PSI Center for Photon Science

CPS Mini Symposium on
Ultrafast Condensed Matter Physics

Tuesday, 15.04.2025 | 14:00 – 17:00 | WBGB/019

SCHEDULE

14:00	Michael Grimes (MM)	Evolution of orbital 3*d* states in photoexcited CuO
14:25	Leonie Spitz (QCD/ETHZ)	Ultrafast phonon driving of the spin-Peierls system $CuGeO_3$
14:50	Aidan McConnell (QPS)	Fast optical control of silicon-based qubit candidates
15:15	**BREAK**	
15.45	Rok Venturini (QPS)	Room-temperature memristive switching of charge density wave states
16.10	Rossella Acampora (ETHZ)	Phonon-polariton nonlinearities in ferroelectric $LiNbO_3$
16.35	Clifford Allington (MM)	Investigations of the “dynamical multiferroicity” with time-resolved X-ray diffraction and magnetic circular dichroism
17.00	**END**	

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Evolution of orbital 3*d* states in photoexcited CuO

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Ultrafast time-resolved X-ray experiments have attracted increasing interest in the study of strongly correlated materials due to their ability to separate orbital, spin and charge degrees of freedom in the time domain. It is predicted that ultrafast resonant inelastic X-ray scattering (RIXS) can access and follow the transient orbital occupancy. CuO is an ideal candidate as a well-studied $3d^9$ system using an above-band gap excitation in the O 2*p* – Cu 3*d* hybridised band. The material is a magnetically frustrated charge transfer insulator with two distinct antiferromagnetic phases [1]. For $T < 213$ K, it exists as a collinear antiferromagnet (CM) which becomes incommensurate above 213 K. Femtosecond resonant magnetic soft X-ray diffraction has revealed a partial and transient transition from the CM to the incommensurate (ICM) phase in the range of 400 fs to 2 ps depending on the fluence of an intense photoexcitation with a 40-fs duration pulse [1]. Furthermore, static RIXS experiments have observed circular dichroism in the CM phase, which is thought to originate from birefringence near the Cu L_3-edge [2].

In this talk, I will discuss a recent study of the crystal field, and charge transfer excitations in CuO using laser-pumped (400 nm) time-resolved RIXS at the Cu L_3-edge in energy gain and loss spectroscopy. Circular dichroism was again observed but the absence of a pump response indicates that it is not of a magnetic origin. We focus on the crystal field *d-d* excitations and their changes as a function of delay time, which directly measures the orbital occupation. CuO shows a distinct fast (< 1 ps) response in in the L_3 pre-edge region for sufficiently high fluence (> 8 mJ/cm^2) with emergent features in the energy gain spectroscopy. On ps timescales, this fluence dependence is not seen which we attribute to the de-excitation across the band gap, *e.g.* due to multiphonon processes. These changes would be indicative of *p-d* excitations directly producing either a hole in the oxygen *p*-shell and doublons in the *d*-states or indirectly causing scattering events in the conduction band. In contrast to recent time-resolved X-ray absorption spectroscopy studies on similar materials we do not that believe direct metal-to-metal charge transfer occurs [3].

[1] Johnson *et al.*, *Femtosecond dynamics of the collinear-to-spiral antiferromagnetic phase transition in CuO*, Phys. Rev. Lett. **108**, 037203 (2012).
[2] Nag *et al.*, *Circular dichroism in resonant inelastic X-ray scattering from birefringence in CuO*, arXiv:2501.16034.
[3] Lojewski *et al.*, *Photoinduced charge transfer renormalization in NiO*, Phys. Rev. B **110**, 245120 (2024).

Ultrafast phonon driving of the spin-Peierls system $CuGeO_3$

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Coherent phonon driving with ultrafast light pulses can be leveraged to modulate magnetic exchange interactions due to their sensitivity to the bond geometry around the magnetic ions. While most research in the field was conducted on long-range ordered magnets, we focus on low-dimensional quantum spin systems which are characterized by the absence of long-ranged magnetic order down to the lowest temperatures. We exploit the strong spin-phonon coupling exhibited by the spin-Peierls material $CuGeO_3$ to study the ultrafast response of the system to coherent phonons driven by THz pulses. The material consists of quasi-1D antiferromagnetic $S = ½$ chains, which are unstable towards a lattice distortion that promotes the formation of a gapped, non-magnetic, collective singlet ground state of spin dimers below a transition temperature $T_{SP} = 14$ K. The THz pulses excite a variety of IR- and Raman-active phonons in the material as probed by time-resolved near-infrared spectroscopy and X-ray diffraction experiments, and we discuss their excitation mechanisms. Additional low-energy modes appear at frequencies below the THz pump spectrum in response to phonon driving, which we could neither attribute to phonons nor to the well-known magnetic excitations of the system. We explore the potential magnetic and magnetoelastic origins of these modes, as well as the possibility of an exclusively nonequilibrium response of the spin system to phonon driving.

Fast optical control of silicon-based qubit candidates

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Single electrons and holes bound to dopants in silicon form wavefunctions analogous to the hydrogen Rydberg states. These states have advantageous properties for quantum computing, including long spin coherence times and compatibility with well-established semiconductor fabrication techniques. Control and understanding of the excited ladder of Rydberg states is important to further development of entangling gates, similar to those used in alkali atoms. Here we use picosecond infrared pulses from a free-electron laser to control and probe both the orbital and spin states of these impurity-bound carriers, highlighting potential improvements for future quantum information processing.

