PIPT 5

Photoinduced Phase Transitions and Cooperative Phenomena

BOOK OF ABSTRACTS

Bled, Slovenia, 8 – 13 June 2014

PIPT5 - 5th International Conference on Photoinduced Phase Transitions and Cooperative Phenomena

8 – 13 June 2014
Bled, Slovenia

Organized by
Jožef Stefan Institute, Ljubljana, Slovenia

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Bled, June 2014

Dear participans welcome to the PIPT5 Conference

On behalf of the Organizing Committee and the International Scientific Committee we would like to extend you all a very warm welcome to the 5-th International Conference on Photoinduced Phase Transitions and Cooperative Phenomena (PIPT5).

PIPT5 is the fifth in the series of triennial International Conference on photoinduced phase transitions, which was launched in 2001 by Prof. Nasu in Tsukuba, Japan. The first meeting was followed by the 2005 conference in Rennes, France (chaired by Prof. Cailleau and Prof. Luty), the 2008 conference in Osaka, Japan (chaired by Prof. Koshihara) and the 2011 conference chaired by Prof. Luty, which took place in Wroclaw, Poland hosted by the Wroclaw Institute of Technology.

The initial meetings covered a wide range of topics from photoinduced transitions in photo-ferroelectrics, amorphous semiconductors, liquid crystals to solid-liquid phase transitions. The development of novel femtosecond techniques and their applications to advanced quantum matter has been pushing the scope into the direction of correlated electron system. Hand in hand with major advances in experimental techniques (e.g. time-resolved X-ray and electron diffraction enabling making of molecular movies, time and angular resolved photoemission providing access to dynamics of electrons in a momentum and energy resolved fashion) numerous theory groups are becoming involved in the emerging field of ultrafast phenomena in cooperative/correlated systems.

The goal of the PIPT5 meeting was to bring together leading experimentalists and theorist working in the field of ultrafast phenomena in cooperative systems, and provide a forum for discussion on mechanisms of ultrafast optically induced phase transitions in condensed-matter systems, as well on potential applications of these processes in modern electronic circuitry. The main focus of the PIPT5 lies on the cooperative phenomena in advanced quantum materials that take place and can be recorded on the fundamental time and atomic length scales following femtosecond optical quenching. Intense light (in a most general sense, with photon energies ranging from meV to several eV) pulses are used to perturb and drive non-thermal phase transitions from different ordered broken symmetry states (e.g. superconductivity, charge or spin density wave order, ferromagnetism). By selectively probing the magnetic, electronic and structural degrees of freedom the underlying relaxation pathways are being probed aiming at elucidating the mechanisms of interplay between various degrees of freedom (e.g. different interactions giving rise to the existence of the unconventional low temperature order). Moreover, quite often novel metastable states are generated by such rapid quenches, which are not present in thermodynamic equilibrium. Such emergent behaviors present novel possibilities for ultrafast switching and memory applications.

When putting together the programme the International Scientific Committee strived to include the most important recent advances in the field including e.g. recent developments in the field of femtosecond structural probes, THz driven phenomena, time and angular resolved photoemission, surface and interface phenomena and photoinduced generation of metastable states. Moreover, one of the goals was to increase the number of junior scientists and, in particular, participating groups from US/Canada.

Our sincere thanks goes to all members of the International Scientific Committee for their help in making the scientific programme very attractive. Furthermore, we would like to thank our sponsors for their contributions. Particular thanks goes to Prof. Koshihara for securing substantial support from the Japanese Science and Technology Agency (JST).

We wish you all a successful conference, both scientifically and socially, and encourage you to take some time off and enjoy Bled and its surroundings.

Sincerely,

Jure Demšar
Dragan Mihailović

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Martin Wolf, Fritz Haber Institute, Berlin, Germany

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ABSTRACTS

Ultrafast control of ferroelectric polarizations by terahertz fields in organic ferroelectrics

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Recently, a new type of ferroelectrics in which ferroelectric polarizations are produced by intra- or inter-molecular charge transfers (CTs) is attracting much attention and called "electronic-type ferroelectrics". Organic molecular compounds, TTF-CA (TTF: tetrathiafulvalene, CA: *p*-chloranil) and α-$(ET)_2I_3$ (ET: bis(ethyleneditio)tetrathiafulvalene), and a hydrogen-bonded molecular crystal, croconic acid ($H_2C_5O_5$), are its proto-typical examples. In this paper, we report a new approach for the rapid control of ferroelectricity using a terahertz (THz) electric field, focusing on these ferroelectric materials. From the results of THz-pump optical-probe and second-harmonic-generation(SHG)-probe spectroscopy, we show that the ferroelectric polarizations dominated by π-electron systems can be controlled in a sub-picosecond timescale via THz-field-induced collective CT processes.

TTF-CA is a mixed-stack CT compound, in which, TTF (donor) and CA (acceptor) molecules arrange alternately forming quasi-one-dimensional(1D) stacks. By lowering temperature, it shows a neutral(N) to ionic(I) phase transition at 81 K. It was revealed that the CT processes within neighboring DA molecules at the NI transition give rise to the electronic-type ferroelectricity. In TTF-CA and its derivative, collective intermolecular CT processes (1,2) and resultant changes of the polarization (3) can be induced by a femtosecond laser pulse, which is called "photoinduced NI transition". Our terahertz-pump optical-reflectivity-probe (4) and second-harmonic-generation(SHG)-probe measurements revealed that the degree of the intradimer charge transfers can be rapidly controlled and a sub-picosecond modulation of the macroscopic polarization is possible by a strong terahertz field (5). In addition, we found that coherent oscillations of dimeric molecular displacements subsequently occurred, which can be explained by the modulation of the spin moment of each molecule (5). Moreover, we have succeeded in rapidly modulating polarizations by a THz field even in the N phase through the field-induced motions of domain walls between N and I domains.

A 2D organic conductor, α-$(ET)_2I_3$, shows a metal to charge-order(CO)-insulator transition at 135 K. In the CO phase, electronic-type ferroelectricity is suggested to appear from the SHG study (6). In this compound, we also observed large changes of the reflectivity and SHG by THz fields. THz-field-direction dependence of transient reflectivity changes revealed that the direction of the ferroelectric polarization is inclined by about 25 degrees from the crystal axis *b*. Such a diagonal polarization can be explained by considering the anisotropy of molecular overlaps in the 2D sheets of ET molecules.

In a hydrogen-bonded molecular ferroelectric, croconic acid (7, 8), we have also succeeded in modulating the ferroelectric polarization more than 10 % by the THz field of ~100 kV/cm. We will discuss the field-induced π-electron and proton dynamics on the basis of the results of THz-pump optical-probe and infrared-probe spectroscopy.

This work has been done in collaboration with T. Miyamoto, H. Yamakawa, T. Morimoto, T. Terashige, D. Hata, M. Sotome, H. Yada, N. Kida, H. Mori (Univ. of Tokyo), M. Suda, H. Yamamoto (IMS), R. Kato (RIKEN), and S. Horiuchi (AIST).

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Optical control of correlated charge driven by 10 MV/cm ac field of 1.5-cycle infrared pulse in organic conductor

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Recent theoretical studies using dynamical mean field theory (DMFT) have shown dramatic highly non-equilibrium phenomena such as a photo-generations of negative temperature state and superconducting state [1, 2]. In those highly-non-equilibrium phenomena, a reduction of transfer integral *t* under high-frequency ($\omega > t/\hbar$) AC electric field $E(\omega)$ [3-5] plays an important role for cw light (Fig. 1(a)), although similar non-equilibrium effects can be also expected for asymmetric near single-cycle pulse by different mechanism [2].

In this study, we have investigated the optical response of the metallic phase of a layered organic conductor α-$(ET)_2I_3$ (ET; bis[ehtylenedithio]-tetrathiafulvelene) exhibiting metal(Fig. 1(b))-ferroelectric CO insulator(Fig. 1(c)) transition (transition temperature T_{CO}=135 K) using near infrared 7 fs (1.5-cycle) pulse. Photoinduced metal to insulator change, that is a reverse process of photoinduced insulator to metal transition [6-7], will be discussed in terms of the reduction of transfer integral driven by 10 MV/cm high-frequency ac field.

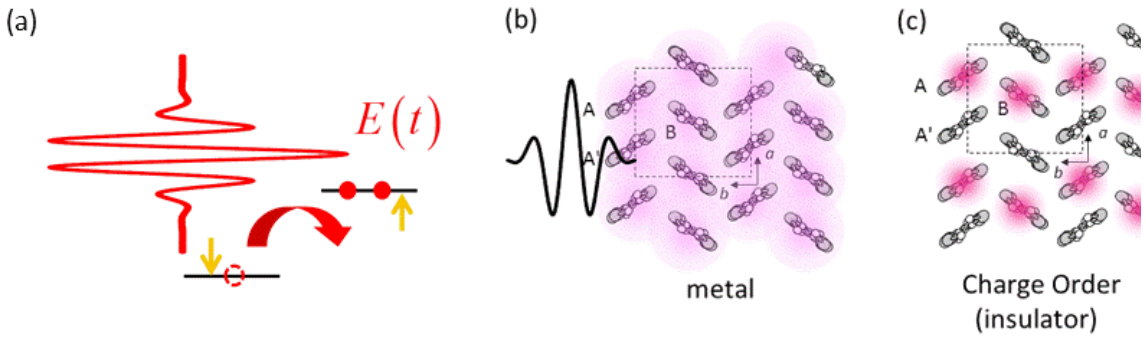

Figure 1: (a) Schematic illustration of the reduction of *t* induced by high-frequency strong ac field , **(b)(c)**Schematic illustrations of **(b)** metallic and **(c)** charge ordered phases of α-$(ET)_2I_3$.

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Sub-cycle charge and spin control with phase-locked multi-terahertz fields

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Femtosecond optical pulses have opened exciting vistas on ultrafast phase transitions. Near-infrared or visible light, however, typically prepares complex excitation scenarios involving multiple degrees of freedom simultaneously. In contrast, electromagnetic radiation in the terahertz (THz) spectral window may couple resonantly and selectively to important low-energy elementary excitations. Modern table-top sources have provided phase-locked field transients covering the entire mid- to far-infrared range and reaching peak amplitudes beyond 11 GV/m [1]. In combination with sensitive electro-optic detection, this toolbox offers an exciting pathway to observe and control spin and charge dynamics on time scales shorter than a single oscillation cycle of the carrier wave. Three recent studies will be reviewed:

In a first experiment [3], we exploit THz electric fields of up to 7 GV/m to investigate a novel non-perturbative regime of sub-cycle coherent charge transport: In the reference system of bulk semiconductors, the THz electric field accelerates electrons through the entire Brillouin zone in a fraction of an oscillation period of light. This dynamics allows us to access the long-sought-after Bloch oscillations in bulk crystals, generating phase-stable high-harmonic transients with spectacularly broad bandwidth, covering the entire THz-to-visible spectral domain between 0.1 and 675 THz, in a single waveform. Quantum interference of different excitation paths of accelerated carriers is controlled via the waveform of the driving field. These results pave the way towards all-optical investigation of electronic bandstructures and correlations throughout the entire Brillouin zone.

While the THz electric field component couples to the charge degree of freedom, in a second study we show that the magnetic component of intense THz transients enables the most direct control of the electron spin [4]: Single-cycle THz pulses switch on and off coherent spin waves in antiferromagnetic NiO, at frequencies as high as 1 THz. Our approach offers a novel ultrafast handle on previously inaccessible magnetic excitations in the electronic ground state of solids.

The third study demonstrates that THz coherent lattice vibrations can induce transient spin order in $BaFe_2As_2$, the parent compound of pnictide superconductors [5]. We trace a multi-THz energy gap characteristic of a spin density wave following excitation with a femtosecond optical pulse. When starting in the low-temperature ground state, optical excitation melts the spin order, followed by an ultrafast recovery. In contrast, the spin density wave gap is induced when we excite the normal state above the transition temperature. Very surprisingly, the transient ordering adiabatically follows a coherent lattice oscillation at a frequency as high as 5.5 THz. Our results attest to a pronounced spin-phonon coupling in pnictides that supports rapid development of a macroscopic order upon small vibrational displacement.

Combining the ideas of all three experiments, intense electric and magnetic THz fields offer unique opportunities to drive charge and spin degrees of freedom selectively and disentangle their interplay in the time domain. Exciting vistas emerge for Zeeman-type excitation of magnons in unconventional superconductors or coherent control of energy gaps in strongly correlated systems.

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Ultrafast Dynamics of Higgs Amplitude Mode in S-Wave Superconductors Induced by Intense Terahertz Pulse Excitation

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Ultrafast photo-control of superconductor is a fascinating subject, whereas various problems remain as open questions: how fast does the order parameter of superconductivity respond to the external perturbation? Can we manipulate the order parameter by optical means? How does the collective mode relevant to the superconductivity emerge in the photo-response? How does it interact with the radiation field? In general, two kinds of collective excitations appear when phase transition occurs associated with the spontaneous symmetry breaking; the gapless phase mode (Nambu-Goldstone (N-G) mode) and the gapped amplitude mode of the complex order parameter. The latter is also called as the Higgs amplitude mode from its analogy to the Higgs boson in elementary particle physics. The nature of the Higgs amplitude mode in superconductors has been intensively studied theoretically, according to which the Higgs amplitude mode can be thought of as the collective Rabi oscillation of the Anderson's pseudo-spins. A variety of collective mode dynamics such as collision-less damping, power-law decay, persistent oscillation, has been investigated [1-6]. Despite the intensive theoretical studies, the experimental investigation of the Higgs mode in superconductors has remained elusive, since the Higgs mode does not couple directly to the electromagnetic field. In this presentation, we report on our recent observation of the Higgs amplitude mode in s-wave superconductors, $Nb_{1-x}Ti_xN$ films [7] by using terahertz (THz)-pump and THz-probe spectroscopy technique [8]. In order to excite the Higgs amplitude mode, we irradiated the sample by an intense monocycle THz pulse whose center frequency was tuned to the superconducting gap energy. In the non-adiabatic excitation regime where the excitation pulse width is shorter than the inverse of superconducting gap energy, a damped oscillation was observed in the transmission of the THz probe pulse as a function of pump-probe delay. The oscillation frequency coincides with the value of asymptotic BCS gap energy after the THz pulse excitation, showing an agreement with the anticipated character of Higgs amplitude mode. When the excitation pulse width is comparable to the inverse of superconducting gap energy and thus the non-adiabatic excitation condition is relaxed, the Higgs mode becomes less prominent. In the presentation, we will report the ultrafast dynamics of Higgs mode and its coherent interplay with the THz electromagnetic wave.

This work is done in collaboration with R. Matsunaga, Y. I. Hamada, A. Sugioka, H. Fujita, K. Makise, Y. Uzawa, H. Terai, Z. Wang, N. Tsuji, and H. Aoki.

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Ultrabright femtosecond electron source captures key molecular motions in the photoinduced insulator-to-metal phase transition of $(EDO\text{-}TTF)_2PF_6$

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The progress in the development of fs-structural probes during the last twenty years has been tremendous. Current ultrafast structural techniques provide the temporal and spatial resolutions required for the stroboscopic observation of atoms in motion. In regards to femtosecond electron sources, different compression approaches have made it possible the generation of ultrashort and ultrabright electron pulses. With an effective brightness only one hundredfold below that of fs-hard X-ray Free Electron Lasers, ultrabright femtosecond electron sources have reveal unprecedented results in the study of photoinduced ultrafast structural dynamics [1, 2]. I will present a brief overview of field along with a recent femtosecond electron diffraction (FED) study of the photoinduced insulator-to-metal phase transition of organic charge-transfer salt $(EDO\text{-}TTF)_2PF_6$ [3]. Here, we implemented a low repetition rate (10 Hz) and ultra-bright femtosecond electron source in order to avoid cumulative heating and photo degradation effects and obtain a movie of the relevant molecular motions driving this photo-induced insulator-to-metal phase transition. We were able to record time-delayed diffraction patterns that allow us to identify time-dependent changes over hundreds of Bragg peaks. Model structural refinement calculations indicate the formation of a transient intermediate structure (TIS) in the early stage of charge delocalization (during the initial 2 ps). The molecular motions driving the formation of TIS were found to be distinct from those that, assisted by thermal relaxation, convert the system into a metallic-like state on the 100-ps timescale. These findings illustrate the potential of ultrabright femtosecond electron sources for capturing the primary processes governing structural dynamics with atomic resolution in labile systems relevant to chemistry and biology.

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Pulsed vs. CW Laser Excitations: Different Controlling Mechanisms of Photoinduced Charge-Order Melting in Molecular Crystals

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We theoretically show different mechanisms of optical control of electronic phases in molecular materials. So far, a variety of phase transitions have been photoinduced with the help of cooperativity originating from electron-electron and/or electron-phonon interactions. Now, we can take advantage of very short and thus strong pulses, which allow us to directly manipulate electrons or their interactions. In this context, photoinduced superconductivity has been proposed by Tsuji *et al*. It is theoretically realized by a band-structure inversion and a resultant negative effective temperature. If continuous-wave lasers are used, its mechanism is based on the fact that the sign of the time-averaged transfer integral involving the Peierls phase can be inverted from the sign of the original transfer integral [1]. If half-cycle or asymmetric mono-cycle pulsed lasers are used, its mechanism is based on the fact that the Peierls-phase difference after the photoexcitation realizes the sign inversion [2]. In any case, the effective value of a single transfer integral is the key quantity.

Here, we demonstrate that materials consisting of intra- and inter-molecular transfer integrals have a choice about which charge transfers are effectively modified. The model system we treat is $Et_2Me_2Sb[Pd(dmit)_2]_2$, in which $Pd(dmit)_2$ dimers are aligned two dimensionally and electrons move within the plane. At room temperature, the dimers are monovalent and the state is a Mott insulator. At low temperatures, the neutral and divalent dimers are regularly aligned, and the state is a charge-ordered insulator. When the neutral dimers are photoexcited by a pulsed laser, a transition into a Mott-insulator phase is experimentally realized [3]. We construct an extended Peierls-Hubbard model that reproduces the electron distributions, the molecular displacements, and the conductivity spectra in these two phases [4]. The time-dependent Schrödinger equation is numerically solved. This model well describes the electron-phonon dynamics during the photoinduced charge-order melting transition [5]. Then, we use continuous-wave and few-cycle pulsed lasers with strong intensities and study the field-intensity dependence of charge transfers to search for possible dynamical localization [6].

Generally, electronic transfer integrals are regarded as renormalized when an oscillating external field is applied, as far as the electronic dynamics averaged over the oscillation period is concerned. The cases where effective transfer integrals vanish are known as dynamic localization. Interdimer charge transfers driven by continuous-wave lasers are thus governed by effective *interdimer* transfer integrals. After the field is switched off, the dynamic localization is no longer relevant. Then, we show that interdimer charge transfers driven by pulsed lasers of energy resonant with an intradimer transition are governed by an effective *intradimer* transfer integral. The total-energy increment depends on how the intradimer transfer integral is renormalized. The same holds for interdimer charge transfers. This interdimer dynamics governed by effective intradimer parameters is evident even for one- and two-cycle pulses, suggesting possible control of photoinduced charge-order melting. These two lasers control the charge transfers in different manners. The difference comes from the molecular hierarchy.

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Spatio-temporal Behavior of Atomic Displacement Parameters during Collective Relaxation of Franck-Condon States

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Recent study of time-resolved x-ray and electron diffraction measurements has shown that the structural dynamics in ultrashort time scales is experimentally observable. The time-evolution of the diffraction patterns reflects the spatio-temporal behavior of the atomic displacement parameters (ADPs) in the Fourier space, and thus ultrafast structural analyses[1] will be used to study the initial dynamics of photoexcited states which is complementary to the conventional pump-probe experiments. In this paper we discuss the way to derive information on the relaxation of Franck-Condon states, coalescence of nuclei in photoinduced nucleation, and their growth dynamics from beam diffraction patterns.

We employ a model of localized electrons in molecular cells on a square lattice coupled with an optical phonon mode given by the following Hamiltonian,

$$H = \sum_{\vec{r}} \left[\frac{p_{\vec{r}}^2}{2} + \frac{\omega^2 u_{\vec{r}}^2}{2} + (\sqrt{2\hbar\omega^3}\, s u_{\vec{r}} + \varepsilon\hbar\omega + s^2\hbar\omega)\tilde{n}_{\vec{r}} + \lambda\sigma_{\vec{r}}^x \right] - \sum_{<r,r'>} \left[\alpha\omega^2 (u_{\vec{r}} - \beta\tilde{n}_{\vec{r}})(u_{\vec{r}'} - \beta\tilde{n}_{\vec{r}'}) + \{V - W(u_{\vec{r}} + u_{\vec{r}'})\}\tilde{n}_{\vec{r}}\tilde{n}_{\vec{r}'} \right], \quad (1)$$

where the detail of the model is described in Refs. [2,3]. We calculated the diffraction intensity $I(\vec{K},t)$ as a function of time by numerically solving the time-dependent Schrödinger equation and the calculated results of the differential diffraction intensity defined by $\Delta I(\vec{K},t) = I(\vec{K},t) - I(\vec{K},0)$ are

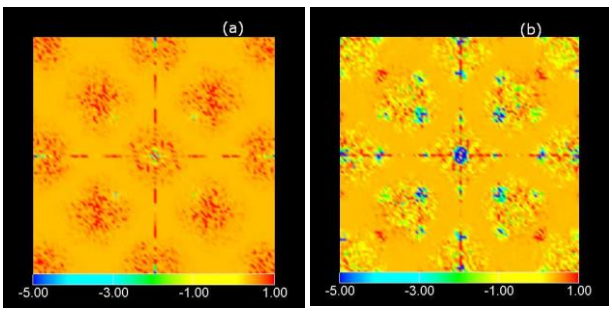

Figure 1: Differential diffraction intensity $\Delta I(\vec{K},t)$ in the

shown in Figs. 1-(a) and (b). We found that these patterns contain information regarding the relaxation of the Franck-Condon state as spatio-temporal variation of the ADPs. In particular, transient ADPs reflect the quantum-mechanical nature of excited wavepackets which is relevant to the formation of the precursor of photoinduced nuclei[3]. In other words, transient ADPs will give us a clue to reveal the origin of the incubation period observed at the earliest stage of photoinduced nucleation. These results exemplify that further study on the mechanism of the photoinduced cooperativity will be possible particularly when ultrashort coherent x-ray or electron-beam pulses are available in the near future.

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Photo-induced dynamics of $Pt(dmit)_2$ salts studied by optical spectroscopy and electron-diffraction technique

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$M(dmit)_2$ (M=Pd, Pt) salts show an unique phase transition due to the strong electron-phonon and electron-electron correlations [1]. We reported the optical spectral change induced by the photoexcitation in the charge separated (CS) phase of the $Et_2Me_2Sb[Pd(dmit)_2]_2$ single crystal, which suggests the occurrence of the photoinduced phase transition (PIPT) [2]. Recently, $A[Pt(dmit)_2]_2$ (A=Me_4P, Me_4As and Me_4Sb) are crystalized and studied by transport and X-ray crystallographic technique [3]. The $Pt(dmit)_2$ system also shows the CS phase with relatively higher transition temperature up to 215 K thanks to the relatively weaker dimerization of the $Pt(dmit)_2$ than $Pd(dmit)_2$ molecules. It is interesting for application to investigate the photoinduced phenomena in this $Pt(dmit)_2$ salt, because of its high transition temperature, as well as unveiling the mechanism of phase transition based on CS mechanism by virtue of the freedom of molecular orbitals.

Since the dimerization of $Pt(dmit)_2$ monomers plays a key role in determining the electronic structure of the salt, the direct observation of the photoinduced structure change and, in particular, the intermolecular distance, could yield powerful new insights into the mechanism of the PIPT. Using the recently developed time-resolved electron diffraction technique [4], we have succeeded in observing directly the photoinduced structural change with sub-ps resolution from the CS phase and also clear coherent oscillation of the intensity of the Bragg peaks with in the THz frequency range. By comparing the results of time-resolved optical spectroscopy and electron diffraction pattern, we are trying to clarify the precise photoinduced dynamics of PIPT in this material.

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Dynamical Role of Hidden Faces in Photo-Functional Materials

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Controlling cooperative phenomena such as phase transition by external stimulus such as electric, magnetic fields is a key subject for materials science, device application and even biological science today. Especially, achieving the photo-tuning of the phase transition which is named as photo-induced phase transition (PIPT) is becoming important target for wide field of science. Because ultra-fast conversion of magnetic, dielectric, structural and optical properties by weak light is expected for PIPT materials as a result of cooperative interactions.
PIPT shows various attractive natures, however, the research of this field is facing difficult and essential problem, i.e. can we realize and identify a new phase of solid based on novel lattice structure which is unique for the photo-excited condition so called as a 'hidden phase'? This 'hidden phase' with electronic and structural order realized only by optical excitation is important merit of PIPT process for achieving ultrafast phase control via pure photonic channel free from thermal effect. Late progress in Laser and Quantum beam technologies are giving us the great chance to solve this problem. Here, we demonstrate that light excitation reveals a 'hidden charge and orbital ordered (CO-OO) phase' which can never be achieved under thermo-equilibrium condition by virtue of ultrafast pulsed X-ray and electron techniques. We also show that 'hidden phase' really becomes the origin of the sensitive photo-induced change in optical property of various inorganic and organic crystals [1,2]. Our new results of the study on 'hidden face' of photo-functional materials will be also briefly discussed.

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Photo-induced phase transition with magnetic change in cyano-bridged bimetallic assemblies

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Research associated with the photo-functional materials is attractive topics. Up to date, we have reported several photo-functional materials using cyano-bridged bimetal assemblies and metal oxides [1-6]. The light-induced excited spin-state trapping (LIESST), the light-induced phase transition from the low-spin (LS) to high-spin (HS) states of some transition-metal ions, has been extensively studied in the field of solid state chemistry. Spin crossover complexes that have been reported are mostly those composed of molecular crystals. However, if spin crossover complexes with 3-dimensional network structures can be synthesized and HS sites can form a magnetic ordering with each other, then the spontaneous magnetization can be expected. From this perspective, we synthesized a new octacyano metal complex, $Fe_2[Nb(CN)_8]\cdot(4\text{-pyridinealdoxime})_8\cdot 2H_2O$ (**1**), and observed ferro-magnetism caused by light-induced spin crossover[4]. The crystal structure of **1** is tetragonal ($I4_1/a$). Fe and Nb ions are bridged via cyano groups to form a 3D network structure (Fig. 1). The molar magnetic susceptibility (χ_M) vs temperature plot of **1** shows the spin crossover transition from Fe^{II}(HS) (S= 2) to Fe^{II} (LS) (S= 0) at 130 K. Next, the photomagnetic effect of **1** was studied. Before irradiation, the LS phase is para-magnetic. By irradiating with a 473-nm cw laser light, spontaneous magnetization appeared (Fig. 1). In the photo-induced ferromagnetic phase, Curie temperature (T_C) and saturation magnetization (M_s) are 20 K and 7.4 μ_B, respectively. The M_s of 7.4 μ_B agrees well with a calculated value of 7.7 μ_B obtained in a case where the spins of photo-induced Fe^{II}(HS) and Nb^{IV} (S= 1/2) are antiparallelly arranged. The superexchange interaction constant (J_{ex}) between Fe^{II}(HS) and neighbouring Nb^{IV} is calculated by molecular field theory, i.e., −6.9 cm^{-1}.

