Cold-Neutron Triple-Axis Spectrometer (CTAX)

Tao Hong, Point of Contact

Spectroscopy Group

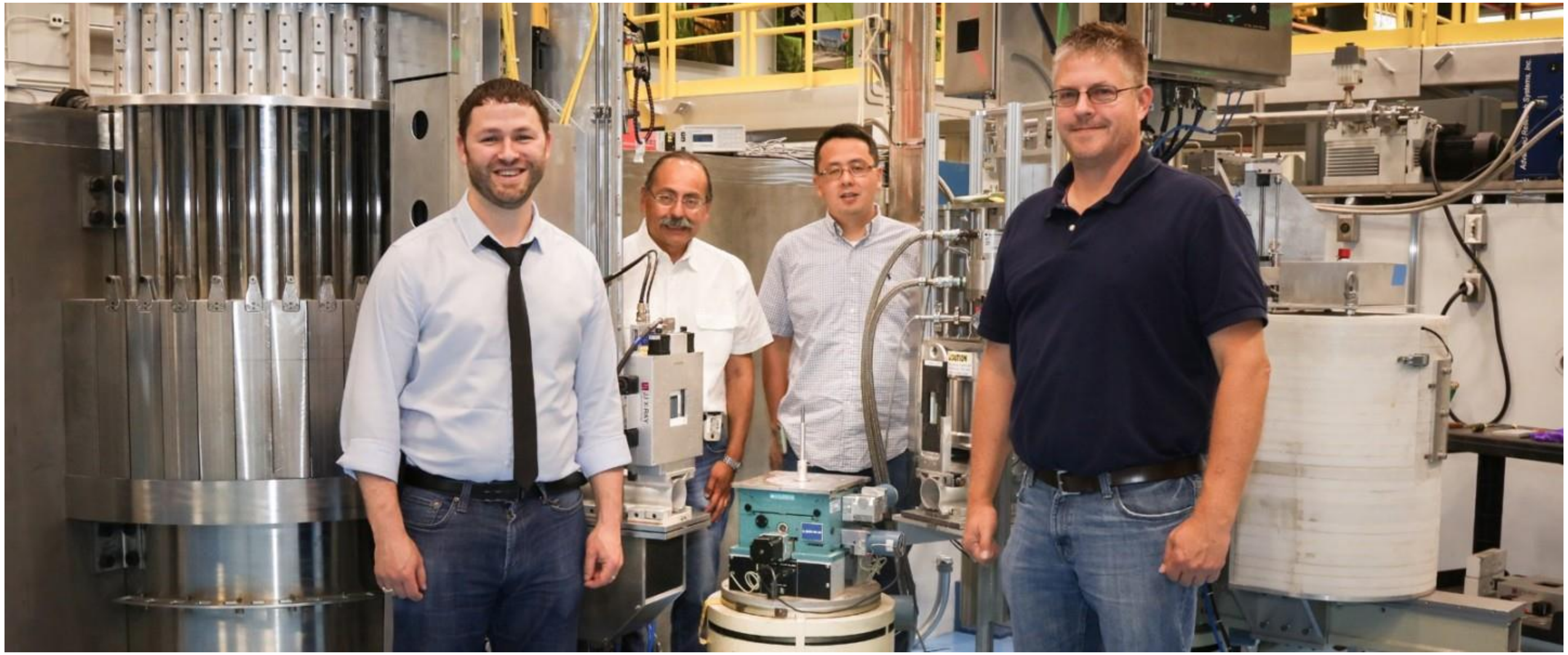
Neutron Scattering Division

Review of the Instrument Suites for Spectroscopy on September 17-18, 2020

CTAX Overview

- The cold-neutron triple-axis (CTAX) instrument is a conventional triple-axis spectrometer.
- Main components of the secondary spectrometer were relocated from Brookhaven National Laboratory after HFBR was shut down.
- It is a collaboration of the Neutron Science Directorate at Oak Ridge National Laboratory and the University of Tokyo, as part of US-Japan Cooperative Program on Neutron Scattering.
- CTAX is currently an instrument in the user program, active since 2012.

CTAX Instrument Team



Instrument Scientists: Jaime Fernandez-Baca (0.33 FTE), Tao Hong (1 FTE) and Travis Williams (0.33 FTE)

Scientific Associate: Mike Cox (0.5 FTE)

Computational Instrument Scientist: Andrei Savici

Instrument Specifications

Beam Spectrum:	Cold
Incident neutron energy E_i range:	2-20 meV
Monochromator:	Variable vertical focusing PG(002)
Final neutron energy E_f range:	$\geq$3.0 meV
Analyzer:	Fixed vertically and variable horizontally focusing PG(002)
Sample scattering angles:	-15°$\leq 2\Theta \leq$115°, with additional restrictions depending on E_i
Collimation before monochromator:	Guide dependent (40’ at 2 meV and 20’ at 20 meV)
Collimation after monochromator:	10’, 20’, 40’, 80’
Detector:	Single He^3 detector
Resolution:	Best energy resolution at the elastic line ~0.1 meV
Peak Flux (n cm^{-2} s^{-1})	$5*10^6$ at E_i=9 meV

