSHG traces. However, above 2.15 eV the modulation...
amplitudes increase monotonically. According to the two photon photoemission study, the resonant transitions take place from the alkali-induced occupied state to the first image state at 2.15 eV and to the second image state at 2.60 eV. Therefore, these resonant transitions are likely responsible for the creation of the coherent vibrational motions.

Time-resolved coherent photoelectron spectroscopy of image-potential states

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Time-resolved two-photon photoemission (2PPE) has proven to be an extremely powerful technique for studying ultrafast dynamics at surfaces. It gives direct access to all relevant quantum states of excited electrons at surfaces (energy, spin and parallel momentum) and allows monitoring their temporal evolution with femtosecond time resolution. Whereas most 2PPE experiments concentrate on measuring the population of excited states and studying incoherent dynamics, 2PPE also offers unique possibilities in accessing the coherent regime of electron dynamics. I will discuss several schemes that have been developed for the latter purpose and illustrate them with experiments involving image-potential states on metal surfaces [1, 2].

In terms of the experiment, the simplest procedure to deduce information about the decoherence of electronic excitations is to compare the linewidths of 2PPE spectra with their intensities as function of pump-probe delay. Another, more direct way, is interferometric 2PPE developed by Petek and co-workers [3]. There, the photoemission probe pulse has a well-defined phase relationship with respect to the pump pulse. In the case of image-potential states it has also been demonstrated that the coherent excitation of several quantum states with one short pump pulse results in the creation of electron wavepackets that resemble the quasiclassical motion of electrons at surfaces [4]. Quantum-beat spectroscopy allowed the resolution of high-order states of the Rydberg series. The dephasing of these quantum beats is mainly due to quasielastic electron scattering processes at steps and defects.

A very recent attempt to access the coherent regime of electron dynamics at surfaces is the generation of a coherent surface current and its detection with 2PPE [5]. In this experiment two femtosecond phase-locked pump pulses with frequencies $\omega$ and $2\omega$ generate a population of excited states which is asymmetric with respect to the directions +k\|$ and -k\|$. In case of the n=1 image-potential state of Cu(100) the electrons excited in this way move with parallel velocities of ~1 Å/fs and carry a net current of up to 10$^7$ A/cm$^2$. The direction of this coherent dc-current can be controlled by the relative phase between the two excitation pulses. The current is detected by time-delayed probe pulses as an anisotropic angle-resolved photoemission yield. This type of detection of coherent currents is not restricted to surface currents. It should also be applicable to coherent currents generated, e.g., in bulk semiconductors and 2PPE should also allow their detection with high sensitivity and temporal resolution.

Poster I
Ultrafast X-ray Measurements at the Sub-Picosecond Pulse Source

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The development of new X-ray sources like free electron lasers, 4th generation synchrotrons and laser driven plasmas will provide new opportunities for producing short pulses with short wavelength radiation. These ultrashort X-ray pulses offer a unique combination of spatial and temporal resolution and thus make it possible to directly observe atomic motion, for example during chemical reactions or phase transitions. Using the Sub-Picosecond Pulse Source in Stanford, USA the collaboration has performed first “optical pump — X-ray probe experiments” on a fs-timescale. The SPPS is the first linear accelerator based femtosecond multi keV X-ray source and stands in the forefront for research, in particular to conduct important R&D work for accelerator concepts, X-ray optics, and experimental techniques for future X-ray FEL experiments. In this contribution we will give an overview of the activities at the SPPS.

We will discuss results on the extremely fast structural changes associated with non-thermal melting of semiconductors after short pulse laser excitation [1]. First results for the detection of optical coherent phonons in laser excited Bismuth will be presented [2]. Beyond the physics of ultrafast melting we discuss the crucial question how to time a linac based source with an optical pump laser [3]. A third time-resolved experiment is set-up to observe the ionic to neutral phase transition in the typical mixed stack charge transfer compound TTF-CA. We show the first preliminary static results from the SPPS and discuss them in the context of the time resolved data from the ESRF, ID09 with 50 ps time resolution [4].

Time resolved Raman spectroscopy

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Optically induced phase transitions, non-thermal processes, and ultrafast optical switching behavior have become, over the past few years, topics of major interest in the condensed matter physics society. To study these phenomena, a variety of ultrafast techniques have been employed over the years, including time resolved reflectivity and transmission experiments and magneto-optical Kerr spectroscopy. These techniques give insight into the electronic and/or charge properties, but usually lack information on structural properties. An exception to this is coherent phonon generation. To address the ultrafast structural properties, there are a number of groups concentrating on the development of time resolved x-ray or electron spectroscopy and diffraction at synchrotrons or XFEL facilities and even by using table top laser systems. This contribution focuses on a promising and powerful alternative to time resolved diffraction techniques in studies on ultrafast structural transitions: Time Resolved Raman Spectroscopy (TRRAS). Apart from the simultaneous study of both the frequency and time domain, TRRAS offers the possibility of monitoring the local temperature, thus providing an unique tool to study non-thermal processes.
Coherent stationary waves in permalloy nano-dots induced by non-adiabatic lattice heating

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Arrays of magnetic dots are receiving a growing interest [1-7] as they form important candidates to high density storage devices. From a fundamental point of view, they constitute a unique tool to study magnetic properties and magnetic effects in low dimensional systems. In particular magneto-optical and microscopy techniques have shown the existence of single-domain and vortex phases on the nanometer scale [8, 9]. Moreover, nanometer-sized metal structures have been recently investigated as model for antennas in the optical region [10, 11].