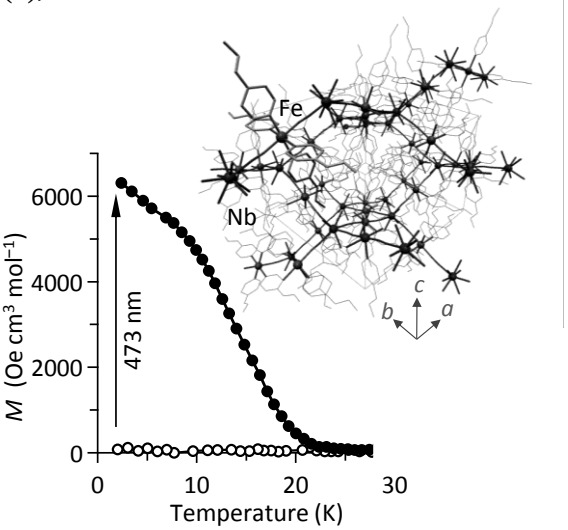

Figure 1. Crystal structure of **1** (inset), and magnetization vs temperature curves of **1** before (○) and after (●) irradiation.

Furthermore, very recently, we developed a new chiral structured magnet of an iron-octacyanoniobate assembly, where Fe ions and Nb ions are three dimensionally bridged by CN ligands[3]. By alternatively irradiating with 473-nm blue light and 785-nm light, the spontaneous magnetization of the material can be reversibly switched, and it exhibits 90-degree switching of the polarization plane of the output second harmonic light by changing the state of the magnet with 473-nm and 785-nm lights.

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Photofunction of organic materials studied by time-resolved infrared vibrational spectroscopy

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Time-resolved vibrational spectroscopy is a powerful tool to investigate photoinduced dynamics of organic materials because intensities and frequencies of vibrational peaks sensitively reflect local charge and structure of molecules. We have developed time-resolved infrared vibrational spectroscopy system using a femtosecond Ti:sapphire laser and studied ultrafast processes in two types of photofunctional materials. One is strongly correlated organic crystals which exhibit photoinduced phase transition (PIPT) [1-3] and the other is metal complexes which are used for organic solar cells and artificial photosynthesis [4, 5]. We found that structural change on the order of tens of picoseconds plays an important role for photofunction in these materials.

Electron-electron and electron-phonon interactions in strongly correlated organic crystals cause photoinduced phase transition. Previously such complicated dynamics have been studied by measuring transient electronic spectra; however, information on structural change was lacking. To address this problem, we have applied time-resolved vibrational spectroscopy to several organic crystals showing PIPT, TTF-CA [1], $(EDO\text{-}TT)_2PF_6$ [2], and $Pd(dmit)_2$ salts [3]. As a result, we found that the structure change takes place with a tens-of-picosecond delay from the electronic state change and noticed that the materials having steric hindrance shows such delayed structural change. We also compared this result to that obtained by more direct method using femtosecond electron-diffraction [6].

Metal complexes consist of a center metal and □-conjugated ligands such as bipyridine. To date, fundamental process of their photofunction has been revealed as follows. Photoirradiation excites the complexes to ^{1}MLCT (singlet metal to ligand charge transfer state) and subsequently the intersystem crossing to ^{3}MLCT takes place within 100 fs. The lifetime of ^{3}MLCT is more than a hundred nanoseconds, and this meta-stable ^{3}MLCT play a key role in the photofunction by transferring the excited electron to a reactive state or another molecule. However, these states do not emit a luminescence, that is, dark state, so that the process has never studied spectroscopically. We found the peaks which are assigned to such dark state by comprehensive studies on transient vibrational peaks of various ligands using time-resolved vibrational spectroscopy [5] and succeeded in direct observation of photochemical reaction process though these peaks in several ruthenium and rhenium complexes.

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Modeling of local phase transformations induced by optical pumping to excitons: applications to neutral-ionic transitions

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In experiments on PIPTs in electronic systems, usually the pumping goes via initial population of high-energy electronic bands. A more special and rare technique is the resonance pumping: either to intramolecular excitons (IME) or to charge-transfer excitons (CTE) – bound states of an electron and a hole. This approach was proved to be particularly useful in studies of neutral-ionic transitions (NIT) in quasi-1D organic compounds.

We present a theory of resulting spacio-temporal effects with a modelling targeting the NIT. We consider cases of IME and CTE corresponding to profound experimental studies of S.Koshihara and H. Okamoto. In our picture, a quasi-condensate of excitons appears as a macroscopic quantum state which then evolves interacting with other degrees of freedom prone to instability. Via these interactions with soft modes, the excitons are subject to self-trapping. That locally enhances their density which can surpass a critical value to trigger the phase transformation, even if the mean density is below the required threshold for the global transition. We recover dynamic interplays of fields such as the collective wave function of excitons, electronic charge transfer and polarization, lattice dimerization. Their mutual interaction results in formation of a symmetry broken ground state which is inhomogeneous if the pumping is below the threshold. In all cases the initial string of the new embedded phase appears as self-trapping; its length is determined by a balance between gaining the potential energy and loosing the quantum kinetic energy of the exciton. The phase transformation proceeds *from large to medium distances, rather than from small to larger ones as in a rather common picture of localized excitons.*

We obtain various transient effects: self-trapping, dynamic formation of domains separated by walls, subsequent merging of domains and collapse of walls, emittance of coherent propagating waves.

Mapping Atomic Motions with Ultrabright Electrons: The Chemists' Gedanken Experiment Enters the Lab Frame

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Electron sources have achieved sufficient brightness to literally light up atomic motions in action. This development provides a direct observation of the very essence of chemistry and the central unifying concept of transition states in structural transitions. Due to the extraordinary requirements for simultaneous spatial and temporal resolution, it was thought to be an impossible quest and has been previously discussed in the context of the purest form of a gedanken experiment. Two new electron gun concepts have emerged from detailed calculations of the propagation dynamics of nonrelativistic electron pulses with sufficient number density for single shot structure determination. The atomic perspective, that these sources have opened up, has given a direct observation of the far from equilibrium motions that lead to structural transitions. It is these motions that lead the system through the barrier crossing region. Recent studies of the structural phase transition in charge ordered organic materials have given the first direct atomic view of barrier crossing processes. The transition is formally a photoinduced charge transfer process in which the change in charge distribution is strongly coupled to lattice modes that stabilize the charge separated state. It was discovered that this nominally 280 dimensional problem distilled down to projections along a few principle reaction coordinates (Gao et al Nature 2013). Similar reduction in dimensionality has also been observed for ring closing reactions in organic systems (Jean-Ruel et al J. Phys. Chem. B 2013). This phenomenon appears to be general and arises from the very strong anharmonicity of the many body potential in the barrier crossing region. The far from equilibrium motions that sample the barrier crossing region are strongly coupled, which in turn leads to more localized motions. In this respect, one of the marvels of chemistry and biology is that despite the enormous number of possible nuclear configurations for any given construct, chemical processes reduce to a relatively small number of reaction mechanisms. We now are beginning to see the underlying physics for these generalized reaction mechanisms. The "magic of chemistry" is this enormous reduction in dimensionality in the barrier crossing region that ultimately makes chemical concepts transferrable. With a large enough basis, it may be possible to characterize barrier crossing processes in terms of reaction modes in analogy to the characterization of equilibrium fluctuations in terms of vibrational normal modes. Additional examples will be presented in which it has been possible to directly observe underdamped modes involved in metal ligand charge transfer processes, as well as structural changes involved in intersystem crossing, to a direct observation of Pauli explosion in alkali halides (Hada et al, Nature Comm, in press) – the reverse of the classic "electron harpooning" reaction concepts that helped establish transition state theory.

Tracking femtosecond dynamics of spins and the lattice with x-ray diffraction

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With femtosecond time resolution, x-ray diffraction offers unique capabilities to observe directly the dynamics of long range order of charge, spin, orbital and lattice in strongly correlated systems. In this talk we highlight a few recent examples of experiments in this area that both demonstrate the capabilities of this method and make quantitative tests on our theoretical understanding in multiferroics, charge density wave systems, and charge–ordered manganites.

In the first example we show how resonantly enhanced magnetic scattering can be used to quantitatively measure the character and magnitude of spin motion in a coherent electromagnon in $TbMnO_3$ driven by a THz frequency electromagnetic field [1]. We observe a 4 degree rotation of the antiferromagnetically ordered spin spiral plane, a result consistent with a previously published model that suggests this may be a viable route for ultrafast domain switching in multiferroics [2].

As a second set of examples we discuss the laser-induced melting of charge order in the charge density wave system $K_{0.3}MoO_3$ and the doped manganite $Pr_{0.5}Ca_{0.5}MnO_3$. Both systems show coherent dynamics that extend into a higher symmetry phase that is distinct from that seen in thermodynamic equilibrium.

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Imaging plasmonic Fabry-Pérot resonances with ultrafast electron microscopy

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Photon induced near field electron microscopy can be used to control and image excited surface plasmons in nanoscale materials with femtosecond temporal resolution [1]. The technique is particularly useful for the investigation of nanophotonic devices and we demonstrate its capabilities by imaging plasmonic Fabry-Pérot resonances that have been excited in a silver nanowire [2,3]. We show that by changing the pump laser properties we can precisely control the plasmons that have been excited in the nanoparticle (see Figure 1). In a similar experiment we also demonstrate the wave-particle duality of the electromagnetic field. By using an imaging spectrometer the energy of the electrons can be captured, while simultaneously projecting a spatial dimension on the detector. The spectrum shows the quantized exchange of photons by the imaging electrons and the spatial dimension captures the Fabry-Pérot interference pattern. Thus both the quantization and interference of the electromagnetic field are captured simultaneously.

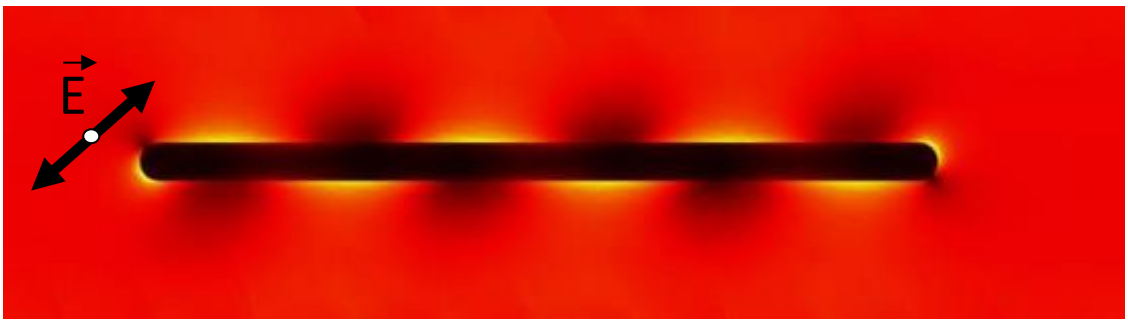

Figure 1: Simulation of the interaction of Ag nanowire with intense 800 nm femtosecond laser pulse. Yellow colour indicates regions of induced electromagnetic fields due to SPP in nanowire. The dark region is the nanowire itself. This simulation represents the image that can be obtained when using the photon-induced near field microscopy technique. The laser pulse is propagating into the page and the electric field is oriented 45^0 relative to the axis of the nanowire.

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All optical control of magnetism: From fundamentals to nanoscale reversal

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From the discovery of sub-picosecond demagnetization over a decade ago to the recent demonstration of magnetization reversal by a single 40 femtosecond laser pulse, the manipulation of spins by ultra short laser pulses has become a fundamentally challenging topic with a potentially high impact for future spintronics, data storage and manipulation and quantum computation.Theoretically, this field is still in its infancy, using phenomenological descriptions of the none-equilibrium dynamics between electrons, spins and phonons.A proper description should include the time dependence of the exchange interaction and nucleation phenomena on the nanometer length scale.A practical challenge is how to bring the optical manipulation of magnetic media to the required nanoscale, which may be possible using plasmonic or wave-shaping techniques. Recent results and an outlookto probe and control magnetic order on the femtosecond time and nanometer length scalewill be discussed.

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Transient magnetism in EuO

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The interplay between itinerant and localized spins in ferromagnetic semiconductors leads to a variety of intriguing phenomena, including the colossal magnetoresistance effects, the formation of magnetic polarons, and the possibility to optically enhance then magnetization. This presentation will review some of our recent work on optically induced magnetic phenomena in the intrinsically ferromagnetic semiconductor EuO, with an emphasis on time resolved x-ray circular dichroism, magneto-optic, and THz experiments.

Dissentangling the nanoscale angular momentum pathways during all-optical magnetic switching

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Understanding the ultrafast interplay between charge, magnetic and lattice degrees of freedom is central to gaining control of condensed matter phenomena as diverse as insulator-metal transitions and magnetic switching. While discovered early [1], perhaps still the least understood is the coupling of magnetism with other degrees of freedom. Magnetism, by symmetry could be expected to couple only weakly to phonons and electrons, however the observed ultrafast demagnetization [1] and all-optical magnetic switching [2,3], have proved this to be far from correct. Ultrafast x-ray pulses offer the fascinating prospect to disentangle, on the nanometer length- and femtosecond timescales, the non-equilibrium angular momentum pathways following optical laser excitation that lead to a reversal of the magnetization [4]. The insights gained in how magnetism evolves on time- and lengthcales associated with the exchange interaction opens a new way of engineering the relaxation path in magnetic systems.

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Ultrafast dynamics in spin crossover cobaltites

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Photonic phase control of electronic properties has been of current interest in condensed matter physics. One of the typical materials is spin crossover (SC) complexes containing Fe^{2+} and they are known as an important example in the photoinduced phase transition (PIPT). This is a photonic change/control of the spin state of *d* electrons in Fe^{2+} between the low spin (LS) state (t_{2g}^6) and the high spin (HS, $e_g^2\ t_{2g}^4$) state.
In addition to those iron complexes, we have also proposed that perovskite-type cobalt oxide systems containing Co^{3+} can be an interesting example showing such a spin state transition by light, and have revealed that some SC cobalt oxides showed a unique ultrafast feature concerning PIPT[1-5]. Among them, in this presentation, we show ultrafast phenomena in $La_{1.5}Sr_{0.5}CoO_4$ and $BiCoO_3$.
$La_{1.5}Sr_{0.5}CoO_4$ is layered perovskite-type and shows a checker-board type charge ordering (CO) as well as SC in the Co^{3+} site. With irradiating fs laser pulses, we observed ultrafast electronic change due to melting of the CO. The real space dynamics of the photoexcited area is discussed in terms of the electromagnetic analysis[5].
$BiCoO_3$ is a ferroelectric (FE) perovskite with HS state and shows SC phenomena from the HS to the LS state by applying external pressure[6]. We demonstrate our recent trial of ultrafast control of the FE behavior in the light of nonlinear pump-probe spectroscopy and discuss the relationship between the FE change and the SC.

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Confinement effects in photostrictive/magnetostrictive core-shell particles based on Prussian blue analogues

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The interest in molecular-based materials for memory devices and magneto-optical switching has been revived after the discovery of high-T_C magnets[1] and various photomagnetic effects under visible light irradiation[2] in Prussian blue analogues. Nevertheless, photo-induced magnetization changes in materials showing long-range magnetic order have been restricted so far to very few examples and low temperatures.[3] An alternative approach is to use the concept of strain-mediated magnetic switching developed for oxide-based and metal/polymer multiferroic composites[4] by combining a magnetostrictive subsystem to a switchable subsystem that undergoes large and reversible photostriction effects.

Polycyanometallates were recently shown to be promising candidates for such photostrictive/piezomagnetic heterostructures.[5] This might be explained by (i) the amplitude of the deformation for the photo-strictive phase, i.e. up to 3% change of the lattice parameter under light irradiation and (ii) the large compressibility of the metallocyanide lattice with a bulk modulus that is one order of magnitude smaller than the one of oxides. In addition, heterostructures with different connectivity (core-shell particles, multilayers, etc) can be grown in a controlled way, leading to heteroepitaxy even in the case of large lattice mismatch between the two phases. In core-shell architectures, we have recently demonstrated the transfer of strain through the interface between the core and the shell upon light irradiation. However, in-situ synchrotron x-ray diffraction measurements also suggested confinement effects, similar to a pressure applied by the shell and opposed to the core dilatation under irradiation (see Fig. 1). This confinement strongly impacts the photo-switching properties of the core particles that are partly or completely blocked depending on the shell thickness.[6] In this work, we will discuss the main parameters that influence the confinement effects, by studying (i) different sizes of the photo-active core particles, (ii) ultra-thin shells and (iii) different strain states associated to different lattice mismatchs between the core and the shell.

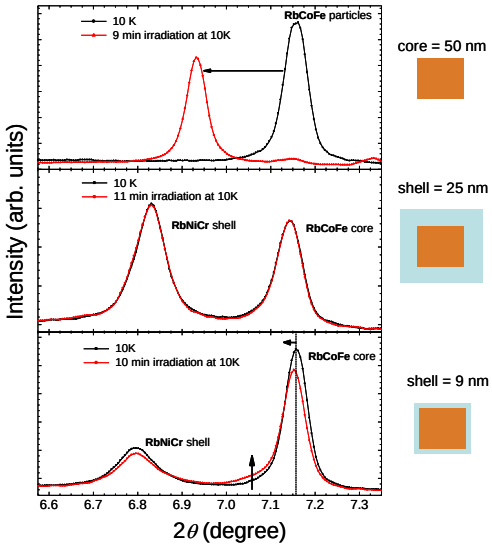

Figure 1: Change in the position of the (400) Bragg peaks before (black) and after (red) light irradiation, λ = 0.31 Å.

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Controlling, Probing and Harnessing the Strongest Force in Magnetism

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The idea to change magnetic properties of media with the help of light has long intrigued people in physics and chemistry. Naturally, this raises the question about the speed limit of the optical control of magnetism [1]. Fundamentally, magnetic order is a macroscopic manifestation of a strong quantum phenomena – exchange coupling between spins. The energy of the interaction is often written as

$$W=-\Sigma_{i,j} J S_i S_j \qquad (1)$$

where J is the exchange parameter, S_i and S_j are the spins of the i-th and j-th adjacent magnetic atoms. This exchange coupling represents the largest interaction in magnetism. It can be associated with an effective magnetic field of 100-1000 T. The strength can be appreciated from the fact that magnetic order in condensed matter survives well above room temperature. Obviously, harnessing the exchange interaction is *the* way to achieve the ultimately fastest magnetic switching. How can we control, probe and harness the exchange interaction for ultrafast magnetic switching? Here we demonstrate that the exchange interaction can be manipulated through ultrafast laser excitation in a large class of transition metal oxides [2]. We show that using ultrashort laser pulses one can monitor laser-induced dynamics of the energy of the exchange interaction with subpicosecond temporal resolution [3]. Finally, we suggest a scenario in which the strength of the exchange interaction is employed to achieve the fastest possible magnetic switching [4].

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Speed limit for FePt spin dynamics on femtosecond timescales

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Magnetization manipulation is an indispensable tool for both basic and applied research and to understand cooperate phenomena in ferromagnetic materials. I will demonstrate some of the knobs to tune dynamics at ultrafast time scales. One possibility is to tune shaping the properties of the electronic system. The dynamics of the response depends on the nature of the heat transfer from the laser heated electrons to the spins, which determines the speed of the ultrafast demagnetization. A signature of the ferromagnetic correlations is found, if the electrons are driven to a strong demagnetization. A second slower process is found after the initial fast drop of magnetization. A special material of interest for magnetic hard disc development is FePt. This material has an interesting modification of its density of states: Pt alloying reduces the number of states at the Fermi level and makes the material "more noble". Consequently, for the same amount of energy deposited, the electron temperatures shoots to much higher values above the Curie temperature at around its phase transition. Using a thermal micromagnetic model based on the Landau-Lifshitz-Bloch (LLB model) equation, we relate the electron temperatures reached by the laser heating with the speed of demagnetization driven to a strong demagnetization much earlier than the pure Fe.

Due to the non-equilibrium electron distribution, also ultrafast currents are generated and contribute to the laser driven spin dynamics. Those currents can be directly accessed via THz radiation emitted from the sample. Similarly to shaping the density of states in the first example, adjacent layers of a noble material like Au with sp-states at the Fermi level, or Ru, which has d-band at around the Fermi level, these materials can shape the THz spin currents opening a way towards THz spintronic devices.

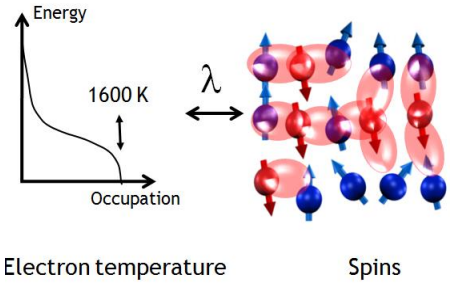

Figure 1: Interplay of electron heating and correlations in the spin-system resulting in critical phenomena around T_C.

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Interfering long-lived spin precessions induced by a THz pulse in $ErFeO_3$

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One of the newly developing topics in terahertz (THz) time domain spectroscopy is the ultrafast excitation of spins with the THz pulse [1,2], where the THz magnetic field component is used for instantaneously tilting the spins. Here, we report the unexpectedly long-lived interfering spin precession induced by THz pulse in an orthoferrite $ErFeO_3$. The spin precession in $ErFeO_3$ single crystal sample (100 □m thick, (001) surface sample) induced by THz pulse was observed through time dependent Faraday rotation of the transmitting visible probe pulse. The Faraday rotation reflects the magnetization dynamics in the thickness direction of the sample, that is, the *c* axis direction. Figure 1(a) shows the time dependent THz induced Faraday rotation obtained with this sample at 70 K. Unlike in the previous report [2], the precession lasts for a long time (typically 1 ns at 4 K) and a 50 ps period beating of the oscillation is also observed. The Fourier spectrum of the oscillation in Fig. 1(b) reveals that the obtained signal consists of two separate sharp spectral peaks. The frequencies of the two peaks are

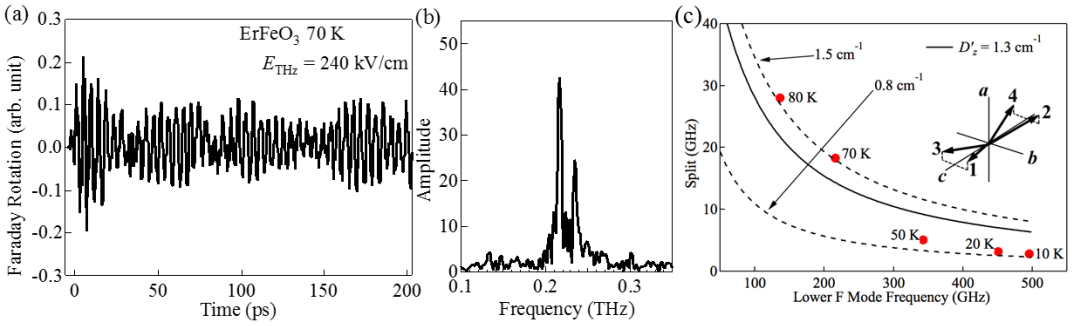

Fig. 1. (a) Time dependent THz induced Faraday rotation signal obtained with (001) $ErFeO_3$ single crystal sample at 70 K. (b) Fourier spectrum of the oscillation shown in (a). (c) Temperature dependence of the splitting. Inset shows the Fe spins.

approximately 0.216 THz and 0.234 THz (close to the F mode resonant frequency at 70 K reported previously [2]). Considering the symmetry of the crystal, it is noticed that the Dzyaloshinskii-Moriya interaction works differently for different pairs of nearest neighbor spins among four spin sublattices (inset of Fig.1 (c)). For example, when focusing on the spin 1, the spins 2 and 4 interact with spin 1 differently. Normally, such differences are ignored for deriving the resonant frequency [3]. By taking this difference into consideration, the resonant frequencies were calculated as a function of lower branch F mode frequency. It was confirmed that in the spin configuration in the inset, the *z* component of the Dzyaloshinskii vector D_z' working on spin pair 1-4 (and 2-3) causes the resonant frequency to split. Figure 1(c) shows the calculated value of the splitting as a function of temperature, which is correlated with the F mode frequency, along with the results obtained experimentally. The figure shows that the observed splitting can be explained reasonably with the proposed splitting mechanism using value of D_z' deduced from ref [3].

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Ultrafast quenching of the exchange interaction in a Mott-insulator

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The control of magnetic order by femtosecond laser pulses is currently a very hot topic in magnetism and condensed matter physics in general. In many situations a description of such ultrafast spin dynamics seems successfully carried out within an effective spin model. Nevertheless, such a model is essentially a low-energy description of the magnetic degrees of freedom only, in which the interaction between spins is described by the exchange interaction. The question how fast such an effective spin model and the exchange interaction can emerge from the nonequilibrium electron dynamics is up to now an unresolved issue in modern condensed matter physics, although an investigation of this ultimate limit of spin dynamics is in range using todays femtosecond laser technology.

In this contribution, we present a theoretical investigation on the question how fast after photoexcitation a modified exchange interaction emerges from the nonequilibrium electron dynamics, and how effective this modification can be [1]. As system of study we choose the prototype Mott-Hubbard insulator. Starting from an initially equilibrium situation, we demonstrate an ultrafast quenching of J_{ex} both by evaluating exchange integrals from a time-dependent response formalism [2], and by explicitly simulating laser-induced spin precession in an antiferromagnet that is canted by an external magnetic field. In both cases, the electron dynamics is obtained from nonequilibrium dynamical mean-field theory [3]. As visualized in Figure 1, we find that the modified J_{ex} emerges already within a few electron hopping times after the pulse, with a reduction that is comparable to the effect of chemical doping.

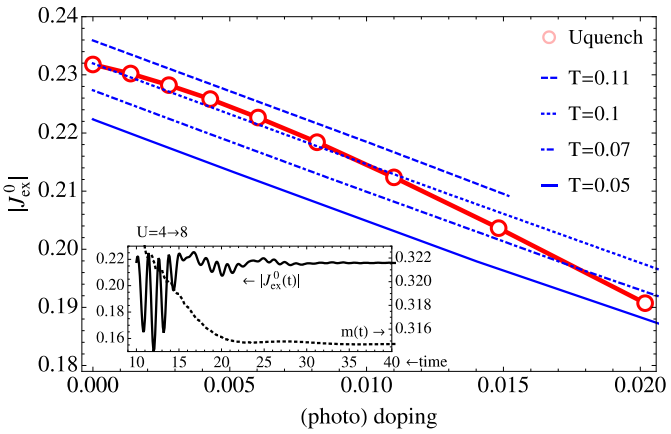

Figure 1: Comparison of the nonequilibrium exchange interaction (open circles) in the quasi-stationary state after an interaction quench in the Bethe lattice with the equilibrium exchange interaction of the chemically doped model (thin lines) for U = 8 and different temperatures. The inset shows the time-evolution of the exchange interaction J_{ex} (black solid line) and staggered magnetization m (black dashed line) after a quench U = 4 → 8. The quasi-stationary modified values both of J_{ex} and m emerge already within a few inverse hopping times.

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Exploring different pathways across the potential energy surface in the early process of photoinduced spin-state switching.