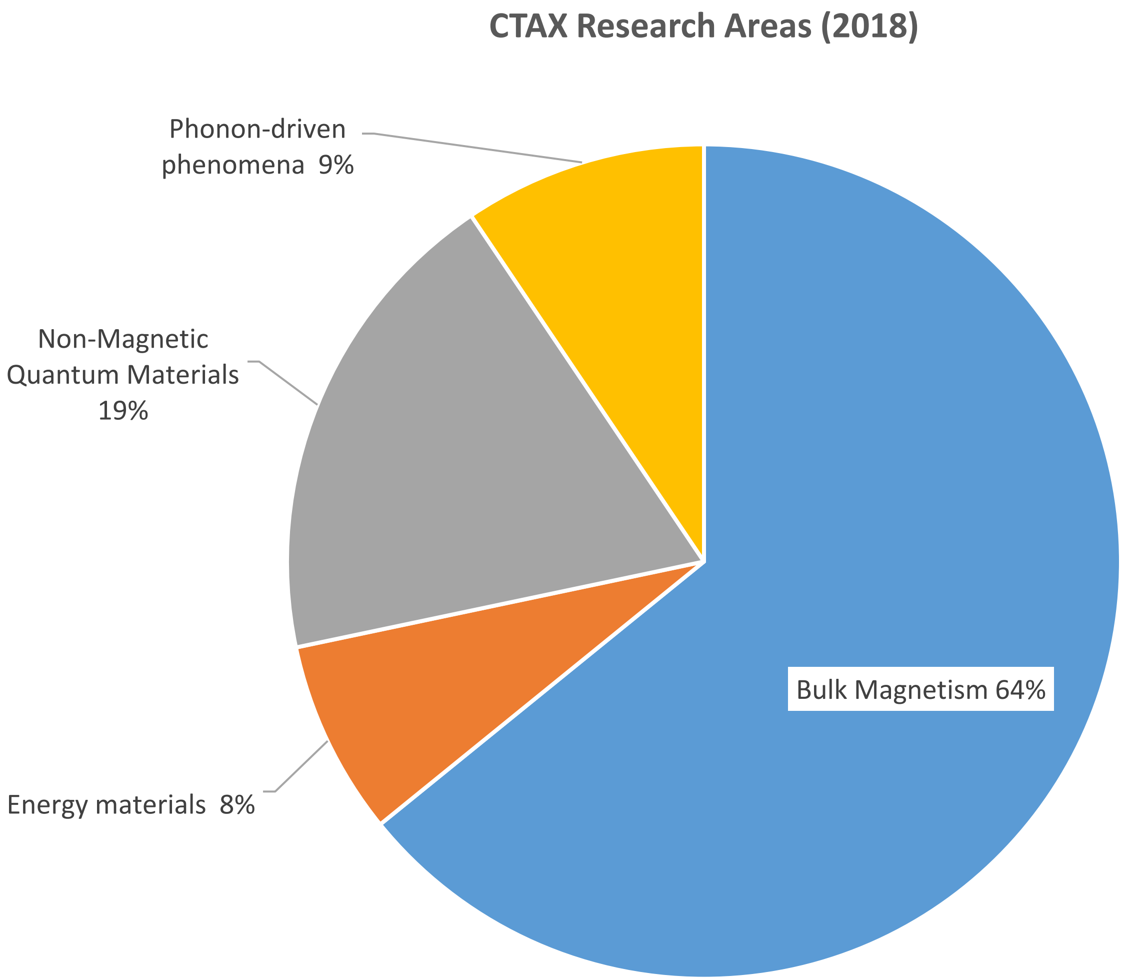
Scientific Mission and Impact

CTAX is ideal for high-resolution measurements of low-energy excitations with high signal-to-noise ratio in materials with emphasis on quantum materials and advanced energy materials.

Topics of study

- Unconventional superconductors
- Lattice dynamics in thermoelectric materials
- Spin excitations in multiferroic materials, quantum and geometrically frustrated magnets
- Exotic ground states and excitations in quantum critical phenomena

CTAX Research Area in FY-2018



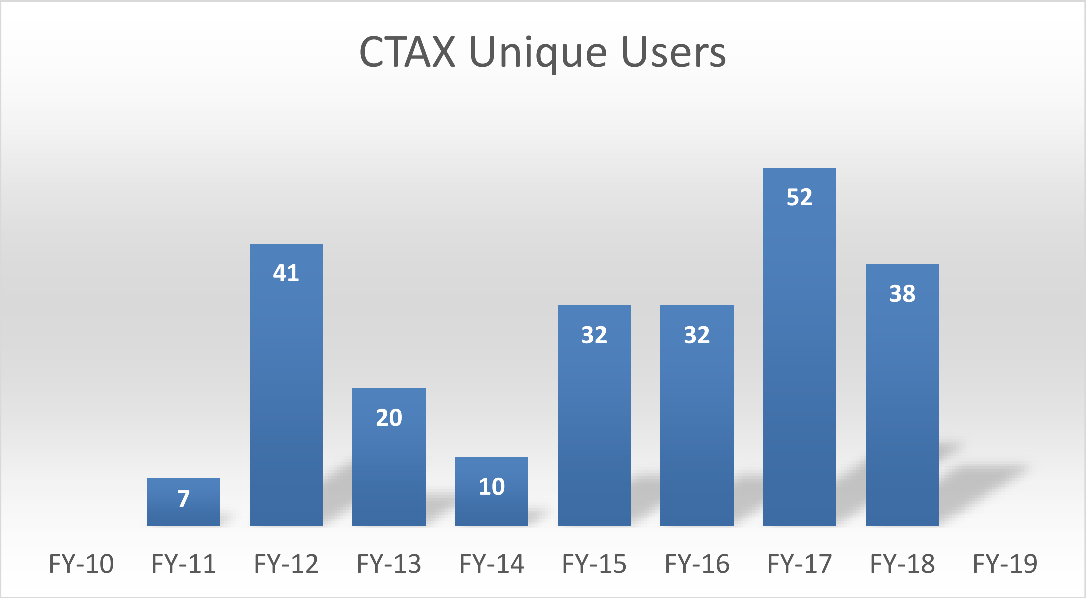
Compare to 2017:

Phonon-driven phenomena up 9%

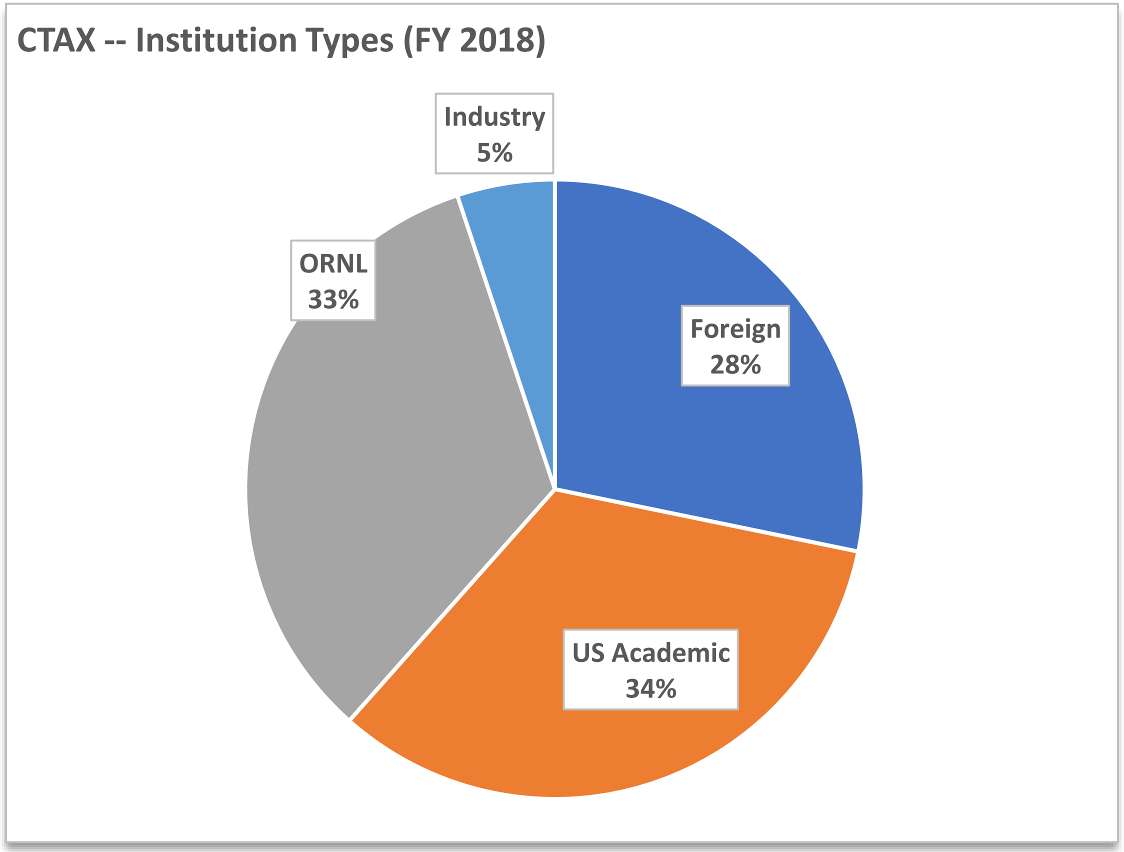
Energy Materials up 8%

Bulk Magnetism down 15%

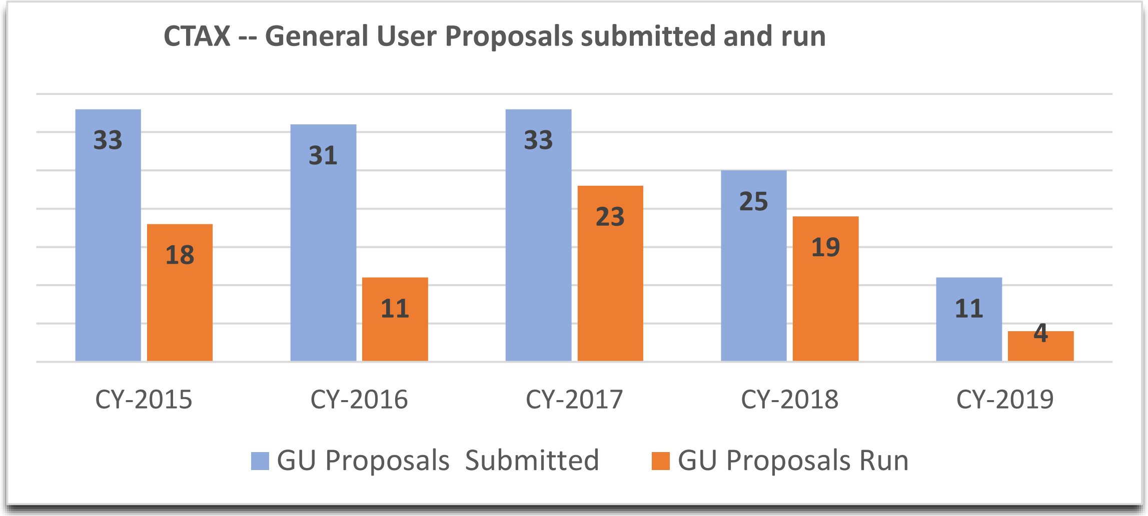
The CTAX Community in FY-2018

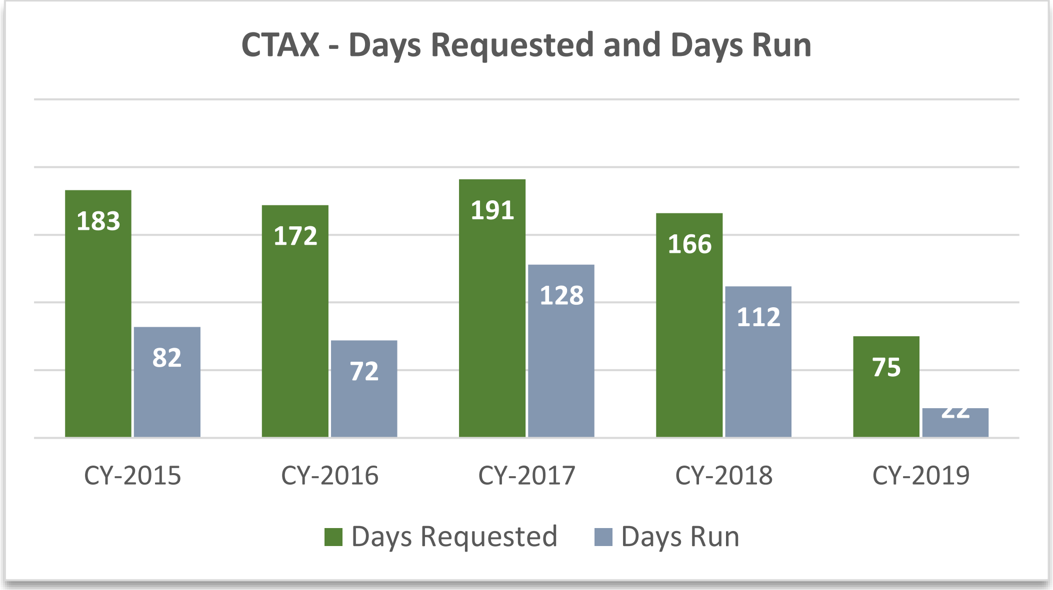


- Top 5 external institutions using CTAX *University of Tokyo, Duke University, Tohoku University, Louisiana State University and University of Virginia*

