

Workshop on Ultrafast Dynamics in Strongly Correlated Systems

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Book of Abstracts

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High-Tc superconductors / 1**The symmetry and strength of the electron-phonon coupling in cuprates single crystals by Ultrafast Electron Crystallography.****Author:** Fabrizio Carbone¹¹ EPFL**Corresponding Author:** fabrizio.carbone@epfl.ch

The phonon-mediated attractive interaction between carriers leads to the Cooper pair formation in conventional superconductors. Despite decades of research, the glue holding Cooper pairs in high-temperature superconducting cuprates is still controversial, and the same is true as for the relative involvement of structural and electronic degrees of freedom. Ultrafast electron crystallography (UEC) offers, through observation of spatio-temporally resolved diffraction, the means for determining structural dynamics and the possible role of electron-lattice interaction. A polarized femtosecond (fs) laser pulse excites the charge carriers, which relax through electron-electron and electron-phonon coupling, and the consequential structural distortion is followed by diffracting fs electron pulses. In this talk, the recent findings obtained on single crystal samples are summarized. In particular, we show the strength and symmetry of the directional electron-phonon coupling in BSCCO; the theoretical implications of these results are discussed with focus on the possibility of charge stripes being significant in accounting for the observed polarization anisotropy. We show that while the average electron-phonon coupling at optimal doping is rather weak, consistent with literature reports, selected atomic motions can be coupled much stronger to polarized excitations. The coupling to the out-of plane motions of oxygen ions is also found to gain strength and exhibit a dramatic temperature dependence at lower doping. The characteristic time for electron-phonon scattering is obtained for these particular modes, and is found to be comparable to the time-scale associated to the magnetic exchange energy J .

Photodoping / 2**Theory of photoinduced electron-phonon-coupled dynamics in two-dimensional charge-ordered systems****Author:** Kenji Yonemitsu¹**Co-authors:** Nobuya Maeshima²; Yasuhiro Tanaka¹¹ Institute for Molecular Science² Institute of Materials Science, University of Tsukuba**Corresponding Author:** kxy@ims.ac.jp

Photoexcitations cause phase transitions from different types of insulators to metals in organic compounds. Their ultrafast dynamics are theoretically studied on the basis of electron-phonon-coupled wave functions in extended Holstein-Peierls-Hubbard models on anisotropic triangular lattices. We focus on 2D 1/4-filled-band charge-ordered insulators, θ -(BEDT-TTF)₂RbZn(SCN)₄ and α -(BEDT-TTF)₂I₃, which have similar Coulomb-driven charge orders and quite different photoinduced dynamics.

On picosecond timescales, couplings with inter-molecular lattice phonons are evident. In the θ compound, the charge order is quickly recovered after photoexcitation because molecular rotations stabilize the charge order in a stripe-by-stripe manner. In the α compound, the charge order is easily melted to create a macroscopic metallic domain because inter-molecular lattice phonons have much weaker effects [1].

On ten-femtosecond timescales, couplings with intra-molecular vibrations are evident. In the α

compound, a coherent oscillation of correlated electrons and subsequent Fano destructive interference with intra-molecular vibrations have been observed, which are well reproduced by calculations based on exact many-electron-phonon wave functions [2].

[1] Y. Tanaka and K. Yonemitsu, JPSJ 79, 034708 (2010).

[2] Y. Kawakami et al., PRL 105, 246402 (2010).

Charge density waves I / 3

Phase rigidity and power law decay for large-amplitude coherent phonons in bismuth

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Fully symmetric A1g coherent phonons of bismuth have been investigated in a wide range of pump pulse intensities by the ultrafast pump-probe technique. It has been shown that in the linear regime, implemented only for low pump intensities, the coherent amplitude is proportional to the pump intensity, whereas the relaxation rate of the coherent state and its frequency remain unchanged. In the nonlinear regime, which can be divided into superlinear and sublinear regions, the relaxation rate of a photoinduced lattice state can be approximated by a two-component response, where only one component depends strongly on the pump intensity. Using coherent control method it has been shown that large-amplitude A1g coherent phonons exhibit the “rigidity” of the phase, which is absent at a small amplitude. The impossibility of changing the phase of coherent oscillations appears at the excitation strength at which their amplitude relaxation law changes from exponential to power. The modification of the phase properties and relaxation law of the excitations of the crystal lattice can be understood in terms of the concept of the condensation of phonons (that is a realization of “coherent crystal”), which occurs with an increase in the excitation strength.

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Search for the photo-induced hidden phase in strongly correlated inorganic and organic systems

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Search for a novel phase and accompanying phase transitions by applying an external stimulus has been widely pursued in materials science. Photons are unique as stimulus in that the energy scale involved (~ eV) is large and the resulting excited states are expected to be very different from normally accessible ones under any other stimuli. Photoexcitation thus may lead a matter to a ‘hidden state’ not accessible by manipulating thermodynamic parameters can be utilized for ultrafast and sensitive phase control via pure photonic channel free from thermal effects. However, scarce report has been made on this issue due to difficulty in making clear the relation between changes in physical properties and the lattice structure which appears only during short lifetime of the photo-induced phase. We report here one clear structural evidence of appearance of a hidden state photoinduced with lifetime of a few nanosecond in a thin film manganite, (Nd_{0.5}Sr_{0.5})MnO₃ becoming an origin of

large change in optical properties. Using picosecond time-resolved x-ray diffraction together with subpicosecond time-resolved optical spectroscopy, we demonstrate the appearance of a transient insulating phase with a new and homogeneous lattice structure distinct from that found in the hitherto known phase diagram after photoexcitation of the manganite film in the charge, spin and orbital ordered state. We also discuss about the appearance of a hidden phase in other organic and inorganic strongly correlated systems.

High-Tc superconductors / 5

Ultrafast electric field gating of quantum transport in a cuprate superconductor

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In cuprate superconductors, tunneling between copper-oxide planes constitutes three-dimensional coherent transport. When a phase gradient of the condensate wavefunction is introduced perpendicular to the planes, the interlayer tunneling amplitude is reduced. As such, c-axis superconductivity becomes controllable by an external electric field resulting in a time-dependent phase modulation. Here, we use a single-cycle terahertz electric field to gate of superconducting transport bi-directionally in La_{1.84}Sr_{0.16}CuO₄. Oscillations between superconducting and resistive states are induced, at a frequency controlled by the electric field strength. In-plane superconductivity remains unperturbed, giving rise to an exotic state in which the dimensionality of superconducting transport is time-dependent. Ultrafast gating of interlayer coupling across individual Copper-oxide planes is of interest for device applications in high-speed nanoelectronics. It also represents a novel example of nonlinear terahertz physics, applicable to nanoplasmonics and active metamaterials.