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Spin-crossover solids are prototypes materials, undergoing photoinduced phase transitions towards long-lived states at low temperature, or short-lived at higher temperature. The seeding process results in an ultrafast local molecular trapping of light-excited states, not so well understood. During the past decades the Light Induced Exited Spin-State Trapping (LIESST) mechanism, as well as its reverse mechanism (reverse-LIESST), have been formerly accepted as a photo-induced process involving an intersystem crossing (ISC) dynamics that allows trapping the high-spin (HS) state at sufficiently low temperatures upon continuous irradiation into the absorption bands of low-spin (LS) state; or into the absorption bands of HS state in case of reverse-LIESST.[1] In the recent literature, ultrafast time-resolved studies on Fe(II) spin-crossover compounds have been mainly devoted to the dynamics of light induced LS($^1A_1$)-to-HS($^5T_2$) switching by pumping the LS state into the spin-allowed metal to ligand charge transfer (^{1}MLCT) bands.[2][3]

In this contribution, we reveal the full pictures of the different dynamical pathways in LIESST and reverse-LIESST processes, showing the latest ultrafast transient absorption results on the spin-crossover complex for $[Fe(ptz)_6](BF_4)$ (ptz = 1-propyltetrazole) having no low-lying MLCT states by directly pumping it into its spin-allowed ligand-field transitions of the HS species at 830 nm ($^5T_2 \rightarrow {}^5E$).[4][5] We will also present LIESST process on the $[Fe^{II}(PM\text{-}AzA)_2(NCS)_2]$ single crystal where detailed information will be given on the electronic and atomic rearrangement after an ultrafast excitation of the LS state into a transient ^{1}MLCT state, where the activation and damping of coherent structural dynamics plays an important role in the trapping process.
Our results evidence totally different kinetics between the LIESST and reverse-LIESST processes. For the reverse-LIESST a double ISC involving only d-d states, namely $^5E \rightarrow {}^3T_1 \rightarrow {}^1A_1$, with a lifetime of the initially excited ^{5}E state of 1.7 ps and of the intermediate $^3T_1$ state of 39 ps.[5] Conversely, for LIESST upon direct pumping into spin-allowed d-d transition of the LS state, ISC from the initially excited $^1T_1$ state to the HS state takes less than 150 fs and is thus equally fast as for irradiation into the ^{1}MLCT bands.

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Photo-induced ordering phenomena in 2D homoligand gold nanoparticles

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We demonstrate the ability of ultrafast electron packets with an energy of 30 keV [1] to perform diffractive imaging from a 2D ensemble of homoligand gold nanoparticles. Hidden local symmetries within disordered systems have been recently classified by means of coherent x-ray scattering intensity correlations around a Debye-Scherrer ring, in the static speckle diffraction pattern of a colloidal glass [2, 3]. We show that a similar approach based on ultrafast electron diffraction can be used to investigate the undisclosed local properties of nanostructured thin films with fs-time resolution, owing to the high cross-section of electrons for interaction with matter and their smaller sample damaging ability [4]. Our results evidence both the dynamics of the 7 nm gold cores and that of the surrounding thiols ligands, revealing a tendency of the latter to order when photoexcited.

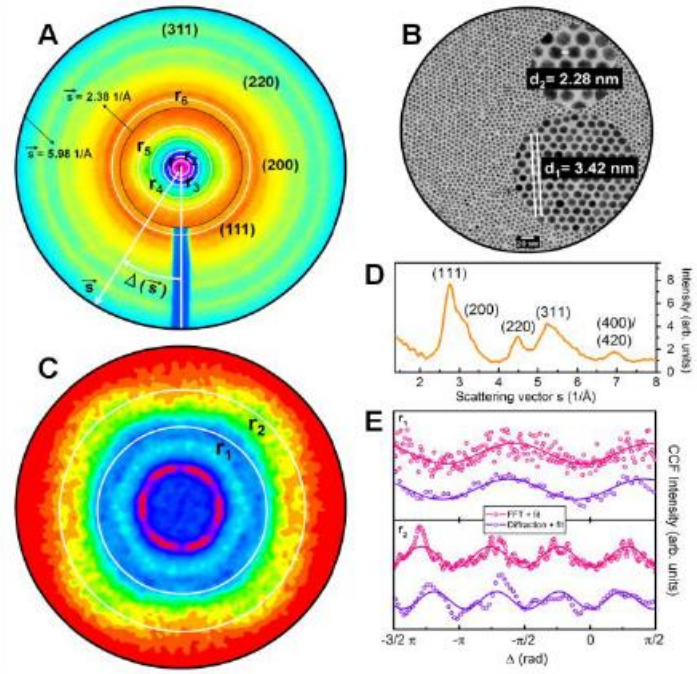

Figure 1 A) Electron speckle diffraction pattern of homoligand gold nanoparticles. B) TEM image of the homoligand gold nanoparticles 2D-assembly. C) 2D-FT of the TEM image displayed in (B). D) Radial distribution function of the polycristalline sample E) Intensity modulations obtained by the angular cross-correlation analysis of the speckle pattern from the rings labeled r1 and r2, indicated respectively in (A) (violet profile) and (C) (pink profile). The modulations are a fingerprint of local symmetries in the polycrystalline nanostructured sample (evidence in the insets of panel B).

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Ultrafast Time Resolved Electron Diffraction at Surfaces: Watching the Atoms Freeze

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Clean crystalline surfaces and adsorbate induced reconstructions on surfaces exhibit a wealth of different phases and phase transitions. We combined a pulsed electron gun under uhv conditions with a fs laser system in a pump probe scheme to study the structural dynamics of such phase transitions on ultra-short time scales. The sample is excited with 800 nm photons with a pulse energy of 1 mJ at 5 kHz repetition rate and a fluence of 1 – 8 mJ/cm^2. Electrons of 7 – 30 keV in a reflection high energy electron diffraction (RHEED) geometry at grazing incident ensure surface sensitivity [1-3].
The huge potential of this technique is demonstrated with the non-equilibrium dynamics of the In induced (8x2) reconstruction on Si(111). This surface exhibits a Peierls-like phase transition at 130 K from a (8x2) ground state, which is accompanied by the formation of a charge density wave (CDW), to a (4x1) excited state. Upon excitation by the fs-laser pulse the (8x2) groundstate is driven into the excited (4x1) state at a sample temperature of 20 K. The surface is only excited electronically, the CDW is lifted by photo doping in less than 3 ps, but the surface remains for almost one nanosecond in a super-cooled excited (4x1) state. An activation barrier of ~40 meV for the collective motion of the In atoms hinders the immediate recovery of the (8x2) groundstate [4]. This metastable situation – far away from equilibrium – is only accessible through the ultra-fast excitation by the fs-laser pulse.
Relaxation to the (8x2) ground state is delayed on a timescale of τ ~ 500 picoseconds and is triggered by remnant (8x2) areas pinned at adsorbates that act as nucleation seeds – the same way that super-cooled water in a bottle freezes upon the insertion of seeds [5]. With increasing density θ_{ad} of adsorbates the recovery to the groundstate proceeds much faster following a $\tau \sim 1/\theta_{ad}$ law. Density functional theory calculations reveal the microscopic scenario of the (4x1)→(8x2) phase transition, which occurs one-dimensionally along the Indium chains. The surface unit cells fall back into their ground state, one at a time, like a row of falling dominoes. The phase front propagates at about 100 m/s.

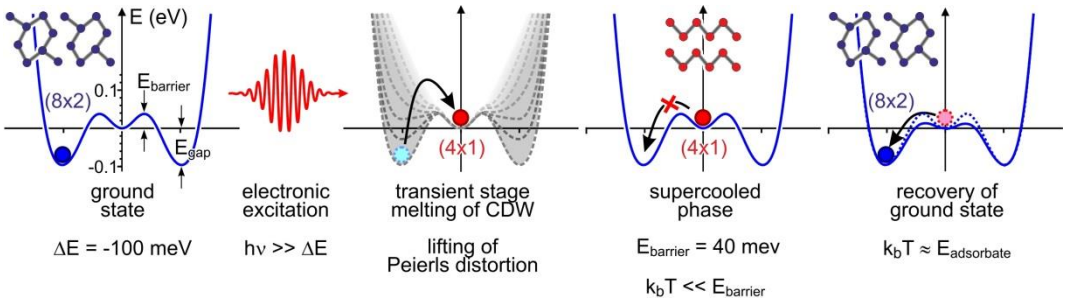

Figure 1: Potential energy landscape of (8x2)→(4x1) phase transition and the photo induced phase transition

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Using Ultrafast Optical Spectroscopy to Explore Magnetoelectric Coupling in Multiferroic Oxide Heterostructures

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Multiferroic oxides have attracted much attention in recent years, due to their potential for controlling magnetism with an electric field and ferroelectricity (FE) with a magnetic field. Existing materials, however, typically display relatively weak coupling between these parameters, making a deeper understanding of magnetoelectric coupling in multiferroics critical for optimizing their performance in applications. In the past few years, we have demonstrated that ultrafast optical spectroscopy is a unique tool for exploring ME coupling in canonical multiferroics such as $TbMnO_3$ and $BiFeO_3$. Here, I will describe more recent work extending these approaches to probe the interplay between FE and magnetic ordering in multiferroic heterostructures. These studies have revealed a long-lived, photoinduced enhancement of the FE polarization in a FE/ferromagnet (FM) heterostructure, as well as the polaronic nature of interfacial magnetic order in a FM/multiferroic heterostructure. Furthermore, our results indicate that femtosecond optical pulses can induce transient magnetoelectric coupling in a FE/FM heterostructure, with implications for high speed magnetoelectric devices. Overall, our studies demonstrate the utility of ultrafast optical spectroscopy for exploring magnetoelectric coupling in multiferroic oxides and their heterostructures.

Photocontrol of Dirac electrons in a bulk Rashba semiconductor

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Dirac electron protected by the time-reversal symmetry can be an efficient spin-current source in spintronics applications. In addition to the surface states of topological insulators, the polar semiconductor BiTe*X* (*X*=Cl, Br, I) was recently identified to host Dirac electrons with helical spin textures in *bulk*, due to its giant Rashba-type spin-orbit interaction [Fig. 1(a)] [1,2]. We demonstrate that these Dirac electrons in the bulk semiconductor can be controlled by photoexcitation through the circular photogalvanic and magneto-photo effects at room temperature. In the experiment, high-quality single crystals of BiTeBr and BiTeI, with the Fermi level close to the Dirac point in the conduction band, were prepared by the vapor transport or Bridgman method. The cleaved samples were illuminated by visible to near-infrared photons (120 fs, 1 kHz), and the resultant short-circuit photocurrent was examined. We found that: (i) Circular photogalvanic effect (PGE) emerges for the interband excitation only at the oblique incidence [Fig. 1(b)]. (ii) This photocurrent flows in the direction perpendicular to both the crystal *c*-axis and the incident plane of photons. (iii) The circular PGE changes its sign by reversing either the photon helicity or the sign of the Rashba parameter. These results indicate that the photocurrent is carried by the Dirac electrons near the Fermi level of the Rashba spin-split bands [3], which are spin-polarized even at room temperature. (iv) The circular PGE shows a distinct onset photon energy and spectral response compared to those of the linear PGE [Fig. 1(c)], revealing their specific optical transition pathways. A strong optical selection rule and possible enhancement of the PGEs would be imposed by the spin splitting both at the valence and the conduction bands. Since the PGE in the Rashba system is associated with the inverse Edelstein effect [4], in which the transient spin population in the momentum space plays a crucial role, the ultrafast optical excitation at a specific photon energy would lead to the full-control of the Dirac electrons. Furthermore, under the in-plane magnetic field (B_y), the photocurrent was found to be modified, both in amplitude and direction, through the Zeeman shift of Rashba spin-split bands [5].

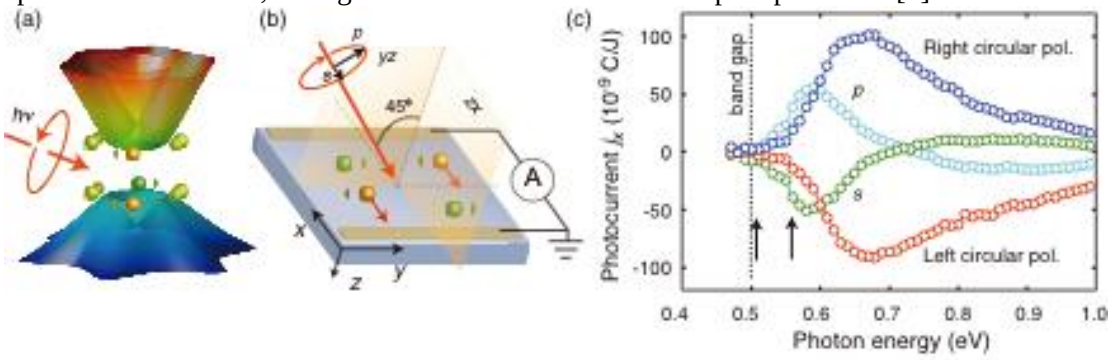

Figure 1: (a) Calculated band structure of BiTe*X* with Rashba-type spin splitting both at valence and conduction bands. Helical spin states and incident photons are illustrated schematically. (b) Experimental geometry for the oblique-incidence photoexcitation. (c) Observed circular and linear PGE in BiTeBr as a function of photon energy and polarization (band gap ~0.5 eV).

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Femtosecond mid-infrared luminescence and its layer-number dependence in graphenes

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The graphene, which has a unique symmetric linear dispersion for electron and hole, provides a promising platform for high speed electronics. From this view point, the dynamics of high energy electrons and coupled phonons have been investigated with various experimental methods, such as transient absorption[1], photoemission[2], and luminescence[3]. However, in most of the transient absorption measurements, the responses are observed at single energy in relatively high energy region (typically from 1 to 1.6 eV). Although luminescence measurements in wider energy range between 0.7 and 1.4 eV [3] are reported, the carrier dynamics below these regions has not been clarified.

In this report, we observed luminescence down to 0.3 eV, corresponding to 0.15 eV electron (see Fig. 1(a)). The sample was excited at 1.57 eV (790 nm), by 70 fs pulses at a repetition rate of 200 kHz, and the infrared luminescence signal was up-converted to visible light, analyzed by a double grating monochromator and detected with a photon counting system. The graphene samples are provided by ACS MATERIAL®.

Luminescence signals at 0.3 eV from graphenes with different layer numbers are shown in Fig. 1(b). In this figure, the layer number dependence is clearly seen, that is, the lifetime in mono-layer graphene is roughly one half of that in graphite. Figure 1(c) shows the photon energy dependence of the decay curves in bi-layer graphene. Here, the lifetime is shorter at higher photon energy, as is commonly observed in graphite [4] and other semiconductors.

We apply the two-temperature model to understand these observations. We assume that the energy of the electron system is transferred to the 0.2 eV optic phonon (G mode) as the first step and that the energy of the hot optic phonon is dissipated to the larger heat bath. This model well reproduces the behavior in graphite. Then the fast decays in bi-layer graphene are reproduced simply by adding an energy dissipation path from the optic phonon system to the substrate, having an infinite heat capacity. This suggests that the interaction with the substrate is the main cooling mechanism in silica-supported graphene.

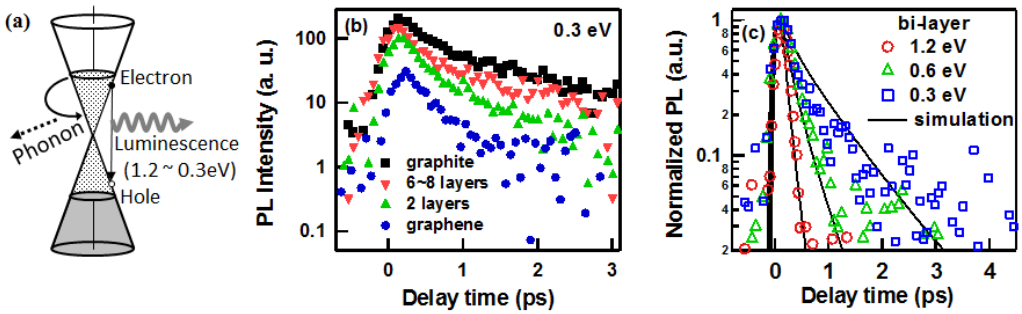

Fig. 1. (a) The Dirac cone and the radiative transition. (b) Time evolution of luminescence in mono, bi- and 6-8 layer graphenes and graphite. (c) Time evolution of luminescence in bi-layer graphene. The black curves represent the results of the two temperature model.

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Transient spin polarized current induced by femtosecond pulse excitation in topological insulators

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In three-dimensional topological insulators (TIs), nontrivial topology in the bulk insulating state results in the surface electronic state that follows the massless Dirac equation with the spin state locked by the momentum direction. Optically excited Dirac fermions provide a good platform for studying the non-equilibrium dynamics of relativistic particles interacting with electromagnetic field. Theoretical reports predict that spin-selective excitations by circularly polarized light induce an asymmetric distribution in the ***k***-space resulting in a macroscopic charge current, which has actually been observed by two-terminal sensing [1]. However, the breaking of the symmetry due to the attachment of electrodes hinders evident understanding of the mechanism. Also, time-resolved techniques are necessary for understanding the transient dynamics of the induced charge current.

In this study, we investigate the transient dynamics of the charge current under the illumination of the near-infrared circularly polarized femtosecond pulse (pulse width: 100 fs) with a contactless measurement in an intrinsic TI material $Bi_{2-x}Sb_xTe_{3-y}Se_y$ $[(x, y) = (0.5, 1.3)$ and $(1, 2)]$ [2]. The transient charge current $\boldsymbol{j}(t)$ evolved in sub-picosecond time region should produce a far-field electric field $\boldsymbol{E}(t) = \partial \boldsymbol{j}(t)/\partial t$ in terahertz (THz) range. We observed the THz waveforms with the electro-optical sampling techniques, enabling us to extract dynamical information on the charge current. Figure 1 shows horizontal (a and c) and vertical (b and d) components of the helicity-sensitive THz waveforms $\boldsymbol{E}_{\mathrm{HS}}(t) = \boldsymbol{E}_{\mathrm{R}}(t) - \boldsymbol{E}_{\mathrm{L}}(t)$ from $Bi_{1.5}Sb_{0.5}Te_{1.7}Se_{1.3}$ for the azimuthal angles $\theta = 0°$ and $-30°$, where the detected electric field induced by right-handed circularly polarized light is denoted by $\boldsymbol{E}_{\mathrm{R}}(t)$, and that induced by left-handed circularly polarized light is denoted by $\boldsymbol{E}_{\mathrm{L}}(t)$. At $\theta = 0°$ (a and b) the emitted wave is entirely horizontally polarized. In contrast, at $\theta = -30°$ (c and d), the THz emission is entirely vertically polarized. The detailed θ dependence indicates that the polarization plane of the THz wave should rotate with 3θ dependence, which apparently reflects the three-fold rotational symmetry in the crystal surface. This nontrivial dependence cannot stem from the conventional photogalvanic effect [1]. Furthermore, we conclude from the time domain analysis of the waveforms that the photocurrent flows only when the excitation light pulse exists in the sample. In the presentation, to account for the unexpected behaviours, we propose a new mechanism of photocurrent based on the formulation of the Floquet-Bloch states, where electronic band structures are asymmetrically modified by light irradiation.

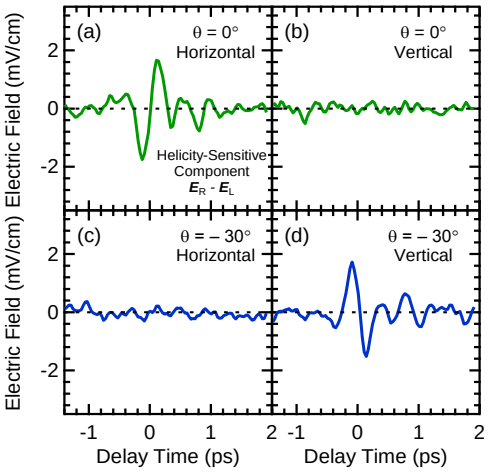

Figure 1 (a)-(d) Horizontal and vertical components of the helicity sensitive terahertz emissions from $Bi_{1.5}Sb_{0.5}Te_{1.7}Se_{1.3}$ at azimuthal angle $\theta = 0°$ (a, b) and $-30°$ (c, d).

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Imaging of photo-generated carrier dynamics in semiconductor using femtosecond time-resolved photoemission electron microscopy

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We constructed time-resolved photoemission electron microscope (TR-PEEM) utilizing a fs laser pulse in our purpose to visualize the dynamical properties of photo-excited states with spatial resolution of 40 nm and with temporal resolution of 230 fs. By utilizing a repetition-rate-variable fs laser system as an excitation source and by carefully optimizing laser parameters, electron dynamics in semiconductor can be imaged while suppressing the charging of samples. In this contribution, we introduce (1) imaging of relaxation/recombination process on a GaAs surface in the sub-ps to ns time-scales. We also demonstrate that (2) much faster recombination on nano-scale surface defects ranging from 0.5 ps to a few ps in comparison to the surrounding clean surface area. We concluded that the rate depends on the density of defect states in the forbidden band. We also show (3) imaging of electron propagation on a GaAs surface [1] as summarized in the following.

Fig. 1(a) provides experimental scheme to estimate the drift velocity and mobility of the photo-injected carriers. The pump pulse focused down to 10 μm x 20 μm on the sample surface with the energy larger than the band gap excites electrons into the conduction band. The spatial distribution of the electron density can be monitored using PEEM by locally different photoemission signal projected by the probe pulse with the energy larger than the workfunction. Fig. 1(b) and (c) are TR-PEEM images obtained at 20 ps and 40 ps after the pump pulse impinging. The bright elliptical contrast at around the middle of the images indicates the distribution of photo-generated electrons, and which is driven by the external electric field gradient between the electrodes (20 V/100 μm). Intensity profiles along the vertical direction in Fig. 1(b) and (c) indicate the motion of the electron bunch (Fig. 1(d)). TR-PEEM images are taken in 1 ps-step up to 50 ps, and the bunch position is plotted with the delay time (Δt) in Fig. 1(e). The slope given by liner fit to the data provides the drift velocity of 7.4 x 10^6 cm/s, and as a results, 3700 cm^2/Vs of mobility is figured out. This is the first observation of estimating the velocity and mobility by directly imaging the electron motion.

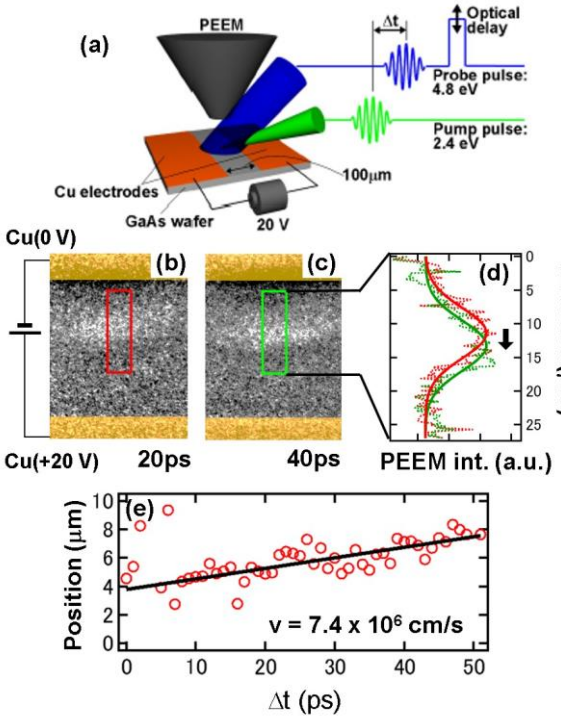

Figure 1: (a): Experimental scheme. (b) and (c): TR-PEEM images 20 ps and 40 ps after pumping, respectively. (d): intensity profiles of the red and green area in (b) and (c). (e): Position of electron bunch vs. Δt.

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Interaction quench dynamics of the Bose-Hubbard model

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We study the non-equilibrium dynamics of the Bose-Einstein condensate formed by cold atoms trapped in an optical lattice. By extending Bosonic dynamical mean field theory (B-DMFT) [1] to the real-time contour, and using a first order strong coupling expansion [2], we are able to investigate the dynamics of the super-fluid order parameter after an interaction quench.

Compared to the time-dependent mean-field approximation, where non-condensate hopping processes are discarded, the B-DMFT retains single-particle fluctuations and thereby the possibility of thermalization. For certain ranges of quench parameters we find rapid thermalization, when starting in the Mott insulator, and damped collapse and revival oscillations of the condensate fraction, when starting in the super-fluid. The condensate dynamics is reminiscent of experimental observations [3].

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Polarized femtosecond spectroscopy for quasiparticle dynamics associated with symmetry breaking in $Bi_2Sr_2CaCu_2O_{8+\delta}$

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The interplay between superconducting (SC) gap and psuedogap (PG) of cuprate superconductors is a key issue in high-T_c superconductivity. Time-resolved optical pump-probe (Pp) spectroscopy of various high-T_c superconductors has revealed dichotomous quasiparticle (QP) dynamics associated with the SC gap and PG excitations. Here the two excitations are characterized by distinct relaxation times, temperature dependencies, and/or sign of the optical signal, depending on the material, doping level, and pump-probe conditions [1-6]. In this work, by performing a concise symmetry analysis of polarized Pp experiments on high-T_c $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) and identifying the connections to Raman, we demonstrate the feasibility to investigate the QP dynamics associated with hidden broken symmetry in high-T_c superconductors.

The polarized Pp experiments using two-color (pump: 3.1 eV, probe: 1.55 eV) 120 fs pulses were performed on slightly overdoped and underdoped Bi2212. In both samples, reflectivity transients (ΔR) are divided into isotropic and anisotropic modes with respect to the probe polarization. Below T_c, the anisotropic mode has two distinct polarizations oriented at ~45° to each other, which are attributed to the SC and PG QPs. Here the anisotropic SC and PG signals are identified to be polarized along the crystalline axes and Cu-O bond directions, respectively. On the other hand, the pump polarization shows no anisotropy, indicating the totally symmetric excitations (the absence of the coherent excitations). We can thus conclude that the nonsymmetric excitations detected by the probe are generated by the presence of a spontaneous breaking of the bulk symmetry. Based on this idea, and in analogy with the electronic Raman, the SC and PG anisotropic polarizations are associated, respectively, with B_{1g} and B_{2g} like symmetries of the dielectric tensor. The polarization analysis of ΔR in detection clearly isolates the individual modes below T_c, accounting for the conclusion and identification. From the temperature dependences of the asymmetric modes, we also found the symmetry breaking to occur near onset temperature of PG. These results lead to a consequence of the underlying symmetry breaking as follows: B_{2g} symmetry breaking can originate in the weak orthorhombicity of the crystal (BiO chain ordering) while the B_{1g} symmetry breaking can be attributed to the softness of the CuO_2 planes towards stripe ordering or similar textures. The apparent absence of the SC response in the B_{2g} channel accounts for the sensitivity of the corresponding Raman vertex to the nodal direction, where the SC gap has nodes (Fig. 1). On the other hand, the presence of the PG response in B_{2g} channel indicates that the PG response can, at least in part, be associated with the nodal QPs.