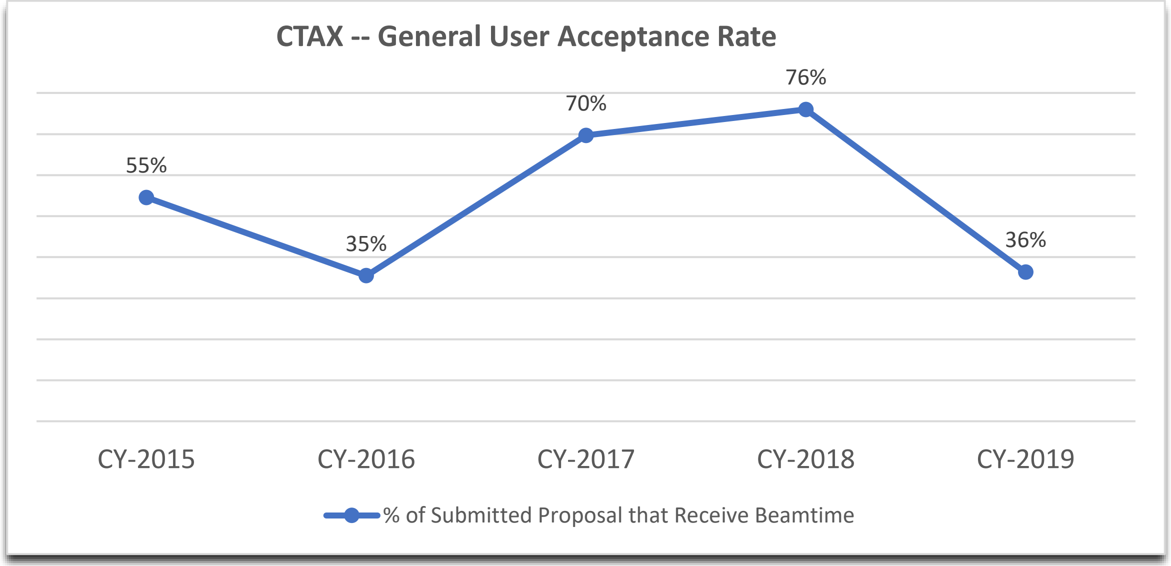


CTAX Instrument Productivity





*Number of completed includes carryover and alternates that were allocated after the BAC

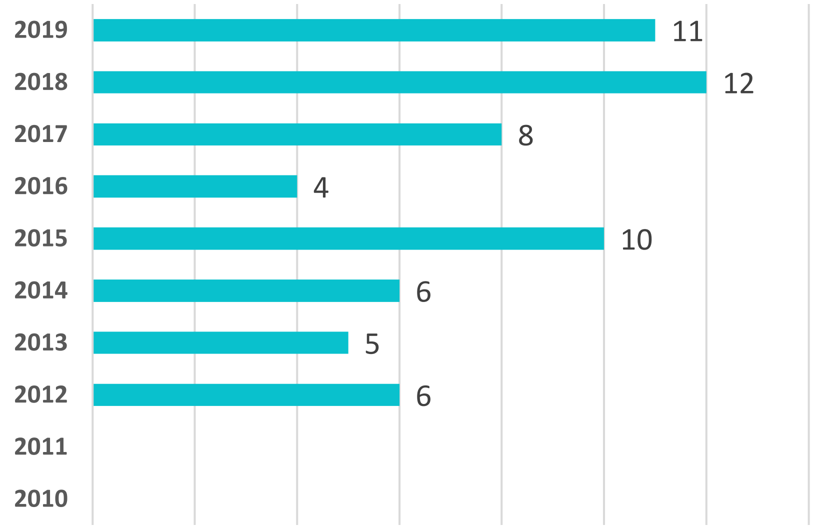


CTAX Instrument Productivity

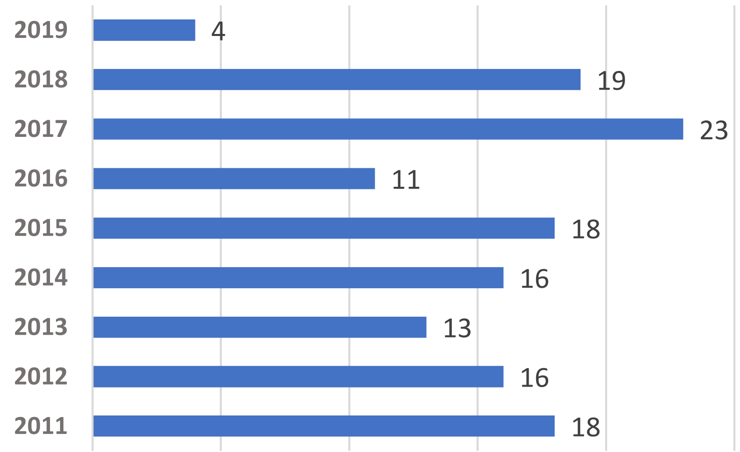
*Total Publications: **62***
*Instrument H-Index: **14***
*Publication Impact: **19%** publications with a high impact factor (recent 3 years)*
*Results from use of both facilities: **44%***
*Results from use of multiple instruments: **71%***
*Unique authors 2017-2019: **158***

Top Author Institutions (2017-2019)
University of Tennessee
University of Tokyo
Michigan State University
NIST Center for Neutron Research
Shizuoka University
Tulane University
Nanjing University

CTAX Publications by CY



CTAX - Experiments by CY



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Software

Data Acquisition

- **SPICE**

  Controlling motors, sample environments, and accessories as well as data acquisition

Experiment Planning

- **Virtual SPICE**

  Simulating scans and checking angle accessibility to select the best configuration of scattering plane and neutron energy

- **TAS Tools (DAVE)**

  Developed at NCNR/NIST for resolution calculation

Data Analysis

- **Graffiti**

  Peak fitting with Gaussian, Lorentzian and squared Lorentzian

- **Reslib for Matlab**

  Resolution convolution

- **SPINW and SpinWaveGenie**

  linear spin-wave calculations

- **Software developed by user groups**

  NeutronPy etc.

