Oak Ridge National Laboratory

ARCS
Wide Angular-Range Chopper Spectrometer

Doug Abernathy, Point of Contact

2020 Review of the Instrument Suites for Spectroscopy

September 17–18, 2020

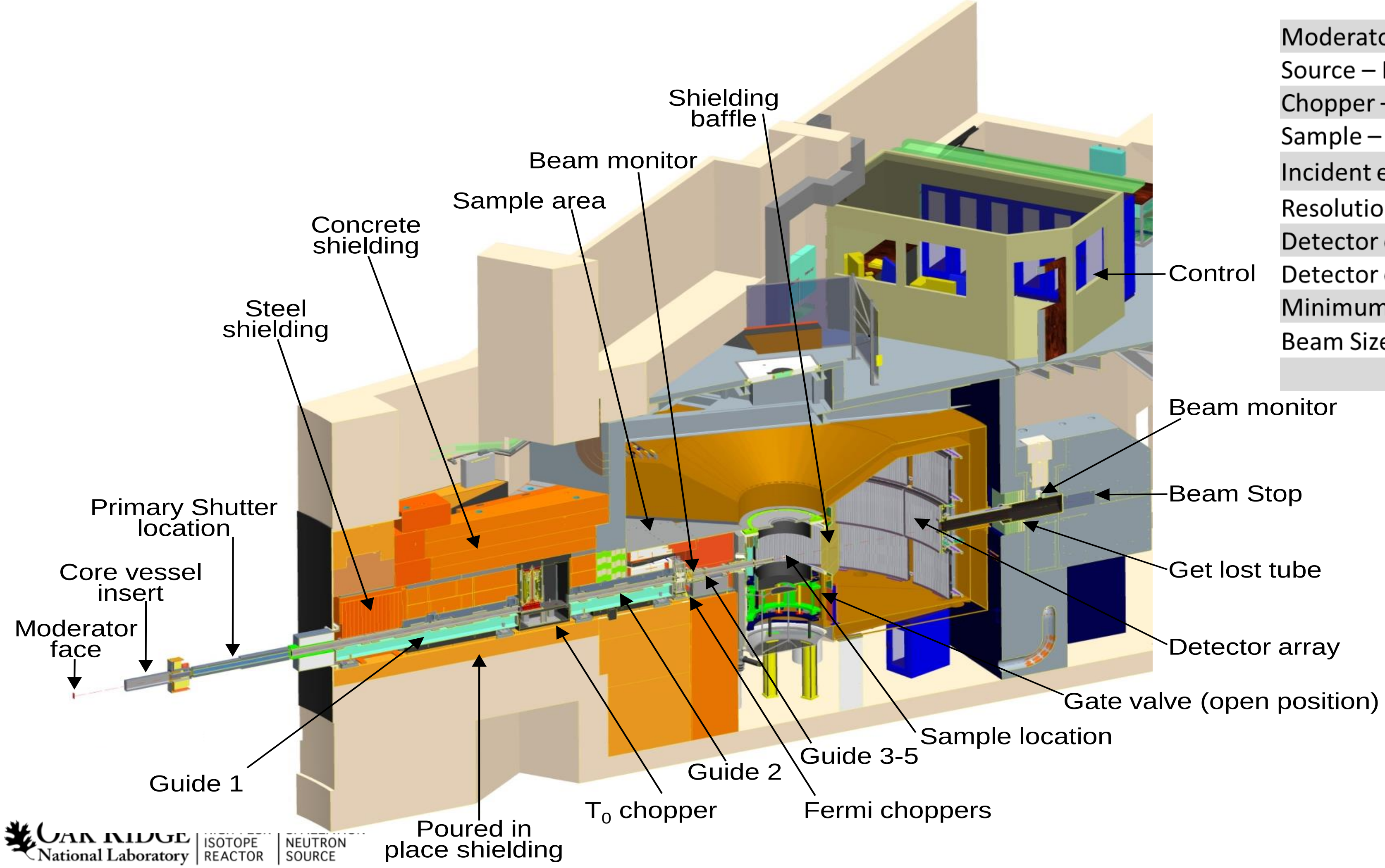
U.S. DEPARTMENT OF ENERGY

Beamline Review Topics

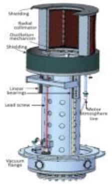
- ARCS overview
- Scientific Mission and Impact
- General user program and beam time usage
- Beamline Productivity
- Adequacy and reliability of software, sample environment and ancillary equipment
- Science Highlights
- Risks
- Future instrument science and development plan
- Response to instrument specific recommendations from last review
- Summary

ARCS Overview

- ARCS is optimized to provide a high neutron flux at the sample position with a large solid angle of detector coverage and uses thermal and epithermal neutrons for studies with moderate resolution and a wide range of wavevectors and energies.



Moderator	Decoupled ambient water
Source – Fermi chopper distance	11.6 m
Chopper – sample distance	2 m
Sample – detector distance	3.0 – 3.4 m
Incident energy range	12 – 1000 meV
Resolution (elastic scattering)	3 – 5% E_i
Detector coverage horizontal	-28°– 135°
Detector coverage vertical	-27° – 26°
Minimum detector angle	3°
Beam Size	5 cm x 5 cm

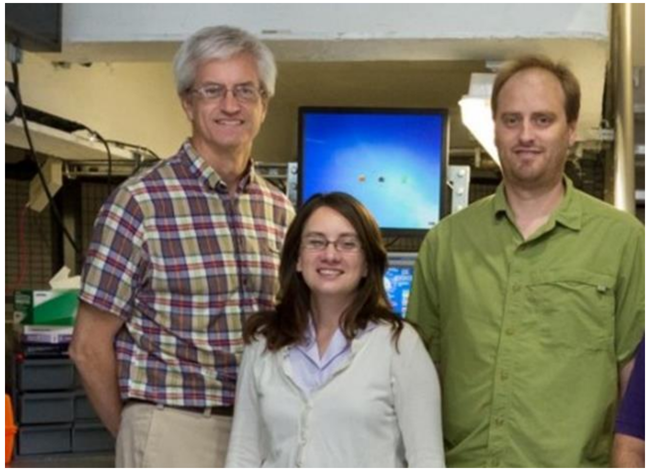


ARCS Instrument Team

- Core team
  - Doug Abernathy
  - Garrett Granroth
  - Rick Goyette
- Additional support
  - Andrei Savici
  - Victor Fanelli
- Former team members since 2017
  - Andy Christianson
  - Lacy Jones
  - Arnab Banerjee
  - Jiao Lin