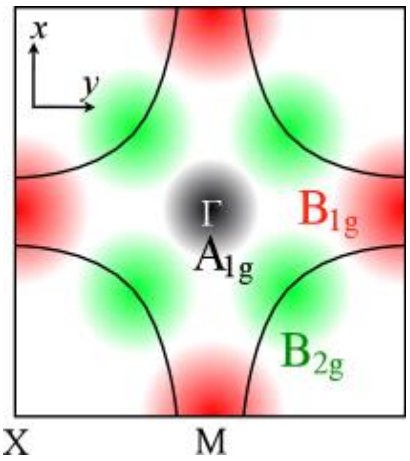

Figure 1: The *k*-space selectivity of the probe according to the Raman-like process [7]

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Light-induced coherence enhancement in cuprates

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Cuprate superconductors are layered compounds consisting of stacks of copper oxygen planes. Three dimensional superconductivity is achieved in these materials via coherent coupling between planes. Stripe order is thought to disrupt this coherence, with the most dramatic example being the complete destruction of superconductivity in the 1/8th doped lanthinum copper oxides. I will present an overview of recent results showing an enhancement of c-axis coherent coupling in two cuprate families--YBCO and LBCO--using different excitation mechanisms. Results will be discussed in the context of accompanying transient changes in structural, charge, and spin properties of these materials.

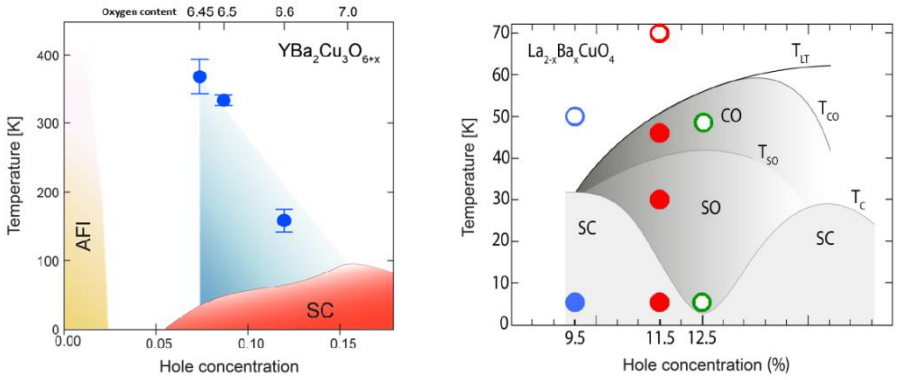

Figure 1: Phase diagrams of transient coherent transport in YBCO and LBCO. **Left panel:** Phase diagram of $YBa_2Cu_3O_{6+x}$ with blue dots indicating the temperature below which a c-axis coherent response can be induced with MIR light. **Right panel:** Phase diagram of $La_{2-x}B_xCuO_4$ with filled dots indicating the temperatures and dopings at which a c-axis coherent response was measured after excitation with 800 nm light.

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Photoinduced antinodal metallicity in the pseudogap state of high-Tc cuprates

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A major challenge in understanding the cuprate superconductors is to clarify the nature of the fundamental electronic correlations that lead to the pseudogap phenomenon. We used ultrashort light pulses to prepare a non-thermal distribution of excitations, and we performed time-resolved broadband reflectivity measurements in order to capture novel properties that are hidden at equilibrium. Our framework unveils a universal pseudogap-like region in the temperature (T) and hole-doping (p) phase diagram, delimited by a well-defined T*neq(p) line. In this region the photoexcitation process leads to a quench of local correlations triggering the evolution of antinodal excitations from gapped (localized) to metallic (delocalized) quasi-particles characterized by a longer lifetime. This photoinduced antinodal metallicity finds a natural explanation in terms of the single-band Hubbard model, in which the short-range Coulomb repulsion leads to a k-space differentiation between "nodal" quasiparticles and antinodal excitations, whose self-energy diverges as in the insulating state.

Ultrafast paired-carrier coherence control in two-leg ladder cuprate

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Controlling interplays among charge, spin, and lattice degrees of freedom in strongly correlated electron (SCE) systems by light is an important topic in condensed matter physics because the emergence of novel photoinduced phenomena makes us expect the creation of a new field involving both fundamental material science and device applications. Copper oxides with a perovskite structure are typical example of a SCE system and those photoinduced phenomena are extensively investigated, as exemplified by high-T_c cuprates. Among this family, ladder-type cuprates have a unique nature of the carrier conductivity, which is a key nature to understanding the high-T_c superconducting mechanism [1,2]. The paired carriers theoretically predicted to be responsible for the superconductivity and the related phenomena [1].

In this study, we investigate the unique carrier nature in two-leg ladder superconducting cuprate $Sr_{14-x}Ca_xCu_{24}O_{41}$ in terms of femtosecond time-resolved spectroscopy and theoretical simulation. We demonstrate not only the photoinduced insulator-to-metal transition with a gigantic reflectivity change [3] but also the opposite phenomenon, i.e. photoinduced *metal-to-insulator* transition, by tuning the paired-carrier coherence which plays an essential role of the carrier conductivity in the ladder system.

In the experimental study, we used the parent compound of $Sr_{14}Cu_{24}O_{41}$(SCO) which shows the charge-density wave in the ladder below 210 K and the hole-doped metallic compound of $Sr_4Ca_{10}Cu_{24}O_{41}$ (SCCO). Immediately after photoexcitation of the resonant with the Cu-O charge transfer excitation on the CDW state in SCO, the large Drude response appeared, indicating the photoinduced CDW-to-metal transition [3]. In contrast to this, we observed the ultrafast suppression of the inherent Drude weight in the hole-doped SCCO. The theoretical simulation results with the two-leg ladder Hubbard model indicate that the observed Drude weight suppression response is ascribed to the destruction of the long-range singlet hole pair correlation corresponding to the coherence among the hole pairs by the photo-generated carriers and the resultant suppression of the metallicity in the system. By the sequential photoexcitation, we can realize the successive photo-tuning of the paired-hole coherence in SCO as the enhancement and suppression of the metallicity. The paired-carrier coherence will be new candidate for degree of freedom in SCE system.

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Ultrafast dynamics of photoinduced Mott insulator-metal transition in an undoped 2D cuprate Nd_2CuO_4

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A number of perovskite-type transition-metal oxides show Mott-insulator to metal transition induced by chemical carrier doping. A photocarrier doping may also cause similar Mott-insulator to metal transition. We have been investigating such photoinduced transitions to metal phases in various kinds of Mott insulators. Recently, we have reported photoinduced Mott-insulator to metal transition in undoped 2D cuprates Nd_2CuO_4 and La_2CuO_4 [1, 2], which are famous as parent materials of high-T_c superconductors. In this paper, we report the initial dynamics of photoinduced Mott-insulator to metal transition in Nd_2CuO_4 by using pump-probe spectroscopy with the time resolution of ~10 fs.

Visible pulse with the duration of ~7 fs was generated by using a non-collinear optical parametric amplifier excited with an output of a Ti: sapphire regenerative amplifier (795 nm, 130 fs). The obtained pulse was divided into pump and probe pulses. The photon energy (1.7-2.3 eV) corresponds to the charge transfer (CT) transition from the O 2p valence band to the Cu 3d upper Hubbard band. Gray shades in Figs. (a) and (b) show the cross-correlation between the pump and probe pulses measured with a β-BaB_2O_4 crystal.

The red line in Fig. (a) shows a time evolution of reflectivity change ($\Delta R/R$) by the CT excitation in Nd_2CuO_4 at 294 K. Excitation photon density (x_{ph}) is ~3.3×10^{-3} photons/Cu. The negative value of $\Delta R/R$ is due to the spectral weight transfer from the CT transition to the mid-gap absorption due to localized photocarriers [2]. After the initial decrease of the reflectivity, an additional slow decrease with the time constant of ~20 fs is observed. Possible origin of this slow component is the destruction of the spin order due to the spin-charge coupling, which is characteristic of 2D Mott insulator.

In the case of the strong excitation x_{ph}~0.055 photons/Cu, on the other hand, the absolute value of $\Delta R/R$ reaches the maximum immediately after the photo-excitation as shown in Fig. (b). This ultrafast decrease of the reflectivity is attributable to the photoinduced metallic state [2]. The analyses of the excitation-photon-density dependence of $\Delta R/R$ reveal that the decay time of the photoinduced metallic state is about 20 fs. Such an ultrafast decay of the metallic state suggests that the excess energy generated by the photocarrier recombination is rapidly transferred to the spin system by emission of magnons via the spin-charge coupling.

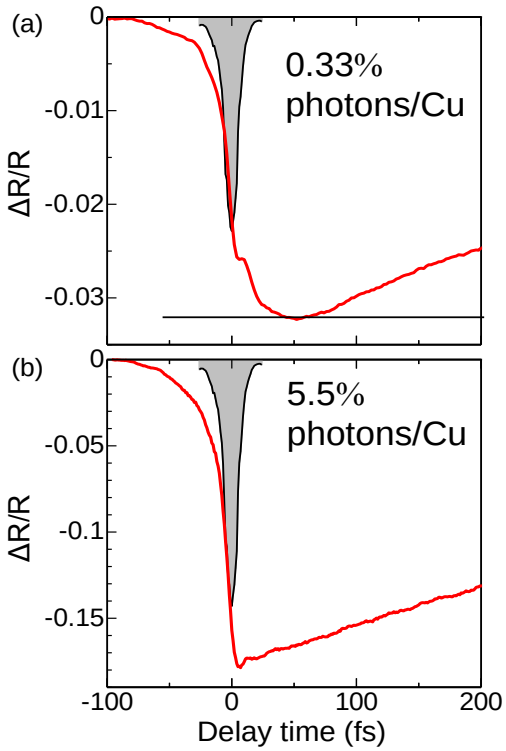

Figure: Time evolutions of $\Delta R/R$ for (a) x_{ph}~3.3×10^{-3} and (b) x_{ph}~0.055 photons/Cu. Gray shades are the cross-correlations between pump and probe pulses.

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Mechanisms of nonthermal destruction of the superconducting state and melting of the charge-density-wave state by femtosecond laser

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We use the femtosecond spectroscopy to study the processes leading to nonthermal condensate destruction and charge-density wave (CDW) melting. Systematic measurements have shown that the destruction is relatively slow (tau ~1 ps) and inefficient in superconductors, exhibiting a strong systematic dependence of the destruction energy U_v on T_c. In contrast, melting of CDW order proceeds rapidly (tau = 50~200 fs) and more efficiently. We propose a model based on a phonon-mediated quasi-particle bottleneck mechanism for the observed behaviour in superconductors whereas the CDW melting is explained in the frame of the Fermi surface disruption by hot quasiparticles.

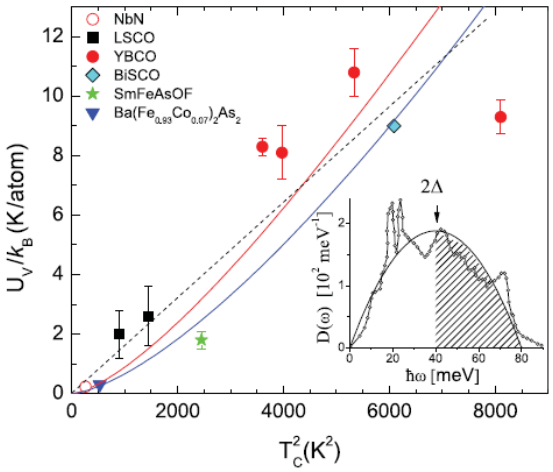

Figure 1: Destruction energy U_v expressed in K pre planar Cu as a function of T_c^2 for the cuprates. The data for NbN, SmFeAsOF and Ba(Fe,Co)As_2 are included for comparison. The dashed line is a square law $U_v=\eta T_c^2$. The inset shows the phonons with $\hbar\omega>2\Delta$ which can break pairs. The measured phonon density of states for YBCO is approximated by a parabola.

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Dynamics of fluctuations in high temperature superconductors far from equilibrium conditions

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Despite the extensive literature on high temperature superconductors, the critical dynamics of an incipient condensate has so far been studied just in equilibrium conditions. Here, I show that resolved THz measurements of $Bi_2Sr_2CaCu_2O_{8+\delta}$ discriminate the temperature regimes where superconductivity is coherent, fluctuating or vanishingly small. Above the transition temperature T_c the recovery to equilibrium conditions displays power law behaviour and scaling properties. The experimental evidence that some of the exponents weakly depend on doping level provide hints of universality in systems far from equilibrium. We find partial agreement between the scaling law of the optimal doped sample and the Time Dependent Ginzburg-Landau (TDGL) model. Inherent limits of TDGL call for non-equilibrium field theories treating fast degrees of freedom and fluctuations on equal footing. These results open a timely connection between superconducting condensates and Bose-Einstein condensates of ultra-cold atoms.

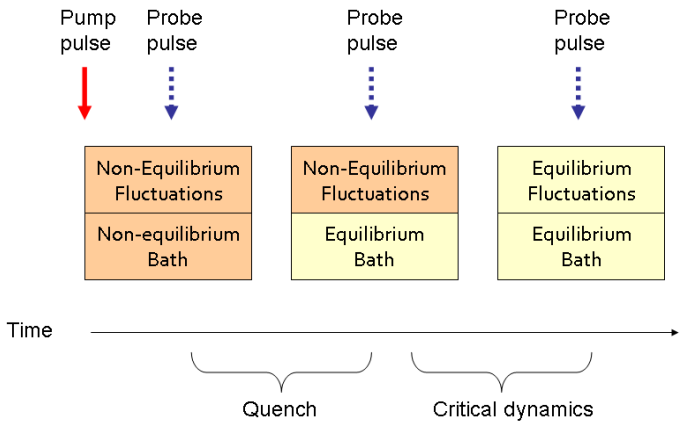

Figure: Fluctuations dynamics for temperature slightly above T_c. First, an optical pump pulse brings the system out of equilibrium conditions. Subsequently, the dissipation induced by the nuclear lattice rapidly quenches the fast degrees of freedom (dubbed bath). From here on, the fluctuations of the order parameter follow a critical relaxation.

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On the contribution of Mott and Peierls instabilities to the semiconductor-metal transition in VO_2

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The complex interplay between strong electron-electron correlations and structural distortions is thought to determine the electronic properties of many oxides, but the respective role of the two contributions is often extremely difficult to determine. Vanadium dioxide (VO_2) is a particularly notorious example. We will report on combined radio-frequency compressed ultrafast electron diffraction[1] (RF-UED) and infrared transmissivity experiments in which we directly watch and separate the lattice and charge density reorganizations that are associated with the optically induced semiconductor-to-metal transition in VO_2. These studies have uncovered a previously unreported photoinduced transition to a metastable intermediate state with the periodic lattice distortion characteristic of the insulator intact, but differing by a 1D rearrangement of charge density along the octahedrally coordinated vanadium dimer chains and a transition to metal-like mid IR optical properties. The results demonstrate that UED is capable of following details of both lattice and electronic structural dynamics on the ultrafast timescale.

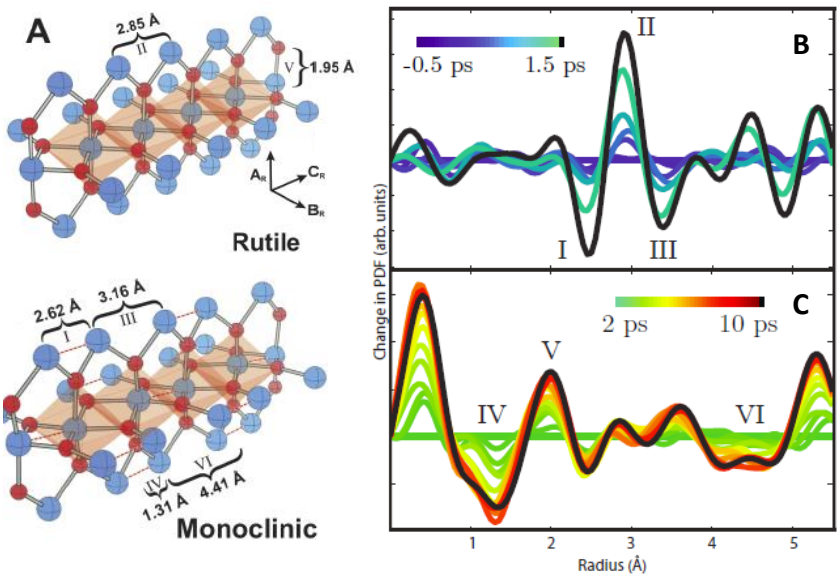

Fig. 1 A) The structure of rutile VO_2 (top) and monoclinic VO_2 (bottom). B) and C) Photoinduced structural dynamics in the VO_2 unit cell. RF-UED measurements of changes to the autocorrelation function of the electrostatic crystal potential. B) Due to the reorganization of lattice structure (the breaking of vanadium dimer bonds). C) Due to a 1D change density reorganization along the octahedrally coordinated vanadium dimer chains (c_R). Peak labels in B) and C) refer to those indicated in panel A).

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Metallization in an instant – The photoinduced phase transition of VO_2

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Even after many decades of research on the transition metal oxide VO_2, the physical nature of the insulator-metal transition (IMT) activated by temperature or light is still highly controversial. From an electronic structure point of view the IMT involves the closing of the insulating band gap and the formation of a metallic electron distribution at the Fermi energy E_F. Since the IMT is accompanied by a structural distortion from monoclinic to rutile symmetry several time-resolved experiments have been conducted in order to elucidate the role of the lattice in the photoinduced transition [1-3]. Other investigations focussed on photoinduced changes of the dielectric function at various wavelengths using THz [4], near-infrared [1,5] and visible radiation [5]. However, the dielectric function does not necessarily reflect the electronic structure close to E_F.

In order to directly investigate the changes of the electronic structure across the IMT of photoexcited VO_2 at E_F, we conducted laser-based time-resolved photoelectron spectroscopy (PES) experiments. In a first step, we verified the closing of the insulating gap and the formation of a Fermi-Dirac distribution around E_F upon heating the sample above 340 K. We monitored the *photoinduced* ($h\nu_{pump}$ = 1.55 eV) electron dynamics near E_F from femtosecond timescales up to 400 ps. As shown in Fig. 1(a), the photoinduced *change* of the photoemission spectra 1 ps after excitation exactly resembles the metallic electron distribution observed in the high temperature rutile phase. This verifies that photoexcitation promotes VO_2 into a transient metallic state within 1 ps – a timescale often chosen in the literature to determine the critical excitation density for the IMT. As illustrated by Fig. 1(b), we find that the photoexcitation of electrons from the valence to the conduction band leads to the instantaneous collapse of the insulating gap and redistribution of spectral weight. Subsequently, the hot carriers thermalize on a timescale of $\tau_h \approx \tau_e \approx 200$ fs forming the spectral function of the high-temperature rutile phase. We find no evidence of coherent phonon oscillations and the observed signatures remain unchanged as the lattice transforms to the rutile phase at later times [1-3]. That is, the electronic structure of photoexcited VO_2 becomes metallic on ultrafast timescales, before any ionic motion takes place and even before the hot carriers relax. We thus conclude that the photoinduced transient phase of VO_2 right after photoexcitation is a monoclinic metal and that the electronic transition clearly precedes the crystallographic one.

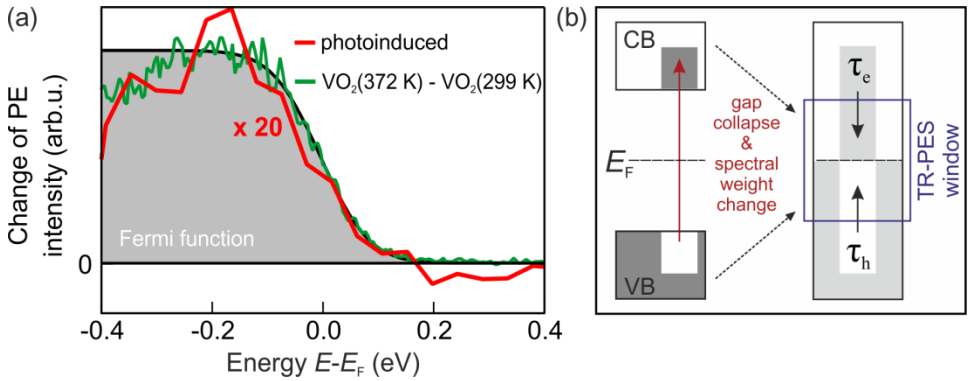

Figure 1: (a) Comparison of the change in PE intensity induced by temperature (green) and photoexcitation (red) and a corresponding fit using the Fermi-Dirac distribution. (b) Sketch of the elementary processes during the photoinduced IMT (see text).

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Relaxation dynamics of photo-induced carriers studied within dynamical mean-field theory

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The early dynamics of photo-excited carriers in correlated materials can reveal coupling of electrons to spin and lattice degrees of freedom, and thus shine light on the origin of complex phases in those systems [1]. On the other hand, having different scattering channels can be used as a tool to direct ultra-fast processes in photo-excited solids in a controlled fashion. We use the nonequilibrium extension of dynamical mean-field theory [2] to study the dynamics of carriers in Mott-insulators. We will first focus on the single-band Mott insulator, where scattering with spin degrees of freedom provides the dominant relaxation channel. While the relaxation in the paramagnetic phase (at high-temperatures) is found to be very slow (slower than the accessible times in the simulation), the presence of an antiferromagnetic background leads to relaxation of the kinetic energy of photo-excited carriers on the timescale of a few hoppings (see Figure 1). This fact can be exploited in the design of functional devices based on correlated hetero-structures: using a real-space nonequilibrium DMFT simulation, we have demonstrated that scattering with antiferromagnetic fluctuations can support ultrafast separation of carriers in Mott-insulating hetero-structures [3]. An important extension of these simulations is the incorporation of non-local correlations (using nonequilibrium dynamical cluster approximation, DCA), which allows to probe the role of short-range spin fluctuations for the carrier dynamics in the paramagnetic phase.

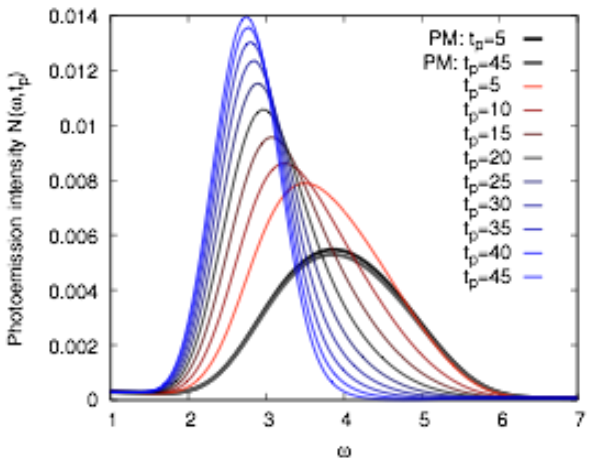

Figure 1: Time-resolved photo-emission spectrum computed for the Mott-insulating phase of the single-band Hubbard model (hyper-cubic lattice, U=8), as a function of frequency and probe time t_p (measured in units of the inverse hopping). The curves show the spectrum of the upper Hubbard band, which is initially unoccupied and then populated by an intense few-cycle laser pulse. In the antiferromagnetic phase, we observe redistribution of the weight within a few hopping times due to scattering with spins (coloured lines), while no substantial redistribution is observed in the paramagnetic phase (bold lines).

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Dynamic scaling of the insulator to metal transition in high quality V_2O_3 thin films

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Insulator-metal transitions (IMTs) are a striking manifestation of the interactions between the various degrees of freedom in complex materials. Vanadium sesquioxide (V_2O_3) is a prototypical IMT material, transitioning from an antiferromagnetic insulator to a paramagnetic metal at T_{IMT}~170K. We present a detailed investigation of the IMT dynamics in thin films of V_2O_3 following optical excitation, measured using THz time domain spectroscopy. Far infrared conductivity dynamics induced below T_{IMT} by ultrafast photoexcitation can be described by nucleation and growth of the metallic volume fraction, which eventually gives rise to the full metallic state of V_2O_3 on a tens of picoseconds timescale. The conductivity rise time is seen to vary with both the initial temperature and the incident pump fluence, revealing a temperature dependent scaling of the nucleation and growth dynamics in V_2O_3 following an ultrafast temperature quench. We will discuss our results in the broader context of phase transition dynamics in the vanadates and related strongly correlated materials.

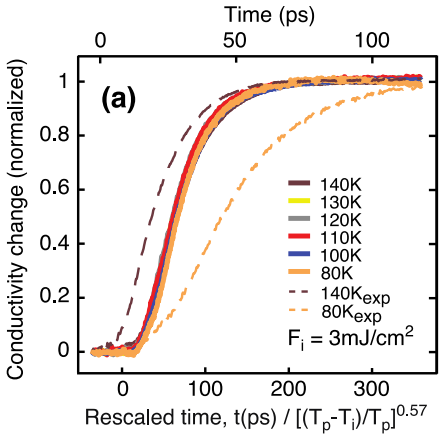

Figure 1: Scaling of the optically induced conductivity dynamics in thin films of V_2O_3 for initial temperatures, T_i, below T_{MIT} = 175K, and for an incident pump fluence F_i = 3mJ/cm^2. Two unscaled curves are shown as dashed lines, corresponding to the top time axis.

Insulator to metal transition induced by an electronic avalanche in the narrow gap Mott Insulators AM_4Q_8 (A=Ga,Ge; M = V, Nb, Ta ; Q = S, Se)

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The narrow gap Mott insulators AM_4Q_8 (A= Ga, Ge; M=V,Nb,Ta ; Q= S,Se) exhibit very interesting electronic properties when pressurized or chemically doped [1]. Optical reflectivity measurements under pressure reveal that this compounds display a prototypical Mott insulator to metal transition.[2] We recently discovered that the application of short electrical pulses is able to drive these compounds out-of-equilibrium and to induce a new phenomenon of volatile or non-volatile insulator to metal transition [3].

The volatile transition appears above threshold electric fields of a few kV/cm, while for higher electric fields, the resistive switching becomes non-volatile. All our results indicate that the resistive switching in the AM_4Q_8 compounds does not match with any previously described mechanisms based on joule heating or electrochemical phenomena [4]. Conversely, our recent works show that the electric-field-induced volatile resistive switching is a direct consequence of an electronic avalanche phenomenon in a Mott insulator [5]. This avalanche breakdown induce the collapse of the Mott insulating state at the local scale and ultimately lead to the formation of a granular conductive filament formed by compressed metallic domains and expanded “superinsulating" nanometric domains [6].

In the non volatile regime, the application of successive very short electric pulses enables to go back and forth between the high and low resistance states. This reversible resistive switching was observed down to 30 nm using an STM tip [6], or on metal-insulator-metal (MIM) devices made of thin layers of GaV_4S_8 which proves the high potential of this new class of Mott-memories for applications [7].

Besides the application of pressure and electric pulses, we have also attempted to destabilize the Mott insulating state of the AM_4Q_8 compounds using ultra-fast light pulses. Results from preliminary optical pump-probe experiments will be presented and discussed in relation with measurements under pressure and electric field.

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Ultrafast modulation of the chemical potential in $BaFe_2As_2$ by coherent phonons

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Time-resolved optical and photoemission spectroscopies have become important tools to probe the microscopic details of electron-phonon coupling. A particularly intriguing aspect of electron-phonon coupling often observed in pump-probe spectroscopy is the generation of coherent optical phonons and their subsequent modulation of electronic properties. This effect not only provides a powerful means to study femtosecond lattice dynamics, but can also be used to coherently control the electronic structure of materials. Since the electrons with the lowest binding energies determine material properties and collective phenomena, the physics will become particularly interesting if transient band shifts and renormalizations are induced near the chemical potential, which itself may then have to adjust to preserve charge neutrality.