Electrons and phonons / 6

Ultrafast changes in lattice symmetry probed by coherent phonons in VO₂

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The effect of an ultrafast change in crystal symmetry on the coherent response of the lattice is investigated using the photoinduced semiconductor-metal phase transition in VO₂. Below the photoinduced phase transition threshold, the four lowest Ag phonon modes of the monoclinic phase modulate the transient reflectivity. As the pump intensity is increased, a photoinduced phase transition occurs resulting in a prompt change in the phonon dynamics. These measurements observe

lattice dynamics on a timescale previously inaccessible to current lattice probes and show that the photoinduced phase transition is driven by excited electrons changing the lattice potential symmetry.

Magnetism I / 7

Lattice and magnetic dynamics of a laser induced phase transition in FeRh

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The interplay between the magnetic, electronic and structural degrees of freedom is often the key to understanding fundamental properties of solid state systems and forms the basis for their use in technological devices. In this context the magnetic alloy FeRh displays a phase transition consisting both of an isotropic lattice expansion and a magnetic transition from an anti-ferromagnetic to a ferromagnetic state, and thus serves as a model system to study the interaction between structural change and ferro-magnetic ordering. The ultra-fast laser-induced dynamics of this transition has previously been studied by optical methods [1,2] and x-ray magnetic dichroism [3]. Here we present the result of a time-resolved x-ray diffraction experiment which allows us to directly study the laser induced lattice dynamics in an FeRh thin-film. In addition measurements of the magneto optical Kerr effect on the same sample allow us to directly compare the structural and magnetic dynamics. The results show how the initial phase nucleation upon excitation with a fs laser starts in the surface region of the film, in agreement with static measurements of the transition, after which the created phase front moves into the now superheated film.

[1] J-U.Thiele et al., Appl.Phys.Lett. 85, 2857 (2004).

[2] G.Ju et al., Phys.Rev.Lett. 93, 197403 (2004).

[3] I.Radu et al. Phys.Rev.B 81, 104415 (2010).

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Photoinduced ultrafast local volume changes in intermediate-valence solid Ce

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We present a theoretical model for the description of the ultrafast structural response of intermediate-valence solids to femtosecond laser excitation. The approach includes the calculation of the free energy of the hot electrons produced by the laser pulse and the determination of the changes in the

thermodynamic and elastic properties of the solid as a consequence of the excitation. In particular and based on the promotional Ramirez-Falicov model, we consider the femtosecond laser heating of α cerium and the subsequent ultrafast lattice expansion dynamics. For this purpose, we determine the thermodynamic properties of cerium at very high electronic temperatures (simulating the laser excitation). The possibility for a nonequilibrium photoinduced inverse volume collapse transition is discussed. We consider both the laser-excited and the unexcited parts of the system in order to account for inertial confinement. The thermodynamic properties are obtained as function of time and used to calculate the shock velocity variation and the time scale for expansion of the heated spot into the surrounding (unheated) part of the sample. A transition on a subpicosecond time scale is predicted.

High-Tc superconductors / 9

Ultrafast transient response and electron-phonon coupling in the iron-pnictide superconductor $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$

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The real-time study of out-of-equilibrium states give access to the dynamical excitation and relaxation of electrons close to the Fermi level, tightly related to the structural and electronic properties of the material. We carried out femtosecond pump-probe reflectivity measurements on a novel pnictide high-Tc superconductor, namely $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, studying the relaxation dynamics and especially the role of coherent optical lattice oscillations observed for the first time in an iron-pnictide material [1]. After ultrafast photoexcitation, hot electrons were found to relax with two different characteristic times, indicating the presence of two distinct decay channels. Our analysis indicates that the fast relaxation should be attributed to preferential scattering of the electrons with only a subset of the lattice-vibration modes with a second moment of the Eliashberg function $\lambda\omega^2 \sim 64$ meV. The simultaneous excitation of a strong fully symmetric A_{1g} optical phonon corroborates this conclusion and makes it possible to deduce the value of $\lambda \sim 0.12$. This small value for the electron-phonon coupling confirms that a phonon-mediated process cannot be the only mechanism leading to the formation of superconducting pairs in this family of pnictides [2].

[1] B. Mansart et al., Phys. Rev. B 80, 172504 (2009)

[2] B. Mansart et al., Phys. Rev. B 82, 024513 (2010)

Ultracold atoms II / 10

Non-equilibrium dynamics of ultracold fermions in optical lattices

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Ultracold atomic gases provide an ideal playground for studying the non-equilibrium dynamics of quantum many-body systems. On one hand, key models for strongly correlated materials can be implemented with an unprecedented level of tunability and control. On the other hand, the dilute nature and exceptionally low temperatures of these gases result in long timescales for dynamical effects (on the order of milliseconds or longer), allowing for time-resolved studies. Furthermore, they exhibit a remarkably high degree of isolation from the environment and can therefore be regarded as closed quantum systems.

In our experiment we take advantage of these unique features to explore the non-equilibrium dynamics of the repulsive Fermi-Hubbard model, which is realized by trapping a repulsively interacting two-component Fermi gas in an optical lattice. Starting from a system in the metal-Mott insulator transition, we generate additional doubly occupied sites by a periodic modulation of the lattice depth. The decay of these high energy excitations is then monitored in a time-resolved manner. Over two orders of magnitude it shows an exponential dependence on the ratio of interaction energy to kinetic energy. We show that the dominant mechanism for the relaxation is a simultaneous many-body process involving several single fermions as scattering partners.

Poster session / 11

Ultrafast dynamics of crystalline bismuth studied by femtosecond pulses in visible and near-infrared range

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Ultrafast dynamics of laser excited bismuth were studied by means of wavelength-resolved femtosecond pump-probe technique. Several values of excitation wavelengths from 400 to 2300 nm and a probe femtosecond continuum in 400-900 nm range were used. The analysis of decay traces revealed three processes with relaxation time values of 1 ps, 7 ps and ~ 1 ns ascribed to groups of photoexcited electrons with essentially different coupling to the lattice. A conclusion was made that the fastest process along with the relaxation of the instantaneous frequency shift equally represent the return of the displaced equilibrium position of bismuth lattice to its unperturbed value. An intermediate 7-ps process was attributed to the crystal heating. The nanosecond component is believed to be due to electrons near the Fermi level. Spectral amplitude of A1g oscillations was also measured. The analysis of its specific shape shows that fully symmetric coherent atomic oscillations affect reflectance of bismuth crystal through the modulation of conductivity. Using different excitation wavelengths we found a sharp decrease (up to 5 times) of the coherent phonon amplitude at excitation photon energies less than 0.7 eV. Relying on this fact we conclude that only a portion of excited electrons (probably in states near the T₆₊ point of the Brillouin zone) alter the potential surface of bismuth sufficiently and cause the shift of A1g mode equilibrium position.

Poster session / 12

Spatially resolved ultra-fast magnetization dynamics tracked via resonant magnetic scattering at FLASH

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Free-electron laser (FEL) sources based on self-amplified spontaneous emission can provide intense and ultra short femtosecond x-ray pulses from the vacuum ultraviolet to the x-ray range. The unique combination of short pulselength and short wavelength allows to investigate a number of dynamic phenomena on otherwise inaccessible scales.