On the other hand arrays of nano-structures are very promising models to investigate mechanical and thermodynamical properties when the typical size of the system is reduced to the nanoscale [12]. In particular cylindrical dots, in thermal contact with the substrate, are very interesting for the study of dynamical heat exchange at the nanometric interface.

In this work we apply time-resolved optical techniques to investigate the dynamical properties of an array of permalloy nano-dots, excited by femtosecond laser pulses. In fact, when the nano-dots are excited by ultrashort pulses, non-adiabatic lattice heating is induced. As a consequence stationary longitudinal and radial sound waves are generated on the 100ps timescale. The oscillation of the radial and length parameters in the sub-angstrom scale results in a modulation of the reflectivity on the first-order of the diffraction pattern.

In Fig. 1 the reflectivity variation as a function of the delay between the pump and probe pulses is reported. The pump energy used in the experiment is about 10nJ/pulse, with a spot diameter of about 40μm and a pulse timewidth of 120 fs. In the inset of Fig. 1 the AFM image of the sample is shown. The mean radius of the dots is 400nm, whereas the period of the lattice is 800nm.

The dot oscillation can be modeled by an harmonic oscillator equation, where the non-adiabatic lattice strain is the source of the oscillation and the equilibrium position is depending on the lattice temperature. The solution of the equation is given by:

\[ x(t) = \frac{c\Delta T(0)\omega_0^2}{\omega_0^2 + \frac{1}{\tau^2} - \frac{2\gamma}{\tau}} \left( e^{-t/\tau} - e^{-\gamma t} \cos \omega t + \frac{\alpha}{\omega} \sin \omega t \right) \]
where $c$ is a constant, $\Delta T_l(0)$ is the lattice temperature at $t=0$, $\omega_0$ is the normal-mode frequency, $\gamma$ is the oscillation damping, $\tau$ is the relaxation time of the dot temperature, $\sigma=(\omega_0^2 - \gamma^2)^{1/2}$ and $\alpha=1/\tau - \gamma$. 

By fitting the data with this function (see solid line in Fig. 1) it is thus possible to estimate both the damping of the oscillation and the time constant of the heat-exchange process between the dot and the substrate. In addition the oscillation frequency is related to the sound velocity in the permalloy. At this light we are able to directly measure mechanical and thermodynamical properties of the system, without perturbing it with an external probe in mechanical contact with the sample. This approach is very promising for the study of the thermodynamical properties of nano-sized systems across a phase transition. In this case the discontinuity in the specific heat can be studied as a function of the size of the system and of the coupling between the dots and the substrate.

Coherent optical phonons in graphite and diamond

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Coherent optical phonons, which are impulsively excited with short optical pulses in semiconductors and semimetals, not only reveal the lattice dynamics, but also the interaction between the phonons and the electronic system on femtosecond timescale. Graphite and diamond, although both polymorphs of carbon, have quite different character (semimetal and insulator), and thus are model systems for investigation of the effect of dimension to the lattice dynamics. Optical phonons of carbon materials have been studied extensively in frequency domain because it carries detailed information on crystallinity, periodicity, local bonds, etc.

In this study, we present femtosecond dynamics of the coherent optical phonons of highly oriented pyrolytic graphite (HOPG) and synthetic single crystal diamond excited with 10 fs laser pulses with center wavelength of 400 nm. Anisotropic reflectivity change is measured using electro-optic sampling scheme, while the delay between pump and probe pulses are scanned at a rate of 20 Hz. The in-plane (E2g2) phonon of graphite is well fitted with a chirped damped-harmonic function. The frequency obtained from the time-domain fitting shows an upshift and a negative phonon frequency chirp with increasing pump power. This observation, is opposite to the downshift of the same phonon upon heating, [1] but has the same sign as the shift observed on intercalation with hole dopants.[2] We tentatively attribute our observation due to transient injection of holes, which stiffen the in-plane C--C bonds. We also observed the lattice dynamics of graphite after He ion irradiation. Although we observe the disordered-induced (D) peak around 1350cm⁻¹ in Raman scattering spectrum, no trace of the disordered-induced mode is seen in the time domain. Yet the frequency E2g2 phonon shows a clear downshift by creation of point defects both in the time domain and in Raman spectrum. Our result supports that the appearance of the D peak includes incoherent process such as double resonant Raman scattering, as is suggested by recent theoretical and experimental studies double resonant Raman scattering.[3]

The optical phonon of diamond has a long lifetime significantly exceeding 10 ps. The amplitude of the coherent phonon is found to be linear with the pump power, in spite that the excitation energy is less than half of the direct band gap. The synthetic diamond used has isolated N atom impurities, which has as absorption band between 400 and 500 nm. Our observations suggest that the electronic excitation via the impurity level contributes to the resonant stimulated Raman process to generate coherent optical phonons in diamond.