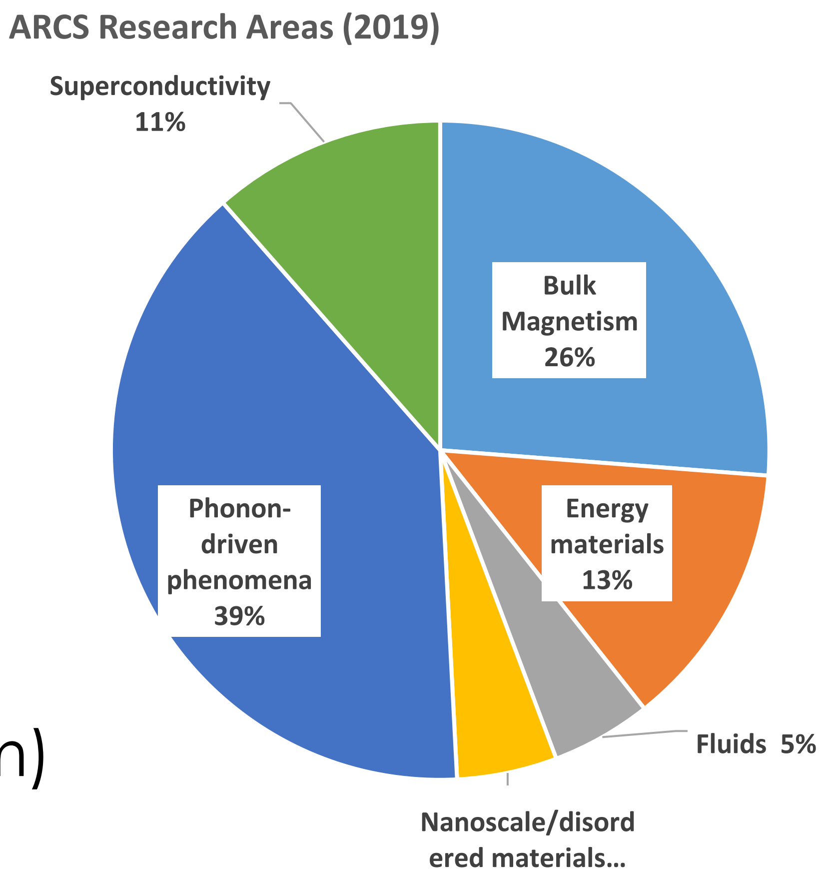


Scientific Mission and Impact

Topics of study

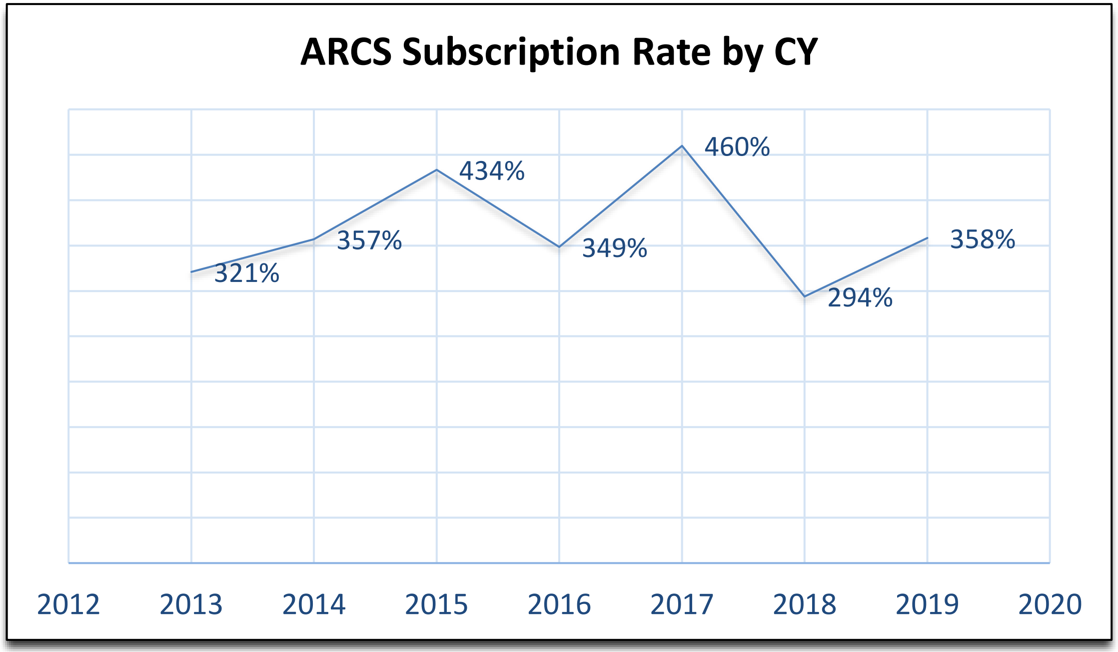
- Unconventional superconductors
- Quantum magnetism
- Itinerant magnets
- Ferroelectrics
- Thermoelectrics
- Multiferroics
- Dynamics in liquids (conventional, quantum)