One of the forefront problems in modern magnetism research, namely ultra-fast demagnetization, related to non-equilibrium magnetization dynamics and manipulation of the magnetic state on ultra-fast time scales, can be addressed with the possibilities of these new sources [C. Gutt, et al., PRB, 79 212406 (2009), C. Gutt, et al., PRB 81, 100401(R) (2010).]. Here results of an optical IR-pump-FEL-probe experiment on a ferromagnetic Co/Pt multilayer with perpendicular magnetic anisotropy are shown. The optically induced ultra-fast demagnetization dynamics have been measured with resonant x-ray small angle scattering element specific at the Co M-edge allowing for simultaneous observation of the local magnetization and the characteristic length scale of the domains via the x-ray magnetic circular dichroism (XMCD) effect.

The experiments were performed at the FLASH facility at DESY in Hamburg.

Poster session / 13

Dual-Fermion approach to Non-equilibrium strongly correlated problems

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We present a generalization of the recently developed Superperturbation solver for the Anderson impurity model for the non-equilibrium case. We show that the general dual perturbation theory can be formulated on the Keldysh contour. Starting from a reference Hamiltonian system, in which the time-dependent solution is found by exact diagonalization, we make a dual perturbation expansion in order to account for the relaxation effects from the fermionic bath. Simple test results for closed as well as open quantum systems in a fermionic bath are presented.

Ultracold atoms II / 14

Cold atoms in a 1D periodically driven system

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We study the driving of a one-dimensional ultracold quantum gases an optical lattice. The driving is a periodic translation of the lattice potential in space. If the lattice itself is quickly shaken this induces effectively a change of the tunneling constant between neighboring lattice sites. This has also been recently verified experimentally. We study how the presence of such a driving affects the different quantum states that can emerge in a one-dimensional system.

New opportunities with x-ray FELs / 15

Resonant soft X-ray scattering system for LCLS experiments and hierarchical temporal scales in laser-induced transient state of nickelate

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Electronic orderings in correlated systems can compete and/or cooperate with electron itinerancy and lead to the emergence phenomena like colossal magnetoresistance (CMR) and high temperature superconductivity (HTSC). To understand how these orderings can show up from strong correlations, resonant soft X-ray scattering (RSXS) spectroscopy has been demonstrated as one of the most powerful probes for addressing this question. Although quasi-static nature of orderings has been widely studied with RSXS, nevertheless, their dynamics remains largely unexplored. In this presentation, we will show the construction of a new RSXS endstation equipped with compact-fast CCD camera that allows us to carry out time-resolved RSXS experiments at the LCLS, and the time-resolved RSXS data on stripe-phase nickelate revealing the hierarchical temporal scales for charge and spin stripes in laser-induced transient state.

Ultracold atoms I / 16

Dynamical control of many-body interactions in lattice fermion systems

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An experimental realization of lattice fermion systems using cold atoms trapped in an optical lattice allows us to study nonequilibrium dynamics of interacting fermions in an ideal situation. It is of particular interest how one can control physical properties of interacting fermions by driving the system with external fields. In particular, we focus on sinusoidal (ac) fields, which can be applied to the system by shaking the lattice potential. Here we analyze the time evolution of the fermionic repulsive Hubbard model nonadiabatically driven by ac fields using the nonequilibrium dynamical mean-field theory with the quantum Monte Carlo method.

The results show that the double occupancy, a measure of the effective interfermion interaction, decreases for ac fields of small amplitude. This can be interpreted as an effective reduction of the hopping parameter (i.e. relative amplification of the repulsion) with ac drives. Strikingly, as one increases the amplitude the double occupancy goes beyond the noninteracting value (0.25), which implies that the repulsive interaction is effectively converted to an attraction. We will discuss the physical mechanism of the dynamically controlled many-body interaction, and argue a possibility of ac-field-induced superconductivity (superfluidity) with the effective attraction.

Ultracold atoms I / 17

Slow quench dynamics in ultracold atoms

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Atomic gases cooled to Nanokelvin temperatures are a new exciting tool to study a broad range of quantum phenomena. In particular, the outstanding degree of control which has been achieved over these quantum systems facilitates access to the dynamics of strongly correlated quantum many body physics. We analyze the effect of different perturbation to a one-dimensional Bose-gas to an optical lattice. The evolution of different observables such as the experimentally accessible onsite particle distribution are studied using the time-dependent exact diagonalization and density-matrix renormalization group techniques.

Mott-Hubbard systems / 18

Nonequilibrium quantum dynamics of a charge carrier doped into Mott insulator

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We study real-time dynamics of a single carrier moving in a strongly-correlated medium under a constant electric field described by a two-dimensional t-J system, i.e., a ladder and 2D square lattice. The understanding of this subject does not only provide an extension of equilibrium studies of a

charge carrier doped into the antiferromagnetic (AFM) background, but it primarily represents a fundamental problem of a quantum particle moving in a dissipative medium where the picture of Bloch oscillations breaks down and a particle acquires a constant non-zero velocity.

Three most important findings of our study are the following:

- (i) By applying exact-diagonalization technique for a ladder, we present a scaling which effectively singles out the heating of the spin background when the carrier repeatedly encircles the ladder. As a consequence, a linear current-voltage (I-V) characteristics emerges and a carrier mobility is calculated.
- (ii) For 2D lattice, we show how the application of a novel numerical method designed to describe properties of a doped charge carrier moving in AFM background, enables calculations of real-time quantum dynamics and can reach quasistationary conditions for different F.
- (iii) We calculate I-V characteristics for 2D lattice which reveal regimes of positive and negative differential resistivity. The former regime provide estimation of carrier mobility while in the latter regime the current decreases with increasing field as $1/F$.

Poster session / 19

Exact Photo-Carrier Creation Rate in the One Dimensional Mott Insulator

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We calculate the photo-carrier creation rate in the half-filled Hubbard model in strong AC electric fields. This is done by combining the imaginary time method with the Bethe ansatz wave function, which was developed in ref. [1] in order to study the many-body Schwinger-Landau-Zener mechanism in DC electric fields. The present result is a many-body generalization of the creation rate obtained by Popov[2]. We discuss the nature of the crossover from the weak field multi-photon processes to the strong field tunneling regime.

[1] T. Oka and H. Aoki, Phys. Rev. B 81, 033103 (2010)

[2] V. Popov, JETP, 34, 709 (1972).