Room-temperature memristive switching of charge density wave states

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Control over the novel quantum states that emerge from non-equilibrium conditions is of both fundamental and technological importance. Metastable charge density wave (CDW) states are particularly interesting as their electrical manipulation could lead to ultra-efficient memory devices [1]. However, the ability to use electrical pulses for non-volatile resistance switching involving CDW states is exceedingly rare and has been limited to cryogenic temperatures. In this presentation, I will focus on a recently discovered layered semiconductor $EuTe_4$ that exhibits the coexistence of distinct CDW orders [2, 3]. We have discovered that electrical pulses can be used for excitation to hidden, yet stable electronic states over a broad temperature range between 6 and 400 K [4]. We find that switching occurs through a non-thermal pathway and is reversible via a thermal erase procedure. As the change in electronic order is accompanied by a change in the material resistance, the electronic device acts as a memristor. Fast and energy efficient CDW switching in $EuTe_4$ holds great promise for novel memory devices and neuromorphic computing applications.

[1] Mraz *et al., Charge configuration memory devices: Energy efficiency and switching speed*, Nano Lett. **12**, 4814 (2022).
[2] Wu *et al., Layered semiconductor $EuTe_4$ with charge density wave order in square tellurium sheets*, Phys. Rev. Mater. **3**, 024002 (2019).
[3] Lv *et al., Unconventional hysteretic transition in a charge density wave*, Phys. Rev. Lett. **128**, 036401 (2022).
[4] Venturini *et al., Room-temperature memristive switching between charge density wave states*, arXiv:2412.13094.

Phonon-polariton nonlinearities in ferroelectric $LiNbO_3$

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In ferroelectric $LiNbO_3$, THz light couples with low-frequency optical phonons and forms phonon polaritons [1,2]. Recent studies have shown that is possible to probe nonlinearities at specific points along the phonon-polariton dispersion curve with different probe wavelengths after broadband excitation with strong THz transients [3,4]. However, extensive measurements of lattice anharmonicities in $LiNbO_3$ are still lacking. To bridge this gap, we characterised the nonlinear behaviour of phonon-polaritons in $LiNbO_3$ using nonlinear THz spectroscopy. We mapped the $LiNbO_3$ phonon polariton E branch by varying the probe wavelength and observed a strong dependence of the nonlinear response on the wavelength of the near-optical pulse, which arises from the momentum selection of the detection process.

[1] Crimmins, Stoyanov & Nelson, *Heterodyned impulsive stimulated Raman scattering of phonon–polaritons in $LiTaO_3$ and $LiNbO_3$*, J. Chem. Phys. **117**, 2882 (2002).

[2] Knighton, Dastrup, Johnson & Johnson, *Measurement of a phonon-polariton dispersion curve by varying the excitation wavelength*, Phys. Rev. B **97**, 214307 (2018).

[3] Ikegaya, Sakaibara, Minami, Katayama & Takeda, *Real-time observation of phonon-polariton dynamics in ferroelectric $LiNbO_3$ in time-frequency space,* Appl. Phys. Lett. **107**, 062901 (2015).

[4] Dastrup, Hall & Johnson*, Experimental determination of the interatomic potential in $LiNbO_3$ via ultrafast lattice control*, Appl. Phys. Lett. **110**, 162901 (2017).

Investigations of the "dynamical multiferroicity" with time-resolved X-ray diffraction and magnetic circular dichroism

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In a recent experiment at LCLS, we investigated the effect of high-field, circular THz excitation on the perovskite material, $EuTiO_3$. For the analogous compound $SrTiO_3$ it has been proposed theoretically [1], and evidenced experimentally [2], that the use of such a driving field resonant with an optical phonon (degenerate in the *ab*-plane perpendicular to the *k*-vector of the light) can create a transient magnetic field originating from the circular orbit of the ions. This effect, termed "dynamical multiferroicity," however, has many unresolved questions regarding the strength and origin of the transient field; including whether any non-Maxwellian contributions are present in the process [3]. To answer these questions, we investigate the response of $EuTiO_3$ during a comparable excitation, where the magnetic nature of the Eu^{2+} vs. Sr^{2+} ion is leveraged. Utilizing time-resolved (resonant) X-ray diffraction, we directly observe the circular motion of the ions by measuring a clear ±p/2 phase shift between the motion of ions along the *a*- and *b*-axes of the material with $\sigma_{THz}^{\pm}$ excitation. From this, we plan to quantify the amplitude of circular atomic motion and compare the predicted field with the simultaneous X-ray magnetic circular dichroism measurements of the Eu^{2+} ion. In this talk, the status of the project and ongoing analyses will be discussed.

[1] Juraschek & Spaldin, *Dynamical multiferroicity*, Phys. Rev. Mater. **3**, 064405 (2019).
[2] Basini, *et al.*, *Terahertz electric-field-driven dynamical multiferroicity in $SrTiO_3$*, Nature **628**, 534 (2024).
[3] Merlin, *Unraveling the effect of circularly polarized light on reciprocal media: Breaking time reversal symmetry with non-Maxwellian magnetic-esque fields*, Phys. Rev. B **3**, 094312 (2024).