Ultrafast dynamics of soft coherent phonon in ferroelectric Pb$_{1-x}$Ge$_x$Te

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The understanding of structural phase transitions in ferroelectric materials is an important issue because of its wide applications in modern rewritable memory media and other electro-optic devices. [1] Moreover, control of structural phase transition in solid in real time scale using ultrashort laser pulses represents a great challenge for creating new crystal phase via nonthermal pathway, where the atom moves to a quasi-equilibrium position in the unit cell.

Structural phase transitions in ferroelectric materials have been investigated extensively by monitoring a so-called ‘soft mode’, i.e., the lowest frequency transverse optical phonon at the wavevector $k=0$, using frequency domain technique, such as Raman scattering and neutron scattering.[2] However, the study of critical dynamics of structural phase transition near the phase transition temperature ($T_c$) is difficult because the soft mode frequency drastically reduced toward zero frequency, if the transition is a second-order. Dougherty et al. has examined the femtosecond dynamics of ‘soft mode’ structural phase transitions in perovskite, and revealed that the soft mode became heavily damped oscillation below $T_c$. [3]

Ferroelectric Pb$_{1-x}$Ge$_x$Te is a narrow band-gap semiconductor. It exhibits structural phase change from rocksalt structure above $T_c$ to rhombohedral structure below $T_c$, and $T_c$ depends on the composition “x” [4, 5]. The A$_1$ mode is a soft mode in Pb$_{1-x}$Ge$_x$Te, which is responsible for the structural phase transition. The Raman data [4] showed 23 cm$^{-1}$ (0.7 THz) peak frequency with 10 cm$^{-1}$ (0.3 THz) bandwidth at 7 K, which predicts that the soft phonon is heavily damped, even far below the $T_c$. We have performed pump-probe reflectivity measurements on a single crystal of Pb$_{1-x}$Ge$_x$Te ($x = 0.07$, $T_c \sim 150 \pm 10$ K) using 800nm optical pulses with a pulse duration of $\sim$140 fs. The pump fluence was reduced to 0.59 mJ/cm$^2$ in order to minimize heating effect, while the probe fluence was kept at 0.06 mJ/cm$^2$. The sample was mounted inside a closed-type cryostat. The transient reflectivity change ($\Delta R/R$) was recorded as a function of the time delay.

Figure1 shows the transient reflectivity obtained at various temperatures. The transient signal arising at time delay zero is the electronic response, and the dip feature (denoted by arrows in Fig.1) followed by the adjacent monocycle oscillation is the contribution from the coherent soft phonon. Such the over-damped oscillation makes the Fourier-transform analysis difficult. With increasing the temperature, the coherent phonon signal gradually decreases till 160 K. The present result suggests $T_c = 150$ K in Pb$_{1-x}$Ge$_x$Te when $x = 0.07$ [5]. Another feature of the data is a sudden decrease of the electronic response around 150 K. We also observed the sudden decrease of the electronic response at 150 K in another Pb$_{1-x}$Ge$_x$Te ($x \sim 0.2$) sample and the coherent soft phonon still maintain above 160 K. Therefore, the sudden decrease of the electronic response would be due to the pseudo-gap existing at 150 K for such kind of narrow band semiconductor. The inset in Fig. 1 is the temperature dependence of the frequency squared of the coherent phonon obtained from the fitting by using a damped harmonic function plus an exponential decay. We display both the present data and the Raman data derived from [4]. The time-domain frequency is close to the Raman data at low temperature, but time-domain frequency could reach much lower value than that of Raman frequency. As the temperature increases, the frequency of the A$_1$ phonon downshifts close to zero around $T_c$. Such continuous reduction of the frequency indicates a second order phase character for
Pb$_{1-x}$Ge$_x$Te. The solid lines are the fitting with Landau mean field theory $f(T) = A(T_c - T)^b$. Here, $T_c = 120$, $b = 1$ for the present study and $T_c = 150$, $b = 1$ for the Raman data are obtained. The lower $T_c$ by ~30 K for the time domain data could be due to the thermal effect from the pump laser.

In summary, ‘soft mode’ dynamics was investigated through time-resolved coherent phonon spectroscopy in ferroelectric Pb$_{1-x}$Ge$_x$Te. The coherent $A_1$ phonon damped rapidly and vanishes above $T=160$ K. The frequency of the coherent $A_1$ mode gradually red-shifted toward zero value when the temperature approaches $T_c$, which confirmed that Pb$_{1-x}$Ge$_x$Te undergoes a second order displacive-type phase transition.