Future Plan

- More user-friendly software for resolution convolution calculation
- Replacement for Graffiti

Sample Environments

- Currently operating
  - Top loading orange cryostat (1.5-300 K)
  - Bottom loading CCR (4-300 K)
  - High temperature CCR (10-625 K)
  - Furnace (30-1600 C)
  - ^{3}He and dilution refrigeration inserts for cryostats and magnets
  - Cryomagnet
    - Horizontal magnetic field (0-6 T): Mag-G
    - Vertical magnetic field (0-8 T): Mag-E
  - Hydrostatic pressure
    - CuBe clamp-type pressure cell up to 1.5 GPa.

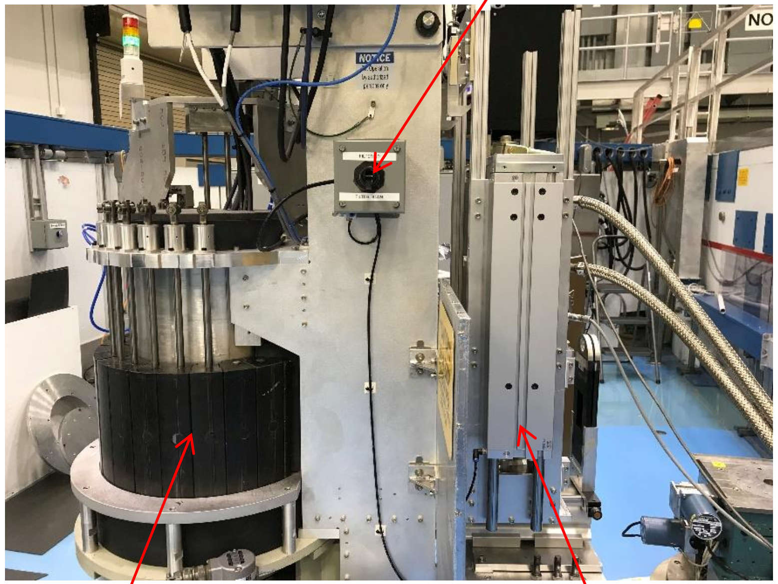
CuBe Pressure Cell

Oak Ridge National Laboratory

CTAX Backend Upgrade

➢ Be/BeO filter vertical translation

- Cooled down to 20 K
- Transmission≈90%
- $I_{\lambda/2}/I_{\lambda}<10^{-4}$

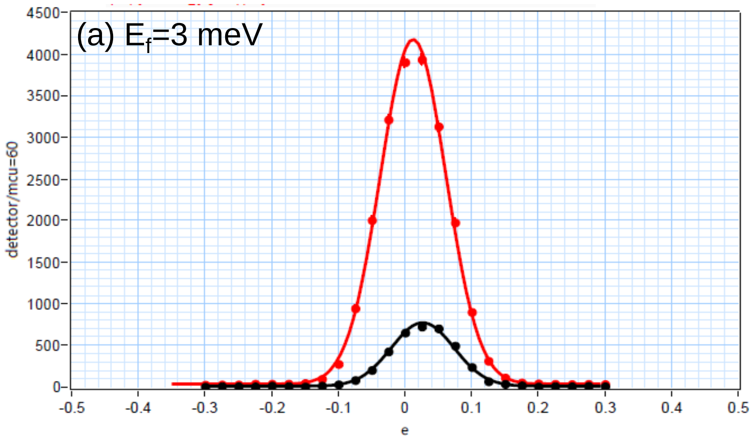


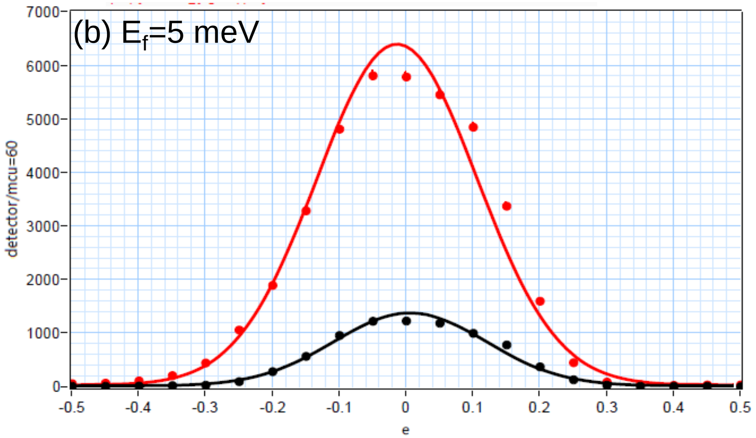
Be/BeO filter assembly

Doubly focusing analyzer assembly provided by Japanese collaborators

➢ Doubly focusing (vertically fixed and horizontally variable) analyzer

Vanadium sample





- Horizontal focus mode
- Flat mode

$I_{HF}/I_{Flat}>5$

ΔE: no change

Future Upgrades

- **MANTA**
  - Next-generation cold-neutron multi-analyzer triple-axis spectrometer (MANTA)
  - Located at current CG-1 guide at the cold guide hall, HFIR
  - Installation will be coupled to the re-optimization of HFIR cold guide hall in 2024-25 during HFIR Be reflector changeout

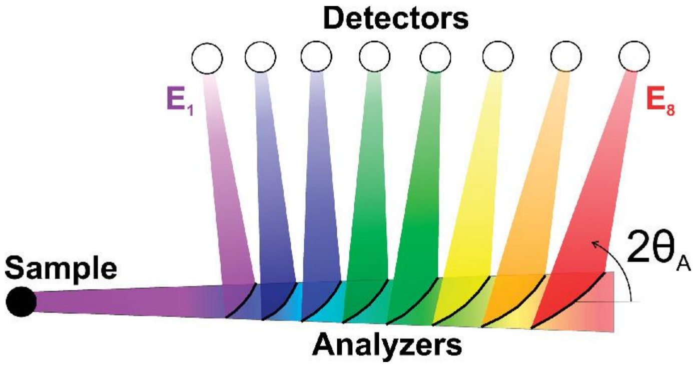
Overview of MANTA

❑ Phase 1, Features of "MANTA"