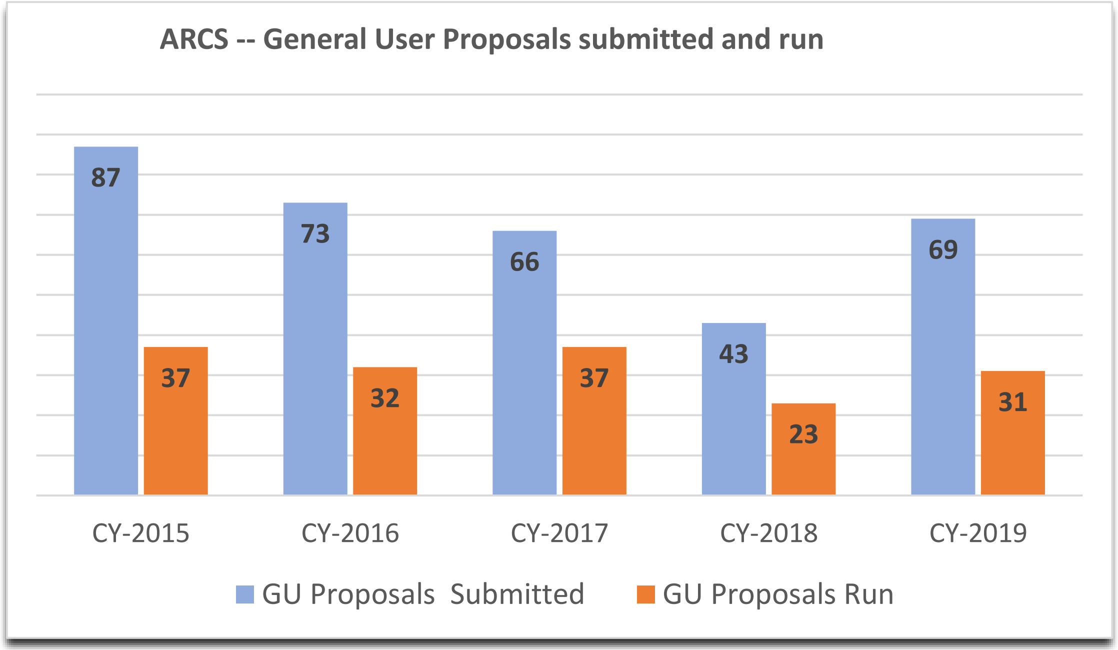


Program & User Community Development

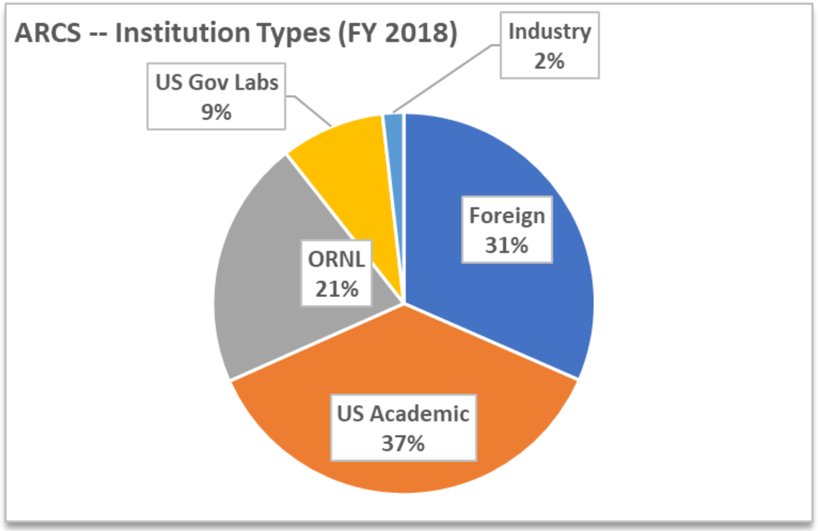
- Supporting dynamic PDF and van Hove function development (workshop, sample environment, software, future upgrades)

General user program and beam time usage





- Subscription rate from days is high. Percentage of GU proposals run is more reasonable, based on counting alternate proposals run.
- ARCS serves a variety of user institutions



TOP 5 EXTERNAL INSTITUTIONS USING ARCS (FY2019)
CALIFORNIA INSTITUTE OF TECHNOLOGY
UNIVERSITY OF CALIFORNIA RIVERSIDE
UNIVERSITY OF TENNESSEE
SWARTHMORE COLLEGE
DUKE UNIVERSITY

Beam time usage: Discretionary Time

- Almost no Initiative DT at ARCS. First is from High Pressure.
- Used to encourage collaboration among local students (UTK), postdocs and other ORNL staff, e.g. studies of resolution correction and the super-resolution concept. Used to test the suitability of samples and to complete data collection for ongoing collaborations to enable publications.
- Success story: Transformational Challenge Reactor team used DT and eventually GU time to study moderator materials at ARCS, SEQUOIA and VISION.

