WORKSHOP #15

Nanoscale Structure in Quantum Materials Under External Stimuli

Organizers: Milinda Abeykoon (NSLS-II, BNL), Emil Bozin (CMPMSD, BNL), João Horta Belo (IFIMUP/FCUP Portugal), Trevor Tyson (NJIT)

Complex interplay of electronic degrees of freedom and the host lattice structure renders the functional properties of quantum materials amenable to external stimuli, such as the temperature and magnetic or electric fields. This is exemplified by magnetic materials, some of which exhibit pronounced magnetostriction effects or changes in local and/or long-range ordered structures induced by an external magnetic field applied at temperatures close to the ordering temperatures. Alignment of the electronic magnetic moments in the direction of applied field results in these intrinsic and reversible structural modifications. Magnetostriction effects are widely utilized in various important technologies including sensors, actuators, and transformers. Similarly, polarization-related atomic-scale responses of dielectrics and ferroelectrics to electric fields is central to their functionality. The charge-storing ability makes dielectrics exceptional candidates for capacitors and radio frequency transmission lines. On the other hand, ferroelectrics are widely used for tunable capacitors, sensors, and energy storage devices. To fully grasp the underlying mechanisms and adopt novel device-design principles, it is critical to enable atomic structure studies in these materials over a wide field-temperature parameter space. The 28-ID-1 (Pair Distribution Function, PDF) beamline at the NSLS-II currently offers an opportunity to study materials under variable magnetic field and temperature, while electric field capabilities are under consideration. 28-ID-1 offers PDF, complementary Small, and Wide-Angle X-ray scattering capabilities enabling parametric studies on various structural scales from sub-nanometer to micron. This workshop will bring together the existing user community, the leading researchers in the field of quantum materials, and prospective new users to benchmark the existing and identify the emerging material classes that would benefit the most from these unprecedented beamline capabilities.

Start Time (ET)	Title	Speaker (Affiliation)
1:30 – 1:45 p.m.	Opening Remarks	Organizers (BNL/NJIT/IFIMUP/FCUP Portugal)
1:45 – 2:25 p.m.	Magnetic-field-induced transitions between competing phases	John Tranquada (Brookhaven National Laboratory)
2:25 – 3:05 p.m.	Structural Analysis of Quantum Materials: From Design to Discovery	Sara Haravifard (Duke University)
3:05 – 3:35 p.m.	Resolving emergent structure states in quantum materials by variable temperature total x-ray scattering in magnetic field	Valeri Petkov (Central Michigan University)
3:35 – 4:05 p.m.	Local Structural Studies in Electric Fields using Total Scattering Techniques	Jacob L. Jones (North Carolina State University)
4:05 – 4:15 p.m.	Break	
4:15 – 4:45 p.m.	Structure-Property Relationships in Rare Earth Intermetallic Compounds with Reversible First-Order Transformations	Yaroslav Mudryk (Ames Laboratory)
4:45 – 5:15 p.m.	Rich surprises in a “simple” material: A pair distribution function story	Ben Frandsen (Brigham Young University)
5:15 – 5:45 p.m.	Concluding Remarks and Discussion	All

ABSTRACTS:

Magnetic-FIeld-Induced Transitions Between Competing Phases

John Tranquada
Brookhaven National Laboratory

Correlated-electron systems frequently exhibit competing electronic phases. In some cases, it is possible to tune between such phases by application of a magnetic field. A now classic example involves colossal magnetoresistance in compounds such as La1-xSrxMnO3, in which a first-order structural transition occurs between a high-temperature paramagnetic insulator and a low-temperature ferromagnetic metal. An applied magnetic field favors the ferromagnetic phase and shifts the transition. More subtle structural effects are possible in layered nickelate and cuprate compounds that exhibit charge-stripe order. For example, in La2NiO4.133 at a temperature corresponding to the start of a Devil's staircase of stripe phases, a magnetic field applied in plane can induce a sharp jump in charge-stripe wave vector [1]. In the cuprate La2-xBaxCuO4 with x = 0.095 and at low temperature, a magnetic field applied perpendicular to the planes can enhance charge-stripe order while simultaneously driving a transition from bulk- to two-dimensional superconductivity [2]. Related behavior has been seen in other cuprates.