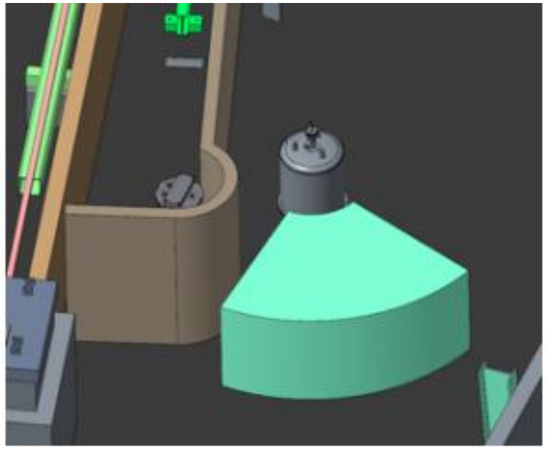
- ❑ New neutron guides optimized for cold neutrons
- ❑ Neutron velocity selector to cut out $\lambda/2$
- ❑ Rowland / Vertical Focusing Monochromator
- ❑ Large sample space for cryogenics and magnets
- ❑ Use CTAX backend as the secondary spectrometer
- ❑ Useful for high Q resolution measurements and eventually Spherical Neutron Polarimetry

❑ Phase 2, Features of MANTA

- ❑ Multi-ANalyzer Triple Axis: MANTA
  - ❑ CAMEA-like multi analyzer
- ❑ Polarization Capabilities
  - ❑ Although an integral part of the design for Phase 1, investment for polarization optics is planned for Phase 2



CAMEA back-end



OAK RIDGE National Laboratory

Review of Scientific Instruments **87**, 035109 (2016); https://doi.org/10.1063/1.4943208

Scientific Highlights

1. Time and Magnetic Field Variations of Magnetic Structure in the Triangular Lattice Magnet $Ca_3Co_2O_6$
– Complex sample environment *i.e.* the horizontal-field cryomagnet Mag-G

2. Novel Excitations near Quantum Criticality in Geometrically Frustrated Antiferromagnet $CsFeCl_3$
– INS study under hydrostatic pressure

3. Neutrons Clarify the Spin Structure of a Promising Metallic Antiferromagnet for Energy-Efficient Spintronics
– Neutron diffraction study on the epitaxial thin-film sample

4. Extended Anharmonic Collapse of Phonon Dispersions in SnS
– Low-energy phonon measurements complementary to other spectrometers

Time and Magnetic Field Variations of Magnetic Structure in the Triangular Lattice Magnet $Ca_3Co_2O_6$

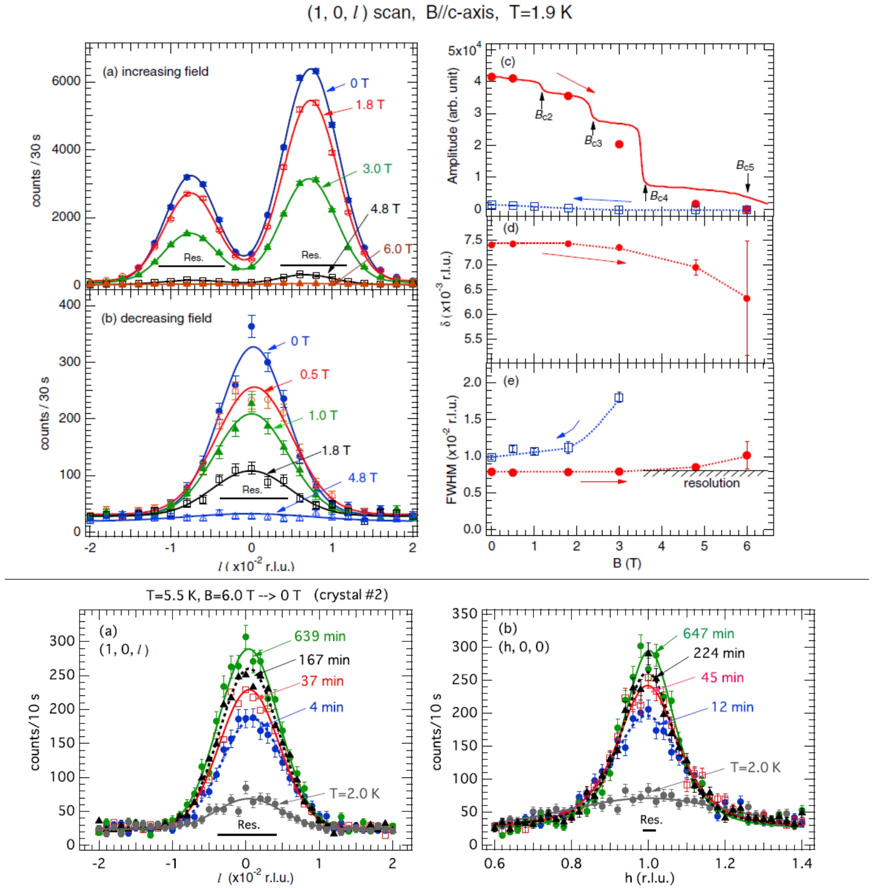


Figure Top panels: Magnetic field variations of neutron scattering intensity along the (1, 0, *l*) at *T*=1.9 K in the (a) increasing, (b) decreasing field processes and of (c) the integrated amplitude, (d) the peak position δ, and (e) the width (FWHM). Bottom panels: Neutron scattering patterns measured in zero field along (a) (1, 0, *l*) and (b) (*h*, 0, 0) at various elapsed times in *T*=5.5 K after removing the magnetic field of 6.0 T.

Reference: K. Motoya, T. Kihara, H. Nojiri, Y. Uwatoko, M. Matsuda, and T. Hong, Journal of the Physical Society of Japan **87**, 114703 (2018).

Scientific Achievement

This work studies the magnetic field and time variations of the magnetic structures on $Ca_3Co_2O_6$ by the magnetization and neutron scattering methods.

Significance and Impact

$Ca_3Co_2O_6$ is one of the very few known examples to observe the time variation of the magnetic structure in a system without randomness or imperfections. The observed long-period magnetic order is attributed to competition between the ferromagnetic intrachain and antiferromagnetic interchain interactions.

Research Details

Magnetic field evolution of the magnetic structures were determined by single-crystal neutron diffraction measurements at HFIR's beam line CG4C-CTAX using the horizontal-field Mag-G with maximum allowable field of 6 T.