Transformational Challenge Reactor Program

THERMAL NEUTRON SCATTERING EVALUATION OF YTTRIUM HYDRIDE: FY2020 PROGRESS

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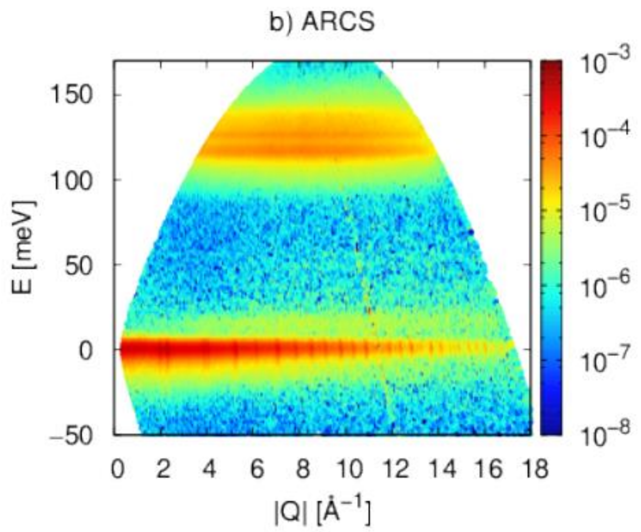
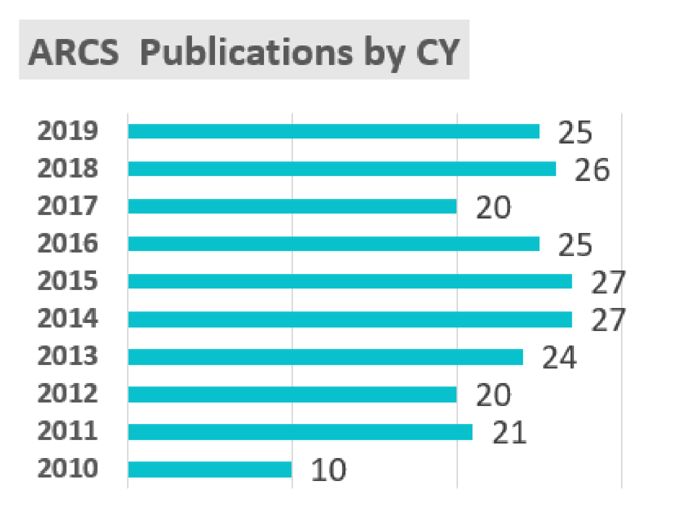
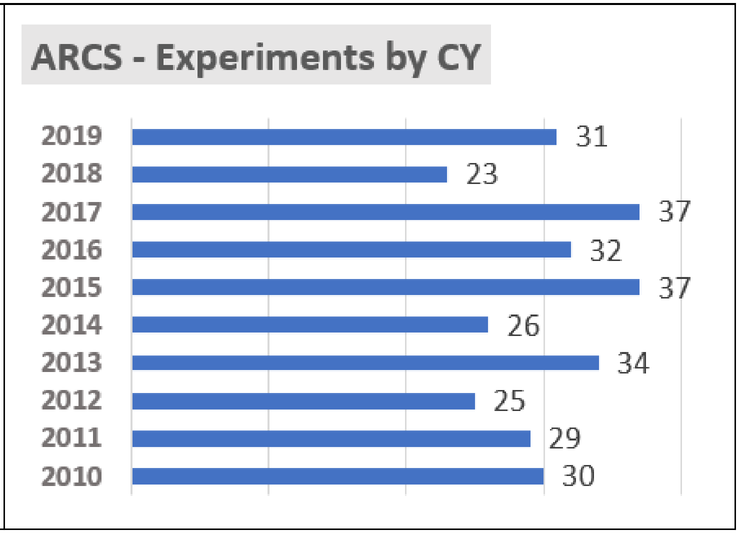
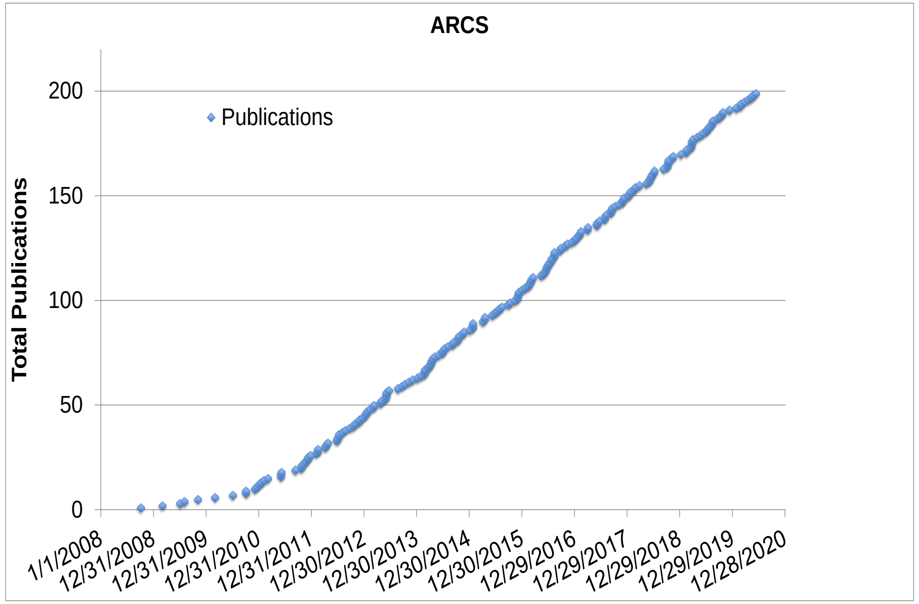
Date Published: September 2020



Beamtime productivity



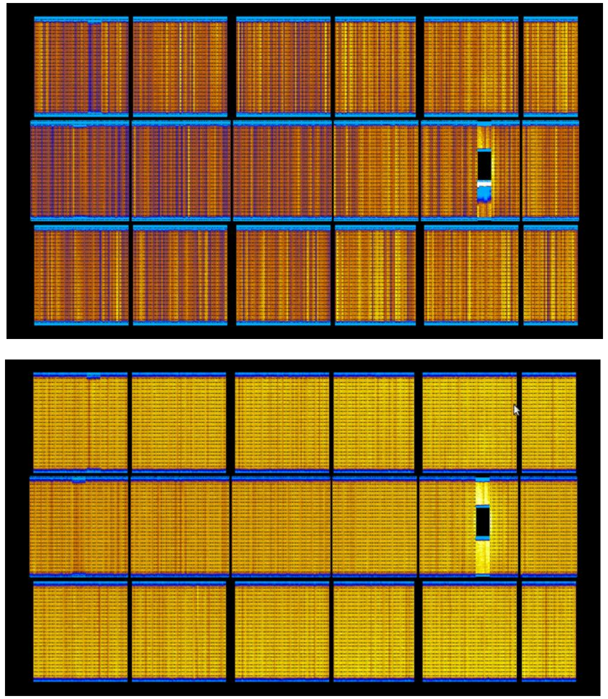




- ARCS continues a steady pace of publications – for CY2017 to CY2019, there were a total of 61 journal publications [19 (31%) IF > 7] and 8 theses.

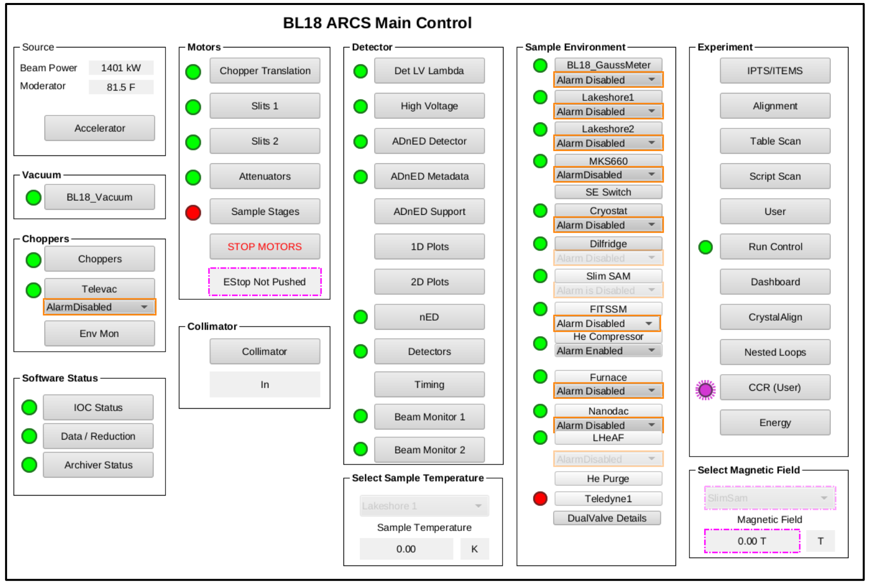
Reliability of equipment – Detector upgrade project