Magnetism II / 20

Non-equilibrium electron and spin dynamics in magnetically ordered films probed by fs x-ray pulses

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Investigations of fs laser excited non-equilibrium states are performed using soft x-ray pulses with 100 fs duration from the BESSY II femtoslicing facility. Time-resolved x-ray absorption spectroscopy (XAS), x-ray magnetic circular dichroism (XMCD), and recently resonant x-ray diffraction enable detailed measurements of the dynamic properties of magnetically ordered systems. Tuning the x-ray energy to individual absorption edges we study the laser induced changes of the valence electrons by XAS. Circularly polarized x-rays are used in XMCD measurements to follow the sub-picosecond quenching of spin and orbital angular momentum through a sum rule analysis. In Ni we found a transient increase of the spin-orbit coupling just after laser excitation, which persists during the demagnetization process. An element-resolved measurement of the magnetization dynamics in NiFe results in two different time scales for the Ni and Fe spins, demonstrating the strong influence of the laser-induced non-equilibrium state onto the exchange interaction.

Poster session / 21

TR-ARPES by High Harmonic light pulses for the study of ultrafast dynamics in correlated materials

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Angle resolved photoelectron spectroscopy (ARPES) has proved to be a leading technique in identifying static key properties of complex electron systems. In a pump-probe scheme using femtosecond light pulses this application can be extended to monitor ultrafast changes in the electronic valence structure in response to an intense optical excitation, i.e. photo-induced phase-transitions of correlated systems [Talk Rossnagel]. Here we present an experimental setup for time-resolved ARPES using femtosecond XUV probe pulses. We will focus on the relevant details and specifications of our system such as time and energy resolution, XUV photon flux and harmonic selection by a multilayer mirror monochromator. The capabilities of the setup will be exemplified by selected data that we obtained when studying phase transition dynamics in 1T-TiSe₂. We particularly show that the application of XUV pulses is highly advantageous in recording photoemission transients covering the full size of the Brillouin zone.

Poster session / 22

Combined femtosecond time-resolved photoemission at the FEL and the HHG

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The combination of time-resolved core-level photoemission spectroscopy using the free-electron laser FLASH and time- and angle-resolved photoemission spectroscopy with a table-top higher-harmonics-generation source opens the way to study the nonequilibrium dynamics of condensed matter systems with full momentum, elemental, chemical, and atomic-site selectivity. We present our experimental approaches and discuss important technical challenges at these light sources such as vacuum space-charge effects and synchronization issues. We show recent results of the atomic-site and momentum specific charge-order dynamics of the Mott insulator 1T-TaS₂ and the Peierls insulator RbTaS₂.

New opportunities with x-ray FELs / 23**The Verwey transition ultrafast: lattice and charge/orbital order go neck and neck****Author:** Niko Pontius¹**Co-authors:** A. Föhlisch¹; B. Bräuer²; C. Schüßler-Langeheine¹; C. Trabant³; C.-F. Chang³; D. Zhu²; F. Sorgenfrei⁴; H. A. Dürr²; M. Beye¹; M. Döhler³; R. Kukreja²; S. Hossain²; S. de Jong²; T. Kachel¹; W. Schlotter²; W. Wurth⁴; W.-S. Lee²¹ *Helmholtz-Zentrum Berlin für Materialien und Energie*² *SLAC*³ *University Köln*⁴ *University Hamburg***Corresponding Author:** pontius@helmholtz-berlin.de

At 120K magnetite Fe₃O₄ undergoes a metal-to-insulator transition, the Verwey transition. It is accompanied by a transition from a charge/orbital ordered state with a monoclinic symmetry to a high temperature cubic phase without electronic order. Until today the question whether this transition is mainly driven by the lattice or by electronic degrees of freedom remains unanswered.

We studied this transition using time-resolved soft x-ray diffraction at the free electron laser LCLS in Stanford. After selectively exciting the electronic system by an infrared fs-laser pulse from below the transition temperature we get direct insight into the “melting” of charge and orbital order by doing the diffraction resonantly at the Iron L3 and the Oxygen K edges. Complementary, the response of the lattice is probed using purely structural reflections.

Surprisingly, the data show that both a change of lattice symmetry and the quenching of charge/orbital order occur unexpectedly fast on a sub 200 fs timescale. Moreover, the measurements suggest the formation of a new transient phase after laser excitation, which has not been observed in thermal equilibrium before.

Charge density waves I / 24**Femtosecond time-resolved photoemission of layered charge-density-wave compounds****Author:** Kai Rosnagel¹¹ *University of Kiel***Corresponding Author:** rosnagel@physik.uni-kiel.de

Charge-density waves (CDWs) are broken-symmetry states of low-dimensional materials that are brought about by strong electron-phonon interaction. Yet, surprisingly, a clear microscopic understanding beyond this statement has not really evolved for this classical paradigm of condensed matter physics. In quasi-two-dimensional systems, for example, the common approaches based on ARPES band structure results—looking for nested sections of the Fermi surface or for a peak in the electronic susceptibility—have almost no predictive power. Apparently, a more successful explanation has to take into account the delicate balance between several factors including not only electronic and phononic structure, but also electron-electron (electron-hole) and electron-phonon interactions. Here, we will explore whether femtosecond time-resolved XPS and ARPES using pulsed extreme ultraviolet radiation generated by a free-electron laser (FLASH) and a table-top high-harmonic-generation source can provide novel insights into the relative roles that the various factors play in CDW formation. We will focus on three conspicuous CDWs in prominent members of the family of layered transition-metal dichalcogenides: the ($\sqrt{13} \times \sqrt{13}$) CDW in the Mott insulator 1T-TaS₂, the

$c(2\sqrt{3}\times 4)\text{rect.}$ CDW in the Peierls insulator RbTaS_2 , and the $(2\times 2\times 2)$ CDW in the possible excitonic insulator 1T-TiSe_2 . Our particular goal will be to reveal the relative importance of electronic (excitonic) or phononic contributions to each CDW transition by relating measured vaporization and relaxation times of CDW-induced spectral features to typical elementary time scales in layered compounds.

Magnetism II / 25

Influence of carrier scattering on the magnetization dynamics of ferromagnets

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We present microscopic theoretical results on the demagnetization dynamics in ferromagnets after excitation by ultrashort optical pulses. To this end, we include carrier scattering in the presence of spin-orbit interaction, and calculate the band and momentum resolved electron and hole distributions at low temperatures by combining the band structure including spin-orbit interaction with the scattering dynamics at the level of Boltzmann scattering integrals. We first discuss a model in which carrier-carrier scattering is included using simplified Coulomb and dipole matrix elements that are obtained from fits to experiments. Although this model gives some qualitative insight into the purely electronic contribution to the magnetization dynamics, for quantitative comparisons with experiment it needs to be supported by ab-initio input data. We do such an ab-initio based calculation for the magnetization dynamics due to the electron-phonon interaction, which is sometimes referred to as the Elliott-Yafet demagnetization mechanism. Here we use density-functional theory results for the momentum-resolved band structure as well as for the electron-phonon interaction, and dipole matrix elements. We show that this Elliott-Yafet mechanism cannot quantitatively explain the ultrafast demagnetization in ferromagnets after excitation with an ultrashort optical pulse.