Novel Excitations near Quantum Criticality in Geometrically Frustrated Antiferromagnet $CsFeCl_3$

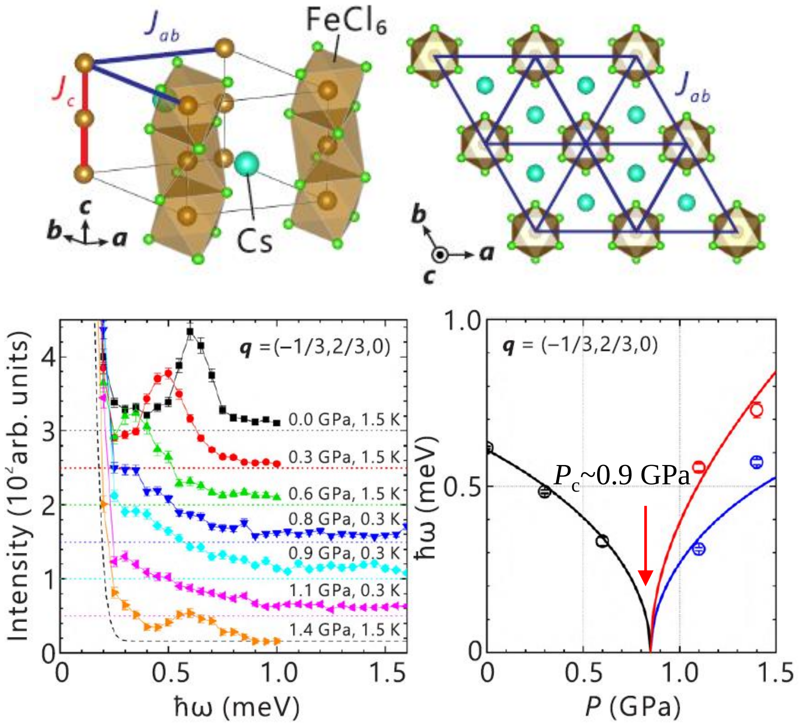


Figure: Top panel: In term of crystal structure of CsFeCl3, magnetic Fe^{2+} ions having pseudospin S = 1 form one-dimensional chains along the crystallographic c axis, and the chains form the triangular lattice in the ab plane. Bottom panel: Pressure evolution of the constant-q scans at (-1/3,2/3,0) and the gap energy calculated by the extended spin wave theory. Above 0.9 GPa, the blue and red curves are the excitations of gapless and gapped modes. The circles are peak energies evaluated from the constant-**q** scans.

Reference: S. Hayashida, M. Matsumoto, M. Hagihala, N. Kurita, H. Tanaka, S. Itoh, T. Hong, M. Soda, Y. Uwatoko, T. Masuda, Science Advances, ***5,*** eaaw5639 (2019).

Scientific Achievement

Inelastic neutron scattering study under hydrostatic pressure reveals novel excitations near quantum criticality in $CsFeCl_3$ as hybridization of the phase and amplitude fluctuations of the order parameter due to nature of the non-collinear ground state.

Significance and Impact

This work is of primary importance to discover a novel quantum hybridized state and to advance the physics of the interplay between the geometrical frustration and the quantum criticality.

Research Details

- **A single crystal (~0.5 g) of CsFeCl3 was aligned and loaded into a CuBe clamp-type pressure cell.**
- **The excitations as a function of applied hydrostatic pressure were measured using a cryostat with either a standard He-4 or a He-3 insert.**
- **Calculations of the spectra were performed by the extended spin-wave theory.**

Neutrons Clarify the Spin Structure of a Promising Metallic Antiferromagnet for Energy-Efficient Spintronics

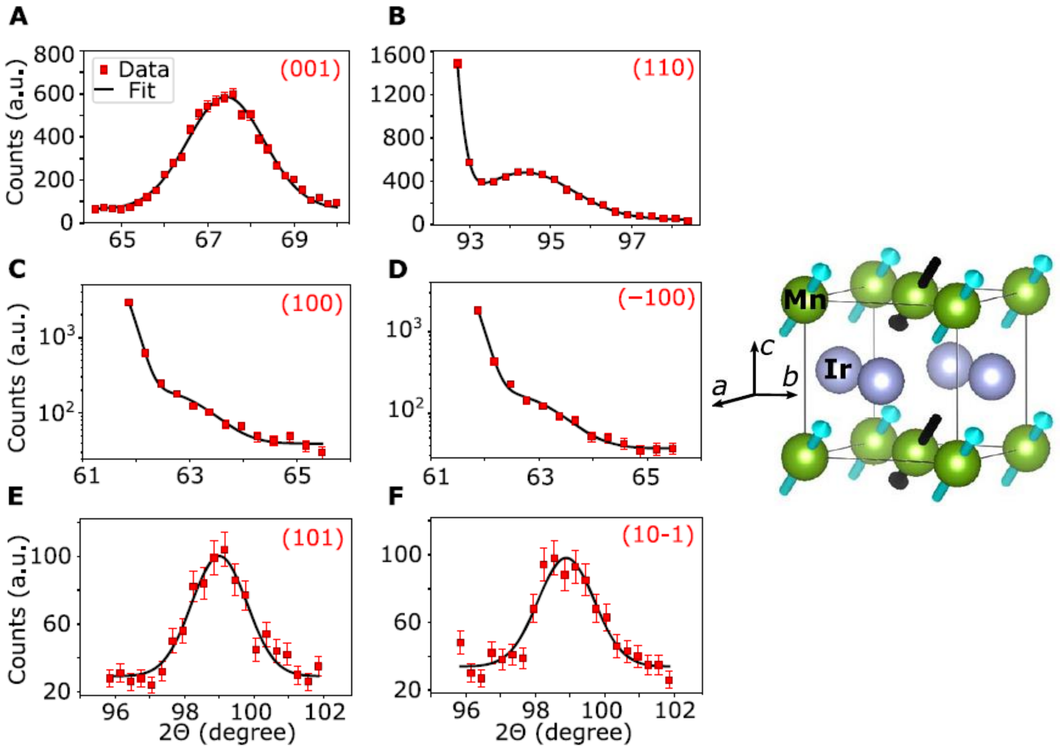


Figure: Neutron diffraction θ-2θ scans at room temperature for several nuclear and magnetic reflections in the epitaxial IrMn thin film. **A** (001). **B** (110). **C** (100). **D** (-100). **E** (101). **F** (10-1). Note that the film's signals are about 5000 times weaker than those from the substrate. CTAX's high signal to noise ratio and high instrumental resolution are critical for quantifying thin film's weak signals.

