U.S. DEPARTMENT OF ENERGY | Office of Science

# Scientific User Facilities in Energy Research

J-PARC Symposium 2019

September 24, 2019

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Associate Director of Science
Basic Energy Sciences

# 1980 USA-Japan Agreement on Cooperation in Science and Technology

AGREEMENT BETWEEN THE GOVERNMENT OF THE
UNITED STATES OF AMERICA AND THE
GOVERNMENT OF JAPAN
ON COOPERATION IN RESEARCH AND DEVELOPMENT
IN SCIENCE AND TECHNOLOGY

The Government of the United States of America and the Government of Japan;

Desiring to create a truly productive partnership and further strengthen the bonds of friendship existing between the two countries;

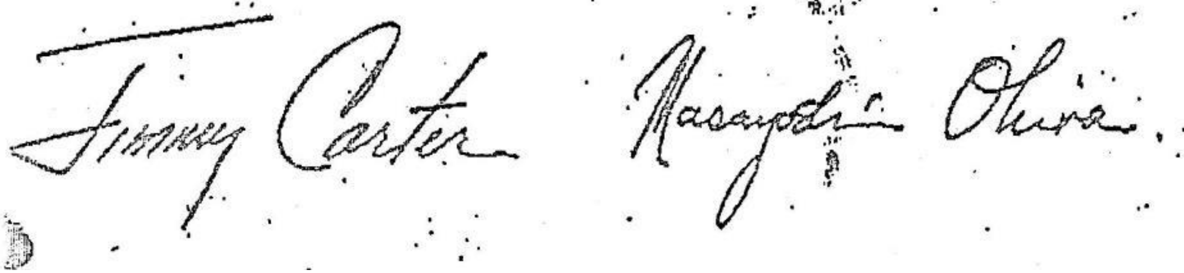
Believing that the future welfare and prosperity of mankind requires untiring efforts in the fields of scientific and technological research and development and that such efforts are not only beneficial to the country involved but also to the entire world;

Realizing the great benefits derived by the two countries from their long and highly successful scien-

DONE at Washington on May 1, 1980, in duplicate in the English and Japanese languages, both being equally authentic.

FOR THE GOVERNMENT OF THE UNITED STATES OF AMERICA:

FOR THE GOVERNMENT OF JAPAN:



# US-Japan Cooperative Program on Neutron Scattering

- Started in 1983 as one of the programs under 1980 USA-Japan Agreement on Cooperation in Science and Technology (Umbrella agreement)
- **US DOE and Japan MEXT (originally MONBUSHO and STA)**
  - ORNL, BNL in the US;
  - ISSP-University of Tokyo and MEXT universities, JAEA in Japan

- Program objective:

  "To establish a joint basic research program in the area of neutron scattering involving scientists from Japan, BNL, and ORNL. Solid state physics, chemistry, biology, metallurgy, etc."

# USA-Japan Agreement on Cooperation in Science and Technology extended in 2014

- April 23, 2014 in Tokyo, “Protocol extending the Agreement between the Government of Japan and the Government of the United States of America on Cooperation in Research and Development in Science and Technology” was signed between Mr. Fumio Kishida, Minister for Foreign Affairs, for Japan and Her Excellency Caroline Bouvier Kennedy, Ambassador Extraordinary and Plenipotentiary to Japan, for the U.S.

# US-Japan Cooperative Program on Neutron Scattering

- **Benefits to the US**
  - Access to instruments, equipment and technical developments provided by the program
  - Steady flow of users (program funds their travel)
- **Benefits to Japan**
  - Access to neutrons at the High Flux Isotope Reactor (20-30 visits/year)
- **Mutual Benefits**
  - Productive collaborations in areas of mutual interest
  - Enhances scientific exchange, funds Japanese experts travel to US.
  - **700 publications since the beginning of the program**
  - **150 publications since 2010**

## Recent scientific and technical development highlights

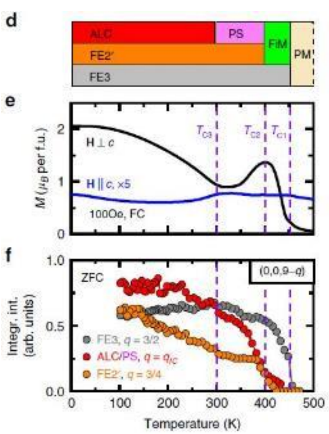
nature COMMUNICATIONS

ARTICLE

https://doi.org/10.1038/s41467-019-09205-x OPEN

Magnetization-polarization cross-control near room temperature in hexaferrite single crystals

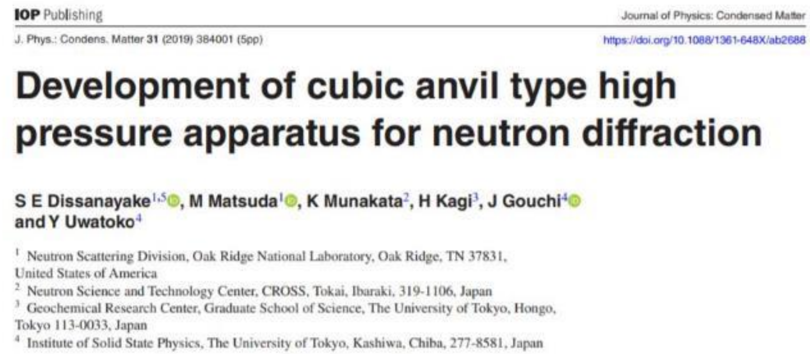
V. Kocsis[1], T. Nakajima[1], M. Matsuda[2], A. Kikkawa[1], Y. Kaneko[1], J. Takashima[1,3], K. Kakurai[1,4], T. Arima[1,5], F. Kagawa[1,6], Y. Tokunaga[1,5], Y. Tokura[1,6] & Y. Taguchi[1]