In this presentation I will report on the electronic structure dynamics in $BaFe_2As_2$ as monitored around the high-symmetry points Γ and M by means of time- and angle-resolved extreme ultraviolet photoemission spectroscopy. A momentum-independent (global) oscillation of the Fermi level at the frequency of the A_{1g}(As) phonon mode is observed (see Figure 1). We assign this behavior to a modulation of the effective chemical potential in the photo-excited surface region that arises from the high sensitivity of the band structure of $BaFe_2As_2$ near the Fermi level to the A_{1g}(As) phonon mode combined with a low electron diffusivity perpendicular to the layers. The results establish a novel way to tune the electronic properties of iron pnictides: coherent control of the effective chemical potential. The results further suggest that the equilibration time for the effective chemical potential needs to be considered in the ultrafast electronic structure dynamics of materials with weak interlayer coupling.

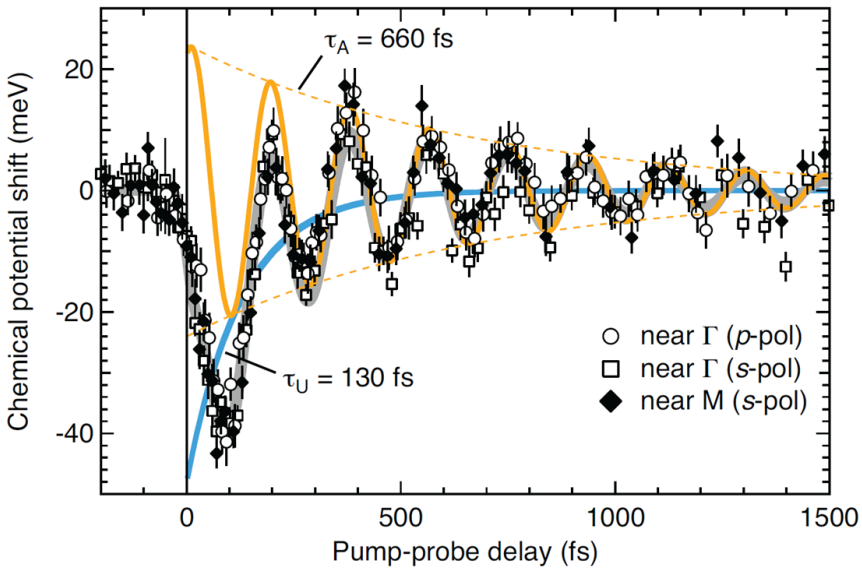

Figure 1: Transient shift in the chemical potential of $BaFe_2As_2$ at Γ and M extracted from trARPES data with s- and p-polarized 22.1 eV probe pulses. The oscillatory behavior is due to the coherent excitation of the A_{1g}(As) phonon mode.

Ultrafast dynamics of insulator-to-metal transitions probed by time-resolved ARPES

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The electronic properties of complex materials are often governed by electron-phonon coupling and many-body correlation effects leading to the formation of broken symmetry ground states. One example are charge-density wave (CDW) materials where at low temperatures a periodic lattice distortion leads to an opening of an electronic gap at the Fermi surface. We use time- and angle-resolved photoemission spectroscopy (trARPES) for a systematic study of the photoinduced non-equilibrium phase transition in the tri-telluride CDW system (RTe_3, R=Te, Ho, Dy) and probe directly the resulting transient evolution of the electronic structure and collective phonon dynamics of the system through their influence on the quasiparticle band structure. In particular, we can directly map the transient changes of the Fermi surface and the opening and closing of the CDW gap (see Fig.1).

Furthermore, we have performed the first direct trARPES measurement of the spectral function of VO_2 during the photoinduced insulator-to-metal transition and find a quasi-instantaneous band gap collapse upon photoexcitation. This ultrafast metallization occurs even faster than hot carrier relaxation and thus also before significant lattice rearrangement has occurred suggesting that the abrupt collapse of the electronic gap is independent of the changes of the crystal lattice structure.

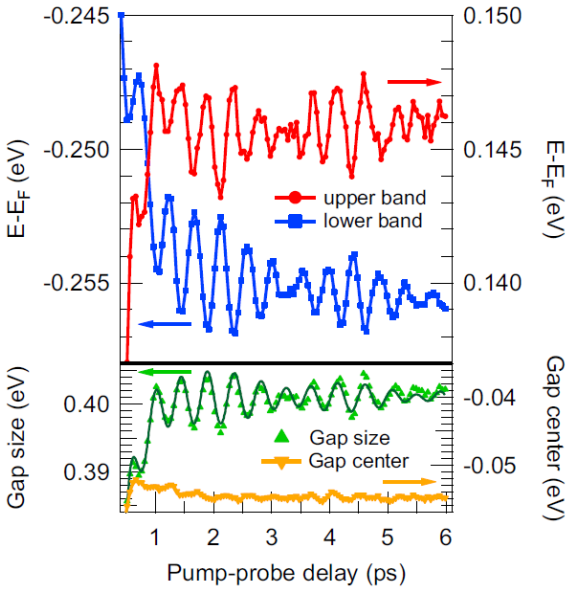

Fig.1 Transient evolution of the CDW gap in $TbTe_3$ probed by time-resolved ARPES. Top: Peak positions of lower (blue) and upper (red) CDW band show an antipodal oscillation. Bottom: The order parameter (CDW gap size) is modulated by the amplitude mode motion at frequencies of 1.75 THz and 2.2 THz.

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Ultrafast Metamorphosis of a Complex Charge-Density Wave

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Complex intertwined electronic and lattice orders are a hallmark of strongly correlated materials. Femtosecond time-resolved optical[1-2] and diffraction techniques[3-5] have recently contributed many important insights into their origin and nature by tracking the photo-induced suppression (melting) of the low temperature order and its recovery.
Here, using femtosecond electron diffraction,[3,5] we directly follow the formation of a new order: the emergence of an incommensurate charge density wave (IC) upon quenching a nearly commensurate (NC), domain-like charge-density wave in the drosophila of ultrafast dynamics 1T-TaS_2. Photo-excitation with an intense near-infrared femtosecond pulse triggers a multi-step process. After a suppression of the old order, which takes place on a sub-picosecond timescale, the IC order nucleates at the domain walls of the NC phase within about one picosecond. Following this rapid nucleation the IC domains grow on the 10-100 ps timescale. For the highest excitation densities the transformation is complete within 50 ps, about six-orders of magnitude faster than any commensurate-incommensurate first order phase transition studied thus far and serves as a reference for how complex coupled electron-lattice orders are transformed on the ultrafast timescale.

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Ultrafast recovery of the CDW phase in $TiSe_2$ due to electron-hole scattering

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Transition metal dichalcogenides are a family of quasi-two dimensional materials, which have often been studied for their charge density wave (CDW) phases and more recently for applications in atomically thin electronics [1]. Among those, TiSe2 offers a puzzling case, since the mechanism responsible for its CDW transition is still debated.
Here we present a time-resolved and angle-resolved photoemission spectroscopy (trARPES) study of the charge density wave (CDW) phase of $TiSe_2$. trARPES measurements were performed at temperatures in the CDW phase and above the phase transition, where CDW fluctuations are intense. Comparison of these two sets of data emphasizes that a similar mechanism is responsible for the suppression of the CDW phase and its fluctuation phase. Photoinduced melting of the CDW phase occurs on an ultrafast time scale, shorter than 200 fs [2]. A detailed analysis of the recovery of the CDW band structure after photoexcitation reveals a slow and a fast component. The fast component, with a relaxation time of about 100 fs, is identified as the signature of strong electron-hole scattering, specific to the semi-metallic band structure of $TiSe_2$ [3]. We conclude that this electron-hole scattering drives the electronic instability leading to the CDW phase transition in $TiSe_2$.

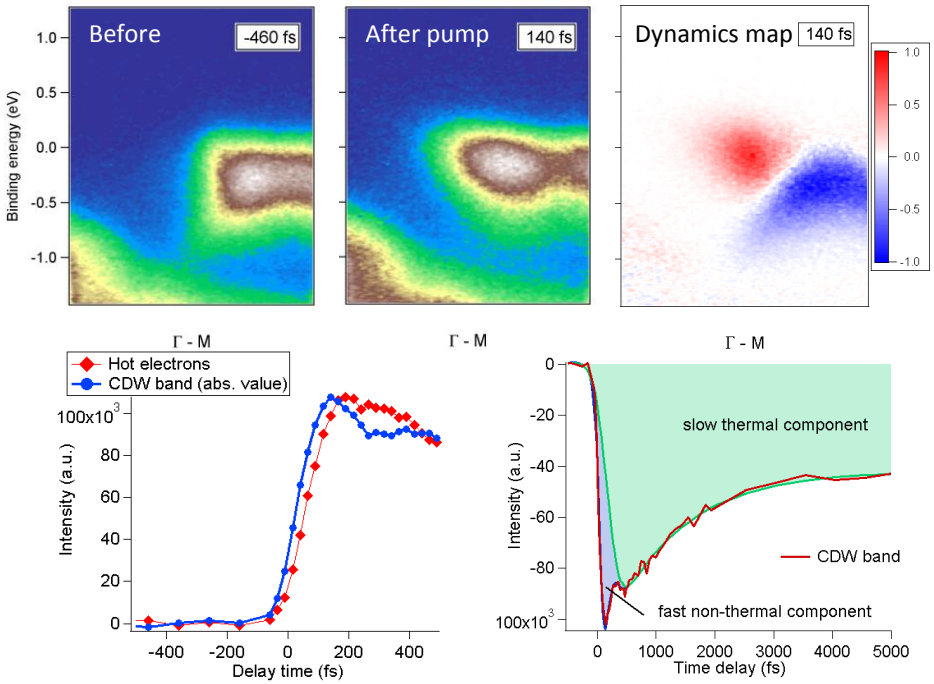

Figure 1: Top: trARPES data of TiSe2 taken at 30 K at the border of the Brillouin zone. Bottom: (left) transient photoemission intensities of the hot electrons and CDW band (30 K); (right) decomposition of the transient photoemission intensity of the CDW band in a fast and a slow component (30 K).

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Photo-induced dynamics in frustrated charge ordered systems

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Charge ordered (CO) states at quarter filling are ubiquitously observed in a number of materials, such as organic compounds, transition-metal oxides, heavy fermion systems and so on. Some charge ordered materials show geometrical frustrated crystal, e.g. two-dimensional triangular lattice. Previous theoretical studies have found competitions between several CO patterns and exotic metallic phase due to the frustration. Recently optical Pump-probe experiments using the ultra-fast laser pulse have been applied to the CO states with geometrical frustration, and have shown CO melting and the photo-induced insulator-to-metal transition in many CO systems [1]. Recent developments of pulse laser techniques, and time-resolved quantum beams diffraction and spectroscopy techniques, enable us to study not only the transient optical responses, but also direct observations of the transient electronic structures. Thus, the nonequilibrium electronic structures are now reinvestigated from the modern viewpoints.

We study photo-induced nonequilibrium dynamics in CO systems based on the two-dimensional spinless Vt models in a triangular lattice where nearest neighbour Coulomb interactions and the electron transfers are taken into account. We use the exact diagonalization to evaluate quantum and many-body correlation effects exactly in finite size clusters. Before photo-excitation, three different CO states are identified: the horizontal, vertical and 3-fold COs [2], characterized by the momentum dependences of the charge correlation functions. We calculate the real-time dependences of the charge correlation functions in a wide parameter region. In all CO states the primary charge correlation functions for the initial CO states decrease after photo-excitations, corresponding to the photo-induced CO melting. In addition, in the horizontal CO states, enhancement of the 3-fold CO correlation is remarkable, and the largest CO correlation is changed from the horizontal into the 3-fold one. This is the photo-induced CO phase transition. On the other hand, in the vertical CO states, we have no remarkable enhancement for any CO correlations after photo-excitation, that is the CO to metal phase transition. Finally, in the 3-fold CO states, the 3-fold CO structure is remained after photo-excitation. We will also talk details of the numerical results for the transient optical responses.

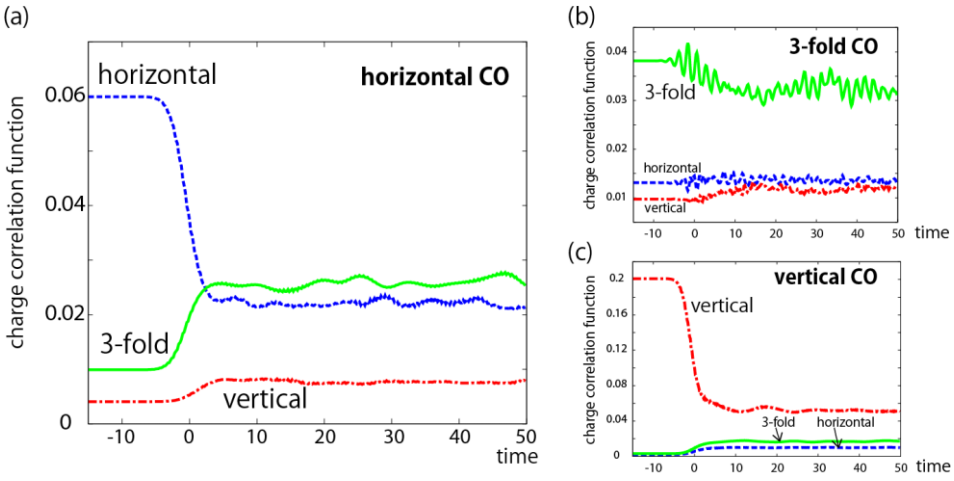

Figure 1: Time dependences of charge correlation functions in (a) horizontal CO, (b) 3-fold CO, and (c) vertical CO states. Red, green and blue lines represent the vertical, 3-fold and horizontal charge correlation functions respectively.

References:
[1] S. Iwai, Crystal 2, 301 (2012)
[2] C. Hotta, et al., JPSJ 75, 123704 (2006)

Non-equilibrium electronic structure of transient states in solid materials driven by femtosecond laser pulses

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Optical excitations in solid materials typically decay on femto- to picosecond time scales due to elementary interactions which lead to a redistribution of the excess energy among the electronic, the lattice, and the spin subsystem, before final dissipation. In the thermodynamic ground state interactions which compete with thermal excitations result in pronounced instabilities, fluctuations, and phase transitions. Here, we analyse the electronic structure of such materials like charge or spin density waves and Mott insulators in a highly non-equilibrium state by femtosecond time- and angle-resolved photoemission. Our results highlight (i) screening of local interactions to obtain a quantitative description of the ultrafast relaxation dynamics and (ii) the coexistence of ordered and disordered constituents under non-equilibrium conditions. The latter finding suggests such non-equilibrium studies as means to probe fluctuations in a superheated state under extreme conditions, which bear the potential to stabilize novel metastable non-equilibrium states.

Quantum excitation and time-resolved PES in charge-density-wave insulators

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The simplest model for a charge-density-wave insulator is a two-band model, where there is a constant site potential arranged on one of the two sublattices of a bipartite lattice. The nonequilibrium response of this system can be found exactly by evaluating the time-ordered product for the evolution of the system as a direct product of two-by-two matrices for each coupled pair of momenta, which are found by using the Trotter formula. We use the exact solution of this problem to study two different types of experiments. The first is the quantum excitation problem, which measures the excitation of electrons from the lower to the upper band as a function of the frequency and amplitude of the driving electric field[1]. The second is the nonequilibrium melting of a CDW insulator as observed with time-resolved PES, where we see the transient closing of a gap, while the order parameter remains finite[2]. In Fig. 1 (a), we show a "spectroscopy" for the quantum excitation, where we plot the number of electrons excited by a modulated Gaussian pulse as a function of the modulation frequency and the amplitude of the field. For low amplitude, absorption requires the frequency to be higher than the gap, while for larger amplitude we first see multiphoton processes followed by complex oscillations. In Fig. 1(b), we plot a false color plot of the time-resolved PES as a function of the delay time between the pump and the probe. One can see the transient melting of the gap while the pulse is active, and its reforming at longer times.

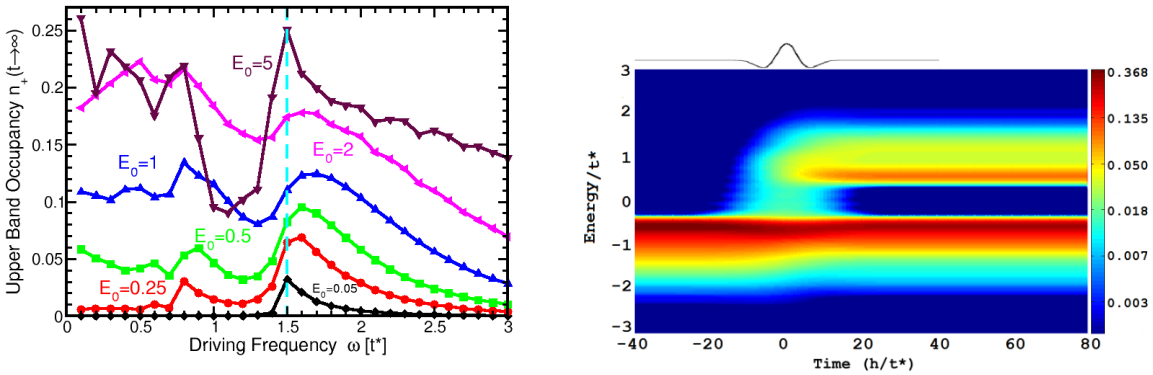

Figure 1: (a) Quantum excitation spectroscopy. Dashed line shows the location of the CDW gap. (b) False color plot of time-resolved PES.

References:

[1] Wen Shen, T. P. Devereaux, and J. K. Freericks, "Beyond Planck-Einstein quanta: amplitude driven quantum excitation," arXiv:1309.2723 (submitted to Phys. Rev. Lett.).
[2] W. Shen, Yizhi Ge, A. Y. Liu, H. R. Krishnamurthy, T. P. Devereaux , and J. K. Freericks, "Nonequilibrium "melting" of a charge density wave insulator via an ultrafast laser pulse," arXiv:1308.6066 (to appear in Phys. Rev. Lett.)

Electronic relaxation in a metal excited by an ultrashort optical pump

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Two temperature model (TTM) and its modifications are very often used for the description of the relaxation of photoexcited electrons in metals in spite of well-known experimental and theoretical constrain of applicability of these models. In this talk I discuss the theory of the electron relaxation in simple metals excited by an ultrashort optical pump. The theory based of the solution of the linearized Boltzmann kinetic equation, where both the electron-electron and the electron-phonon collision integrals are included. The TTM follows from the theory as the limiting case when the thermalization due to the electron-electron collisions is fast with respect to the electron-phonon relaxation. It is demonstrated that the energy relaxation has two consecutive processes. The first and most important step describes the emission of phonons by the photoexcited electrons. It leads to the relaxation of 90% of the energy before the electrons become thermalized among themselves. The second step describes electron-phonon thermalization. The second stage is difficult to observe experimentally because it involves the transfer of only a small amount of energy from electrons. Thus the theory explains why the divergence of the relaxation time at low temperatures has never been observed experimentally.

Photoinduced Electronic Domain Formation and Its Many-body Properties Expected for a Two-dimensional Charge-ordering System

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One of the important aspects of photoinduced phase transitions (PIPTs) is the ultrafast nature observed typically for strongly correlated electron systems. Its typical time scales are femto seconds, that are, the time scales of electron transfers. We interpret this phenomenon as a photoexcitation of an electronic domain and focused on its properties as an elementary excitation with spatial extent, confining the investigations to one-dimensional systems so far [1].

We here report our new results obtained recently for a two-dimensional system. Motivated by a discovery of a ultrafast PIPT in a molecular solid, α-(BEDT-TTF)$_2$I$_3$, from its charge-ordered (CO) ground state [2], we investigate a spinless fermion model on a square lattice, focusing on charge degrees of freedom of an original quarter-filled system. Another particular viewpoint is the so-called frustration, incorporated into this model by the addition of a next-nearest repulsion term. We have special interest in the effect of the frustration on the domain formation, which is accurately treated by exact diagonalization for a 6×6 lattice. In our scenario, the feature of the ultrafast PIPT appears among a special type of optically excited states. First, they have a nature of multi-electron excitations. We emphasize that they are induced by only one photon, and hence can be identified in the ordinary linear optical conductivity spectrum. In Fig. 1 (a), we show the spectrum calculated for a combination of the two parameters, V and V', which are nearest-neighbor and next-nearest-neighbor repulsions scaled by the transfer energy. As is specified in the spectrum by hatching, a part exhibits numbers of electron excitations, N_{ex}, that exceed three (see the examples in Fig. 1 (b)), keeping finite spectral intensities. We also remark that such part has emerged by the introduction of V', suggesting the role of the frustration for the domain formation.

In addition to the multi-electron excitation, we analyze the domain excitations from other viewpoints. For example, photoinduced changes in charge-correlations show a large suppression of the original checkerboard type of CO and a meaningful appearance of another type of CO (so-called three-sublattice structure). We also calculate photoinduced spectra based on obtained excited states, and find that the electronic structures are not only significantly reconstructed as a result of one-photon absorption but also show metallic features for the domain excitations. These results demonstrate the importance of the domain excitations and are interpreted to have direct connection to the observed ultrafast PIPTs in the material.

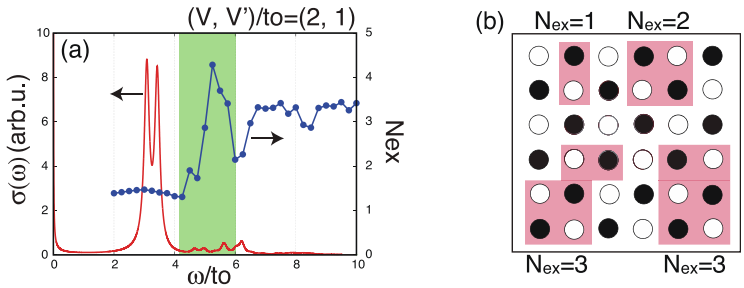

Figure 1: (a) Optical conductivity spectrum and frequency dependent N_{ex} . (b) Various domains in the lattice.

References:

[1] K. Iwano, Phys. Rev. Lett. **97**, 226401 (2006) and K. Iwano, Phys. Rev. Lett. **102**, 106405 (2009).
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How much time necessary to photo-generate Fermi surface from true electron vacuum.

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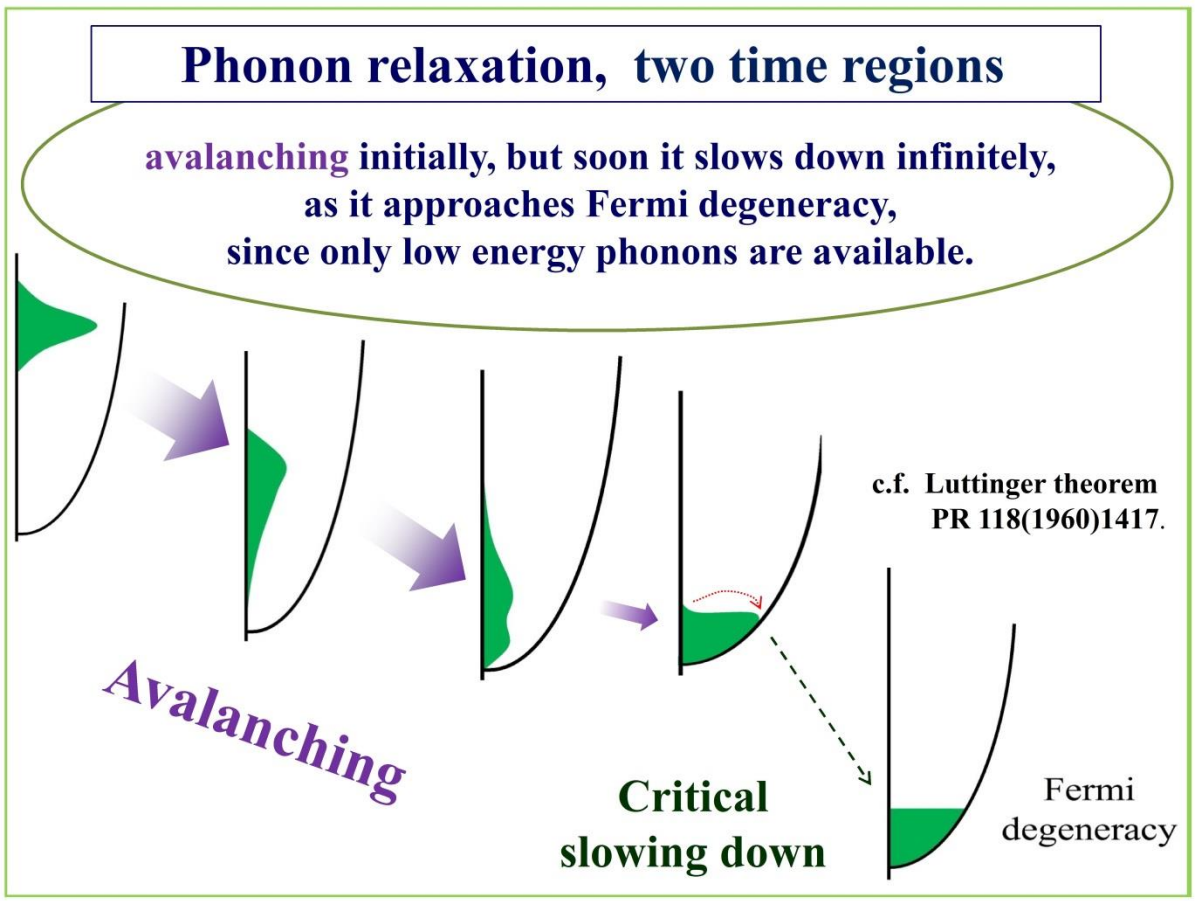
Rapid relaxation dynamics of optically excited electrons in metallic systems, has already been widely investigated. In most cases, however, only a few electrons are optically excited, while, the main part of electrons is still in the original ground state, works as an infinite heart reservoir, resulting in quite rapid relaxation of newly given energy and momentum.
What occurs, if a small but macroscopic number of electrons are excited at once, into a truly vacant conduction band without electronic heat reservoir, at absolute zero temperature? Two successive laser pulse excitations of GaAs or InP, and subsequent time-resolved photo-emission spectrum measurement of the conduction band electrons by Kanasaki-Tanimura, can partly answer this most simple, but ultimate photo-induced phase transition problem.
Coulombic inter-electron scatterings within the conduction band, being completely elastic, can give no net energy dissipation. While, the phonon relaxation of conduction band electrons has two time regions. Initially, it is rapidly avalanching, but, as the system approaches to the complete Fermi degeneracy, the relaxation slows down infinitely according to the Luttinger theorem. Hence, it never Terminates.



Femtosecond Switching to a Stable Hidden Quantum State in an Electronic Crystal

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Hidden states of matter with novel and unusual properties may be created if a system out of equilibrium can be induced to follow a trajectory to a state which is inaccessible or does not even exist under normal equilibrium conditions. We report on a hidden topologically protected electronic state in a layered dichalcogenide 1*T*-TaS crystal reached as a result of a quench caused by a single 35-fs laser pulse. The properties of the H state are markedly different from any other state of the system: it exhibits a large drop of electrical resistance, strongly modified single particle and collective mode spectra and a marked change of optical reflectivity. Particularly important and unusual, the H state is stable for an arbitrarily long time until a laser pulse, electrical current or thermal erase procedure is applied, causing it to revert to the thermodynamic ground state. Major observed events can be reproduced by a kinetic model describing the conversion of photo excited electrons and holes into an electronically ordered crystal, thus converting a Mott insulator to a conducting H state. Its long-time stability follows from the topological protection of the number of periods in the electronic crystal.