1. J. M. Tranquada et al., Phys. Rev. B 55, R6113 (1997).
2. Jinsheng Wen et al., Phys. Rev. B 85, 134513 (2012).

Structural Analysis of Quantum Materials: From Design to Discovery

Sara Haravifard
Duke University

One of the critical aspects of understanding novel emergent phenomena in Quantum Materials is to shed light on their structural properties and the role they play. From the initial steps of design to the prospect of discovery of exotic properties in these systems, the necessity for providing clear picture for structural evolutions has been proven in an array of systems. In this talk we present our recent efforts in understanding the connection between crystal structure and magnetism in frustrated quantum materials. We further show how such studies can be used to map the phase diagram in candidate materials.

Resolving Emergent Structure States in Quantum Materials by Variable Temperature Total X-ray Scattering in Magnetic Field

Valeri Petkov
Central Michigan University

Materials with properties rooted in the quantum world exhibit coupled electronic, magnetic and lattice degrees of freedom leading to fascinating physics and practical applications. Moreover, the coupling renders their ground state susceptible to perturbations, providing an opportunity to investigate it by varying important physical parameters such as temperature, pressure, electric and magnetic field. In the talk, we will present results of recent variable temperature and magnetic field total x-ray scattering studies on the spin-lattice coupling in the archetypal magnetocaloric Gd5(Ge,Si)4 [1], metal-insulator transition in pure and Ti-substituted bilayer Ca3Ru2O7 [2], magnetic field driven orthorhombic to hexagonal phase transition in strongly correlated MnAs [3] and 3D charge density wave in “dual” heavy fermion liquid UPt2Si2 [4]. In these systems, the lattice degrees of freedom appear as local lattice distortions bridging proximal electronic phases with distinct ground states.

[1] V. Petkov, T. D. Rao, AM Milinda Abeykoon, Jorge R. Galeano-Cabral, and K. Wei, Phys. Rev. Materials 6, 104407 (2022).
[2] V. Petkov, T. Durga Rao, A. Zafar, AM Milinda Abeykoon, E. Fletcher, J. Peng, Z.Q. Maoand, X. Ke, J. Phys.: Condens. Matter 51, 015402 (2023).
[3] V. Petkov, A. Zafar, T. D. Rao, and AM Milinda Abeykoon, Phys. Rev. B (under review)
[4] V. Petkov, R. Baumbach, AM Milinda Abeykoon and J.A. Mydosh, Phys. Rev. B (under review).

Acknowledgments: The reported research was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under Award No. DE-SC0021973 and used resources of the National Synchrotron Light Source at the Brookhaven National Laboratory provided by the DOE Office of Science under Contract No DE-SC0012704.

Local Structural Studies in Electric Fields using Total Scattering Techniques

Jacob L. Jones
Department of Materials Science and Engineering, North Carolina State University
Science and Technologies for Phosphorus Sustainability Center, North Carolina State University

The importance of structure-property relationships in materials research cannot be overstated. At the crystallographic level, the structure relates directly to nearly all physical properties ranging from thermal expansion and elastic compliance to dielectric polarization, piezoelectricity, and pyroelectricity. In ferroelectric crystals, in particular, there is great complexity in the crystal structures investigated today with examples including disordered atomic displacements, chemical disorder, octahedral tilting, and incommensurate modulations. A growing emphasis on in situ characterization, e.g., crystallographic study during application of electric fields or mechanical stress, further increases the challenges in determining and describing the structural features of interest.

In this talk, I will first describe the application of Pair Distribution Function (PDF) methods for characterizing local structures in dielectric, piezoelectric, and ferroelectric ceramics [1]. PDFs can be determined from X-ray or neutron total scattering measurements and provide information about structure that is invisible to Bragg diffraction. The interesting local structures in a variety of compounds and solid solutions measured by our group and other researchers will be described. I will then present

the development and application of in situ PDF methods for determining how insulating materials respond to electric fields, providing critical insight to understanding of dynamic structure-property relationships [2-3].