Poster session / 26

Correlated Spin Dynamics in the 1D Kondo-Lattice including Triplet Scattering

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We present a theoretical investigation of spin correlations and ordering effects in a one-dimensional Kondo-lattice system, which we study for parameters that are typical of ferromagnetic semiconductors. We set up the general dynamical equations of motion for the spin correlations between itinerant and localized spins including interactions between correlations and densities at the scattering level. By computing the correlated dynamics the relevant two-particle correlation functions starting from an uncorrelated initial state, we explore ground state properties of the one-dimensional Kondo-lattice model. We find that the system exhibits long-range correlations at low temperatures, and a maximum of the correlation length at finite temperatures, which can be associated with the Kondo effect. For very low temperatures, Cooper-pair-like correlations, i.e., correlations between opposite spins and momenta with opposite signs, develop.

Poster session / 28

Time-resolved X-ray diffraction and heat propagation on LSMO/STO superlattices

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We report on the first time-resolved X-ray diffraction experiment at the EDR beamline at BESSY II performed on epitaxial La_{0.8}Sr_{0.2}MnO₃ (LSMO) and SrTiO₃ (STO) superlattices grown on single crystal STO substrate.

A Ti:Sapphire laser operating at 208 kHz frequency with 350 fs pulse duration was used as a pump beam. The X-ray pulses from a bending magnet were monochromatized using a single Ge crystal and used as a probe. Superlattice diffraction peak profiles were recorded as a function of pump-probe delay. The measured peak shift, which is proportional to the strain in the superlattice, is a measure of the thermal expansion. Heat transport in both the superlattice and the substrate has been studied on timescales of 50 ps to 4 μ s. The experimental results agree well with theoretical simulations of heat transport in such systems. The transient thermal expansion was determined with an accuracy of 10^{-7} , which corresponds to a temperature rise of 0.01°C.

High-Tc superconductors / 29

Momentum-resolved ultrafast electron dynamics in superconducting BSCCO

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In comparison to metals or semiconductors the decay of hot quasi-particles proceeds in superconductors not only by scattering of quasiparticles among each other and with phonons, but also through Cooper formation. Analysis of Cooper pair formation in the time domain has the potential to shed light on the interaction which is responsible for it. This interaction is well understood for conventional BCS superconductors, but remains controversial for High-Tc-Superconductors (HTSC). The decay of laser-excited quasi-particles in HTSC has been investigated by optical pump-probe experiments. It proceeds on femto- and picosecond time scales [1,2]. Two decay processes were distinguished in the superconducting state at temperatures $T_0 < T_c$ for a pump fluence of $\sim 10 \mu\text{J}/\text{cm}^2$. The slower time of several ps increases towards T_c and was attributed to Cooper pair reformation. The faster time is several 100 fs and is rather independent on T_0 [1]. At higher fluence a significant part of the superconducting condensate is evaporated and hot quasiparticles are excited which leads in the optical experiment to a fast contribution in the monitored relaxation [2].

In comparison to optical techniques angle-resolved photoelectron spectroscopy (ARPES) is sensitive to the electron momentum parallel to surface. Therefore, time-resolved ARPES might provide new,

relevant information for HTSC. In an earlier study we investigated BSCCO by time-resolved ARPES and estimated after high fluence excitation the electron-phonon coupling constant [4]. Recently, we achieved a sensitivity to low fluence excitation down to $6 \mu\text{J}/\text{cm}^2$ and analyzed the quasi-particle decay as a function of momentum from the nodal towards the anti-nodal point of the gap function $\Delta(E, k)$. We observe a hot quasi-particle population that increases towards the anti-node as expected from $\Delta(E, k)$ and conclude that quasi-particles off the node become metastable due to phase space restrictions in a d-wave superconductor. The decay times agree with Cooper pair recombination, however, they bear no momentum dependence, which will be discussed in the context of the boson bottleneck active in Cooper pair recombination.

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Ultracold atoms II / 30

Matrix Product Operator Simulations of Dissipative Quantum Many Body Systems

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The ability to simulate quantum many body systems in and out of equilibrium and subject to dissipative dynamics is of substantial current interest. Important applications are the description of heating processes in ultracold atomic gases in optical lattices, the modeling of transport through correlated nanostructures, and several proposals have been put forward which exploit dissipative dynamics in order to stabilize exotic quantum phases.

In this contribution we present first results obtained with our implementation of a Matrix Product Operator (MPO) approach to simulate the relevant Lindblad equation. Our implementation exploits systematically symmetries and conservation laws and is therefore able to simulate larger systems and more accurately than previous implementations. We illustrate its strength by performing simulations for local and global heating processes in one dimensional optical lattices, which reveal in interesting interplay between quantum and dissipative dynamics.

Photodoping / 31

Dynamics of orbital and spin ordering in manganites –A direct view from time-resolved resonant soft x-ray scattering

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Charge, orbital and spin orderings play an important role in transition metal oxides. In colossal magnetoresistance (CMR) manganites, the melting of the CE-type charge, orbital and spin ordering is believed to be directly relevant to the colossal change in resistance which can be induced by various external perturbations. Here I will present our recent results on the spin and orbital ordering dynamics in $\text{Pr}_{1-x}\text{Ca}_x\text{MnO}_3$ (PCMO) both at 50% ($x=0.5$) and 30% ($x=0.3$) doping using laser pump Ultrafast X-ray probe time-resolved resonant soft x-ray scattering. Our studies reveal the different response of the ordering under laser excitation for these two dopings, as well as new ordering states which can't be accessed by heating or photo doping.

Charge density waves II / 32

Ultrafast Dynamics of CDW state in TbTe3 via time-resolved resonant diffraction

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TbTe3 is a model system that exhibits a two dimensional incommensurate charge density wave state due to the Fermi surface nesting. Ultrafast pump-probe dynamics of the CDW state in such system has attracted significant interest in the field. Although coherent phonon mode and the amplitude mode, a collective excitation of the CDW state, has been observed by time-resolved pump-probed reflectivity and ARPES measurements, direct observation of the CDW state through ultrafast x-ray diffraction is not yet available. Using the ultrafast XFEL pulse, we performed resonant diffraction experiment on the TbTe3 to monitor the ultrafast response of the CDW diffraction peak. Results and comparison with the information obtained by time-resolved ARPES will be discussed.

Poster session / 33

Novel Quadratic X-Ray Magneto-Optical Effect for Time-resolved Experiments at the M Edges of the 3d Transition Metals

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We have observed a quadratic x-ray magneto-optical effect in near-normal-incidence reflection at the M edges of iron [1]. The effect appears as the magnetically induced rotation of $\sim 0.1^\circ$ of the polarization plane of linearly polarized x-ray radiation upon reflection. A comparison of the measured rotation spectrum with results from x-ray magnetic linear dichroism data demonstrates that this is the first observation of the Schaefer-Hubert effect in the x-ray regime. Ab initio density-functional theory calculations reveal that hybridization effects of the 3p core states necessarily need to be considered when interpreting experimental data. The discovered magneto-x-ray effect holds promise for future ultrafast and element-selective studies of ferromagnetic as well as antiferromagnetic materials.