![Fig. 1. Transient reflectivity at various temperatures. The inset shows the frequency squared of the coherent soft phonon. Solid circles represent the present study and open circles are Raman data [4]. The solid lines are fittings with $f(T) = A(T_c - T)^b$ and the dashed line is the extension of fitting trace of Raman data.](image)
Coupled decay of impulsively excited phonon-pair combination states in KTaO$_3$

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Impulsive second-order Raman scattering processes provide the possibility to induce macroscopically detectable dynamics of statistical crystal lattice properties in solid state. First theoretical investigations by Xu and Nori predicted vacuum squeezing of impulsively excited phonon states [1] and stimulated the experimental observation of this effect by Garret et al. in a femtosecond pump-probe study [2]. The key feature of the vacuum squeezing effect is the suppression of the phonon amplitude noise, i.e. of the variance of the atomic displacements, to below the vacuum limit temporarily within a phonon oscillation period. Beyond these investigations, Bartels et al. showed that covariances of phonon-pair combination state amplitudes can also be detected in such an experiment [3]. They appear as oscillations at the sum- and difference-frequencies of the contributing phonon states in spectrally and time-resolved experiments.

Both groups have chosen KTaO$_3$ as the sample material because for symmetry considerations only second-order Raman scattering is allowed and first-order Raman processes are ruled out. The investigations focussed on the identification of the excitation and detection processes so that no distinct analysis of the phonon-pair combination states decay processes has been carried out so far. By means of a temperature dependent analysis we show that impulsively excited phonon-pair combination states decay in a correlated process. Their coupled decay is theoretically described by a three-phonon interaction model which results in a strongly increased lifetime of the difference-frequency combination states as compared to the sum-frequency combination states at low temperatures.

The interaction of two phonon eigenstates in thermal equilibrium – represented by harmonic oscillators with amplitudes $Q_1$, $Q_2$ and eigenfrequencies $\omega_1$ und $\omega_2$ – with an ultrashort laser pulse via second-order Raman scattering results in the impulsive excitation of correlated phonon-pair combination states. Here, sum frequency states (at frequencies $\omega_1 + \omega_2$) arise from the simultaneous creation or annihilation of two phonons while difference frequency states (at frequencies $\omega_1 - \omega_2$) are the result of the simultaneous phonon creation and annihilation. Consequently, their amplitudes are

$$A^+ = C \cdot (1 + \langle n_1 \rangle + \langle n_2 \rangle) \quad \text{and} \quad A^- = C \cdot (\langle n_1 \rangle - \langle n_2 \rangle)$$

respectively, where $n_i$ are the occupation numbers of the harmonic oscillators in thermal equilibrium prior to the optical excitation (for details, see Ref.3).

To describe the coupled decay of the impulsively excited phonon-pair combinations we now introduce a three-phonon interaction model by expanding the Hamiltonian of the crystal lattice up to the first anharmonic term. This is cubic in the harmonic oscillator displacements $Q_i$ and thus allows for phonon-phonon interactions – in contrast to the harmonic crystal lattice Hamiltonian. Taking into account possible three-phonon interaction processes and both, energy and wavevector, conservation we obtain the following probabilities for the decay of sum- and difference-frequency phonon combination states which again depend on the phonon occupation numbers [4]:

$$P^+ \propto (\langle n_1 \rangle + 1) \cdot (\langle n_2 \rangle + 1) - \langle n_1 \rangle \langle n_2 \rangle = \langle n_1 \rangle + \langle n_2 \rangle + 1 \quad \text{and} \quad P^- \propto \langle n_1 \rangle \cdot (\langle n_2 \rangle + 1) - \langle n_1 \rangle \langle n_2 \rangle + 1 = \langle n_1 \rangle - \langle n_2 \rangle$$
The lifetimes of the phonon-pair combination states are then defined as the inverse of these probabilities. To verify the above models of generation and coupled decay, we perform a temperature-dependent femtosecond pump-probe study on a (100)-oriented KTaO$_3$ single crystal. The laser source is a home-built 300 MHz repetition rate Ti:sapphire oscillator which delivers pulses of 25 fs lengths to the sample. For the observation of phonon-pair combination states we employ a grating monochromator in a spectrally-resolved detection scheme. 

Time-domain signals exhibit a beating of several oscillation components which – after Fourier transformation and by a comparison to a second-order Raman spectrum of KTaO$_3$ – can be identified to result from the excitation of distinct phonon-pair combination states. The amplitudes and lifetimes of these states cannot be extracted directly from the Fourier transforms because the shape and linewidths of the Fourier peaks are mainly determined by the combined density-of-states of the involved phonons. Therefore, we use the method of short-time Fourier transformation (STFT) to unambiguously determine amplitudes and lifetimes from the transient dynamics. For clarity, we focus here on the TO$_4$ ± TA phonon-pair combination states at frequencies of 13.5 and 16.5 THz, respectively.

