Residual Stress and Ferroelastic Domain Reorientation in Declamped {001} $Pb(Zr_{0.3}Ti_{0.7})O_3$ Films

Lyndsey M. Denis-Rotella,[1] Giovanni Esteves,[2] Julian Walker,[1] Hanhan Zhou,[2] Jacob L. Jones,[2] and Susan Trolier-McKinstry[1]

[1] Materials Science and Engineering Department and Materials Research Institute, Penn State

[2] Materials Science and Engineering Department, North Carolina State University

Ferroelectric films are often constrained by their substrates and subject to scaling effects, including suppressed dielectric permittivity. In the present work, the thickness dependence of intrinsic and extrinsic contributions to the dielectric properties was elucidated. A novel approach to quantitatively deconstruct the relative permittivity into three contributions (intrinsic, reversible extrinsic and irreversible extrinsic) was developed using a combination of X-ray diffraction and Rayleigh analysis. *In situ* synchrotron X-ray diffraction was used to understand the influence of residual stress and substrate clamping on the domain state, ferroelastic domain reorientation, and electric-field-induced strain. For tetragonal {001} textured $Pb_{0.99}(Zr_{0.3}Ti_{0.7})_{0.98}Nb_{0.02}O_3$ thin films clamped to a Si substrate, a thickness-dependent in-plane tensile stress developed during processing which dictates the domain distribution over a thickness range of 0.27 μm to 1.11 μm. However, after the films were partially declamped from the substrate and annealed, the residual stress was alleviated. As a result, the thickness dependence of the volume fraction of *c*-domains largely disappeared, and the out-of-plane lattice-spacings (*d*) for both *a* and *c*-domains increased. The volume fraction of *c*-domains was used to calculate the intrinsic relative permittivity. The reversible Rayleigh coefficient was then used to separate the intrinsic and reversible extrinsic contributions. The reversible extrinsic response accounted for ~50% of the overall relative permittivity (measured at 50 Hz and AC field of $0.5 \cdot E_c$) and was thickness dependent even after poling and upon release.

Emergent topological phenomena in (111) pyrochlore iridate thin films.

Dr. Xiaoran Liu, Rutgers University

— Abstract: The interplay between electron-electron correlations and strong spin-orbit coupling has lead to tremendous predictions for correlated topological states of matter. As the archetypal candidates for magnetic Weyl semimetal, the pyrochlore iridates have been proposed to host more exotic topological phases in their thin films, manifested as the emergence of zero-field Hall conductance induced by the Berry curvature of bands. In this talk, I will show combined experimental and theoretical evidences for the observation of these emergent topological states in high-quality (111) pyrochlore iridate thin films, where intrinsic anomalous Hall effect with extremely large coercivity is demonstrated on the low-temperature magnetic phase with the peculiar all-in-all-out antiferromagnetic ordering. The nontrivial band topology is revealed as a result of the surface state induced by the termination of the film. These results highlight the intrinsic bulk-surface correspondence of the Ir sublattice in pyrochlore and open new directions for novel correlated topological materials.

Magnetically and Electrically driven Phenomena in Complex Oxide Heterostructures

Derek Meyers[1], Jieun Kim[2], Lane Martin[2]

[1]Physics Department, Oklahoma State University, Stillwater, OK 74078
2Materials Science and Engineering Department, University of California, Berkeley, Berkeley, CA 94720

In this talk we will discuss the current utilization of externally applied electric and magnetic fields for x-ray scattering experiments on novel magnetoelectric effects in oxide superlattices and films. Examples of 2D anti-ferromagnetism driven by small applied magnetic fields and electrically driven relaxor phase behavior at the morphotropic phase boundary will be emphasized. Present data analysis and modeling schemes will be discussed along with the potential for future improvements. Finally, potential applications of these multimodal measurements with the addition of sub-micron beam profiles and the increased brilliance promised by the APS upgrade will also be explored. Such capabilities are anticipated to allow more advanced device structures to be investigated.

The authors acknowledge support for this work from the Gordon and Betty Moore Foundation's EPiQS Initiative, under Grant No. GBMF5307.

Title: Machine learning augmented scattering spectroscopy for emergent quantum materials

Prof. Mingda Li, Massachusetts Institute of Technology

Abstract: Machine learning has demonstrated great power in materials science but encounters a few challenges when coming to spectroscopic data, such as data scarcity, noise and difficulty in interpretation. In this presentation, after a brief primer on deep learning, with an emphasis on the link to spectroscopies, I will provide two of our recent works on inelastic scattering and absorption spectroscopy. By building a full 3D rotational-transnational equivariant neural network, we showed that the elementary excitations of phonons can be well captured by only using atomic species and coordinates information, even when the output dimension like phonon density of states greatly exceeds the input dimension. On the other hand, by seeking for the connection between x-ray absorption spectra and the topological class, we show that the materials topological information - a global quantity in reciprocal space - can well be coded implicitly in local real space information [1]. We conclude by showing a variety of problems machine learning may solve in neutron and x-ray spectroscopic researches.