[1] Hou, Zhao, Paterson, Li, and Jones, https://doi.org/10.1016/j.jeurceramsoc.2017.12.003
[2] Usher, Levin, Daniels, and Jones, http://dx.doi.org/10.1038/srep14678
[3] Usher, Forrester, McDonnell, Neuefeind, Page, Peterson, Levin, and Jones, https://doi.org/10.1063/1.5037609

Structure-Property Relationships in Rare Earth Intermetallic Compounds with Reversible First-Order Transformations

Y. Mudryk
Ames Laboratory

Strong non-linear responses to external stimuli are commonly realized across first-order phase transformations. Such magneto-structural or magneto-elastic transformations in solid state compounds, driven by applied magnetic field, may produce fundamentally interesting and technologically relevant phenomena, such as colossal magnetoresistance or giant magnetocaloric effect. For magnetic field to be an effective thermodynamic driver a strong coupling of magnetic and crystallographic sublattices, which exists in magnetic rare earth intermetallics, is required. First-order nature of the transition can drastically increase magnetocaloric effect but, often, it does so at the cost of hysteresis losses. Recently, R2In compounds with R = Pr, Nd, and Eu were found to exhibit nearly anhysteretic first order magnetoelastic transitions. Here we will review the basic physical behaviors of R2In compounds, discuss what distinguishes from other magnetically responsive materials, and present important open science questions that remain unanswered. We will compare their structural behaviors in the vicinity of the magnetic ordering with the behaviors of other rare earth compounds that undergo strong magneto-structural transformations, Gd4ScGe4 and Nd7Pd3. Finally, we will discuss opportunities for basic characterization of R2In and similar compounds using temperature-, magnetic field-, and, potentially, time-dependent in situ mapping of their crystal structure.

This research was performed at Ames National Laboratory and supported by the Division of Materials Science and Engineering of the Office of Basic Energy Sciences, Office of Science of the U.S. Department of Energy (DOE). Ames National Laboratory is operated for the U.S. DOE by Iowa State University of Science and Technology under Contract No. DE-AC02-07CH11358.

Rich Surprises in a "Simple" Material: A Pair Distribution Function Story

Ben Frandsen

Brigham Young University

Structurally and chemically, the semiconductor manganese telluride (MnTe) would appear to be a rather simple—one might almost say boring—material. A binary compound with a high-symmetry hexagonal crystal structure, even the fact that it becomes antiferromagnetically ordered below 307 K does not seem particularly remarkable, as it adopts a very standard A-type antiferromagnetic structure. Nevertheless, this seemingly simple material possesses outstanding properties that make it a promising candidate for multiple applications, including as a high-performance thermoelectric material and a platform for antiferromagnetic spintronics. It can also be considered a structural component of the candidate magnetic topological insulator $MnSb_2Te_4$. Regarding the thermoelectric properties, it was recently pointed out that robust nanoscale magnetic correlations present in the paramagnetic phase can significantly enhance the thermopower, leading to an impressive thermoelectric figure of merit when lightly doped. Intrigued by the role played by short-range correlations, we initiated a comprehensive atomic and magnetic pair distribution function (PDF) study of MnTe using x-ray and neutron total scattering, collecting data across a wide temperature range and with an in situ magnetic field using the unique capabilities at beamline 28-ID-1 at NSLS-II. The data have yielded rich insights into MnTe, revealing a much more complex picture of nanoscale structural and magnetic correlations that have direct bearing on the observed macroscopic properties. In this talk, I will discuss how our x-ray PDF analysis, combined synergistically with magnetic PDF analysis of neutron total scattering data, has led to a deeper understanding of the large thermoelectric figure of merit, uncovered an exceptionally large magnetovolume effect driven by a novel magnetostructural coupling mechanism, and revealed an emphanisis-like distortion of the local structure that is similar to the well-known effect in other thermoelectrics like PbTe but seems to have a completely different origin. The abundance of interesting results in MnTe demonstrates that even seemingly simple materials may have hidden secrets rich in information when analyzed with the proper tools.