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Charge density waves II / 34

Dynamics of the electronic and lattice parts of the order parameter in density wave systems

Author: Jure Demsar¹

Co-authors: A. Tomeljak ; G. Moriena ; G. Sciaini ; H. Berger ; H. Schäfer ; K. Biljakovic ; M. Beyer ; M. Eichberger ; M. Krumova ; R. J. D. Miller ; V. V. Kabanov

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Numerous advanced materials, like e.g. high-temperature-superconductors or colossal magneto-resistance compounds, owe their unique macroscopic properties to the existence of a delicate balance among different interactions on nanoscale. Thus, the study of interplay between various degrees of freedom in such system is crucial for their understanding. Charge-Density-Wave (CDW) systems, with their inherently multi-component order parameter (a modulation of the electron density accompanied by a periodic lattice distortion (PLD)), present an ideal model system for studying these cooperative phenomena. Utilizing femtosecond time-resolved optical spectroscopy [1,2] and femtosecond electron diffraction [3] an interplay between the electronic and lattice parts of the order parameter has been studied. The results suggest, that following photoexcitation with an intense optical pulse, the electronic and lattice parts of the order parameter are decoupled on the timescale shorter than the characteristic vibrational periods. The implication of these results on the interpretation of the nature of the low lying collective modes is addressed.

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Poster session / 35

Time-resolved studies of the superconducting order parameter in a BCS superconductor NbN

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Numerous femtosecond time-resolved studies of relaxation phenomena in high temperature superconductors have been performed over the last 20 years. Several competing theoretical models have been proposed, yet an overall consensus on the underlying relaxation processes is still lacking, largely due to the fact that also their ground state properties are still under debate. On the other hand, no systematic study of the dynamics in the conventional BCS superconductor existed. Here we present the first spectrally and time-resolved studies of the THz conductivity dynamics in a prototype BCS superconductor NbN. The excellent agreement of the equilibrium complex THz conductivity (0.2 - 4 THz) with the BCS theory, enabled us to apply this analysis also in the non-equilibrium state and to study the time evolution of the order parameter (superconducting gap, $2\Delta_{\text{NbN}}(4\text{K}) = 1.5$ THz) as a function of temperature and excitation density over two orders of magnitude.

Unlike in high temperature superconductors [1], the absorbed energy density required to suppress superconductivity is found to match the superconducting condensation energy. The analysis of the excitation dependent Cooper-pair breaking rate with the phenomenological Rothwarf-Taylor model [2] enabled us to determine the microscopic quasiparticle recombination rate and the value of the electron-phonon coupling constant. Probably the most fascinating observation, however, is the apparent imbalance between the photoinduced reduction of the condensate density, and that of the superconducting gap, which is observed throughout the pair-breaking process (~ 10 ps) and superconducting state recovery dynamics (~ 100 ps), which is discussed in terms of the enhanced pairing strength in the non-equilibrium superconductor.

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Magnetism I / 36

Ultrafast heating above the spin reorientation phase transition in the Co/SmFeO₃ heterostructure

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The magnetization direction of ferromagnetic (FM) films can be pinned by coupling them to an antiferromagnet (AFM). It has been demonstrated recently [A. Kimel et al., Nature, 429, 850 (2004)] that a subpicosecond laser-excitation of an antiferromagnetic orthoferrites RFeO₃, where R is a rare-earth ion, may result in ultrafast modification of the magnetic anisotropy followed by a reorientation of the antiferromagnetic spins over 90-degrees within a few picoseconds, which is much faster than a typical nanoseconds long spin precession period in a ferromagnetic material. What would be the response of an FM-layer if such a spin-reorientation in the AFM takes place in a coupled AFM-FM structure? How fast would the FM-layer reorient?

Here we report on our investigations of the laser-induced dynamics of Co spins in a Co/SmFeO₃ heterostructure using X-ray magnetic circular dichroism (XMCD) and time-resolved Photoemission electron microscopy. Orientation changes of the Co-spins faster than 100 ps is observed. The possible formation of exchange springs in the SmFeO₃ to prevent large orientation changes will be discussed.

This work is supported by EC FP7 [Grant No. NMP3-SL-2008-214469]

New opportunities with x-ray FELs / 37**The SwissFEL X-ray Laser and its Applications in Correlated Electron Materials****Author:** Rafael Abela¹**Co-author:** Bruce Patterson ¹¹ *PSI***Corresponding Author:** rafael.abela@psi.ch

Beginning in the year 2017, the SwissFEL X-ray laser at the Paul Scherrer Institut will provide users with ultra-bright, 20 femtosecond pulses of X-rays, with photon energies 180 – 12'400 eV, with a high degree of transverse coherence and at a repetition rate of 100 Hz. Shortly later, the performance will be extended to include simultaneous operation of two beamlines, circular polarization, high longitudinal coherence from seeding and a synchronized source of energetic terahertz pulses. The status of the project will be presented, and experimental methods will be proposed to perform novel time-resolved spectroscopy and diffraction/imaging on correlated electron materials.

Mott-Hubbard systems / 38**A DMFT investigation of relaxation phenomena in Mott-insulating states****Author:** Martin Eckstein¹¹ *ETH Zurich*

Nonequilibrium dynamical mean-field theory (DMFT) provides a nonperturbative way to compute the time evolution of correlated electrons on a lattice, by mapping the lattice model to an impurity model. Recently, we have implemented an impurity solver based on the non-crossing approximation and its extensions, which allows us to address the regime of strong interaction in Hubbard-like models. In this talk I will discuss applications of this method to study the pump-excitation and subsequent relaxation of Mott-insulating states. The thermalization of photo-excited carriers in the Hubbard model is shown to resemble the decay of doublons observed in experiments with ultra-cold atoms.

Ultracold atoms I / 39**Nonlinear Current Response of an Isolated System of Interacting Fermions****Author:** Marcin Mierzeiewski¹**Co-authors:** J Bonca ²; L Vidmar ³; P Prelovsek ²¹ *Institute of Physics, University of Silesia, 40-007 Katowice, Poland*² *J. Stefan Institut, SI-1000 Ljubljana, Slovenia and Department of Physics, FMF, University of Ljubljana, Jadranska 19, SI-1000 Ljubljana, Slovenia*³ *J. Stefan Institut, SI-1000 Ljubljana, Slovenia*

Nonlinear real-time response of interacting particles is studied on the example of a one-dimensional tight-binding model of spinless fermions driven by electric field. We show that for a nonintegrable

case at finite temperatures the major effect of nonlinearity can be taken into account within the linear response formalism extended by a renormalization of the kinetic energy due to the Joule heating. On the other hand, integrable systems show on constant driving a different universality with a damped oscillating current whereby the frequency is related but not equal to the Bloch oscillations. Results for a single carrier moving in a two-dimensional t-J system under a constant electric field will be discussed as well.