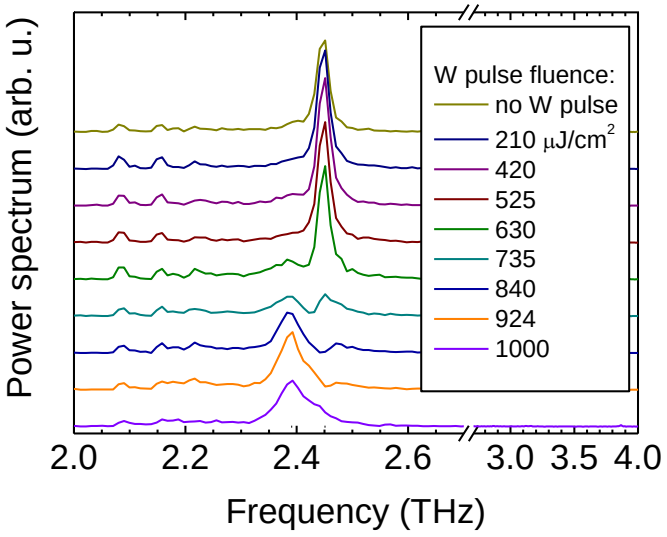

Figure 1: Collective mode spectra after exposure to 50-fs write (W) pulses with different fluences. The spectra show bimodal switching behaviour between 2.39 and 2.46 THz. For fluences near the threshold two distinct modes are observed, but no spectral density in between. Above 1 mJ/cm^2 a complete switching to the H state is observed.

[1] L. Stojchevska *et al.*, Science **344**, 177 (2014).

Towards a Microscopic Picture of the Photo-Induced, Metastable State in TaS_2

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The recent discovery of a photo-induced, metastable state in TaS_2 [1] promises a deeper understanding of non-equilibrium phase changes in complex materials. Notably, this state can only be induced with a sub-ps laser pulse, which reversibly switches from the insulating equilibrium state to the conductive metastable state. Here, we employ angle-resolved photoemission spectroscopy (ARPES), femtosecond time-resolved ARPES and low-energy electron diffraction (LEED) to investigate this photo-induced state. ARPES evidences that the metastable state is characterized by an ungapped Fermi surface near the Γ-point. trARPES suggests that the transition is driven by a transient imbalance of electron and hole populations while LEED does not resolve dramatic structural changes. We discuss similarities and differences to known phases of TaS_2.

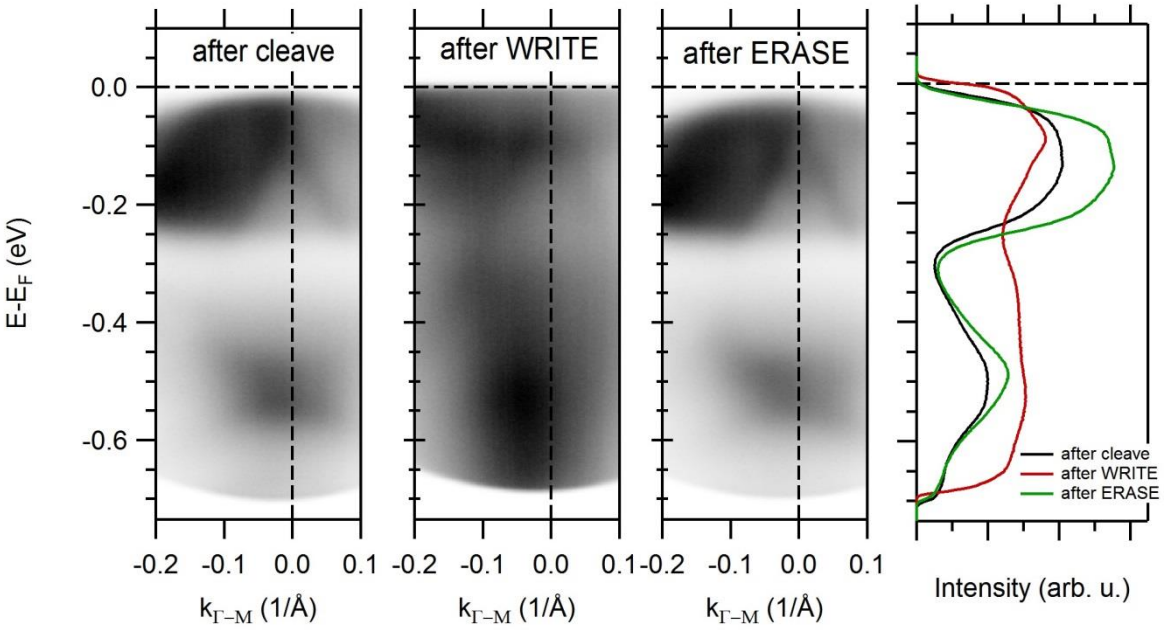

Figure 1: 6eV ARPES intensity of TaS_2 at T=9K near normal emission after cleaving, creating the metastable state with a single sub-50fs 800nm, 2mJ/cm^2 pulse (WRITE) and thermally annealing it with a 2.5s 1W 80MHz pulse train (ERASE). The right panel compares the spectral intensities, which have been integrated along $k_{Γ-M}$ from -0.2 to 0 Å^{-1} and clearly evidences an opening and closing of band gap near the Γ-point.

References:

[1] L. Stojchevska et al., Science **344**, 177 (2014)

Modeling of evolution of a complex electronic system to an ordered hidden state: application to optical quench in TaS2.

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Femto-second optical techniques addressing PIPTs put an ambitious goal to attend "hidden" states of matter which are inaccessible and even unknown under equilibrium conditions. For the first time among electronic systems, the reversible switching to a hidden truly stable spontaneously ordered state has been achieved recently in 1T-TaS2. The hidden state is lasting until a laser pulse, electrical current or thermal erase procedure reverts it to the thermodynamic ground state. The discovery, as well as earlier observations [2] of spacio-temporal dynamic transitions, called for developing a theoretical approach which we shall review here.
In application to events in TaS2, the theory [1] focuses upon dynamic evolution of electrons and holes as mobile charge carriers, crystallized electrons modifiable by intrinsic defects (interstitials and voids), and the CDW background. Mutual transformations among the three reservoirs of electrons, together with the heat production, are dictated by imbalances of three partial chemical potentials. The exchange rate among any two reservoirs vanishes when the corresponding chemical potentials become equal. The theory fits and explains all major observations. The approach sheds a light on very complicated and not yet resolved physics of this material including interplaying effects like CDW, Wigner crystal, polarons, Mott state. Our analysis is intermediate between the purely phenomenological [2] and a microscopic approaches. Both attempts suggest a direction to describe in details evolution of various PIPT.

References:

[1] L. Stojchevska, I. Vaskivskyi, T. Mertelj, P. Kusar, D. Svetin, S. Brazovskii, and D. Mihailovic. "Ultrafast switching to a stable hidden quantum state in an electronic crystal." Science, **344**, 177 (2014). More on theory in arXiv:1401.6786v3.
[2] R. Yusupov, T. Mertelj, V.V. Kabanov, S. Brazovskii, J.-H. Chu, I. R. Fisher, and D. Mihailovic, "Coherent dynamics of macroscopic electronic order through a symmetry breaking transition", Nature Physics, **6**, 681-684 (2010).

Optical exploration of hidden phases in correlated electron materials visualized by femtosecond electron crystallography

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Several classes of transition metal compounds, such as oxides and chalcogenides, exhibit sharp electronic phase transitions near room temperature, which have been contemplated for possible applications in nanoelectronics and photonics. It is generally believed that the competitive nature of Mott and Peierls physics is what drives the strongly first-order phase transition, which is highly tunable by intercalation-, substitutional doping, or applying pressure [1]. In the steady state measurements, the insulator-metal switching is often linked to subtle atomic lattice distortion, which allows specific controls, but also imposes structure-bottleneck for high speed operation. Using femtosecond electron crystallography and photonic tuning, we characterize the transformation in the nonequilibrium regime for several correlated transition metal compounds. We establish that the impulsive light-induced charge doping and highly athermal coupling between charge carriers and atomic lattice could generally provide an alternative pathway for a more efficient switching, bypassing the lattice enthalpy requirement.

We use 1T-TaS_2 as an example to highlight the richness of optically induced domain, lattice, and CDW dynamics. In the steady state, the insulator-pseudogap-normal state switching is coupled to various charge-ordered states, from commensurate (C), to triclinic (T) and hexaganally composed near-commensurate (NC), to incommensurate (IC) CDWs, as shown in Fig. 1. Using infrared laser pulses as photonic controls, which lead to charge carrier doping, similar hierarchy in topological evolution can be induced, but via an athermal pathway. Thus we have unveiled hidden phases, not accessible under thermodynamic conditions. These hidden phases are connected by "stair-cases", which are visible in the phase diagram under photo-doping x, and also in the time evolution, where sub-picosecond sharp jumps between different topologically stable phases are manifestation of soliton-like collective domain proliferation responsible for topological evolution. Femtosecond electron crystallography allows a complete mapping of these subtle structural adjustments, shedding insight into the organizing principle of C to IC transition and the emergence of pseudogap induced by charge doping. The sharp staircases may provide high-fidelity control over the transition between different hidden phases with distinct electronic properties, which could be utilized for efficient ultrafast electronic and topological switching with proper photonic control sequences.

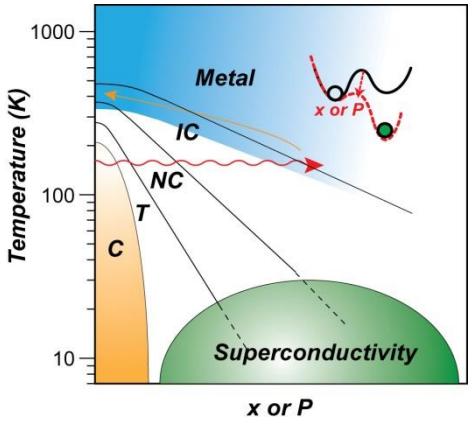

Figure 1: The phase diagram of 1T-TaS2 under chemical doping (x) or applying pressure (P). In the pristine 1T-TaS_2 (x, or P =0) crystal, the CDW undergoes four consecutive phase transitions.

References:

[1] T.-R. T. Han et al., Phys. Rev. B 86, 075145 (2012); Z. Tao et al., Phys. Rev. Lett. 109, 166406 (2012).

Ultrafast optical response and structural dynamics of the photoinduced phase transition of phase change materials

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Phase change materials, such as $Ge_2Sb_2Te_5$ (GST), have several metastable amorphous and crystalline phases at room temperature with vastly different optical and electronic properties. The fast and reversible optically-induced switching between those states has found numerous applications in modern data storage technology [1]. Despite the extensive research on tuning material properties for applications, many aspects of the microscopic origin of the optical contrast and the ultrafast response to femtosecond light pulses are still subject to debate. Specifically, the mechanism of the switching is poorly understood, with theoretical calculations suggesting a non-thermal pathway for ultrafast amorphisation [2].

We investigate the evolution of optical and structural properties of crystalline GST upon excitation with a femtosecond laser pulse by applying two complementary pump-probe techniques. We follow the transient optical properties by femtosecond reflectivity and transmission spectroscopy and directly probe the response of the lattice with time-resolved electron diffraction (diffraction images of the two phases and their radial averages are shown in Figure 1a).

We excite the material below and above its threshold for structural phase transition. For reversible excitation, we retrieve ground state properties such as the equilibrium electron-phonon coupling (determined by the timescale of the decay of Bragg peaks in Figure 1b).

Above the switching threshold, we use single-shot optical and diffraction measurements to follow the transient properties of GST during the phase transition. In the initial optical response, we observe a saturation in magnitude when crossing the fluence for permanent change, what, in earlier work, has been attributed to an ultrafast phase transition [3] or non-thermal melting [4], respectively.

We show, that the contrast in the early optical response is purely related to electronic excitation and melting of the material only takes place on a few-picosecond timescale. Ultimately, thermal effects take over and therefore dictate the final state of the material.

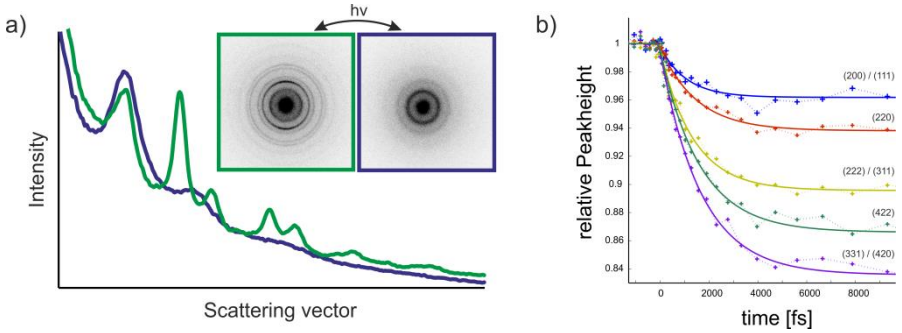

Figure 1: a) Diffraction images and radial averages of crystallised (green) and amorphous (blue) GST and b) relative intensity of the Bragg-peaks after reversible photo-excitation with an IR laser pulse.

References:
[1] D. Loke et al., Science **336** (2012)
[2] A. Kolobov et al., J. Phys.: Condens. Matter **19**, (2007).
[3] K. Sokolowski-Tinten et al., PRL **81**, (1998).
[4] J. P. Callan et al., PRL **86**, (2000).

Mecano-elastic switching in solids- Alternative route to PIPT

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During a transformation in a material triggered by a fs laser pulse the system is driven along a complex pathway towards a new macroscopic state. Different kinds of degrees of freedom play their part on significantly different time and length scales[1]. This leads to successive steps in the dynamical picture of the photo-induced transformation. The role of coherent optical phonons has been long under scrutiny within PIPT community, whereas that of acoustic phonons and cell deformations, albeit looked upon, has not been emphasized enough. Crystal deformations do not occur during the first steps since the process involves propagation of a strain wave, essentially determined by sound velocity. This coherent process has the long coherence time of acoustic phonons and long range of elastic interactions. The coupling between the strain wave and the order parameter field raises a challenging question whether such coupling can induce self-amplification of the material transformation.
Crystals composed of bistable molecules where the cooperativity is mainly governed by the change of molecular "volume" between the two states provide an excellent test bed for addressing this question.. We have investigated a cooperative spin crossover material, exhibiting a first order phase transition between low spin (LS) and high spin (HS) phases, by optical pump-probe spectroscopy over several time decades, and on samples of very different size, from nano- to macro-crystals. When a certain fraction of molecules, initially in LS state, is photo-converted to HS state (of higher molecular "volume") the pressure suddenly increases and drives lattice expansion on an acoustic time scale of size/speed (100 ps-100 ns). A self-amplification process is observed above a threshold excitation density. For low excitation density, the photo-switched molecules recover to the LS ground state, but above a critical excitation density the increase of HS fraction on the acoustic time scale is five fold. This illustrates the efficiency of cooperative mechano-elastic process during photo-induced transformation in a material. Our study brings the photo-switching of materials into new perspective, notwithstanding its common perception, uniquely related to electronic or optical phonon dynamics.

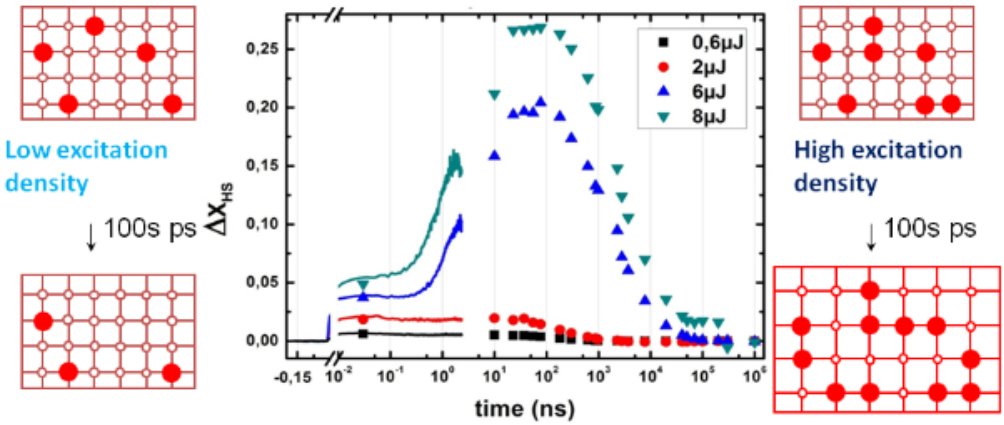

Figure 1: Variation of HS fraction after a fs excitation.

References:
[1]Lorenc, et al, Phys. Rev. Lett. 103, 028301 (2009).

Irreversible Photoinduced Phase Transitions Studied By Single-Shot Pump-Probe Spectroscopy

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We have observed a permanent photoinduced insulator-to-metal phase transition in a strained epitaxial thin film of $La_{0.67}Ca_{0.33}MnO_{3-\delta}$ on $NdGaO_3$. When deposited, the films are metallic at zero field below 250 K, and become insulating upon annealing[1]. An ultrafast laser pulse can switch the film between the as-grown and annealed states. The conductivity and optical properties of the switched state is tuneable based on the incident pulse fluence. Using single-shot pump-probe spectroscopy, we have tracked the evolution of the optical properties of the photoinduced state of the thin film.

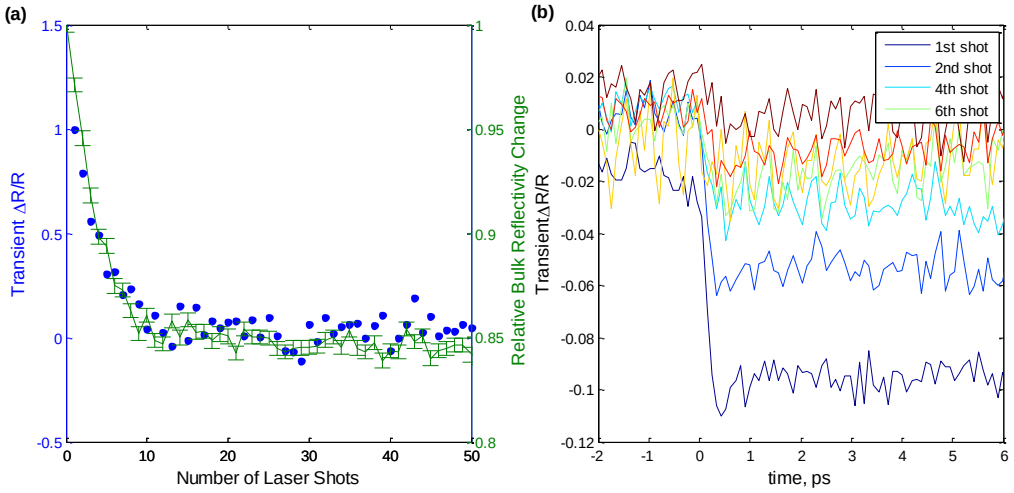

Figure 1: (a). Steady-state and transient reflectivity change at 720 nm over the first 20 laser shots. This wavelength is the peak of a localized electronic resonance in the insulating state that disappears in the metallic state. After the first ten laser shots, the sample is completely switched to the new phase. Pump-probe traces and the steady-state reflectivity evolve at the same rate. (b) Transient reflectivity change into the new state, showing a 500 fs transient peak followed by a steady change that persists for more than 10 ps.

References:

[1] LF Wang et al, AIP Advances, 3, 052106 (2013)

Ultrafast Dynamics probed with pulsed X-rays

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The recent development of X-ray Free Electron Lasers (XFELs) is opening new opportunities in ultrafast structural studies. The short and high intensity pulses are indeed perfectly suited for time resolved (pump and probe) techniques.

In this talk, a short review of XFEL properties will be given together with the opportunities and challenges of these new machines.

In the second part of the talk few examples of ultrafast dynamics will be presented.

Nonequilibrium electron density maps of ionic crystals from femtosecond x-ray powder diffraction

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X-ray diffraction represents a key method for spatially resolving electron distributions in crystalline materials. So far, electron density maps have mainly been derived from stationary diffraction patterns, providing specific and precise information on both equilibrium atomic positions and chemical bonding via valence electrons. Nonequilibrium excitations and phase transitions of crystals frequently imply atomic motions and changes of electron distributions occurring in the ultrafast time domain. All-optical experiments monitoring the related changes of the dielectric function give insight into the dynamics but not into transient structures on atomic length scales. In contrast, femtosecond x-ray diffraction methods which presently undergo a rapid development, provide direct access to transient atomic positions, electron distributions and the underlying microscopic interactions [1]. In this talk, new results are presented on transient electron distributions of ionic materials mapped with the help of femtosecond x-ray powder diffraction. Experiments are based on a pump-probe approach in which an optical pulse initiates structural dynamics and a hard x-ray pulse from a synchronized laser-driven plasma source is diffracted from the excited powder sample. Such experiments have revealed the interplay of lattice and charge motions which occur on distinctly different length scales in the prototype material KDP (KH_2PO_4) [2]. Here, electron relocations induced by strong external optical fields will be discussed [3,4]. This interaction mechanism allows for generating coherent superpositions of valence and conduction band quantum states and inducing fully reversible charge dynamics. While the materials $LiBH_4$ and $NaBH_4$ display electron relocations from the $(BH_4)^-$ ions to the neighboring Li^+ and Na^+ ions, LiH exhibits an electron transfer from Li to H. The latter is a manifestation of electron correlations and in agreement with theoretical calculations. A second study addresses electron transfer processes in crystals consisting of ionic transition metal complexes [5]. The metal-to-ligand charge transfer is complemented by a charge transfer from the counterions to the ligands as observed via transient electron density maps. The results demonstrate the many-body character of electron relocation which originates from the long-range Coulomb interactions between the different ionic constituents.

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Domain Alignment and Switching in Correlated Materials by THz Fields

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The strong link between the electronic and crystallographic degrees of freedom in correlated materials opens the possibility of unique routes towards material control that are not possible in simpler material systems. Of particular interest is the use of low frequency transient fields to alter the underlying microstructure of a material through direct interaction with the various coupled degrees of freedom of the material. $La_{0.5}Sr_{1.5}MnO_4$ (LSMO) is a prototypical insulating correlated material which exhibits strong coupling and orbital ordering. On cooling below T_{oo} = 220K, atomic orbitals order in one of two possible orientations forming random domains [1]. These domains can be oriented with an applied electric field [2] and thus could be used for data storage. In addition, as these domains arise from electronic correlations, domain switching has the potential to be very rapid; thus such materials could be used as a high speed alternative to magnetic storage.

In this work we demonstrate the use of the polarization of THz light to control the domain orientation in LSMO. The long wavelength light prohibits direct absorption and causes the light to more closely resemble an applied field. We observe that the degree of induced domain alignment increases linearly with applied THz field strength for temperatures sufficiently below T_{oo}. We also observe a non-linear enhancement in the degree of domain alignment in the vicinity of T_{oo}, where the system becomes very susceptible to the applied electric field (Figure 1). Furthermore, the THz-induced domain modifications we observe are completely reversible.

These experiments suggest that THz can be used to manipulate ordered phenomena in correlated materials without the need for any special material resonance effects. By comparing the domain response of the LSMO under linear and circularly polarized fields, we propose a material/light interaction analogous to that found in high-harmonic generation, and suggest that high speed THz switching may be possible.

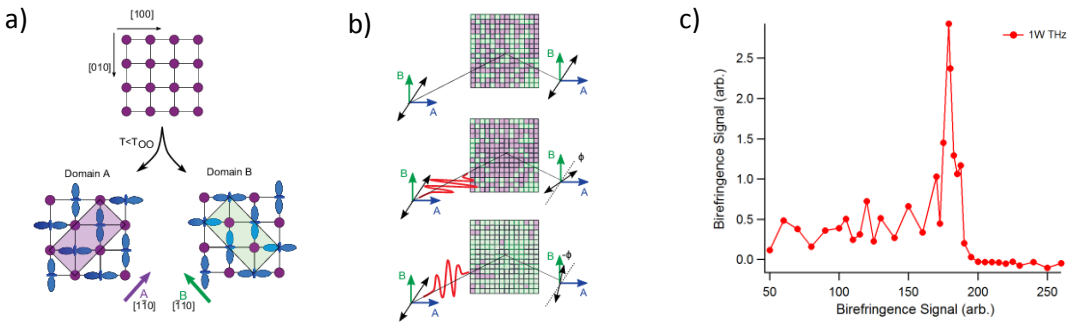

Figure 1: a) The two possible domain orientations in orbitally-ordered LSMO. b) Schematic of THz-induced birefringence used to measure the degree of orbital alignment. The domain orientation is set by the THz polarization. c) The temperature dependence of the THz-induced domain alignment, measured through an optical birefringence effect, with clearly visible enhancement close to T_{oo}.

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Charge recombination and relaxation in photoexcited Mott-Hubbard insulators

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Recent femtosecond pump-probe experiments on Mott-Hubbard (MH) insulators, in particular undoped cuprates, reveal charge recombination, which is in the picosecond range, much faster than in clean band-gap semiconductors although the excitation gaps in MH insulators are larger. We present a theory of recombination [1] of the excited bound holon-doublon pair, based on strong correlations and on the Hubbard and charge-transfer microscopic model which shows that such fast processes can be explained even quantitatively with the multi-magnon emission. We find that the recombination rate is exponentially dependent on the charge gap and on the exchange energy, whereby numerical model results are supported by an exactly solvable exciton-boson model.

Related is the question of the theoretical approach and understanding of the transient response of charge carriers, as probed by probe optical absorption in pump-probe experiments. We develop a linear-response formalism for the time dependent optical conductivity $\sigma(\omega,t)$ within a strongly correlated system [2] and discuss the questions of the time dependent optical sum rules and possible dissipation-less Drude weight. As an example we consider a single highly excited particle in the spin background. Of particular interest is the approach to the equilibrium response and the dependence on the initial pump perturbation.