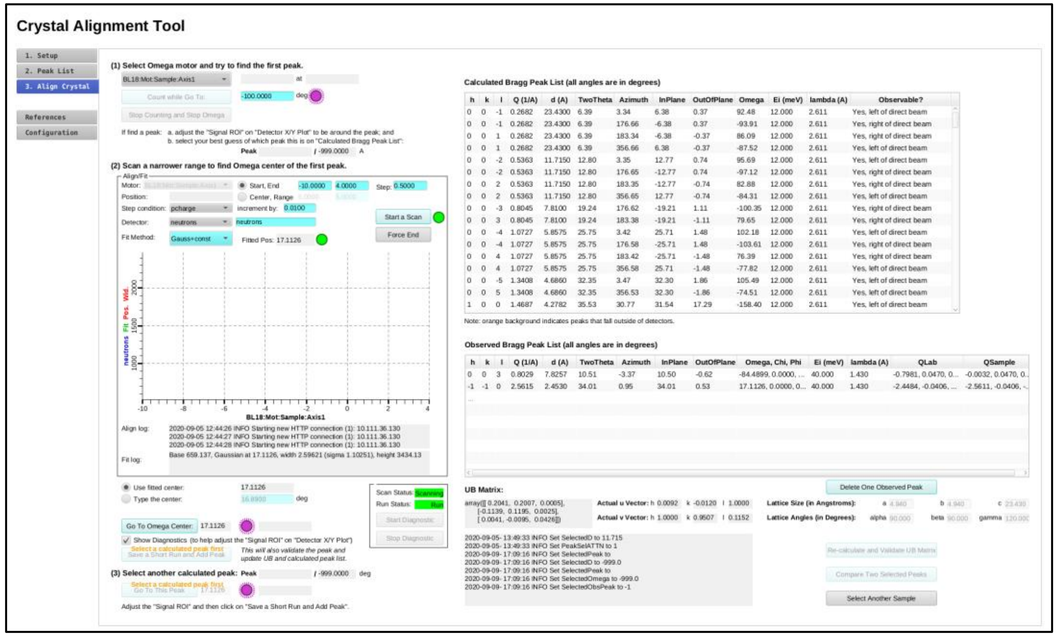
- Changeout of the detector electronics and controls for the ARCS ^{3}He LPSD array was completed in 2018 during IRP outage.
  - Replaced all electronics boards with current standards.
  - Reconfigured for external high voltage.
  - Replaced all cabling.
- New design of pre-amp boards
  - More resistant to damage during venting.
- Detector packs are individually powered
- Finer control of the discrimination
  - Reduced the variation in apparent efficiency across the detector array tubes and
  - Gained ~15% in overall count rate as determined by the standard high-intensity vanadium calibration runs



New data acquisition system

- ARCS updated to the EPICS/CSS/Phoebus standard data acquisition system (IRP 2018)
  - Common look and feel across the DGS instruments
  - Single crystal alignment interface
  - Incident energy selection tool with automatic "Flux" and "Resolution" modes. Uses look-up tables based on measured vanadium flux and resolution.
- ARCS able to participate in DGS developments
  - auto-reduction and visualization
  - Python scripting





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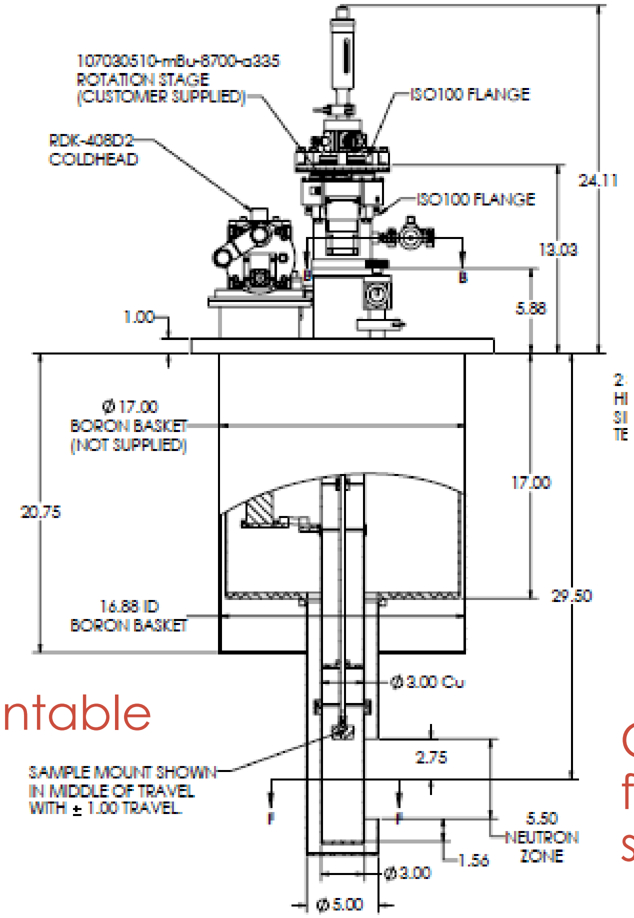
Adequacy of sample environment

- Continue to use workhorse SE
  - Bottom-loading CCR with low background (5K – 300K/450K hot stage)
  - Top-loading 100mm CCR (5K – 700K)
  - MICAS vacuum furnace(300K -1500K(V)/1800K(Nb))
- Neutron ElectroStatic Levitator (NESL)
  - Runs about one time per cycle
  - New hire in the SE high temperature team
- New 70mm top-loading CCR (5K – 700K)
  - Optimized for background and cooling speed
  - Delivery delayed by pandemic
- Automatic helium exchange gas handling
  - Automate the pump/purge and backfill process
  - Computer control pump out gas for high temperature operation



