APS meV-IXS Program Overview

Ahmet Alatas, Ayman Said
Advanced Photon Source, Argonne IL

Inelastic x-ray scattering is a unique technique that has increasing impact in addressing lattice dynamics related issues in wide range of scientific areas including condensed matter physics, geophysics and biology.

In this talk, an overview of the high-energy resolution momentum-resolved IXS program at the APS will be presented. This includes the current status of the spectrometers at sectors 3 and 30. The current beamline capabilities and the recent upgrades will also be discussed [1-3]. Moreover, future developments planned for sector 30 under APS-U enhancement project will be discussed.

[1] Said A.H. *et.al.,* J. Synch. Rad. **27**, 827, (2020)
[2] Toellner T.S. *et.al.*, J. Synch. Rad. **18**, 605, (2011)
[3] Sinn H., J. Phys. Cond. Matt. **13**, 7525, (2001)

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Anomalous suppressed thermal conduction by electron-phonon coupling in CDW TaS2

Jiawang Hong

School of Aerospace Engineering, Beijing Institute of Technology, Beijing 100081, China.

Charge and thermal transport in a crystal is carried by free electrons and phonons, the two most fundamental quasiparticles. Above the Debye temperature, phonon-mediated thermal conductivity is typically limited by mutual scattering of phonons, resulting in lattice thermal conductivity decreasing with inverse temperature, whereas free electrons play a negligible role. In this talk, we will discuus an unusual temperature-independent lattice thermal conductivity in charge-density-wave tantalum disulfide. From inelastic X-ray scattering measurements and first-principles calculations, it is found that the conventional phonon–phonon scattering is alleviated by its uniquely structured phonon dispersions, and unusually strong electron-phonon coupling arises from its Fermi surface strongly nested at wavevectors in which phonons exhibit Kohn anomalies. The finding reveals new physics of thermal conduction, offers a unique platform to probe electron phonon interactions, and provides potential ways to control heat flow in materials with free charge carriers.

Soft phonon modes in materials with competing phases

F. Weber[1], A.H. Said[2], R. Heid[1], S. Rosenkranz[3]

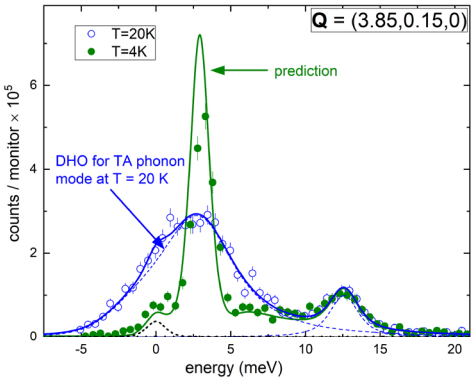
[1] *Institute for Quantum Materials and Technologies, Karlsruhe Institute of Technology, Germany*

[2] *Advanced Photon Source, Argonne National Laboratory, Lemont, Illinois, 60439, USA*

[3] *Materials Science Division, Argonne National Laboratory, Lemont, Illinois, 60439, USA*

I will review work on two compounds featuring intertwined structural and superconducting phase transitions: The *A15* superconductor V_3Si features a structural/martensitic transition temperature of T_s = 18.9 K and T_c = 16.8 K whereas *2H*-$NbSe_2$ is a seminal charge-density-wave (CDW) compound in which charge order, T_{CDW} = 33 K, coexists with superconductivity, T_c = 7.2 K. For both compounds inelastic x-ray scattering (IXS) provides unique possibilities study lattice dynamics and, thereby, obtain a detailed understanding of the electron-phonon coupling (EPC) relevant for the intertwined phases.

Phonon measurements in single crystals of V_3Si were complicated in the past because of unfavourable neutron scattering properties. Hence, only few studies of the lattice dynamical properties with momentum resolved methods were published, in particular below the superconducting transition temperature T_c. In our combined experimental and theoretical investigation of lattice dynamics in V_3Si [1] we focus on the evolution of the transverse acoustic soft phonon mode of the structural phase transition at T_s = 18.9 K, and discuss its relevance with regard to the value of T_c. Furthermore, we explain superconductivity-induced anomalies in the line shape of several acoustic phonon modes using a model proposed by Allen et al.[2] (see Figure on the right).