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Ultrafast dynamics of photoinduced phase transitions in cuprates and manganites

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In this talk, we report dynamical aspects of ultrafast photoinduced phase transitions in typical correlated electron systems of transition metal oxides (cuprates and manganites) focusing on the following three topics.

- (1) Photoinduced transitions from Mott-insulator to metal in the undoped layered cuprates (Nd₂CuO₄ and La₂CuO₄) [1, 2].
- (2) Photoinduced transitions from antiferromagnetic charge order phase to ferromagnetic metal phase in manganites (Gd_{0.55}Sr_{0.45}MnO₃ and related compounds) [3-7].
- (3) Ultrafast control of magnetization by photocarrier injection in manganites using heterostructures of manganites and titanates (La_{0.9}Sr_{0.1}MnO₃/SrTiO₃ and related systems) [8].

From the results of femtosecond pump-probe spectroscopy and transient magneto-optical Kerr effect measurements, we will discuss the charge, spin, and lattice dynamics in these photoinduced transitions.

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Magnetism II / 42

Two-stage thermalization of nearly integrable systems

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A quantum many-body system which is prepared in the ground state of an integrable Hamiltonian will not directly thermalize after a sudden small parameter quench away from integrability. Rather, it will be trapped in a prethermalized state and can thermalize only at a later stage, as observed after an interaction quench in the Hubbard model. We show that the prethermalization stage can be described by a generalized Gibbs ensemble built from approximate constants of motion in the

vicinity of the integrable point. For the second stage we derive a quantum Boltzmann equation that describes the crossover from the prethermalized to the thermalized state. For nearly integrable systems this two-stage scenario provides an understanding of thermalization from a pure initial state to a final thermal ensemble state.

Charge density waves II / 43

Structural response to non-thermal melting of a charge density wave.

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Recent developments in time resolved techniques such as angle resolved photo emission Spectroscopy and x-ray diffraction have opened new opportunities to probe directly dynamics of the electronic and structural order on femtosecond time-scales. Charge Density Waves (CDW) comprise a class of collective phenomena arising from a correlation between the electron density and the underlying lattice. 1T-TiSe₂ is one example of CDW materials. It has a quasi two-dimensional structure where Ti atoms are sandwiched between two layers of Se atoms. Below 200 K, it undergoes a second order structural phase transition into a commensurate CDW state with a (2a x 2a x 2c) superlattice.

The origin of this phase transition, although extensively studied both experimentally and theoretically, is not yet unambiguously determined. Lack of parallel areas in the Fermi surface at 2kF points eliminates FS nesting as a possible scenario for the CDW formation. There exist several competing hypotheses for the mechanism driving the CDW formation in 1T-TiSe₂. One of the most successful of these is the condensation of excitons, which becomes possible due to a low free carrier density and a consequently poorly screened Coulomb interaction. An alternative is the band Jahn-Teller effect: a lowering of the average energy of the valence and conduction bands in the vicinity of the Fermi surface as a result of lattice distortion.

Here we apply time-resolved optical reflectivity and x-ray diffraction with femtosecond resolution to study the dynamics of the structural order parameter of the charge density wave phase in TiSe₂. We find that the energy density required to melt the charge density wave phase non-thermally is four times lower than for thermal suppression of the superlattice. These results lend support to models suggesting that the charge density wave in TiSe₂ is driven by the exciton condensation.

Charge density waves II / 45

Momentum-dependent snapshots of a melting charge density wave

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Charge density waves (CDWs) underpin the electronic properties of many complex materials. When electrons are uncorrelated, CDW order is driven by the electron-phonon interaction alone, producing linearly coupled lattice and charge-density modulations whose dynamics are understood in terms of collective amplitude and phase modes. However, if electronic correlations dominate, lattice and

charge order may de-couple as the CDW can respond on a purely electronic time scale. 1T-TaS₂ is a two-dimensional compound with a CDW of controversial origin, where a nesting Fermi-surface geometry coexists with strong electronic correlations. We use time and angle resolved photoemission spectroscopy with sub-30-fs XUV pulses to map the time- and momentum-dependent electronic structure in this material. This allows us to simultaneously resolve the collapse of the Mott gap at the Fermi level, the synchronous collapse of splitting between occupied sub-bands associated with the electronic component of CDW order, and the subsequent unfolding of the Brillouin zone on a structural time scale. Our results highlight the importance of strong electronic correlations in all aspects of the low-temperature ordered phase of this material.

Mott-Hubbard systems / 46

Non-equilibrium Mott-Hubbard Systems in Strong External Laser Fields

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We consider the Hubbard model at half filling, driven out of equilibrium by a strong external laser field, represented by a classical single-mode, time-periodic electromagnetic field. We show that in an appropriate U(1) gauge the low-frequency, long-wavelength limit may be taken in a controlled way, such that no spurious infrared singularities (orthogonality catastrophe) occur, in contrast to the familiar Peierls substitution. We generalize the Dynamical Mean Field Theory (DMFT) for the Hubbard model to non-equilibrium in a time-periodic field, using the Floquet mode expansion and the Keldysh technique. Spectral densities, electronic distribution functions, relaxation rates and the DC conductivity out of equilibrium are calculated for both, the metallic and the insulating phase. In the metallic pseudogap phase, enhanced quantum coherence is predicted due to a polariton-like coupling of the electronic excitations to the discrete electromagnetic mode. This leads to a resonant revival of the Kondo-like manybody resonance at the Fermi level. In the Mott insulating phase, the external field drives a non-equilibrium insulator-metal transition as a function of the laser frequency as well as a collapse of the Mott-Hubbard gap for sufficiently high laser intensities, in qualitative agreement with experiments.

Electrons and phonons / 47

Nonequilibrium electron dynamics—photovoltaic Hall effect, ac-induced repulsion-attraction conversion

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Nonequilibrium can give rise to a variety of intriguing phenomena that are unimaginable in equilibrium. Here I shall focus on two such phenomena, both generated by intense laser light in electron systems and/or cold atom systems:

(i) Can we look for the abilities of strong laser light beyond just high-energy excitations? We propose here to evoke a topological system for controlling topological properties by dynamical (i.e., photoinduced) nonequilibrium. Namely, we predict a “photovoltaic Hall effect” in zero uniform magnetic field should occur in graphene when a circularly polarised light is applied[1]. While the spin Hall effect is known to occur when a spin-orbit interaction opens a gap in the Dirac cone, the circularly polarised light opens a dynamical gap when the k-points encircling the Dirac point acquire an Aharonov-Anandan phase. The effect is genuinely nonlinear as well as dynamical, since the photovoltaic Hall current is carried by the Floquet states drastically modified by strong laser light, and

is quadratic in the laser intensity. The required intensity of the laser field is within experimental feasibility. We can also propose an all-optical detection of the photovoltaic Hall effect, where the Faraday rotation is used to measure the optical Hall conductivity[2]. The effect is rather universal extending to multi-layer graphene and other multi-band systems[2].