[1] N. Andrejevic, J. Andrejevic, C.H. Rycroft and M. Li. arXiv:2003.00994.

Probing the nematicity and superconductivity by the technique of elasto-XRD

Prof. Jiun- Haw Chu, University of Washington

Electronic nematicity refers to a self-organized electronic state that breaks rotational symmetry without long range translational order. In the iron-based superconductors, the nematic transition temperature can be continuously tuned by doping and pressure, which extrapolates to zero as the superconducting Tc is tuned to optimal. In the vicinity of the nematic quantum critical point, superconductivity is extremely sensitive to the anisotropic strain. Remarkably, the Tc can be fully reduced to zero under less than a percent anisotropic strain. In this talk, I will use this strain induced superconductor-metal transition as a showcase for the technique of elasto-XRD – the x-ray diffraction measurement with in-situ tunable stress. I will also discuss how the elasto-XRD reveals the complex electronic and lattice response of a sample within the nematic phase when the structural twin domains are removed by the externally applied stress.

Multi-control and Multi-probe of the Metamagnetism in the Pseudospin half Mott Insulator Sr_2IrO_4

Han Zhang[1], Lin Hao[1], Junyi Yang[1], Josh Mutch[2], Zhaoyu Liu[2], Qing Huang[1], Kyle Noordhoek[1], Andrew F. May[3], Jiun-Haw Chu[2], Jong-Woo Kim[4], Philip J. Ryan[4,5], Haidong Zhou[1] and Jian Liu[1*]
[1]Department of Physics and Astronomy, University of Tennessee, Knoxville, TN, 37996
[2]Department of Physics, University of Washington, Seattle, WA, 98195
[3]Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN, 37831
[4]Advanced Photon Source, Argonne National Laboratory, Argonne, IL, 60439
[5]School of Physical Sciences, Dublin City University, Dublin 11, Ireland

Emergent phenomenon cooperatively driven by electronic correlation and spin-orbit coupling is at the forefront of condensed matter physics. Here we show that, by applying anisotropic strain up to only 0.05%, we can in-situ modulate the metamagnetic transition field of spin-orbit-coupled Mott insulator Sr_2IrO_4 by almost 300%. Our simultaneous measurements of resonant x-ray scattering and transport reveal that this drastic response originates from the complete strain-tuning of the transition between the spin-flop and spin-flip limits, and is always accompanied by large elasto- and magneto-conductance. This enables electrically controllable and electronically detectable metamagnetic switching, despite the antiferromagnetic insulating state. The resulted strain-magnetic field phase diagram reveals that the strain introduces C_4-symmetry-breaking magnetic anisotropy to the square lattice via pseudospin-lattice coupling, directly demonstrating the pseudo-Jahn-Teller effect of the spin-orbit-coupled correlated system. The extracted coupling strength is much weaker than the superexchange interactions, yet crucial for the spontaneous symmetry-breaking, affording the remarkably efficient strain-control.

Dr. Thanh Nguyen
Massachusetts Institute of Technology

Title: New class of Kohn anomalies in Weyl semimetals

Abstract: The way electrons interact with phonons determines many physical processes that can occur within electronic devices and material systems, but generally, this subject have been difficult to investigate due to the weak strength of electron-phonon interaction. An unusual kind of electron-phonon interaction in particular is the Kohn anomaly, first discovered in the 1950s, whereby phonon softening is observed due to the divergence of the electron screening. Kohn anomalies have been previously observed in several material systems, ranging from elemental metals to carbon-based allotropes such as graphene, but not yet in topological materials whose topology imparts an inherent robustness against perturbations. In this talk, I will describe the first observation of a *chiral* Kohn anomaly in the Weyl semimetal tantalum phosphide using inelastic x-ray and neutron scattering, which were guided upon by theoretical calculations [1]. In particular, the strong agreement between theory and calculations with regards to the unique features of the new type of Kohn anomaly in this topological material can help shed light on the strength of the electron-phonon coupling, devices using chirality degrees of freedom and on the fundamental processes that would underlie some of these exotic materials.

[1] Nguyen, T. et al. Topological Singularity Induced Chiral Kohn Anomaly in a Weyl Semimetal. Phys. Rev. Lett. 124, 236401 (2020).