Reference: J. Zhou, X. Wang, Y. Liu, J. Yu, H. Fu, L. Liu, S. Chen, J. Deng, W. Lin, X. Shu, H.Y. Yoong, T. Hong, M. Matsuda, P. Yang, S. Adams, B. Yan, X. Han, J. Chen, Science Advances, **5**, eaau6696 (2019).

Scientific Achievement

It has been demonstrated that epitaxial $L1_0$-IrMn thin films can host an unconventional spin structure at room temperature that gives rise to a very large spin-orbit torque.

Significance and Impact

The spin-orbit torque provides an energy-efficient path for switching magnetization for magnetic memory. This work reveals the critical roles of spin structure on large spin-orbit torques generated by metallic antiferromagnets that hold great potential for future spintronics.

Research Details

- **Transport properties of patterned films were studied in the National University of Singapore to determine the spin-orbit torque.**
- **High resolution neutron diffraction experiments on thin film sample with thickness of 25 nm were conducted at CTAX and PTAX, HFIR and CORELLI, SNS to determine the film's spin structure.**
- **Ab initio calculations, performed at Weizmann Institute of Science, has attributed the large spin-orbit torque to the spin structure.**

Extended Anharmonic Collapse of Phonon Dispersions in SnS

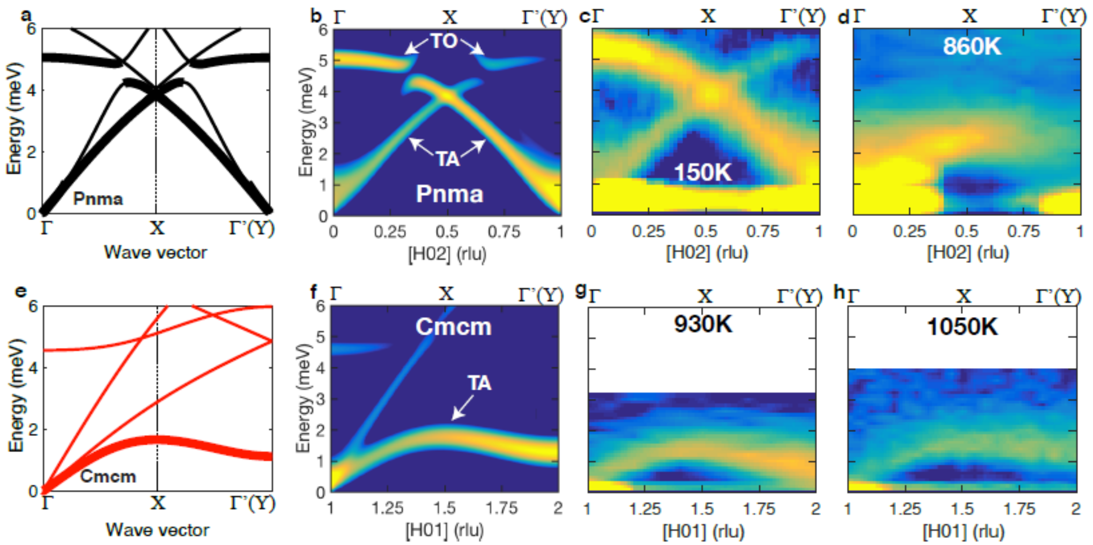


Figure: Evolution of SnS phonon dispersions and dynamical susceptibility $\chi''(\mathbf{Q}, E)$ across the structural phase transition. Upper panels are for the Pnma phase whereas lower panels show the Cmcm phase. Low energy dispersion computed in the Pnma phase with the harmonic approximation (a) and the Cmcm phase with the temperature dependent effective potential method at 800 K (e), for wave-vectors along [100] (Pnma notation). Panels (b) and (f) show the corresponding $\chi''(\mathbf{Q}, E)$ computed along [H02] and [H01] (Pnma notation). Panels (c,d,g,h) highlight the temperature evolution of dynamics susceptibility measured by INS with low-energy TA and TO phonons polarized along *c* in the Pnma phase (c,d) and Cmcm phase (g,h). Data were collected on CNCS (c), HB3 (d) and CTAX (g,h).

Reference: T. Lanigan-Atkins, S. Yang, J. L. Niedziela, D. Bansal, A. F. May, A. A. Puretzky, J. Y. Y. Lin, D. M. Pajerowski, T. Hong, S. Chi, G. Ehlers and O. Delaire, Nature Communications **11**, 4430 (2020).

Scientific Achievement

Inelastic neutron scattering uncovers a spectacular, extreme softening and reconstruction of an entire manifold of low-energy acoustic and optic branches across a structural transition, reflecting strong directionality in bonding strength and anharmonicity.

Significance and Impact

This work provide a detailed microscopic understanding of phase stability and thermal transport in technologically important materials, providing insights on ways to control phonon propagation in thermoelectrics, photovoltaics and other materials requiring thermal management.

Research Details

- **INS experiments were conducted at CTAX and PTAX, HFIR and CNCS, SNS to map out the phonon excitation spectra across the structural phase transition.**
- **In addition to INS data, high-resolution Raman spectra on single crystal of SnS were collected to probe zone-center optical modes.**
- **The anharmonic first-principles simulation was performed based on density functional theory to calculate the dynamical susceptibility of lattice dynamics.**

Risks

- Low neutron flux due to the misalignment of the CG4 guide.
- Low data collection efficiency due to single He-3 detector.
- Lack of instrument staff (full time second instrument scientist)
- Lose scientific competitiveness without MANTA

Overall Summary

- CTAX has proven to be an instrument with a solid user base.
- There are now active user groups performing experiments on the instrument in research areas of quantum materials, energy materials, functional materials and thin film heterostructures.
- Five of published papers were in journals with high impact factors (>7) since 2018.
- CTAX already supports a good diversity of sample environments, such as ultra-low temperature, high-magnetic field and high-hydrostatic pressure.