Normalized amplitudes obtained by applying the STFT-method to time-domain signals at temperatures between 10 and 320 K are shown in Fig.1 (a). At low temperatures, the TO$_4$ – TA difference-frequency state disappears because the thermal phonon occupation numbers $n_1$ and $n_2$ vanish for $T \to 0$ K. The experimental data of sum- and difference-frequency states are in excellent agreement with the theoretical values (dashed line) calculated from Eq. (1).

In addition, Fig.1 (b) depicts the temperature-dependent lifetimes of the TO$_4$ ± TA phonon-pair states which are also obtained from the STFT-approach. Obviously there is a strong difference in the lifetimes of the sum- and the difference-frequency states, especially at low temperatures. This observation establishes that the decay of the phonon-pair combination state is dominated by a coupled process and not by the population decay of one of the contributing phonon states. Moreover, the temperature dependence follows the calculated values of the introduced three-phonon interaction model throughout the whole temperature range thus demonstrating its validity to describe the coupled decay of the phonon-pair combinations.

Generation of $K_{\alpha}$-radiation from a tape target by a femtosecond Ti:Sapphire laser at 10 kHz repetition rate

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A compact, laser-based hard x-ray source at a repetition rate of up to 10 kHz is presented. The x-ray radiation is efficiently generated by focusing an ultrashort laser pulse of about 30 fs pulse duration and 1 mJ pulse energy onto a metal containing tape target. Here we report on results for different tape targets and repetition rates up to 10 kHz. We also analyze the keV x-ray emission and demonstrate the monochromatic focusing with a toroidally bent crystal.

An oscillator-amplifier system consisting of a cavity-dumped Ti:Sapphire oscillator which generates seed pulses at a variable repetition rate between 1 kHz and 4 MHz and a 9-pass CPA Ti:sapphire amplifier has been developed. To minimize thermal problems resulting from the high pump powers applied a liquid nitrogen cooled Ti:Al$_2$O$_3$ laser crystal is used in the amplifier. After a prism compressor the beam has a pulse duration of 28 fs and a pulse energy of 1.1 mJ at a repetition rate of 10 kHz, which can be varied between 1 and 15 kHz. The laser beam $M^2$ factor is measured to $M^2 = 2$. Varying the pulse energy of the laser from 0.1 to 0.8 mJ focal intensities in the range $5 \times 10^{14}$ to $5 \times 10^{15}$ W/cm$^2$ are generated.

The target consisting of commercial chrome and iron audio cassette tapes is moved at high speed, ensuring that every laser pulse is applied to a fresh surface on the target. The tapes used for these experiments contain either chrome or iron, enabling the generation of Cr$K_{\alpha}$ and K$\beta$ radiation at $h\nu = 5.41$ keV and 5.95 keV, respectively, as well as Fe$K_{\alpha}$ and K$\beta$ radiation at $h\nu = 6.40$ keV and 7.06 keV, respectively. The generated x-ray radiation is detected by a thermoelectrically cooled Si pin diode. With this diode x-ray spectra are taken at a repetition rate of 1 kHz to 10 kHz showing characteristic line emission as well as a bremsstrahlung continuum. At 10 kHz repetition rate about $6 \times 10^9$ Fe$K_{\alpha}$ photons/s are generated. No significant spectral differences are observed in the spectra for the different repetition rates.

For the focusing of the x-rays a toroidally bent Si(311) crystal is implemented, intended for the monochromatic reflection of Cr$K_{\alpha 1}$ radiation at $h\nu = 5414.72$ eV. The crystal is bent for a 1:1 imaging of the x-ray source generating a focus on a thermoelectrically cooled, front illuminated deep depletion CCD. Moving the camera along the beam path the parameters of the x-ray beam are determined. By turning the crystal the angle of incidence of the x-ray radiation varies and therefore the reflected wavelength changes. The set-up can therefore also be used as a monochromator and the spectral distribution of the Cr$K_{\alpha}$ emission has been measured. With the resolution of this monochromator the spin-orbit splitting of the Cr 2p$1/2,3/2$ levels can be resolved and the Cr$K_{\alpha 1}$ line at 5414.72 eV and the Cr$K_{\alpha 2}$ line at 5405.51 eV appear as separated lines with half widths (FWHM) of 4.05 and 3.88 eV, respectively, see Figure.
Poster II
Ultrafast studies of carrier cooling in CdSe quantum dots

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Semiconductor quantum dots (QDs) have uniquely tunable optical properties, offering novel possibilities in optoelectronics, such as light emitting diodes, solar cells and nanocrystal lasers. One of the key aspects underlying the application of QDs to e.g. solar cells is the step immediately following photo-excitation: the cooling of the photo-generated hot electron and hot hole to their lowest energy states. Carrier cooling in QD’s has thought to be slow, limited by a ‘phonon bottleneck’ [1], as the discrete energy levels (similar to those in a hydrogen atom – see figure) imply that energy transfer from the carriers to the lattice must occur in discrete steps, requiring the simultaneous emission of a large quantity of phonons, a process of low probability.