One-Dimensional Nature of Superconductivity in $LaAlO_3/SrTiO_3$

Prof. Jeremy Levy, University of Pittsburgh

I will describe a series of experiments that have helped to unravel the mystery of superconductivity in $SrTiO_3$ (STO). Using heterostructures formed with a thin layer of $LaAlO_3$ (LAO), we can control the conductivity of the interface with few-nanometer precision and create quasi-0D, 1D, and 2D structures. We have identified a regime in which electron pairing exists far outside the superconducting state, and have also performed experiments that probe the 1D-2D crossover in superconducting structures. Surprisingly, we have found that superconductivity in LAO/STO nanostructures appears to be localized at the conducting boundary, where ferroelastic domain walls are known to exist. This direct connection between structural domains in STO and electron pairing and superconductivity brings us close to understanding how electron pairing works in this dilute superconducting semiconductor, and provides a model system for understanding how nematicity may play a role in other higher temperature superconductors.

Title: A multi-modal approach to understanding structure-property relationships in relaxor ferroelectrics

Jieun Kim[1], Hiroyuki Takenaka[2], Yubo Qi[3], Abinash Kumar[4], Derek Meyers[5], Yun-long Tang[1], Jong-Woo Kim[6], Philip Ryan[6], Ramamoorthy Ramesh[1,7], James LeBeau[4], Andrew M. Rappe[8], Lane Martin[1,9]

1. Department of Materials Science and Engineering, University of California at Berkeley, CA, USA
2. Department of Physics, University of California at Santa Cruz, CA, USA
3. Department of Physics, Rutgers University, NJ, USA
4. Department of Materials Science and Engineering, Massachusetts Institute of Technology, MA, USA
5. Department of Physics, Oklahoma state University, OK, USA
6. Advanced Photon Source, Argonne National Laboratory, IL, USA
7. Department of Physics, University of California at Berkeley, CA, USA
8. Department of Materials Science and Engineering, University of Pennsylvania, PA, USA
9. Materials Sciences Division, Lawrence Berkeley National Laboratory, CA, USA

Abstract

Relaxor ferroelectrics have attracted significant interests in the past decade due to their intriguing dielectric and piezoelectric properties in single crystal forms. A multi-modal approach is particularly important in understanding structure-property relationships in relaxor ferroelectrics due to the complex nature of relaxor behavior that originates from competition of various forces over a large range of length scales. Through a multi-modal approach that combines synchrotron-based temperature- and field-dependent X-ray techniques, molecular dynamics simulations, scanning transmission electron microscopy, and dielectric spectroscopy, etc., we aim to elucidate the structure-property relationships in epitaxial thin films of relaxor ferroelectrics, with a special emphasis on understanding the role/response of mesoscale domain structures to external stimuli.

In this talk, we will introduce our recent advances in understanding relaxor behavior through such a multi-modal approach, with a particular emphasis on *in operando* measurements enabled by recent developments at beamline 6-ID-B in APS. We will also discuss future directions that advances in X-ray instrumentation may enable us to study.

A first-principles description of stronger correlations: Novel superconductors to topological phases and ultrathin 2D films

Arun Bansil
Physics Department
Northeastern University
Boston, Massachusetts USA

I will discuss how advanced density functionals are enabling new insights into the electronic structure, phase diagrams and magnetism of a wide variety of materials that have been traditionally considered to be so strongly correlated as to lie outside the scope of first-principles predictive treatment. A spectacular example is provided by the cuprate high-Tc superconductors in which the density functional theory fails to correctly predict the half-filled parent compounds to be insulators. In sharp contrast, however, the recently constructed SCAN functional not only reproduces the insulating character and magnetism of the half-filled cuprates, but also captures the transition to the metallic state with doping without invoking any free parameters such as the Hubbard U. [1] I will also comment with examples drawn from our recent work on the opportunities for a new generation of predictive modeling in correlated materials more generally, including the topological phases of quantum matter and ultrathin 2D films, which are drawing intense current interest [2-5]. Work supported by the U.S. Department of Energy.

[1] Y. Zhang *et al*., *Proceedings of the National Academy of Sciences 117*, 68 (2020).
[2] A. Bansil, H. Lin and T. Das, *Reviews of Modern Physics 88*, 021004 (2016).
[3] C. Hu *et al*., *Science Ad*vances 6, eaba4275 (2020).
[4] I. Belopolski *et al*., *Science 365*, 1278 (2019).
[5] Z. Hennighausen *et al.*, *Nanoscale 11*, 15929 (2019).

Biosketch of Professor Arun Bansil, Northeastern University

Bansil is a University Distinguished Professor of physics at Northeastern University (NU). He served for over two years at the US Department of Energy managing the flagship Theoretical Condensed Matter Physics program (2008-10). He is an academic editor of the international *Journal of Physics and Chemistry of Solids* (1994-), the founding director of NU's Advanced Scientific Computation Center (1999-) and serves on various international editorial boards and commissions. He has authored/co-authored over 400 technical articles and 17 volumes of conference proceedings covering a wide range of topics in condensed matter and materials physics, and a major book on *X-Ray Compton Scattering* (Oxford University Press, Oxford, 2004). Bansil is a Highly Cited Researcher (ISI Web of Science/Clarivate Analytics).