2H-$NbSe_2$ turned out to be a seminal example of CDW order driven by momentum-dependent EPC. I will review EPC properties via studies of the soft phonon mode [3,4] and the Fermi surface [5] along with models explaining the surprisingly low CDW transition temperature [6,7]. Measurements in the superconducting phase of *2H*-$NbSe_2$ [8] will be discussed in comparison to results in V_3Si [1] revealing the detailed sampling of the Fermi surface [5] by phonons with strong EPC.

[1] A. Sauer *et al.*, Physical Review B **99**, 134511 (2019).
[2] P. B. Allen *et al.*, Physical Review B **56**, 5552 (1997).
[3] F. Weber *et al.*, Physical Review Letters **107**, 107403 (2011).
[4] F. Weber *et al.*, Physical Review B **87**, 245111 (2013).
[5] F. Weber, *et al.*, Physical Review B **97**, 235122 (2018).
[6] F. Flicker *et al.*, Nat Commun **6**, 7034 (2015).
[7] M. Leroux *et al.*, Physical Review B **92**, 140303 (2015).
[8] F. Weber *et al.*, Physical Review B **94**, 140504(R) (2016).

Anharmonic phonons near lattice instabilities: beyond harmonic quasiparticles

Olivier Delaire, Duke University

A detailed understanding of atomic vibrations in solids is needed to refine microscopic theories of thermal transport and thermodynamics, and is of practical interest for the design of future materials. In particular, quasiparticle interactions arising from phonon-phonon, electron-phonon, or spin-phonon couplings are key to rationalize numerous functional properties, ranging from multiferroics for information processing and superionics for solid-state batteries, to thermoelectrics for waste-heat harvesting, or metal-insulator transitions for ultrafast transistors. Near phase transitions associated with phonon instabilities, one needs to properly account for the effect of strong anharmonicity, which disrupts the quasiharmonic phonon gas model through large phonon-phonon coupling terms. Large phonon amplitudes can also amplify the electron-phonon interaction and lead to renormalization of a material's electronic structure. These interactions, often neglected in textbooks, remain insufficiently understood but could open the door to new and improved material functionalities.

State-of-the-art inelastic x-ray scattering techniques are key to probe atomic dynamics in small samples and complement more traditional neutron scattering approaches. By mapping phonon spectral functions across reciprocal space, and comparing with theoretical models, phonon anharmonicity and couplings to other degrees of freedom are revealed in great detail. Our investigations bring direct insights into phonon scattering mechanisms, including anharmonicity, electron-phonon coupling, spin-phonon coupling, or scattering by defects and nanostructures. Our first-principles simulations enable the quantitative rationalization of these effects, for example with ab-initio molecular dynamics simulations and anharmonic renormalization at finite-temperature. This presentation will highlight results from our investigations of atomic dynamics in several classes of materials impacted by lattice instabilities, such as halide perovskite photovoltaics [1], ferroelectrics and multiferroics [2], thermoelectrics [4,5], superionic conductors [6,7], and VO_2 across its metal insulator transition [8,9].

[1] T. Lanigan-Atkins*, X. He*, M. J. Krogstad, D. M. Pajerowski, D. L. Abernathy, Guangyong NMN Xu, Zhijun Xu, D.-Y. Chung, M. G. Kanatzidis, S. Rosenkranz, R. Osborn, and O. Delaire, "**Two-dimensional overdamped fluctuations of soft perovskite lattice in $CsPbBr_3$**", Nature Materials (2021), https://doi.org/10.1038/s41563-021-00947-y

[2] D. Bansal, J. L. Niedziela, S. Calder, T. Lanigan-Atkins, R. Rawl, A. H. Said, D. L. Abernathy, A. I. Kolesnikov, H. Zhou, and O. Delaire, "**Magnetically-driven phonon instability enables the metal-insulator transition in *h*-FeS**", Nature Physics 16, 669–675 (2020). https://doi.org/10.1038/s41567-020-0857-1

[4] J. Ding, T. Lanigan-Atkins, M. Calderon-Cueva, A. Banerjee, D. L. Abernathy, A. Said, A. Zevalkink, and O. Delaire, "**Anharmonic phonons and origin of ultralow thermal conductivity in Mg_3Sb_2 and Mg_3Bi_2**", Science Advances, 7, no. 21, eabg1449 (2021). https://doi.org/10.1126/sciadv.abg1449