Demountable thin tail

Cutout for foil 1st shield

Close

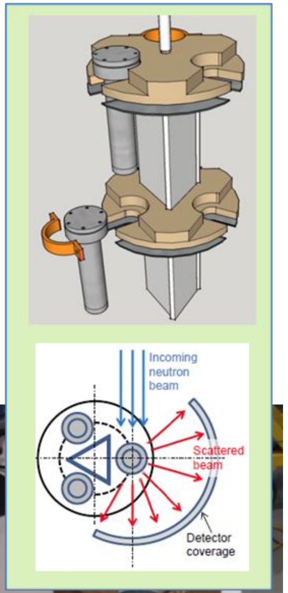
Flow

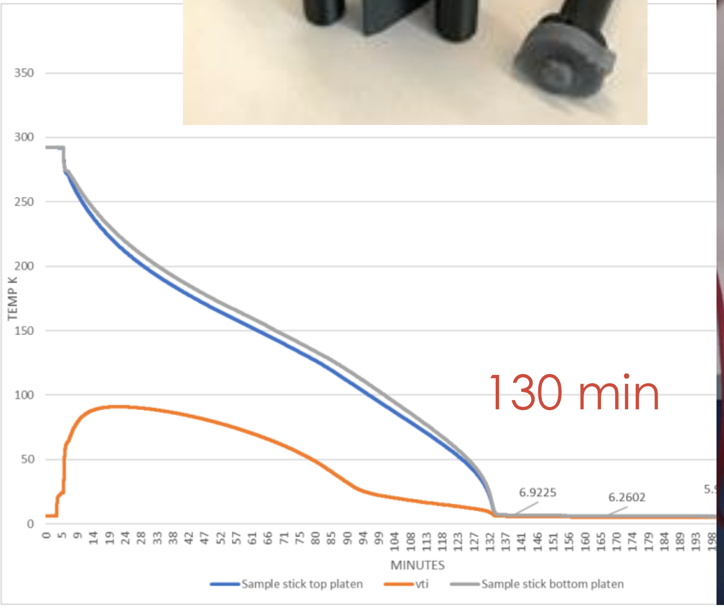
Pump

Purge & Backfill

Multi-sample changer for top-loading CCR

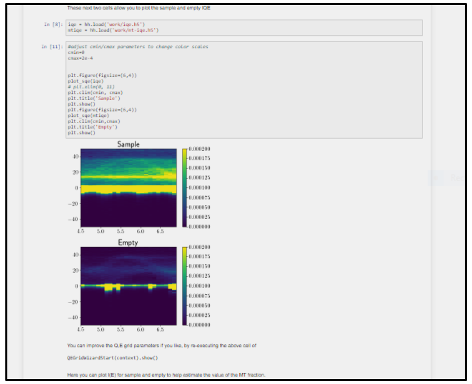
- Improve powder data collection efficiency with a 6-sample stick
  - No obstruction for entire ARCS array
  - Motorized rotation (existing) and vertical translation (on stick) to put samples in the beam
  - Existing 100mm CCR used with offset flange
  - Standard aluminum can bodies (max. 5/8" OD)
- Design is waiting for manufacturing
  - Novel 3D printing concept using B_4C/aluminum composite (ExOne - co-development with ORNL)
  - Short lids with neutron absorbing caps to clamp for heat transfer
  - Test stick of equivalent mass run in 100mm CCR (6K – 500K)
  - Cool down is reasonable, ~2x standard stick

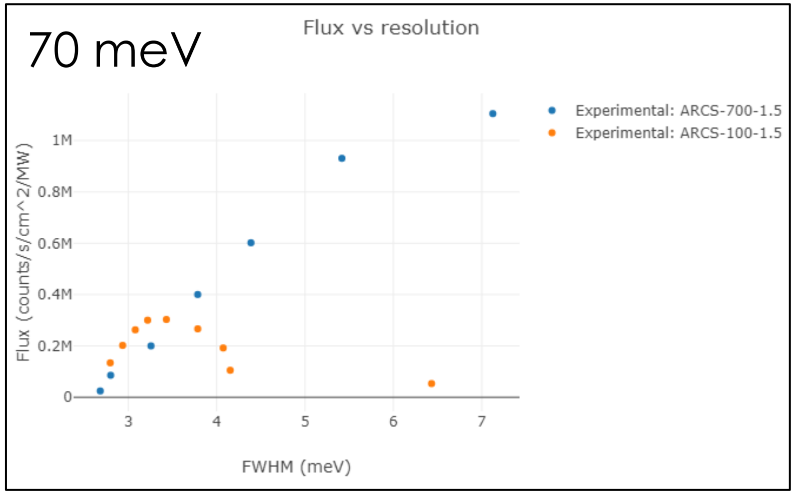




Adequacy of software

- ARCS participates in new visualization and analysis developments
  - Depending on the user group, use legacy (DAVE Mslice, Horace) or Mantid tools
  - Mslice replacement GUI for single crystals will accelerate transition to Mantid methods
  - Other tools: DGSplanner, autoreduction with single crystal displays, UB generation, etc.
- Phonon DOS: *getdos* (J. Lin)
  - Re-coding of incoherent approximation evaluation with multiphonon and multiple scattering correction
  - Combines multiple incident energy data
  - User-friendly Jupyter notebooks
  - Opportunity to engage with other Spectroscopy CIS (YQ Cheng) and software (Oclimax).
- Resolution characterization and modeling
  - Vanadium scattering data provide detailed information about the flux and energy resolution available for users (Lin, Physica B: Cond. Mat. 2019).
  - Validates PyChop model (https://rez.mcvine.ornl.gov)