We have investigated carrier cooling in CdSe QDs using Terahertz time-domain spectroscopy and time-resolved (femtosecond) fluorescence spectroscopy. This unprecedented combination of techniques allows us to unambiguously determine the cooling rate for holes and electrons independently for the first time. We conclude that cooling proceeds without a phonon bottleneck for both holes and electrons, present strong evidence of the involvement of Auger processes in the cooling mechanism (which circumvents any phonon bottleneck), and determine electron-hole coupling times in QDs.

Ultrafast Electron Dynamics in C₆F₆/Cu(111)

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Ultrafast charge transfer processes in molecules adsorbed on metal surfaces can be investigated by Auger-resonant-Raman (ARR) [1] and femtosecond time-resolved two-photon-photoemission (2PPE) [2] spectroscopy.

In ARR spectroscopy a localized core level electron is resonantly excited into a bound, unoccupied molecular resonance by absorption of an x-ray photon. The decay of the remaining core-excited state takes place on a femtosecond timescale and serves as internal clock for the charge transfer process into the metal substrate. At the time of the Auger-like decay process the excited electron can either be still localized in the molecular resonance or may have already relaxed into the substrate fill the core hole. This gives rise to two distinctively different decay channels, termed coherent Auger-Raman and incoherent Auger, respectively. Electron autoionization spectra show that these two decay channels are separated in energy by the so-called spectator shift. The electron in the molecular resonance will contribute to the shielding of the two hole final state via Coulomb interaction, resulting in larger kinetic energies of the emitted Auger electrons. By tuning the photon energy across the molecular resonance the coherent Auger-Raman channel is distinguishable from the incoherent Auger channel due to the linear dispersion of its kinetic energy with the photon energy. The charge transfer times are extracted from the intensity ratio of coherent to incoherent decay channels using a rate equation model.

In 2PPE spectroscopy the charge transfer is investigated directly in the time domain utilizing the well-known pump-probe technique. In this case delocalized electrons from the metal substrate are photo-injected into the unoccupied molecular resonance using a femtosecond UV laser pulse. A second, time-delayed laser pulse in the visible spectral range probes the temporal evolution of the excited state electron population. The second pulse lifts the electrons above the vacuum level where they are detected in an electron spectrometer. The resulting electron population of the molecular resonance as a function of the pump-probe delay is analyzed within a rate equation model yielding the charge transfer times.

To systematically compare both approaches we investigated the system C₆F₆/Cu(111) serving as a model system for organic/metal interfaces. The charge transfer times of the lowest unoccupied molecular resonance are summarized as a function of C₆F₆ coverage in the figure below. This work reveals a qualitatively different coverage dependence of the decay rate of the molecular resonance. In the 2PPE measurements the charge transfer times increases from 7 fs for 1 ML up to 37 fs above 3 ML in agreement with previous studies [3]. In contrast, the decay times in the ARR measurements are almost constant ranging from 1.47 fs for 1ML to 2.30 fs for 10 ML coverage. In addition the decay rate for monolayer coverages measured by ARR is larger by a factor of five compared to 2PPE.

These findings are attributed to the fact that ARR spectroscopy detects intra-molecular charge delocalization as well as the delocalization between the excited molecule and its environment. The almost constant charge transfer time therefore points to a relaxation (delocalization) process within the molecule or in the adlayer itself, which cannot strongly depend on the coupling to the substrate. The careful analysis of the time-depend 2PPE data revealed an intraband scattering process within the delocalized state as well as the population decay by interband scattering to the substrate [4]. The relatively large variation of the decay time of the lowest unoccupied resonance can be rationalized
by the change of the wave function overlap of the excited state electron with the underlying substrate [3].

We therefore conclude that - in the case of organic molecules containing several internal degrees of freedom - the two techniques measure different relaxation channels. In the case of C₆F₆/Cu(111) ARR detects the ultrafast intra-molecular relaxation on a time scale of a few fs. In contrast, 2PPE is sensitive to the (slower) charge transfer into the metal substrate via interband scattering. The project was funded by the DFG through SPP 1093.

Fig. 1. Coverage dependence of the charge transfer times of the lowest unoccupied molecular resonance in C₆F₆/Cu(111) analyzed with Auger-resonant-Raman (ARR) and two-photon photoemission (2PPE) spectroscopy. The solid lines guide the eye. The inset shows a sketch of the different excitation schemes for ARR and 2PPE spectroscopy.