(ii) A completely different, but again a genuinely nonequilibrium effect, is predicted as a “repulsion-attraction conversion” in the inter-fermion interaction[3]. For cold atom systems in optical lattices it has been known and verified that an ac modulation of the optical lattice potential renormalised the hopping energy (J) between lattice sites by Bessel’s function which oscillates with the (amplitude/frequency) of the modulation[4]. We have shown, from a time-dependent DMFT simulation, that (a) an originally repulsive inter-fermion interaction (Hubbard U) effectively changes sign to become attractive (as directly detected from the enhanced double occupancy) when we suddenly switch on (i.e., quench) the ac modulation into a region where the hopping is negative. (b) Physically, this comes from the ac-quench causing a population inversion (a negative T) on the flipped band dispersion, as confirmed from the energy and momentum distributions. This opens up a novel possibility of a dynamically induced superconductivity, since an attraction (for which $T_c \sim 0.1 J$) is obviously favourable for realising superconductivity, for which we discuss how to ramp up the ac, or how to choose appropriate electron systems.

The works described here are collaborations with Takashi Oka, Naoto Tsuji and Philipp Werner.

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Poster session / 48

Electron dynamics in Fe₃O₄(100)/MgO(100) thin layer investigated by spin-resolved photoemission

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We present dynamics studies by spin-resolved photoemission (S-PES) experiments on magnetite (Fe₃O₄), a potentially spintronic suitable material. This ferrimagnet with a high Curie temperature has been theoretically predicted to be a half-metallic material with a conductive minority-spin channel and a semiconducting majority-spin channel, resulting in 100 % spin polarisation at the Fermi level (EF) [Z. Zhang and S. Satpathy, Phys. Rev. B 44, 13319 (1991)]. But the situation remains unclear on the experimental side. We used laser-based S-PES on ex situ prepared Fe₃O₄(100)/MgO(100) thin layers. The laser photon energy (6.2 eV) being close to the photoemission threshold only a narrow region near EF can be measured. However, only the low-lying Fe t_{2g} d-bands (from EF to about 1 eV below EF) are of central interest. In “static” S-PES with 6.2 eV photon energy a change in the spin polarization sign is observed due to the presence of t_{2g} minority band. Further details on this energy region are resolved by reducing the photon energy down to 4.6 eV. Dynamical studies show that the lifetime of excited electrons in Fe₃O₄ is much longer than in an “ordinary” metal. From the spin

analysis of excited electrons we deduce that the demagnetization does not occur in the femtosecond range as it is the case for instance in Ni.

Magnetism I / 49

Ultrafast manipulation of electrons and spins with x-rays

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Polarized soft x-rays have been used over the past 20 years to obtain fascinating new insights into nanoscale magnetism. The separation of spin and orbital magnetic moments, for instance, enabled detailed insights into the interplay of exchange and spin-orbit interactions at the atomic level. The now available polarized soft x-ray pulses with only 100 fs duration allow us to observe the magnetic interactions at work in real time. The ultimate goal of such studies is to understand how spins may be manipulated by ultrashort magnetic field, spin polarized current or light pulses. In this talk I will show how intense fs x-ray pulses now available at LCLS enable us to obtain nanometer scale snapshots of the evolving spin distribution and offer novel ways of magnetic switching without damaging the sample.

Poster session / 50

Quantum quenches in the anisotropic spin-1/2 Heisenberg chain

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We study the unitary time evolution of antiferromagnetic order in anisotropic Heisenberg chains that are initially prepared in a pure quantum state far from equilibrium. Our analysis indicates that the antiferromagnetic order imprinted in the initial state vanishes exponentially. Depending on the anisotropy parameter, oscillatory or nonoscillatory relaxation dynamics is observed. Furthermore, the corresponding relaxation time exhibits a minimum at the critical point, in contrast to the usual notion of critical slowing down, from which a maximum is expected.

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Charge density waves I / 51

Transient electronic structure and coherent phonon dynamics during photoinduced insulator to metal transitions

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The electronic properties of complex materials are often governed by strong electron-phonon coupling and many-body correlation effects leading to phenomena like metal insulator transitions or superconductivity and the formation of broken symmetry ground states. This interplay between electronic and phonon degrees of freedom is of particular importance in thermally or optically driven transitions in charge-density wave (CDW) materials.

Time- and angle-resolved photoemission spectroscopy (trARPES) probes the evolution of single particle spectral function after optical excitation and makes the collective dynamics of a system (e.g. coherent phonons) directly visible through their influence on the quasiparticle band structure. Using this technique, we present a systematic study of TbTe₃, a metal which exhibits a Fermi surface nesting driven CDW transition [1]. Time-resolved data taken at different positions in the Brillouin Zone and at different temperatures enable us to observe collective modes at 3.5 THz and 2.5 THz and their highly anisotropic (k-dependent) coupling to the electronic system in real time. The 2.5 THz mode, which occurs only in the CDW band near the gapped region, is identified as the amplitude mode and governs the retarded (>100fs) collapse of the CDW gap in TbTe₃. This is in clear contrast to the previously observed quasi-instantaneous melting (\ll 50fs) observed for the Mott insulator TaS₂. In addition, a systematic study of the pump fluence dependence in the gapped region of TbTe₃ documents the crossover from a regime where mainly the amplitude mode gets excited to a regime where the CDW gap closes at least partially.

Employing a novel position-sensitive Time-of-Flight spectrometer (pTOF) [2], we are able to investigate the dynamics of both occupied and unoccupied electronic states over a continuous (2D) area of the reciprocal space. We can thereby follow the evolution of the Fermi surface and can precisely determine the position of the CDW gap even in the unoccupied band structure. For example, we can identify, for the CDW compound HoTe₃ the gap associated with the second CDW transition [1b] and investigate its dynamics in real time.

Using the well known photoinduced semiconductor-metal transition in VO₂ we address the dynamics of transient changes in crystal symmetry, which accompany most photoinduced phase transitions. We show that an ultrafast change in symmetry can be probed by the coherent (phonon) response of the lattice. Below the threshold fluence, the four lowest Ag phonon modes of the monoclinic phase modulate the transient reflectivity of VO₂. As the pump intensity is increased, a photoinduced transition is induced by excited electrons resulting in an abrupt change in the phonon dynamics which correspond to an ultrafast change of the lattice potential.

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Poster session / 52

Femtosecond magnetic order dynamics of a multiferroic phase transition

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We report on an experimental demonstration of the ultrafast switching of magnetic order in a single crystal of CuO. In the experiment, a femtosecond laser pulse initiates a transition from a collinear antiferromagnetic structure to a spiral, multiferroic magnetic structure. The first steps occur on time scales ranging from 400 fs to 2 ps, depending on the strength of the excitation. At the strongest excitation levels, the time scale is limited by the period of long-wavelength magnetic excitations in CuO. This has implications for the design of devices that rely upon high speed control over magnetism, particularly in multiferroic materials.

Photodoping / 53

Femtosecond magnetic order dynamics of a multiferroic phase transition

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