[5] T. Lanigan-Atkins*, S. Yang*, J. L. Niedziela, D. Bansal, A. F. May, A. A. Puretzky, J.Y.Y. Lin, D. Pajerowski, T. Hong, S. Chi, G. Ehlers, and O.Delaire. "**Extended anharmonic collapse of phonon dispersions in SnS and SnSe**", Nature Communications 11 (1), 1-9 (2020). https://doi.org/10.1038/s41467-020-18121-4

[6] J. L. Niedziela, D. Bansal, A. F. May, J. Ding, T. Lanigan-Atkins, G. Ehlers, D. L. Abernathy, A. Said & O. Delaire, "**Selective Breakdown of Phonon Quasiparticles across Superionic Transition in $CuCrSe_2$**", Nature Physics, 15, 73–78 (2019). https://doi.org/10.1038/s41567-018-0298-2

[7] J. Ding, J. L. Niedziela, D. Bansal, J. Wang, X. He, A. F. May, G. Ehlers, D. L. Abernathy, A. Said, A. Alatas, Y. Ren, G. Arya, and O. Delaire, "**Anharmonic lattice dynamics and superionic transition in $AgCrSe_2$**", PNAS 117 (8) 3930-3937 (2020). https://doi.org/10.1073/pnas.1913916117

[8] S. Wall*, S. Yang,* L. Vidas, M. Chollet, M. Glownia, M. Kozina, T. Katayama, T. Henighan, M. Jiang, T. A. Miller, D. A. Reis, L. Boatner, O. Delaire+ and Mariano Trigo+, "**Ultrafast disordering of vanadium dimers in photoexcited VO_2**", Science 362, pp. 572-576 (2018), https://doi.org/10.1126/science.aau3873

[9] J. D. Budai*, J. Hong*, M. E. Manley, E. D. Specht, C. W. Li, J. Z. Tischler, D. L. Abernathy, A. H. Said, B. M. Leu, L. A. Boatner, R. J. McQueeney, and O. Delaire, "**Metallization of vanadium dioxide driven by large phonon entropy**", Nature 515, 535–539 (2014). https://doi.org/10.1038/nature13865

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Topological Kohn anomaly and machine-learning augmented phonon DOS prediction

Mingda Li (mingda@mit.edu)

Abstract: Inelastic X-ray scattering (IXS) at meV scale represents a powerful experimental technique that studies materials elementary excitation with ultrahigh resolution. In this presentation, I will introduce how quantum field theory and machine learning can aid IXS discoveries of quantum materials. In one example, quantum field-theoretical calculations predicted the presence of a new class of Kohn anomaly in topological Weyl semimetal materials, that the electronic topology leaves hallmark phonon softening at specific points in Brillouin zone [1]. This theoretical prediction enables a direct experimental probing of Kohn anomaly with IXS, with ~1% error comparing with theory. In another example, using the symmetry-preserved neural networks, we have built a machine-learning-based predictor that can output phonon density-of-states in crystalline solid by directly using atomic structure files. The approach reaches ab initio accuracy for 80% of materials with significantly lowered computational cost, especially for alloys [2]. I will conclude by showing the increasingly important roles theory and machine learning may play to study quantum materials through the probe of IXS [3].

[1] T. Nguyen†, F. Han†, N. Andrejevic†, R. Pablo-Pedro†, A. Apte, Y. Tsurimaki, Z. Ding, K. Zhang, A. Alatas, E. E. Alp, S. Chi, J. Fernandez-Baca, M. Matsuda, D.A. Tennant, Y. Zhao, Z. Xu, J.W. Lynn, S. Huang and M. Li*, "Topological Singularity Induced Chiral Kohn Anomaly in a Weyl Semimetal", Phys. Rev. Lett. 124, 236401 (2020). Editors' Suggestion. (†Equal contribution)

[2] Z. Chen†, N. Andrejevic†, T. Smidt†, Z. Ding, Y-T Chi, Q. Nguyen, A. Alatas, J. Kong and M. Li*, "Direct prediction of phonon density of states with Euclidean neural networks", DOI:10.1002/advs.202004214, Advanced Science (2021). (†Equal contribution)