Time Resolved THz Spectroscopy on Graphite and Carbon Nanotubes

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Carbon nanotubes are quasi-one dimensional structures obtained by folding graphene along a defined wrapping vector. Accordingly to their symmetry, they can be classified as large gap, small gap or gapless. The band picture describes qualitatively the electronic structure but cannot account for several optical and transport properties. Notably, the photogenerated electron-hole pairs of semiconducting tubes form excitons with large binding energy [1]. Moreover, the conducting electrons of small gaps or metallic tube are easily localized by defects or impurities [2]. New insights on these issues can be obtained by means of Time Resolved THz Spectroscopy.

This technique monitors the change of mid-infrared dielectric function after absorption of a pump pulse. It has been fruitfully employed to characterize electron-hole plasmas [3] and excitons [4] in semiconductors and is here applied for the first time to carbon nanotubes. The experiment shows that: i) the lack of free carriers response in large gap tubes is in agreement with excitons generation ii) delocalized electrons in small gap tubes decrease the THz absorption iii) localized excitations generate a weak signal that decays on the time-scale of the electronic thermalization.

Spin-dependent electron dynamics in front of an ultrathin ferromagnetic film

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Recent experiments demonstrate that significant demagnetisation of ferromagnetic thin films upon laser excitation can be achieved within a few hundred femtoseconds (e.g. [1]). Within this time scale the excited electronic system and the underlying lattice are not in equilibrium and it seems that the transient hot electron population is responsible for the change of the magnetisation. Which microscopic processes lead to the loss of magnetization remains controversial (e.g. [2]).

Our approach to tackle this question is to directly aim to find possible decay channels for demagnetization processes by studying the electron dynamics in a ferromagnet spin-dependently. As a model system we employ image-potential states in front of ultrathin Fe films on Cu(001). These unoccupied states are best analysed by two-photon photoemission (2PPE) (for a review on the decay of electronic excitations at metal surfaces and an introduction to 2PPE see [3]). An electron excited by a first laser pulse is trapped in the image potential in front of the surface. The evolution of the transient population can be sampled by a second time-delayed probe pulse. This allows to quantify inelastic scattering processes which always lead to a decay of the population. Quasielastic scattering events in contrast may only destroy the phase coherence. For the welldefined image-potential states these dephasing rates can be identified by analyzing the delaydependent linewidth [4]. A spin-sensitive electron (SP-LEED) detector enables us to distinguish between majority and minority electrons. We were able to observe the exchange-splitting of the first and the second image-potential state directly. Figure 1 shows energy-resolved 2PPE spectra at zero time delay between pump and probe pulse, spin integrated and spin resolved; the exchange-splitting between the majority and minority components is clearly discernible. A schematic representation of bichromatic 2PPE is shown in the inset.
In time- and spin-resolved measurements, spin-dependent lifetimes were found and a linewidth analysis of the energy-resolved 2PPE-spectra revealed also spin-dependent dephasing, which indicates spin-dependent elastic scattering processes. A scattering process which might very well stand behind these quasielastic scattering processes is magnon emission and absorption.

Time-Resolved Five-Wave Mixing at Silicon Surfaces and Interfaces

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Optical techniques have been used successfully to obtain important information on the dynamics of excited states in condensed matter systems. In particular, four-wave mixing (4WM) and pump-probe spectroscopy are well established for investigating the coherent dynamics of bulk semiconductors. Compared to bulk and quantum well structures, the electron dynamics at semiconductor surfaces and interfaces are not well understood. At present, most of the experimental information in this field has been obtained by means of time-resolved two-photon photoemission.

In this contribution, a complementary and purely optical technique is presented. It represents a synthesis of four-wave mixing and optical second harmonic generation (SHG). The resulting five-wave mixing process (5WM) may be viewed as scattering of SHG from a transient excitation grating which has been generated by two laser beams. It is characterised by a $\chi(4)$ tensor. Like the $\chi(2)$ process SHG it is dipole-forbidden in the bulk of centrosymmetric materials, that makes this technique surface- or interface-specific. Depending on the particular setup, it can be used to monitor the temporal evolution of photoexcited polarizations and populations at surfaces and interfaces.

To demonstrate this technique, experimental results on the temporal dynamics of the diffracted second harmonic intensity at silicon surfaces are shown. Ultrashort laser pulses produced by a cavity-dumped Ti:sapphire laser are used to excite the dangling bond states of Si(001) and Si(111) surfaces. At both surfaces, the self-diffracted signal induced by two ultrashort laser pulses shows a rapid coherence decay on a timescale comparable to the duration of the incident pulses.
The Si(001)c(4x2) surface is studied using a five-wave mixing set-up which measures the diffracted second-harmonic intensity induced by three femtosecond laser beams. By varying individual time delays between the pulses, this technique can not only be used to investigate the dynamics of one-photon transitions, but allows although one to monitor the temporal evolution of populations. With the support of model calculations based on optical Bloch equations, the observed ultrafast response as a function of particular delay can be assigned to scattering of the excited electrons within the $D_{down}$ surface band on a timescale of 50-500 fs. These times are consistent with two-photon photoemission experiments at silicon surfaces, where scattering leads to intraband relaxation of excited populations within a few 100 fs.