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A Gap Opening Transition in a Quantum Phase of Photoinduced Quasiparticles in the Partly Filled Mott Insulator UO_{2+x}

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The most common type of photoinduced phase transition would be the triggering of a known, transformation from one crystallographic phase of a material to another. There is also, however, the possibility of a second type. In systems with low values of the electron-phonon coupling constant, λ, photoinduced quasiparticles such as polaronic excitions should exhibit significant mobility because their accompanying local lattice distortions will be small. This raises the possibility the quasiparticles they may aggregate and self organize into ordered quantum phases coincident with the lattice of the host crystal. This phenomenon is quite common in quantum phases of charge-spin inhomogeneities in transition metal-based Mott insulators that are chemically doped so that their valence band is only partly filled, i.e., the stripes in cuprate superconductors, nanophases in manganites, and similar structures in related materials. In fact, photoexcitation of slightly underdoped cuprates has been found to push them over the edge into the superconducting regime,[1] indicating that the photoinduced quasiparticles are identical to or at least emulate the stable, chemical ones that are the origin of the compelling properties to these compounds. We have now observed this phenomenon in the *f* electron Mott insulator UO_2 in its undoped form, where photoexcitation via the metal-to-metal charge transfer transition gives extremely complicated behaviour that is highly unusual or even unique. Interaction of the quasiparticles is implied by an increase in their lifetime, which are in the μsec regime below 100 K, with fluence. A spike in their lifetime around 25–30 K is assigned to their interaction with the spin-ordering transition to the AFM state, with a decrease from the 30.8 K Neel temperature resulting from the disruption from the pump pulse. That the coupling is to the gradient or derivative of the order parameter for this transition rather than to the transition itself is consistent with the exclusion of the photoinduced quasiparticles from the AFM domains, with mixing occurring only in the fluctuation region of the transition. This spike is almost an order of magnitude larger when the transition is to the predominantly 6*d* region of the upper Hubbard band rather than to the lower energy, predominantly 5*f* region, despite the long lifetime that should allow complete equilibration of the mid-gap state being probed. Another difference between the two excitations is that excitation to the 5*f* gives a plateau and cusp in the lifetime at 50–60 K. A discontinuity at this temperature is also observed in a phonon that originates in the excitation. A cusp of this type has previously been assigned to a gap opening phase transition, both superconducting and CDW.[2] Insofar as the only known transition of UO_2 is the AFM one and the phonon does not belong to the UO_2 spectrum, it must be occurring in a quantum phase of the photoinduced quasiparticles. Optical pump–THz probe measurements, although noisy, show an increase in the real component down to 20 K that flattens at lower temperatures concomitant with an increase in the imaginary component. This is consistent with the quasiparticles forming a condensate over this temperature range.[3] Combined with the unique results from structural and magnetic measurements on chemically doped UO_{2+x} compounds, neutron scattering and x-ray scattering and absorption, magnetic susceptibility, and epr, this system may be exhibiting Bose-Einstein-Hubbard condensate type behaviour originating in a novel mechanism – a coherent excitation – that is active through temperatures much higher than typical for BECs composed of atoms.[4]

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Ultrafast Photoinduced Electron Dynamics in Mott Insulators and Correlated Metals

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Ultrafast carrier dynamics in strongly correlated electron systems have significantly attracted much attention, because a number of time-resolved experimental techniques and theoretical calculation methods for non-equilibrium states are rapidly developed in the last decade. A lot of experiments and theoretical analyses [1-3] for transient carrier dynamics have been done in several classes of correlated electron systems. In this talk, we introduce recent our theoretical studies in the photo-induced transient electron dynamics in correlated electron systems.

1) Transient dynamics of hole carriers injected into a Mott insulator with antiferromagnetic long range order are studied based on the two dimensional *t-J* model [4]. Time dependences of the optical conductivity spectra and the one-particle excitation spectra are calculated based on the Keldysh Green's function formalism combined with the self-consistent Born approximation. We show that at early stage after dynamical hole doping, the Drude component appears, and then incoherent components originating from hole-magnon scatterings start to grow (see Figure 1). Time profiles are interpreted as doped bare holes being dressed by magnon clouds, and are relaxed into spin polaron quasi-particle states. The characteristic relaxation times for Drude and incoherent peaks strongly depend on the momentum of the dynamically doped hole and the exchange constant.

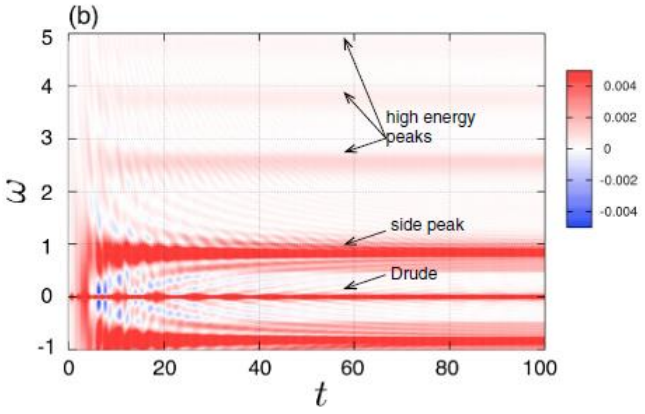

Figure 1: Transient optical conductivity spectra in two

2) The photo-excited real time dynamics in the two-leg ladder Hubbard model are studied [5]. Real-time evolutions of photoexcited electronic states are examined by the exact-diagonalization method in finite size clusters. Iin a doped metallic phase, low-energy optical responses are remarkably suppressed by photo-irradiation, in contrast to the photoexcitation in insulating states. After photo-irradiation, the hole pair-field correlation, which is known to play crucial roles on the electronic structure in ladder system, becomes to be short range, being similar to the insulating phase. We also examine the double-pulse excitations, and propose the optical control of the pair coherence. The results show good agreements with the recent pump-probe experiments in ladder cuprates [6] .

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Photo-induced electron dynamics in one-dimensional extended Hubbard model

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One-dimensional extended Hubbard model at half filling shows a spin-density-wave (SDW) state and charge-density-wave (CDW) state depending on the relative strength of on-site Coulomb interaction (U) and inter-site Coulomb interaction V. We investigate the photo-induced electrons dynamics and nonequilibrium process of the model driven by transient laser pulse by using the time-dependent Lanczos method.

In the case of large U and U~2V, the SDW and CDW phases are separated by a first order phase transition. When the system is subjected to the irradiation of a laser pulse in the SDW phase near the phase boundary, a sustainable charge order enhancement can be realized with proper laser frequency and strength, while local spin correlations remains [1]. Analogously, from the CDW side, the suppression of long-range charge order is accompanied with a local spin correlation enhancement. We analyse the conditions and investigate possible mechanisms of the emerging order enhancements.

We also investigate the ultrafast optical response of the model exposed to two successive laser pulses [2]. We find that following the first pulse, the excitation and deexcitation process between the ground state and excitonic states can be precisely controlled by the relative temporal displacement of the pulses. The underlying physics can be understood in terms of a modified Rabi model. Our simulations clearly demonstrate the controllability of ultrafast transition between excited and deexcited phases in strongly correlated electron systems.

These works are done in collaboration with Hantao Lu, Janez Bon\v ca, Sigetoshi Sota, and Hiroaki Matsueda.

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Equilibrium and Nonequilibrium "Condensations" in Polariton Many-Body Systems

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Semiconductor microcavity systems strongly coupled to quantum wells are now receiving a great deal of attention because of their ability to efficiently generate coherent light [1] by the Bose-Einstein condensation (BEC) of an exciton-polariton gas in a low-excitation regime. Since the exciton polaritons are composite quasi-bosonic particles, many fundamental features arise from their original constituents, i.e., electrons, holes, and photons. As a result, not only equilibrium phases typified by the BEC but also nonequilibrium lasing phases in a high-excitation regime can be achieved.

We developed a theory which can treat such equilibrium and nonequilibrium phases in a unified way. Our comprehensive framework is constructed by extending a Keldysh Green's function approach for two-level systems [2]. In our approach, coupled equations of the electron (hole) distribution, polarization function, and coherent photon field of the cavity are solved with steady-state assumptions. One of the main results are shown in Fig. 1 with increasing the pumping parameter (the chemical potential if all energetic losses were ignored). In the calculations [3], we assumed that the cavity is resonant with the 1S exciton and all energetic losses are caused by the cavity losses. One can notice that the photon density exhibits a secondary threshold-like behavior (Fig. 1(a)) and the oscillation frequency is blue shifted from the lower polariton into the bare cavity resonance (Fig. 1 (b)).

We also found that the principle of determining the steady state is quite different between the two: one is a crossover from the exciton-polariton BEC into the photonic BEC [1], whereas the other is that from the exction-polariton BEC into a laser operation, which is characterized by the kinetic hole burning. Here, unlike the normal photon lasing, we note that the obtained lasing is not described solely by the Maxwell-semiconductor-Bloch equations (MSBE) but still influenced by the BCS-gap equation. In this sense, "BCS-coupled lasing" is achieved.

The light-induced band renormalization causes gaps in the conduction and valence bands, which indicates the exsistence of bound electron-hole pairs in contrast to earlier expectations. We also show that these two types can be distinguished by the gain spectra. This work is supported by KAKENHI 26287087.

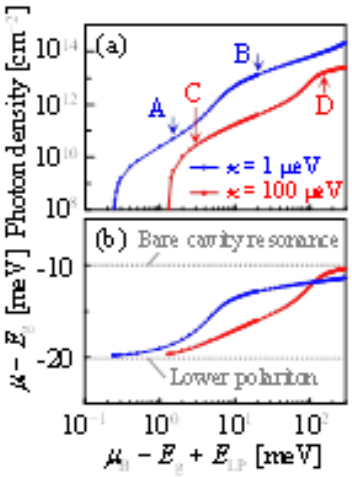

Figure 1: Calculated photon density and oscillation frequency as a function of the pumping parameter.

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Unusual Two-stage Dynamics of the Spin-Lattice Polaron Formation

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We follow the formation of a spin-lattice polaron after a quantum quench that simulates absorption of the pump-pulse in the time-resolved experiments. We discover a two-stage relaxation where spin and lattice degrees of freedom represent an integral part of the relaxation mechanism. In the first stage the kinetic energy of the spin-lattice polaron relaxes toward its ground state value while relaxation processes via spin and phonon degrees of freedom remain roughly independent. In the second, typically much longer stage, a subsequent energy transfer between lattice and spin degrees of freedom via the charge carrier emerges. The excess local spin energy radiates away via magnon excitations.

Additionally, I shall present recent research on non-equilibrium extension of linear response theory applied to optical conductivity of a charge carrier, coupled to phonons, in a spin background.

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POSTER ABSTRACTS

Dynamics of photoinduced semiconductor-to-metal transition in an optical switching nano-oxide Ti_3O_5

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Nano-granular trititanium pentaoxide (Ti_3O_5) has been attracting a great deal of attention as a novel functional material, which shows photo-reversible semiconductor-metal phase transition (β-Ti_3O_5 ↔ λ-Ti_3O_5) [1]. This is the first demonstration as an optical switching oxide at room temperature, and then various applications are expected particularly for fast-writing and high-density optical storage devises in the next generation. Furthermore, this material has a unique characteristic that the direction of the transition can be switched by adjusting the excitation fluence. However, the mechanism of this interesting power-modulated reversibility has not been clear. In this presentation, we focus on the semiconductor-to-metal transition (β-Ti_3O_5 → λ-Ti_3O_5) and investigated the dynamics by time-resolved diffuse reflection spectroscopy. By observing over a wide time range from femtoseconds to microseconds, the overall relaxation behaviour has been revealed.

Figure 1 shows the temporal evolution of the transient diffuse reflectance (ΔDR) at 633 nm associated with the phase transition induced by 120 fs pulse at 800 nm. Here, a pellet composed of β-Ti_3O_5 nano-particles, was excited below the threshold of the persistent transition. Since the decrease of DR probed at this wavelength corresponds to a creation of λ-Ti_3O_5, this evolution means that λ-Ti_3O_5 is transiently created. The whole dynamics consisted of the five processes, (i)-(v) shown in Fig. 1. From a spectral analysis, we concluded as follows: (i) instantaneous generation of λ-Ti_3O_5 fractions and excitation of free carriers (0 - 0.2 ps), (ii) partial annihilation of the λ-Ti_3O_5 fragments (0.2 - 1 ps), (iii) cooperative expansion of the survived λ-Ti_3O_5 domains (1 - 10 ps), (iv) extinction of the excited free carriers (10 ps - 20 ns) and (v) annihilation of the transiently created λ-Ti_3O_5 domains (20 ns - 1 ms). Here, it should be noted that the fast response (~200 fs) in the appearance of λ-Ti_3O_5 indicates that it is non-thermal. It should also be noted that the λ-Ti_3O_5 domain formed around 10 ps stably survives for up to several hundreds of microseconds. Although not shown here, the lifetime gets longer for stronger excitation. Therefore, it is expected that the stable λ-Ti_3O_5 domain formation observed here corresponds to the starting point of the persistent transition. In other words, the switching time of the persistent phase change is about 10 ps.

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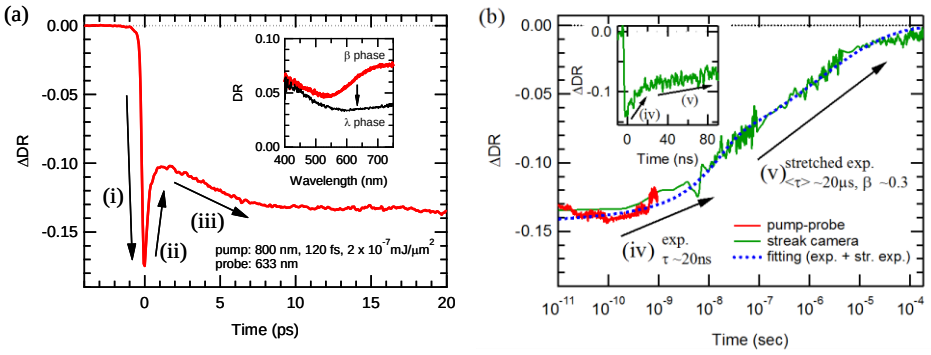

Figure 1: Temporal evolution of the transient diffuse reflectance (ΔDR) (a) till 20 ps and (b) till 200 μs in logarithmic time scale. The inset in (a) is the spectral change of diffuse reflectance (DR) associated with the phase transition from β-Ti_3O_5 to λ-Ti_3O_5. The inset in (b) is the ΔDR curve till 100 ns in linear time scale.

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Nonthermal destruction of SDW order in iron based pnictides by ultrashort laser pulses

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The laser-induced out-of-equilibrium phase transitions in collectively ordered systems have been the subject of number works published in the last few years [1-4]. To our best knowledge a photoinduced spin density wave (SDW) to normal state transition and recovery were not investigated a lot[4].
The standard pump-probe technique was therfore applied to perform a systematic study of the time and energy density needed to nonthermally destroy the SDW state in iron pnictides of 122 family by ultrashort laser pulses. Recovery of the SDW state after a quench was also studied by the 3-pulse pump-probe experiments.
The recovery dynamics of the SDW state in terms of the time-dependent Ginzburg-Landau theory is going to be briefly discussed.

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Initial dynamics of the photoinduced charge-density-wave destruction in Indiumwires on Silicon(111)

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Recently we have investigated, using time-resolved electron diffraction, the dynamics of the charge-density wave (CDW) recovery in quasi-one-dimensional Indiumwires adsorbed on Si(111) [1]. The low temperature (8x2) reconstructed CDW ground state has been excited by femtosecond laserpulses resulting in the excited (4x1) reconstructed state. In these experiments the phase transition is solely driven electronically at temperatures well below the transition temperature. This leads to a long living metastable state that is not reached thermally. We found that the time constant of the CDW recovery is determined by the number of adatoms which act as seeds for the (8x2) formation [1].

In the above experiment the initial stages of the CDW destruction could not be resolved. The temporal resolution of the experiment is limited by the velocity mismatch between the probing electrons and the laser excitation: As the electrons are diffracted at a glancing incident angle the temporal resolution is given by the electron travelling time across the sample. To overcome this limiting factor we employed a laser pulse front tilter in order to match the excitation pulse front to the electron probing time [2]. Using this setup the temporal resolution is tweaked below 2 ps [3].

With this setup it is possible to resolve the initial dynamics of the photoinduced (8x2) to (4x1) phase transition. The temporal evolution of the diffraction spot intensity is not described by a simple excitation/de-excitation process. In fact, a two peak structure is observed. Integer order diffraction spots show an intensity increase on a 5 ps timescale. This is followed by an intensity decrease which is overcompensated by a second intensity increase on a longer timescale. For fractional order spots this behavior is also observed, however, less distinct. The observation is maybe explained by a multi-process excitation where an extreme softening of the soft-phonon-mode connected to the (8x2)-groundstate occurs. Another possibility is the excitation of two independent processes that are delayed with respect to each other but conjointly contribute to the transient intensity change in the diffraction spots. Although a complete understanding of the underlying processes is missing so far, these results will be important to obtain a deeper insight in the microscopic processes leading to the above mentioned metastable state that results from the photoexcitation.

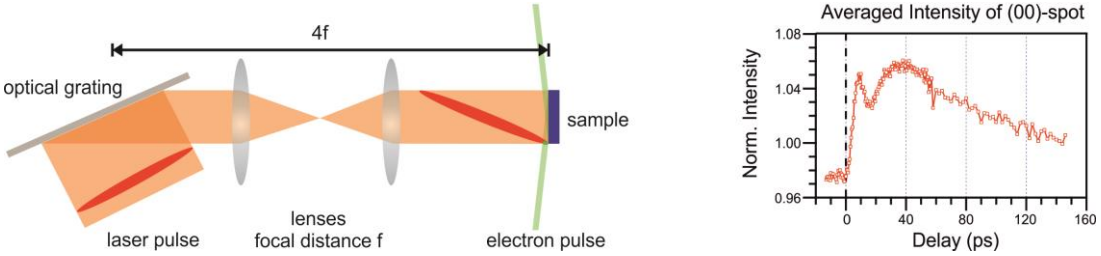

Figure: Left panel: Pulsefronttilter (zero dispersion delay line) in order to match excitation pulsefront to the electron probing time at the sample. Right panel: Transient (00)-spot intensity.

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Structure and Dynamics with Ultrafast Electron Microscopies

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This poster will describe how combining ultrafast lasers and electron microscopes in novel ways makes it possible to directly 'watch' the time-evolving structure of matter (through diffraction and imaging), both at the level of atomic-scale structural rearrangements in the unit cell and at the level of a material's nano- microstructure.

Successful efforts to develop and fully characterize an ultrafast electron diffraction (UED) instrument based on radio-frequency (RF) compressed, 100keV electron pulses will be presented [1]. The concepts involved in recompressing femtosecond laser produced electron bunches with RF cavities will be described along with their practical implementation at McGill University. Novel methods to characterize the temporal impulse response function in pump-probe geometry using laser fields and streak cameras were used [2]. At pC bunch charges, the time resolution of this instrument is <300 fs FWHM, currently limited by RF/laser synchronization jitter not the recompressed electron pulse duration which we estimate to be <100 fs FWHM [1]. This instrument approaches the capabilities of xray free electron lasers (XFELs) for diffraction experiments.

Second, dynamic transmission electron microscopy (DTEM) will also be described [3]. We have used DTEM to make direct, real space images of nano-microstructural evolution during laser-induced crystallization of amorphous semiconductors at unprecedented spatio-temporal resolution [4]. This is a remarkably complex process that involves several distinct modes of crystal growth and the development of intricate microstructural patterns on the nanosecond to ten microsecond timescales, all of which can be imaged directly with DTEM. The insights that DTEM provides has led to a new understanding of the crystallization dynamics and the underlying mechanisms.

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Ultrafast Imaging of the Photo-Induced Magnetization in $DyFeO_3$

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Ultrafast kinetics of the first-order phase transitions is an issue of intense research in contemporary physics and chemistry. In magnetism this is an especially intriguing subject since understanding the kinetics of the phase transitions is crucial for achieving the fastest possible magnetic recording and information processing. Rare-earth orthoferrites is a broad class of antiferromagnetic materials. Due to a wide variety of magnetic phase transitions as well as strong magneto-optical and opto-magnetic effects [1-3] the orthoferrites are very convenient materials for time-resolved studies of ultrafast photo-induced phase transitions [4,5]. Among all rare-earth orthoferrites $DyFeO_3$ is especially intriguing due to its unique property to possess first-order Morin phase transition [6] during which the material acquires the net magnetization upon a temperature increase. The goal of this study was to investigate ultrafast kinetics of the Morin transition induced by a femtosecond laser pulse.

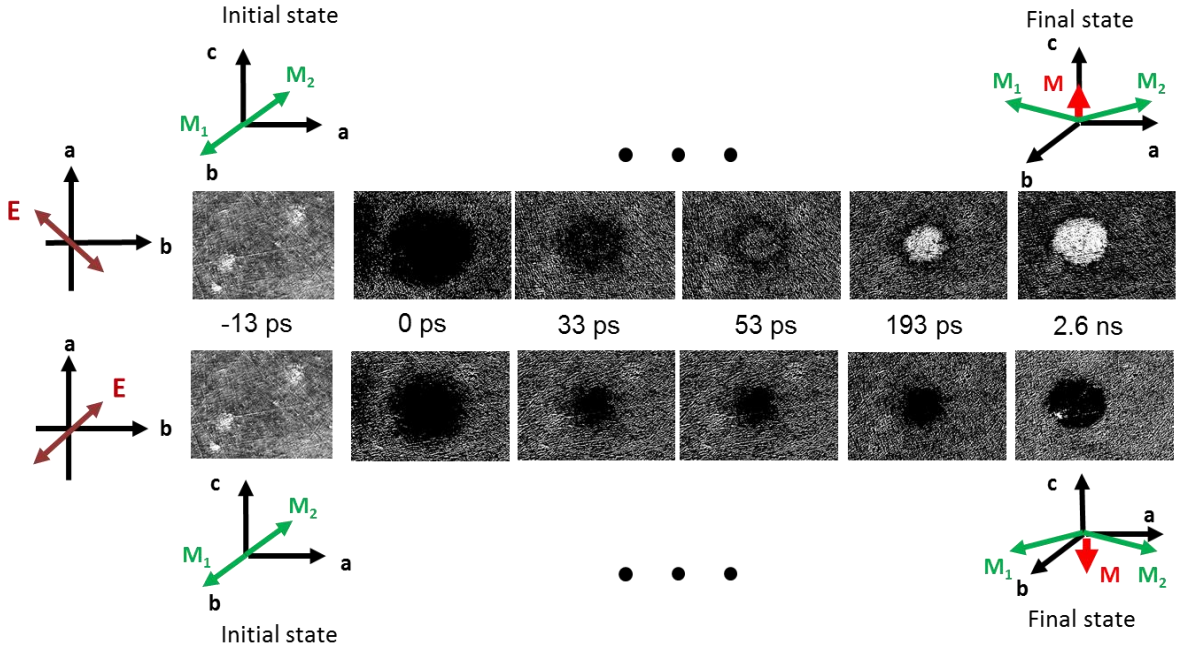

Figure 1: Ultrafast photographs of light-induced magnetization dynamics at T=18 K for two orthogonal linear pump polarizations.

The magnetic order of $DyFeO_3$ is determined by the network of Fe^{3+} spins, while Dy^{3+} spins are not ordered above 4 K. Up to the Morin point T_M=32 K the spins of Fe^{3+} are characterized by a collinear antiferromagnetic alignment along the b-crystallographic axis. At the Morin point T_M=32 K the magnetic structure experiences the first order phase transition so that a temperature increase is accompanied by an abrupt jump of the spins over 90-degrees and alignment of those along the a-axis. The jump is accompanied by a canting of the antiferromagnetically coupled spins over a small angle of 0.5 degree and a generation a net magnetic moment along the c-axis. Here using ultrafast time-resolved magnetic imaging we demonstrate that a 60 fs laser pulse with central wavelength 800 nm is able to trigger the Morin phase transition and generate the net magnetization in $DyFeO_3$. Interestingly, the direction of the magnetization in the final state can be effectively controlled by the polarization of

the pump (see Fig. 1) or by a combination of the polarization and the external magnetic field. Although the role of the trigger of the phase transition is played by a femtosecond laser pulse, the phase transitions itself proceeds long after the event of the femtosecond excitation being stretched up to 60 ps. Here based on the analysis of the temperature and field dependences we reveal the mechanism of the photo-induced phase transition.

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Control of the overlap between stable and metastable HS states in spin-crossover compounds – towards hidden phases

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In recent years, molecular optical switches have attracted considerable attention as they hold great promises in molecular electronic and photonic devices. The reversible photo-switch of the magnetic states[1] is widely studied in spin-crossover systems able to exhibit high-spin (HS) ↔ low spin (LS) conversion under thermal perturbation. This has led to the introduction of the T(LIESST) measurement to estimate the limit temperature above which the photoinduced HS metastable state is erased.[2] The creation of a database[3] allowed to increase T(LIESST) value from 60 K to 130 K for pure iron(II) SCO materials[4] and 180 K in molecular cluster.[5] This open the opportunity to study the particular situations where the metastable HS state reaches, and even overlaps, the thermal spin-crossover regime.[6,7]

In this presentation we will show several examples of such overlap obtained by rational metal dilution approach. The influence of this dilution on the relaxation kinetics and the overlap will be particularly discussed on cooperative materials. Theoretical investigations will be also presented on the basis of the macroscopic master equation.[7]

The control of such overlap aims at generating hidden phases only accessible by light irradiation.

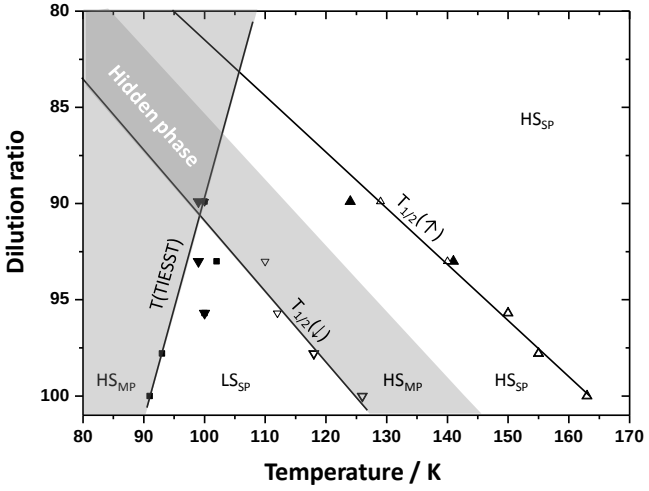

Figure: pseudo phase diagram of the evolution of T(LIESST) and the thermal spin-crossover temperatures in warming and cooling modes as function of metal dilution ratio. It shows the clear overlap of the thermal hysteresis and the photo-induced state giving rise to hidden phases.

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Ultrafast modulation of ferroelectric polarization by THz waves in a charge-ordered organic crystal α-$(ET)_2I_3$

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Recently, it has become possible to generate THz waves with the maximum electric field much higher than 100 kV/cm. Such strong THz waves are now being used to excite electron and/or lattice systems in solids. Last year, we reported that ferroelectric polarizations can be rapidly modulated by a THz electric field in an organic molecular compound, tetrathiafluvalene-*p*-chloranil, TTF-CA [1]. This compound is known to show 'electronic-type' ferroelectricity. Here, we focus on another molecular compound, α-$(ET)_2I_3$. This compound is a famous organic conductor, which shows a metal-insulator transition at T_c=135 K, below which the charge-ordered phase is stabilized. In the charge-ordered phase, the inversion symmetry of the charge distribution is lost. The second-harmonic generation (SHG) measurements revealed that the ferroelectric polarization emerges [2]. The ferroelectricity of this compound is suggested to originate from intermolecular charge-transfers, so that ultrafast responses to external electric fields are expected. In this study, we have investigated THz-field-induced changes of the polarization and charge distribution using THz-pump optical-probe and SHG-probe spectroscopy.

Figures 1(b) and (c) show the time evolutions of the reflectivity changes $\Delta R/R$ at 10 K (the charge-ordered phase) induced by the THz pump pulse (Fig. 1(a)) with the electric field E_{THz} parallel to (//) *a* and *b* axis. The photon energy of the probe pulse is 0.65 eV, which corresponds to the charge transfer transition. The reflectivity changes show instantaneous responses to the THz electric field at around the time origin. Subsequently, they show prominent oscillations, which are related to coherent molecular oscillations. The time evolutions are well reproduced by the sum of the THz electric wave form and three forced oscillations of lattice modes. Large amplitudes of the lattice oscillations suggest that the charge order is strongly stabilized by molecular displacements.