[3] Z. Chen†, N. Andrejevic†, N. C. Drucker, T. Nguyen, R.P. Xian, T. Smidt, Y. Wang, R. Ernstorfer, A. Tennant, M. Chan, and M. Li*, "Material Learning on Neutron and X-Ray Scattering", Chem. Phys. Rev. **2**, 031301 (2021). Editor Featured article; AIP SciLight. (†Equal contribution)

Inelastic x-ray scattering study of topological phonons

Hu Miao[a], Haoxiang Li[a], Tiantian Zhang[b], Mark Dean[c], Ayman Said[d]

[a] *Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA*

[b] *Department of Physics, Tokyo Institute of Technology, Okayama, Meguro-ku, Tokyo 152-8551, Japan*

[c] *Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, New York 11973, USA*

[d] *Advanced Photon Source, Argonne National Laboratory, Argonne, Illinois 60439, USA*

Topological states in quantum materials are defined by bulk wave functions that possess nontrivial topological invariants. While edge modes are widely presented as signatures of nontrivial topology, how bulk wave functions can manifest explicitly topological properties remains unresolved. Here, using high-resolution inelastic x-ray spectroscopy (IXS) combined with first principles calculations, we report experimental signatures of topological phonons, including double Weyl, nodal line, and twofold quadruple Weyl phonons[1-4]. Our results establish IXS as a powerful tool to uncover topological wave functions, providing a key missing ingredient in the study of topological quantum matter.

References:
1. Zhang et al., PRL 120, 016401 (2017).
2. Miao et al., PRL 121, 035302 (2018).
3. Zhang et al., PRL 123, 245302 (2019).
4. Li et al., PRB 103, 184301 (2021).

Understanding vibrational contributions to phase stability and transport in actinides

Michael E. Manley

Materials Science and Technology Division
Oak Ridge National Laboratory

While inelastic neutron scattering drove early studies on the lattice dynamics of actinides, issues with neutron absorbing isotopes, the absence of large crystals, and safety issues associated with handling large quantities of radioactive materials left a large gap in our understanding. Plutonium metal, for example, exhibits a rich phase diagram that includes six different allotropes spanning density variations of more than 30% and sits at the crossover from itinerate to localized 5f electrons in the periodic table – and yet very basic questions about its vibrational and electronic degrees of freedom remained unresolved for decades. For the actinide oxides, on the other hand, phonon thermal transport under irradiation is of critical importance to advanced nuclear reactor designs, but fully understanding phonon transport requires knowledge of the phonon dispersion curves including the line shapes. The measurement situation for lattice dynamics in the more challenging actinides improved dramatically with the advent of high energy resolution synchrotron based inelastic X-ray scattering (IXS) spectrometers, which have no isotope requirements and crystal sizes can be as small as a few microns – allowing measurements of crystal grains within a polycrystal sample. The first measured phonon dispersion curves for plutonium metal and the first phonon density of states of plutonium dioxide were both obtained using IXS rather than neutron scattering. IXS has also opened measurements of phonons at high pressures and in thin films using grazing incidence. The high atomic numbers of the actinides make them particularly well suited to IXS measurements in small geometries since the X-ray scattering per atom increases as the square of the atomic number. In this talk, I will highlight progress made in our understanding phase stability and thermal transport in actinides, with a particular emphasis on key insights obtained using IXS.

Phonon dispersions and elasticity of *d*-block transition metal single crystals at high pressure

Jung-Fu Lin
Department of Geological Sciences, The University of Texas at Austin
afu@jsg.utexas.edu

The *d*-block transition metals of the periodic table display many interesting yet complex physical properties due to their partially-filled *d* outer electronic shells. In this study, we have used high-pressure diamond anvil cell coupled with meV-resolved inelastic X-ray scattering to tune and investigate the *d*-electron interactions and the atomic vibrations in representative single-crystal transition metals. Optical and acoustic phonon dispersions of the single crystals are directly measured along certain crystallographic directions in helium medium with hydrostatic helium medium. Together with theoretical calculations, these results are used to address the contributions of the electron-phonon coupling and Fermi surface nesting to the Kohn anomaly, and the elastic anomaly associated with the electronic topological transition (ETT) at high pressure. Technical developments and scientific prospects in high-pressure meV-resolved IXS will also be discussed.