Due to the purely optical character of the 5WM setup, this technique opens the opportunity to study the largely unexplored field of electron dynamics of buried semiconductor interfaces. The model system of our choice, CaF$_2$/Si(111), is introduced.

We have studied the diffusion of atomic oxygen on a vicinal Pt(111) surface induced electronically by femtosecond laser pulses. The dissociative adsorption of O2 on a vicinal Pt(111) surface was used to selectively decorate the step sites with atomic oxygen. Diffusion was induced from the steps onto the terraces at a substrate temperature of 80K by applying 50-fs, 800-nm pump pulses with absorbed fluences of F = 2-6 mJ/cm². The oxygen coverage at step sites has been monitored in situ during adsorption and induced diffusion by exploiting the sensitivity of optical second harmonic generation (SHG) on surface symmetry. Due to the high sensitivity of the SHG-detection, hopping rates down to 10^-7 per laser shot could be determined. They show a very strong nonlinear dependence on the pump pulse fluence (~F^15).

Under the conditions of our experiment diffusion is induced by multiple excitations from the laser-excited hot electron distribution of the metal, similar to the meanwhile well-studied phenomena of desorption induced by multiple excitation (DIMET). The nonlinear fluence-dependence makes it possible to explore the nature of the excitation process in the time-domain by measuring the hopping rate as a function of delay between two cross-polarized pump pulses. This two pulse correlation has a width of 1.45ps.

The main microscopic factor that determines the width is the coupling of the frustrated translations of the adsorbate to the bath of excited electrons. For a quantitative modeling of the data we approximate this coupling with an electronic friction coefficient. In contrast to the situation in most laser-induced desorption experiments, a friction coefficient that depends on the excitation density is required in order to describe the whole data set. We interpret this dependence in terms of an indirect electronic excitation mechanism of the frustrated translation leading to diffusion. We suggest that the electronic excitation of the substrate couples primarily to the O-Pt stretch vibrations which then excite frustrated translations via an anharmonic coupling of modes.

Fig. 1. Second-harmonic signal (a) and temperature (b) as a function of time. First, oxygen is dosed at a temperature of 160 K. After cooling to 80 K, diffusion is induced by applying the pump laser, leading to a recovery of the SH-signal due to the depopulation of the step edges.

Temporal dependence of optical reflectivity on thin (Pr,Ca)MnO3 films

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We will examine some of the properties of mixed valence manganites with perovskite structure. The research of manganites started with the seminal paper of Jonker and Santen (G.H. Jonker, J. Van Santen Physica 16, 337 (1950)), where the existence of ferromagnetism in solid solutions of LaMnO3-CaMnO3, LaMnO3-SrMnO3 and LaMnO3-BaMnO3 was reported. Theoretical work at approximately the same time explained the ferromagnetic phase (C. Zener, Phys. Rev. 81, 440 (1951)) fairly well, and thus one of the most interesting properties of these materials found a fairly good rationalization in the early studies. As a consequence of the initial theoretical success, studies of the manganites continued at a slower pace in the following years. More recently, their extraordinary magnetoresistive properties have been discovered by Kusters et al. (R. M. Kusters, et al. Physica B 155, 362 (1989)) and von Helmholt et al. (R. von Helmholt et al. Phys. Rev. Lett. 71, 2331 (1993)). The resistivity curve shows a clear metal-insulator-transition in the vicinity of the Curie temperature Tc. There is a resistivity peak at the transition point. This peak becomes smaller and shifts to higher temperatures when external magnetic field is applied to the sample. High negative magnetoresistance (R(B)-R(0))/R(B) is therefore observed around the Curie temperature. This property is especially interesting for use in magnetic storage media such as hard disks. The big boost to the field of manganites was produced by the discovery of the so-called "colossal" magnetoresistance (CMR) effect which was three orders of magnitude larger than a typical magnetoresistance in most other conducting materials. It has been shown (S. Jin et al., Science 264, 413 (1994)) that CMR ratios as large as 127,000% near 77 K can be obtained (That corresponds to a 1000-fold change in resistivity.). Xiong et al. (G. C. Xiong et al., Appl. Phys. Lett. 66, 1427 (1995)) reported thin-films studies of Nd0.7Sr0.3MnOx and in his case magnetoresistivity was as high as 10⁶, a truly colossal factor. Triggered by such huge numbers, the theoretical and experimental study of manganites re-ignited and is currently carried out by numerous groups around the world.

We measured the ultrafast photoinduced reflectivity dynamics in (Pr,Ca)MnO3 thin films as a function of temperature, from 10 K to room temperature. In the bulk there are three phase transitions in this temperature range and the photoinduced reflectivity data on thin films show observable changes in dynamics near these temperatures. Below 60 K the relaxation dynamics is consistent with the existence of ferromagnetic domains in the antiferromagnetic phase.