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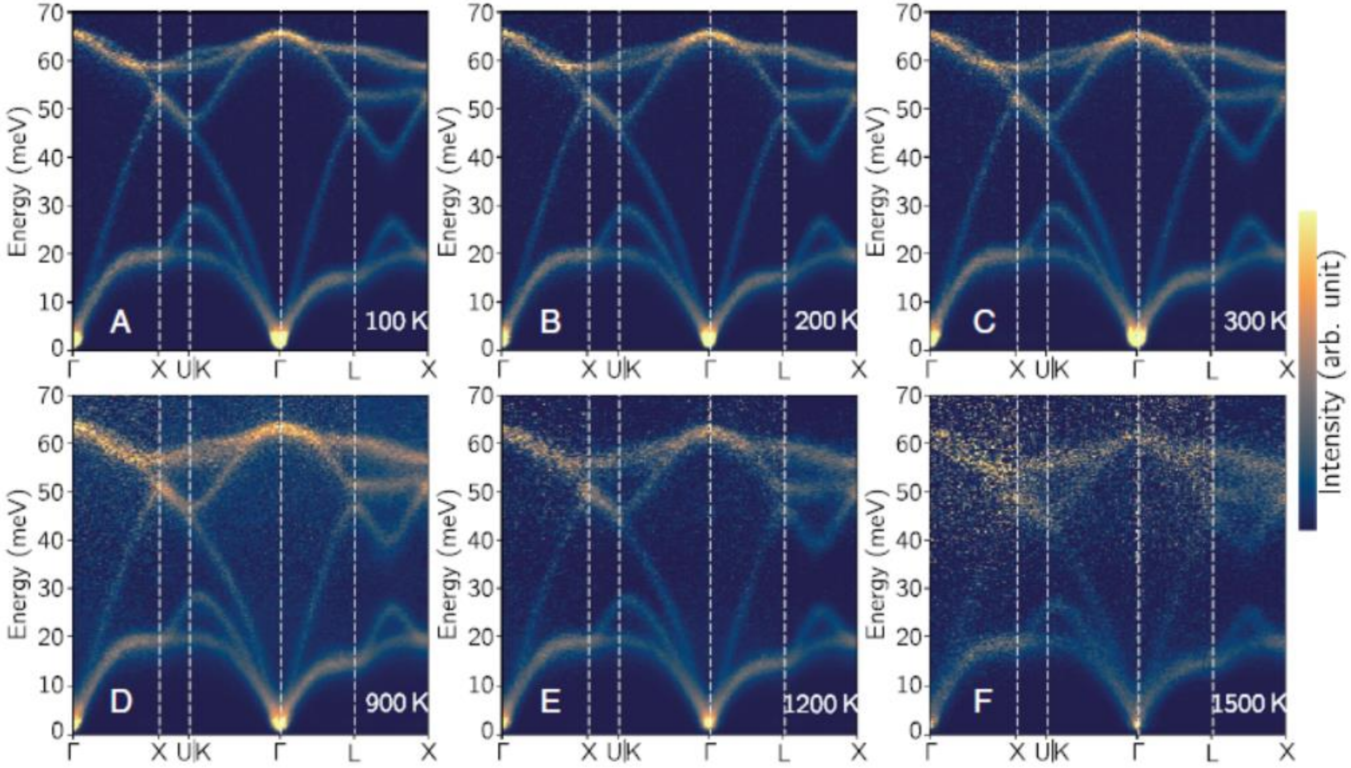
Beyond quasi-harmonic theory: Silicon anharmonicity and quantum effects

- Forefront users
  - Advancing data analysis and model techniques
  - Consistent collaborators
  - Push the instrument to implement improvements

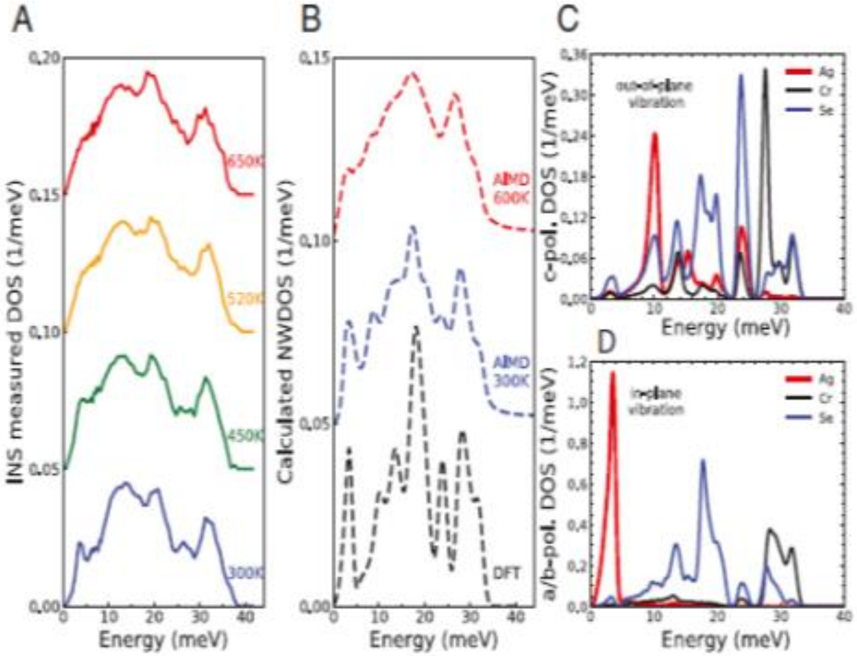


Experimental phonon dispersions of silicon. S(Q,E) were reduced, multiphonon-subtracted,and “folded” into one irreducible wedge in the first Brillouin zone.

D. S. Kim, O. Hellman, J. Herriman, H. L. Smith, J. Y. Y. Lin, N. Shulumba, J. Niedziela, C. W. Li, D. L. Abernathy, B. Fultz, "Nuclear quantum effect with pure anharmonicity and the anomalous thermal expansion of silicon," Proceedings of the National Academy of Sciences of the United States of America **115**(9), 1992-1997 (2018).

Vibrational studies of ionic transport materials

- Example of using instruments for their strengths
  - ARCS for lattice framework dynamics
  - CNCS for low energy modes and QENS
- Additional techniques used
  - X-ray elastic and inelastic for structures and dynamics
  - Raman scattering
  - broad range of computational techniques
  - Ionic mobility and electrochemical characterization
- Groups less experienced with neutrons were included in the experiments