The chiral qubit: quantum computing with Dirac/Weyl semimetals

Prof. Prof. Dmitri Kharzeev, Stony Brook University

The quantum chiral anomaly enables a nearly dissipationless current in the presence of chirality imbalance and magnetic field -- this is the Chiral Magnetic Effect (CME), observed recently in Dirac and Weyl semimetals. It is proposed (work with Qiang Li) to utilize the CME for the design of qubits potentially capable of operating at THz frequency, room temperature, and the coherence time to gate time ratio of about 10,000. The proposed "Chiral Qubit" is a micron-scale ring made of a Weyl or Dirac semimetal, with the $|0\rangle$ and $|1\rangle$ quantum states corresponding to the symmetric and antisymmetric superpositions of quantum states describing chiral fermions circulating along the ring clockwise and counter-clockwise. A fractional magnetic flux through the ring induces a quantum superposition of the $|0\rangle$ and $|1\rangle$ quantum states. The entanglement of qubits can be implemented through the near-field THz frequency electromagnetic fields, as suggested by recent experiments.

Title: multi-modal-based optimization method for tomographic reconstruction.

Dr. Wendy Di ZiChao, Argonne National Laboratory.

Abstract: A recurring challenge in modern imaging problems is how to combine complex data with large-scale models to create better predictions. Recent advances in integration of different types of models from the perspective of both the data fidelity and modality have been achieved which make the solution of such large-scale, complex problems more tractable and accurate. In this talk, we will show 1) how to integrate different modalities and formulate an optimization approach to simultaneously reconstruct the composition and absorption effect in the sample, and 2) how to combine the complementary information from multichannel data to model and calibrate experimental error using an optimization-based approach.

Title: Understanding strong correlations of quantum materials from first-principles.

Prof. Hyowon Park, University of Illinois, Chicago.

Abstract: Strongly correlated materials are promising candidates for future applications since their emergent electronic properties such as the Mott transition, spin-state transition, and charge ordering are strongly coupled to each other. One of the grand challenges in modern material science is the theoretical understanding of strong correlations occurring in quantum materials using first-principles. Dynamical mean field theory (DMFT) has been a successful first-principles method for the study of the electronic structure in strongly correlated materials, especially when it is combined with density functional theory (DFT). In this talk, we will present the DFT+DMFT study of the spin-state transition occurring in $LaCoO_3$ and the oxygen vacancy effect on $LaNiO_3$. We find that the homogeneous spin-state excitation in bulk $LaCoO_3$ at elevated temperature shows the dynamically fluctuating nature of high-spin and low-spin configurations accompanied by the volume expansion. Similarly, as the charge ordering phase of the tensile-strained $LaCoO_3$ film found in the resonant X-ray scattering experiment, we can stabilize the mixed spin-state phase with alternating high-spin and low-spin phases accompanied by the charge ordering in bulk $LaCoO_3$. The nature of the high-spin site becomes a Mott insulator while the low-spin site behaves as a band insulator and this mixed spin phase becomes energetically very close to the homogeneous spin phase. Also, oxygen vacancy plays an important role in bulk $LaNiO_3$ as the metal-to-insulator transition occurs accompanied by magnetism as the oxygen vacancy level gets increased. Using DFT+DMFT, we studied the change of the electronic structure of $LaNiO_3$ with oxygen vacancies and successfully compared to spectroscopic data measured in PES and XAS experiments. We clarified the nature of the insulating state in vacancy-ordered $LaNiO_{2.5}$ as the site-selective Mott phase occurs from both structural and charge-transfer effects of oxygen vacancies, which can not be explained from the result of the rigid band shift based on the homogeneous vacancy scenario.

Tuning electronic and magnetic orders with strain and magnetic field

Shua Sanchez, University of Washington

Strongly correlated electron materials feature a strong coupling between electronic, magnetic and structural degrees of freedom. Here we discuss two recent results from an iron-based high temperature superconductor, EuFe2As2, in which we use simultaneous applied strain and magnetic field to separately tune different orders. The undoped parent compound exhibits unusually strong magnetoelastic coupling due to the interaction of Eu and Fe magnetic moments; we applied field to tune the relative angle of these moments, probed using x-ray magnetic circular dichroism (XMCD), and use applied strain to tune the system between a continuous spin canting and a metamagnetic transition. In the Co-doped sample, superconductivity interacts with Eu ferromagnetism to yield field-induced superconductivity. We tune the superconductivity with strain via its coupling to the lattice and use XMCD to explore its field enhancement due to reorientation of Eu moments.