The magnitude of $\Delta R/R$ for E_{THz}//*b* is twice as large as that for E_{THz}//*a*. To obtain detailed information of the polarization direction, we measured the THz-field-direction dependence of $\Delta R/R$ (Fig. 2(b)). The $|\Delta R/R|$ value has the maximum at 23 degree from *b* axis (the open arrow in Fig. 2(a)), which is considered to be the direction of the ferroelectric polarization. This diagonal direction of the polarization can be explained by the anisotropy of the intermolecular transfer energies.

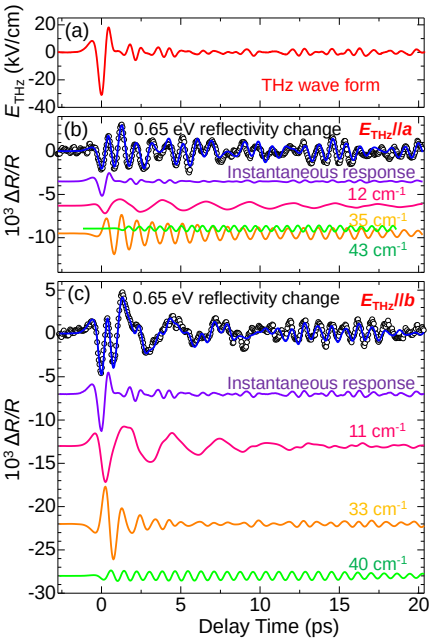

Figure 1: the result of pump-probe measurement

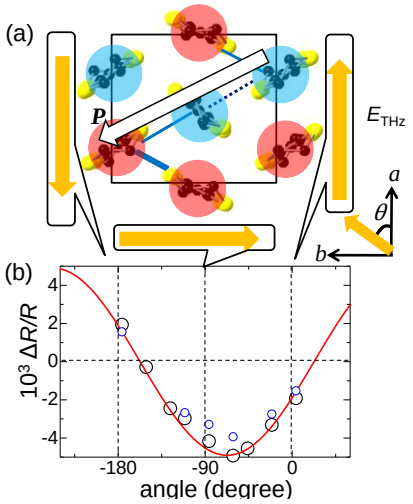

Figure 2: (a) the ferroelectric polarization direction and (b) THz-field-direction dependence of reflectivity changes

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Two step-like transitions in photomagnetic cobalt hexacyanoferrates due to internal stresses

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The optical control of the physical properties of a material has drawn considerable attention during the past years for a fundamental point of view (access to non-equilibrium states) but also for applications (memories, optical switches, etc). In transition metal oxides and molecular-based solids,[1,2] light irradiation can trigger electronic excitations, which can be trapped[1] or propagate in a collective and ultrafast way[3] within the crystal, giving rise to new transient or long-lived phases. The lifetime of the light-driven phase is mainly function of the amplitude of the local structural distortion associated with the electronic excitation. Its spatial expansion is then mediated by elastic interactions in strongly coupled electron-lattice systems.

The photo-switching properties of cobalt hexacyanoferrates, of $A_xCo[Fe(CN)_6]_y.\ zH_2O$ formula (A: alkali metal ion), have been extensively documented in bulk phases, and to a lesser extent in nanoparticles and thin films. These compounds exhibit a charge transfer-induced spin state transition (CTIST) as a function of temperature or, at low temperature, under red light irradiation:[2]

$Co^{3+}(t_{2g}^6,\ S=0)$-$Fe^{2+}(t_{2g}^6,\ S=0) \rightarrow Co^{2+}(t_{2g}^5e_g^2,\ S=3/2)$-$Fe^{3+}(t_{2g}^5,\ S=1/2)$, where HS and LS denote high spin and low spin, respectively. A key factor in the photo-process is the spin transition associated with a local expansion of the $Co(N_{6-x}O_x)$ octahedron. Recent works have evidenced a complex multimetastability, which is function of the photo-thermal history of the sample, and presumably originate from coherent or disordered tiltings of the $[Fe(CN)_6]$ octahedra[4] in this double perovskite structure. Preliminary experiments have also suggested the possibility of self-organisation processes leading to clustering effects[5] largely dependent on the particle size.[6]

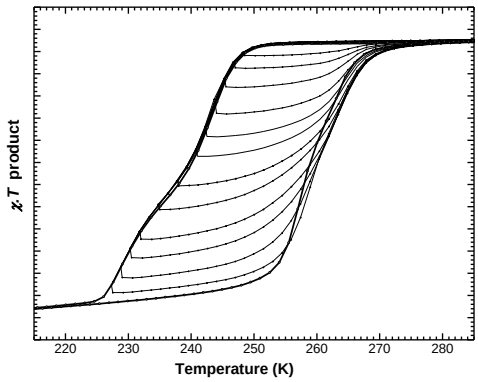

Figure 1: Experimental FORCs derived

In this work, we investigated the thermal hysteresis properties of $Na_xCo[Fe(CN)_6]_y.\ zH_2O$ compounds using the First Order Reversal Curve (FORC) method, first introduced to analyze magnetic domains and further adapted to spin crossover solids. From the two step-like transition of Fig.1, we derived a description of the spin-like domains in terms of energy gap between HS and LS states and intra-domain elastic interactions. Comparison with synchrotron x-ray diffraction measurements demonstrate that internal stresses are an important ingredient for a complete description of the thermal hysteresis loop, as they lead to a substantial decrease of the energy gap and an increased cooperativity.

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Photoinduced phase transitions in $Bi_2Sr_2CaCu_2O_{8+\delta}$

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Possibility of photodestruction of many body quantum states allows realization of truly non-equilibrium phase transition[1]. Pseudogap [2] and superconducting gap[2], [3] could both be destroyed by an ultrashort laser pulse. We present ultrafast study of the recovery of both states conducted systematically with temperature and fluence. The pseudogap state recovers exponentially; recovery time appears to be independent on fluence and weakly dependent on temperature. Superconducting state recovery is exponential for moderate excitation and delayed superexponential for strong excitation. The applicability of Ginzburg-Landau theory is discussed.

The authors acknowledge financial support from European Research Council.

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Properties of the hidden photoinduced state in 1*T*-TaS_2

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Hidden topologically protected (H) state in charge density wave (CDW) material 1*T*-TaS_2 is the first observation of the stable photoinduced state. It has unique properties unlike any other thermodynamic phase of the material [1]. However detailed information on characteristics of the H state is still missing.

Here we report on relaxation dynamics of the H state near the transition temperature that is reflected in DC resistivity measurements. Stability of the H state sharply increases with decreasing of the sample temperature. Relaxation process may be described by a stretched exponential function that is in an agreement with the scaling theory for near commensurate to commensurate CDW transition. Furthermore, jumps in R(t) curve may be attributed to "Devil staircase" that represent favourable values of the wavevector of CDW.

Stability of the H state may be enhanced by applying an in-plane pressure. We introduce 2D-pressure by using substrates with different expansion coefficients. Resulting strain on the sample/substrate interface allows us to apply both positive (compressive strain) and negative (tensile strain) pressure.

0.2% tensile strain increases transition temperature for 15 K in comparison with 0.05% compressive strain.

The authors acknowledge financial support from European Research Council and use of equipment of CENN Nanocenter.

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Controlling exchange interactions with electrical field pulses

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Magnetic long-range order and the dynamics of spins in magnetic materials is governed by the exchange interaction, the strongest force of magnetism. Unlike the spin, which is a fundamental property of electrons, the exchange interaction between spins emerges from the Pauli principle and the electrostatic Coulomb repulsion. Therefore, the exchange interaction is sensitive to purely nonmagnetic perturbations. This fact implies intriguing and largely unexplored possibilities for the ultrafast control of magnetism by femtosecond laser-pulses, which is currently a very active research area. In this contribution we investigate how the exchange interaction can be modified by off-resonant driving with electric field pulses. We develop the framework of Floquet theory for insulating Hubbard clusters and demonstrate that weak off-resonant driving enhances (reduces) the exchange interaction for driving frequencies below (above) gap. Such field strengths are directly relevant for experiments on magnetic oxides. Good agreement is found between such cluster calculations and nonequilibrium dynamical mean field theory simulations for extended systems. Furthermore, in the regime of strong electric fields, which is experimentally accessible with cold atoms in optical lattices, we observe that even the sign of the exchange interaction can be reversed, yielding a ferromagnetic exchange interaction in the Hubbard model. By simulating a one-dimensional Hubbard chain, we demonstrate that the reversal of the exchange interaction can be experimentally detected by following the time-evolution of the spin degrees of freedom, which will be reversed when the exchange interaction changes sign.

Photoinduced dynamics in K-TCNQ studied by time-resolved infrared spectroscopy

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A crystal structure of the K-TCNQ consists of one dimensional stacking of the $TCNQ^-$ ions. K-TCNQ shows so called spin-Peierls (SP) phase in which adjacent TCNQ ions are dimerized below the transition temperature (T_C = 395 K). Very recently, photoinduced phase transition (PIPT) has been reported by Okamoto and his coworkers [1][2]. They report the CT excitation between the adjacent TCNQ molecules destabilizes the SP phase and makes the undimerized photoinduced phase. However, the dynamics of the dimerization of the TCNQ molecules has never been observed directly.

Time-resolved infrared spectroscopy is a powerful technique which directly enables us to detect the photoinduced change of the molecular vibration with ps time-resolution. Using this system, we measured time-dependence of TCNQ a_g ν_4 mode after the photoirradiation. This is an electron-molecular-vibration (emv) coupled mode known as a good indicator of the degree of the dimerization of the TCNQ ion [3].

We analysed the time-resolved reflectivity spectrum of the a_g ν_4 mode. Until today, based on the photo-induced change in the spectral shape, it has been a natural idea that dimerization is strongly affected by charge transfer excitation. In contrast, just after photo excitation (at Δt=1ps), we found that the oscillator strength of the emv mode was scarcely changed by the photo-excitation while the spectral shape of the reflectivity is largely suppressed. This result suggests that the degree of dimerization in the photo-excited state generated just after excitation is fluctuated but its intensity itself is not so weakened, rather larger than that in the high temperature state in K-TCNQ. The detail about the spectral analysis and its time profile will be discussed.

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Irreversible Photoinduced Phase Transitions in Bismuth Studied By Single-Shot Pump-Probe Spectroscopy

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Bismuth is a longstanding testbed for ultrafast spectroscopy, detection, and control of large-amplitude coherent phonons. Using single-shot pump-probe spectroscopy, we can drive and observe irreversible processes in solids. We have generated extremely large-amplitude coherent phonons in bulk and thin film bismuth, leading towards a symmetric phase transition. In bulk and thicker films, excited carrier propagation away from the crystal surface limits electronic excitation. In thin films, confinement of excited carriers leads to melting at fluences slightly lower than the onset of the symmetric phase. Following the state of highly excited bismuth out to 1 ns after excitation, along with simulations based on the two-temperature model reveals the role of excited carriers relaxation and transport in lattice softening and heating.

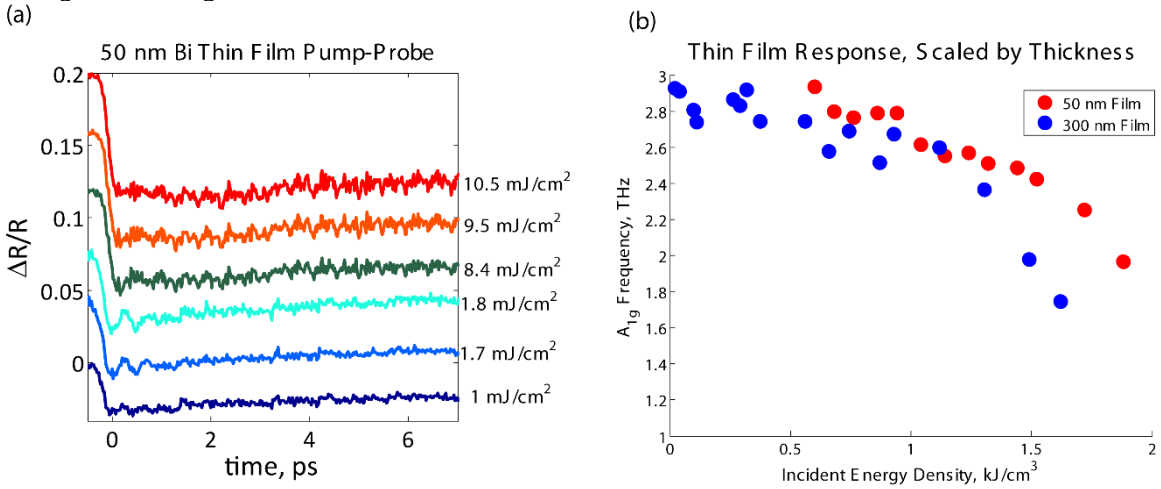

Figure 1: (a). Single-shot response of 50 nm thick Bismuth thin film as a function of fluence. At high fluence, the optic phonon mode frequency approaches zero, supporting evidence of approach to and into the symmetric pahse. (b). Bond softening as a function of film thickness. In thicker films, the incident fluence required is much higher to achive the same degree of bond softening, due to carrier diffusion into the film in the first few ps after excitation. Scaling the incident fluence by film thickness accounts for the difference in degree of bond softening.

Femtosecond pump-probe spectroscopy for a charge-density-wave system of $DyTe_3$ resolved by the probe polarization

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Ultrafast pump-probe spectroscopy has been used to investigate various ordered electronic states such as superconductivity, spin-density-wave and charge-density-wave (CDW). Non-equilibrium dynamics of the photo-induced quasi-particles (QP) were characterized by relaxation time, temperature dependence, the wavelength and intensity of light, and so on. Although dependence on the probe-photon polarization of the QP dynamics can provide important information in terms of a spatially symmetry, the detailed investigation has been reported only in the superconductors [1]. The QP dynamics resolved by the light polarization in other ordered states has remained unclear so far.

In this study, we have performed pump-probe spectroscopy for a CDW system of $DyTe_3$ with different probe polarizations. $DyTe_3$ is a quasi-two dimensional compounds which consists of square planar Te sheets (*a*-*c* plane) and insulating $DyTe_3$ layers. The CDW formation occurs along *c* axis at T_{c1} ~ 305 K and *a* axis at ~ 50 K. The optical measurements were performed using 120 fs pulses centered at 400 nm for a pump (F = 84 μJ/cm^2) and 800nm for a probe from a cavity-dumped Ti:sapphire oscillator with a repetition rate 270 kHz. The pump and probe beams were coaxially overlapped and irradiated perpendicular to the *a*-*c* plane.

Figure 1(a) and (b), respectively, show the temperature dependences of the isotropic (A_{1g}) and anisotropic (B_{2g}) components of the transient reflectivity ΔR/R for the probe, which were obtained by decomposing the probe-polarization-angle dependence of ΔR/R in terms of D_{4h} group symmetry of the Te plane. In the A_{1g} response, a positive component develops with decreasing temperature, while a negative component dominates in the B_{2g} response. Figure 1(c) presents the temperature dependences of the decay time obtained by fitting the rapid decline of transient with a single-exponential function. The decay time of the B_{2g} component shows a clear divergence as T_{c1} is approached from below and agrees well with the theoretical model [3]. This indicates that the QP dynamics of CDW is enhanced in the B_{2g} response rather than the A_{1g} response.

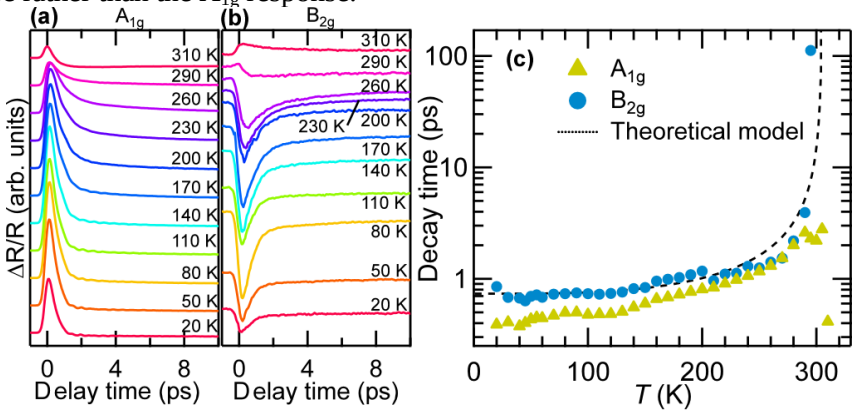

Figure 1: (a),(b) ΔR/R transients of the A_{1g} and B_{2g} components for various temperatures, respectively. The data are shifted for clarity (c) Temperature dependence of decay time on the A_{1g} and B_{2g} components. The dashed line displays the result fitted by the theoretical model [3].

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Ultrafast dynamics of optically pumped spin-cycloid multiferroic

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Multiferroics have attracted intense recent interest largely due to possible applications of these materials, particularly if multiferroic domains can be controlled in faster or more efficient ways relative to simpler ferroic materials. Despite this, the limiting timescales of domain transformation in multiferroics are mostly unknown. It has been shown previously that properties of multiferroic $BiFeO_3$ can be influenced on a picosecond timescale by pumping with laser pulses in the optical range [1]. The multiferroic coupling in $BiFeO_3$ is, however, not direct and thus applications of this material may turn out to be limited.

Here we study the ultrafast dynamics in $TbMnO_3$, a model spin-cycloid multiferroic where the ferroelectric polarization arises directly from the frustrated magnetic order. Using this material it has been demonstrated that excitation using THz pulses can lead to the rotation of the spins towards the opposite domain [2], and that 400 nm excitation leads to a transient reduction of the magnetic order [3]. Here we discuss the magnetic and structural response of multiferroic $TbMnO_3$ to excitation with 800 nm radiation, probed using optical methods and ultrafast resonant x-ray diffraction. The intensity of the (0q0) magnetic diffraction peak, sensitive specifically to the ordering of Mn spins, decays exponentially with a characteristic timescale of 5-25 ps depending on the excitation fluence. Above a threshold fluence the magnetic peak disappears, suggesting the destruction of long-range magnetic order. The rise of the optical reflectivity after the excitation follows exponential increase with similar timescales as seen in the x-ray data. The 30 GHz oscillation visible in the optical reflectivity is present in both room temperature and the multiferroic phase and is attributed to a strain wave. The data suggest that heat-mediated electron-phonon coupling plays a significant role.

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Femtosecond charge transfer from MoS_2 to organic acceptor

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MoS_2 has been shown to have a strong photovoltaic effect [1] and can be used in high sensitivity photodetectors [2]. However, because of the high excitonic binding energy [3] we can expect that the efficiency of such devices can been improved by using MoS_2 in a donor-acceptor configuration.
I will present the femtosecond study of the photo-excitation dynamics of MoS_2 intermixed with different organic acceptor materials. Excitons are efficiently dissociated to free charges on the time scale about few ps and electrons are transfer to acceptor material.

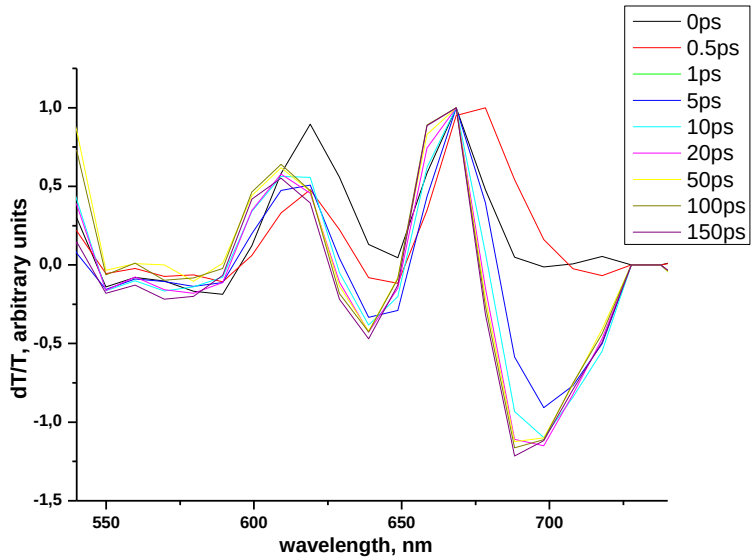

Fig. 1. Differential transmission spectra of mixture of MoS_2 with acceptor material at different time delays

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Kinetics and Dynamics of the Photo Induced (8x2)↔(4x1) Peierls-like Phase Transition of the In/Si(111) System

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The indium induced reconstruction on Silicon(111) is a famous prototype for a quasi one-dimensional wire-type arrangement of atoms on a surface. At room temperature, the indium atoms are arrangened in zig-zag chains, forming a (4x1) structure. Cooling the system down below a critical temperature of about 130 K, leads to a structural phase transition: the periodicity doubles and a (8x2) reconstructed ground state occurs which is accompanied by a metal-to-insulator transition. However, for a long time the driving force for this transition was not entirely clear. Whereas some postulated the formation of a charge density wave (CDW), others expected an order-disorder scenario. We employed high resolution LEED to follow the phase transition during slow heating and cooling rates. A robust hysteresis with a width of almost 10 K was obserserved, clearly indicating the presence of a significant energy barrier. The phase transition can thus be classified as first order type which additionally rules out a simple order-disorder scenario [1].

Ultra-fast time-resolved surface sensitive electron diffraction was used to investigate the transient non-equilibrium dynamics of the (8x2)↔(4x1) phase transition. Weak excitation of the (8x2) reconstructed groundstate with fs-laser pulses revealed the existence of a supercooled (4x1) excited state at 30 K. We confirmed that the laser induced thermal heating was less than 15 K, i.e., the sample was heated to 45 K, which is far below the transition temperature of 130 K. This indicates that the phase transition was purely driven electronically.

While the (8x2) structure was completely lifted, it was found that the supercooled excited (4x1) phase exists for several hundreds of picoseconds (Fig. 1, left). The subsequent relaxation into the (8x2) state is hindered by an energy barrier of 40 meV [2]. Pre-existing adsorbates on the surface can locally pin the (8x2) phase. After excitation, these remnant (8x2) areas act as nucleation seeds that trigger the recovery of the ground state [3]. Hence, the macroscopic relaxation proceeds even faster, the higher the density of the adsorbates (Fig. 1, right). From the recovery time as function of adsorbate density we estimate a phase front velocity of a few hundreds m/s. This scenario was predicted and confirmed by density functional theory calculations and molecular dynamics simulations.

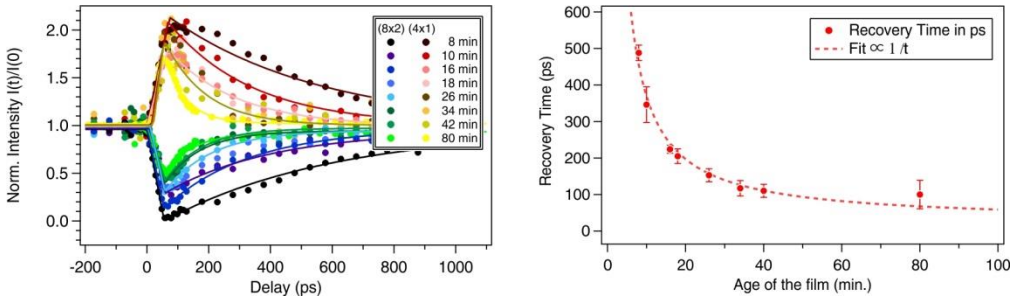

Figure: Left: Normalized spot intensity of the (4x1) and (8x2) spots. Right: Recovery time as function of the age of the film.

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Ultrafast dynamics of the Pb/Si(557) nanowire system: analysis of many-body interactions in the time domain

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Very good candidates to study the exotic physics of quasi 1D systems are self-assembled atomic wires. Particularly attractive are metallic wires adsorbed on semiconducting surfaces which are not only geometrically strongly anisotropic, but also exhibit quasi-1D Fermi surfaces. In metallic wires, rather strong correlation effects are expected due to the reduction of electrostatic screening. However, the correlations are modified in quasi-1D electron systems owing to the coupling to the environment. Interactions wire to wire and wire to substrate should also manifest themselves in the electronic structure and in an anisotropic electronic interaction and relaxation dynamics after photoexcitation.

As a prototypical system we studied the Pb/Si(557) nanowire system with the intention to explore the impact of structural and electronic band anisotropy to the ultrafast hot electron decay dynamics. To investigate elementary and microscopic interactions responsible for the population decay of optically excited electrons as a function of electron momentum we used femtosecond time- and angle-resolved two photon photoemission spectroscopy (2PPE). Two bands in the unoccupied electronic band structure are identified for which the dynamics in various points of the in-plane *k*-space was studied. We find indications of a momentum dependent population probability, but at present the band structure effects cannot be excluded.

As already reported, the electrical conductivity in the system switches from 2D to 1D behaviour upon crossing the critical temperature due to the temperature dependent structural transition [1],[2]. This structural instability opens the opportunity to study different aspects, for example temperature-dependent dynamics or the optically driven insulator-metal transition in the future.

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Symmetry-breaking phase transitions in cuprates

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The physics of the symmetry-breaking phase transitions (SBT) has attracted much of attention since it leads to better understanding of the underlying interactions and establish the range of universality of th e critical phenomena.

Conventional pump(P)-probe(pr) spectroscopy is used quite extensively for the investigation of the rel axation of the elementary excitations under near-equilibrium conditions. However, this technique does not allow us detailed investigations of the SBT. The use of an addition D pulse to the P-pr sequence en ables us to quench the system and then probe its state by the P-pr sequence at any time after the D puls e [1]. Therefore, we can observe in real time the evolution of the system through the SBT and compare it with predictions of the time-dependent Ginzburg-Landau (TDGL) theory [2].

P-pr transient reflectivity experiments have recently shed a light on the mechanism of destruction of su perconductivity [3, 4]. After absorption of photons from a laser pulse, the energy is transferred to the c ondensate, causing destruction of the superconducting state up to a certain depth. Then we observe rec overy of the system to the superconducting state. Taking into account the effect of fluctuations [5] all ows us to get systematics between the results of simulations and experimental data for recovery proces s. Despite a small kink for simulated trajectories at 10-20 ps in $La_{2-x}Sr_xCuO_4$ our model appears to be r easonable. However, in $Bi_2Sr_2CaCu_2O_{8+x}$ these kinks are more pronounced. Therefore, there is a physi cs beyond our model, that should be taken into account. For instance, the Kibble-Zurek theory of vorte x formation [6,7] could explain the kinks. However, the resolution of the experiment is not good enou gh to observe the vortices.

The authors acknowledge European Research Council for financial support.

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Ultrafast charge recombination and relaxation in photoexcited Mott-Hubbard insulator

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Recent femtosecond pump-probe experiments on Mott-Hubbard insulators reveal charge recombination, which is in picosecond range, much faster than in clean band-gap semiconductors although the excitation gaps in Mott-Hubbard insulators are larger. A calculation of the recombination rate of the excited holon-doublon pairs, based on the two-dimensional model relevant for undoped cuprates, which shows that such fast processes can be explained even quantitatively with the multi-magnon emission will be presented. We find that the recombination rate is exponentially dependent on the Mott-Hubbard gap and on the magnon energy, with a small prefactor which can be traced back to large charge-spin coupling. However, such mechanism cannot explain ps recombination times in the one-dimensional Mott-Hubbard insulators that were recently measured on a organic salt. Therefore also a more general formula for calculation of recombination times, based on a general exciton-boson coupling, will be presented as possibly relevant for the recombination due to the creation of vibrational excitations.

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