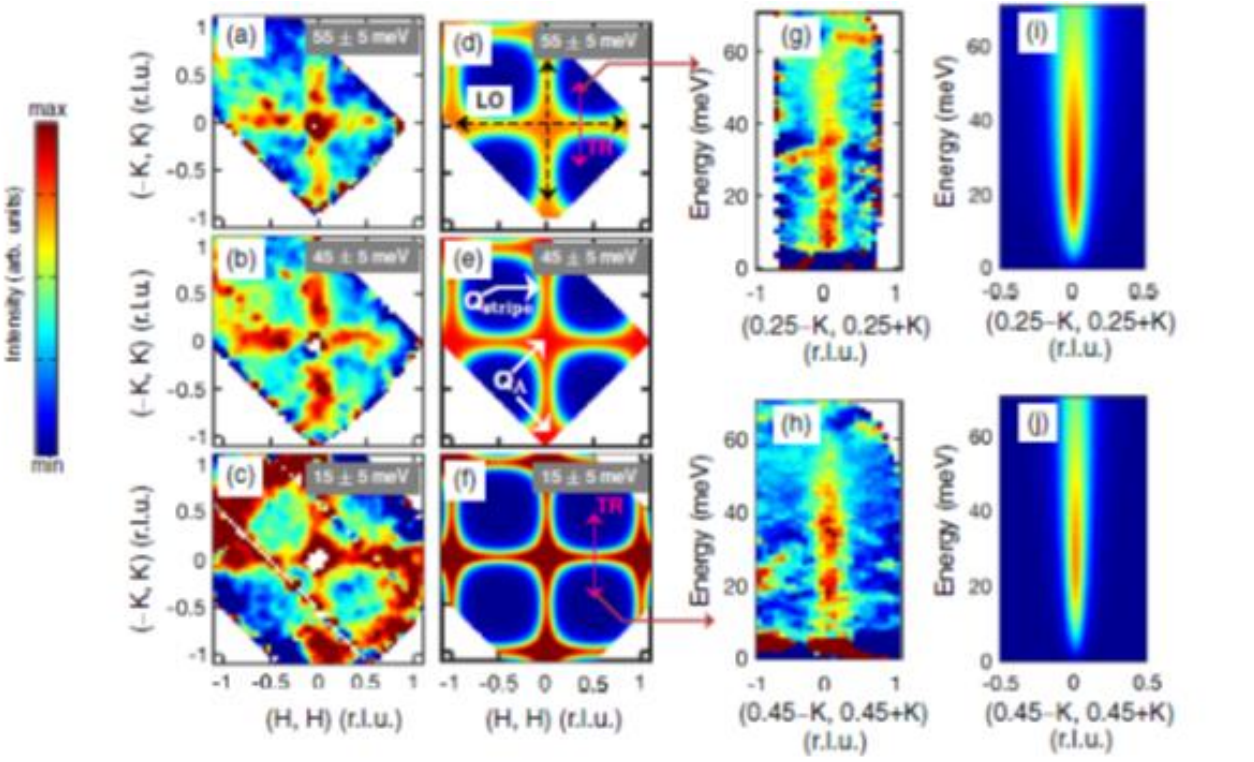


AgCrSe2 phonon DOS from experiments and simulations.

J. Ding, J. Niedziela, D. Bansal, J. Wang, X. He, A. F. May, G. Ehlers, D. L. Abernathy, A. H. Said, A. Alatas, Y. Ren, G. Arya, O. Delaire,"Anharmonic lattice dynamics and superionic transition in AgCrSe2," Proceedings of the National Academy of Sciences of the United States of America **117**, 3930-3937 (2020).
J. Niedziela, D. Bansal, A. F. May, J. Ding, T. Lanigan-Atkins, G. Ehlers, D. L. Abernathy, A. H. Said, O. Delaire, "Selective breakdown of phonon quasiparticles across superionic transition in CuCrSe2," Nature Physics **15**(1), 73-78 (2019).
T. Krauskopf, S. Muy, S. P. Culver, S. Ohno, O. Delaire, Y. Shao-Horn, W. G. Zeier, "Comparing the descriptors for investigating the influence of lattice dynamics on ionic transport using the superionic conductor Na3PS4-xSex," Journal of the American Chemical Society **140**(43), 14464–14473 (October 2018).
S. Muy, J. C. Bachman, L. Giordano, H. H. Chang, D. L. Abernathy, D. Bansal, O. Delaire, H. Satoshi, R. Kanno, F. Maglia, S. Lupart, P. Lamp, S. H. Yang, "Tuning Mobility and Stability of Lithium Ion Conductors based on Lattice Dynamics," Energy & Environmental Science **11**, 850-859 (2018).

Single crystal magnetism: Highly frustrated square-lattice itinerant magnet $CaCo_{2-y}As_2$

- Established team
  - Known for crystal growth and modeling magnetic system
  - Adept at background subtractions
- Classic techniques still used
  - “c-axis along the beam”



Spin fluctuations in $CaCo_{2-y}As_2$: Experimental INS data (Ei = 75 meV) vs diffusive model.

A. Sapkota, B. G. Ueland, V. K. Anand, N. S. Sangeetha, D. L. Abernathy, M. B. Stone, J. Niedziela, D. C. Johnston, A. Kreyssig, A. I. Goldman, R. J. McQueeney, "Effective One-Dimensional Coupling in the Highly Frustrated Square-Lattice Itinerant Magnet CaCo2-yAs2," Physical Review Letters **119**, 147201 (2017).

Risks

- Vulnerabilities as a “mature” instrument
  - Poor access to possible failures, principally the neutron guide system
- Not delivering on recent investments
  - New equipment to make data collection more efficient (new CCR, multi-sample stick)
  - Improved software with community buy-in
  - Translation of cutting-edge research in data treatment and analysis into usable tools (resolution corrections, machine-learning/AI)
- Not able to use higher power effectively
  - Understaffing in support (Sample Environment) and beamline scientists
  - Software to shorten the time from data collection to visualization and modeling

Future instrument science and development plan

- Continue developments for efficient operations
  - Use 6-sample stick to evaluate if a true sample changer system is needed (monitor VISION and NOMAD changers)
  - Automatic reduction and visualization improvements
- Continue to advance sample environment capabilities
  - Large volume diamond anvil cell pressure experiments
- Continue software developments (DGS team)
  - Documentation and tutorials for new data reduction and visualization methods
  - More routine resolution corrections adapted to different applications
    - Analytical resolution convolved with models – powders and single crystal
    - Detailed Monte Carlo based full experiment modeling
- Develop science case and technical possibilities for higher resolution option
  - Quantify flux and resolution trade-off through analytic and MC models
  - Continue evaluation of dynamic PDF and van Hove function methods
  - New applications: Higher resolution full Brillouin Zone mapping of phonons

Response to previous review

- Replace the data acquisition system.
  - Done.
- Develop and deploy a multi-sample changer for samples at room temperature and below.
  - In progress.
- Complete an analysis of the benefit of a disc chopper for energy resolution improvements that could improve dynamical pair distribution function analysis.
  - Not completed. High incident energy for dynamic PDF indicates a new Fermi chopper design is needed. Parameters will have to weigh the flux versus resolution trade-off, still to be modeled. A scientific case for the investment will have to be made to compete for funds.

Summary

- ARCS continues to provide reliable measurements across a wide variety of scientific areas into its second decade of operation
- Increased productivity will come from continued development of sample environment and software
- Upgrade projects keep ARCS effective
  - Detector upgrade
  - DAS upgrade
  - Sample environment ongoing (new top-loader, multisample changer)
- Long-term: what will push ARCS into new fields?
  - Innovative analysis software (resolution, AI/machine learning/data mining, …)
  - Option for higher resolution for access to Fourier transform methods or other applications