Magnetic phase diagram in hexaferrite

T-dependence of field cooled M in H=100Oe

Neutron diffraction peaks representing different magnetic phases



IOP Publishing — Journal of Physics: Condensed Matter

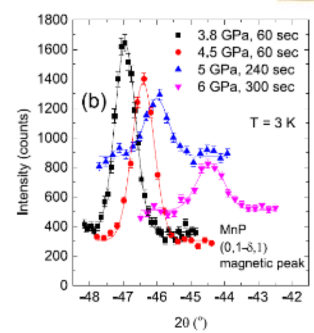
J. Phys.: Condens. Matter 31 (2019) 384001 (5pp) — https://doi.org/10.1088/1361-648X/ab2688

Development of cubic anvil type high pressure apparatus for neutron diffraction

S E Dissanayake[1,5], M Matsuda[1], K Munakata[2], H Kagi[3], J Gouchi[4] and Y Uwatoko[4]

[1] Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, United States of America
[2] Neutron Science and Technology Center, CROSS, Tokai, Ibaraki, 319-1106, Japan
[3] Geochemical Research Center, Graduate School of Science, The University of Tokyo, Hongo, Tokyo 113-0033, Japan
[4] Institute of Solid State Physics, The University of Tokyo, Kashiwa, Chiba, 277-8581, Japan

A cluster of 6 anvils pressing from 3 orthogonal directions provide homogeneous pressure, 4 of these are shown in the left panel. Right panel shows the assembled cubic anvil cell

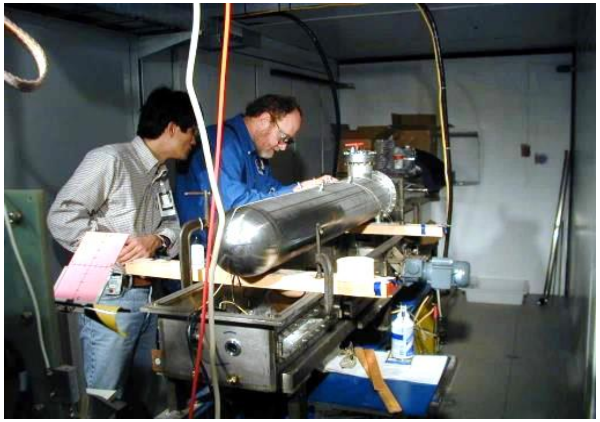



Magnetic Bragg peaks in the pressure-induced superconductor MnP at T=3K.

# US-Japan Cooperative Program on Neutron Scattering

## Leveraging strengths to find solutions to common challenges

Joint mercury target tests at BNL for pulse strain response (2001)




Japanese target experiment at SNS Target Test Facility (2009)

Regular collaboration meeting (2017)

SNS gas wall experiment using mercury cavitation device in J-PARC (2009)

SNS experiments at LANL using swirl bubblers from Japan (2005-2011)

SNS swirl gas bubblers, leverage Japanese development (2018). This will enable SNS to operate at 2 MW after PPU.

# Energy: A Common Challenge for US and Japan

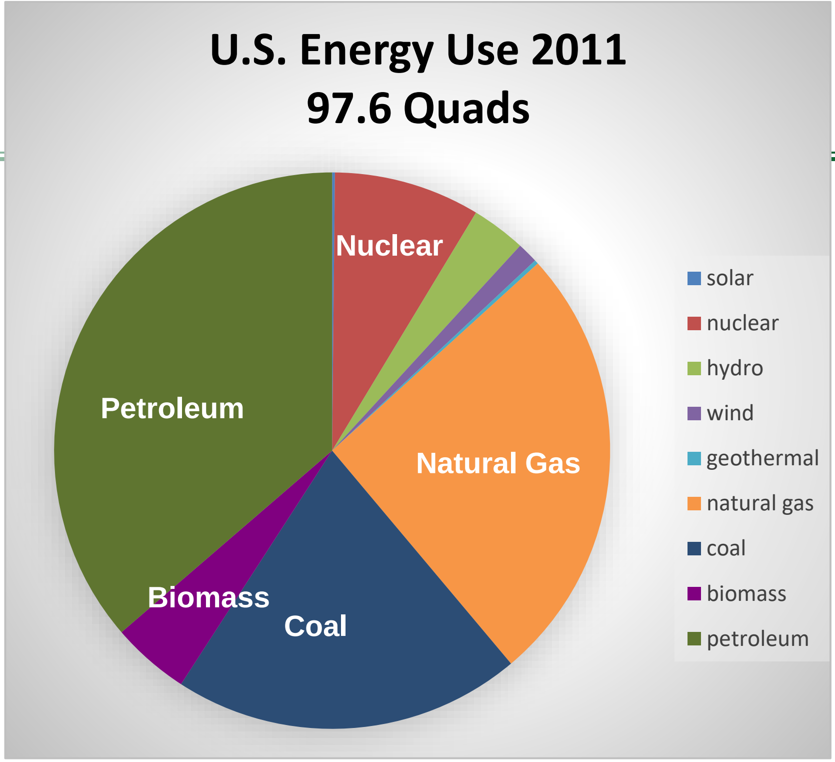
## Leveraging strengths to find solutions

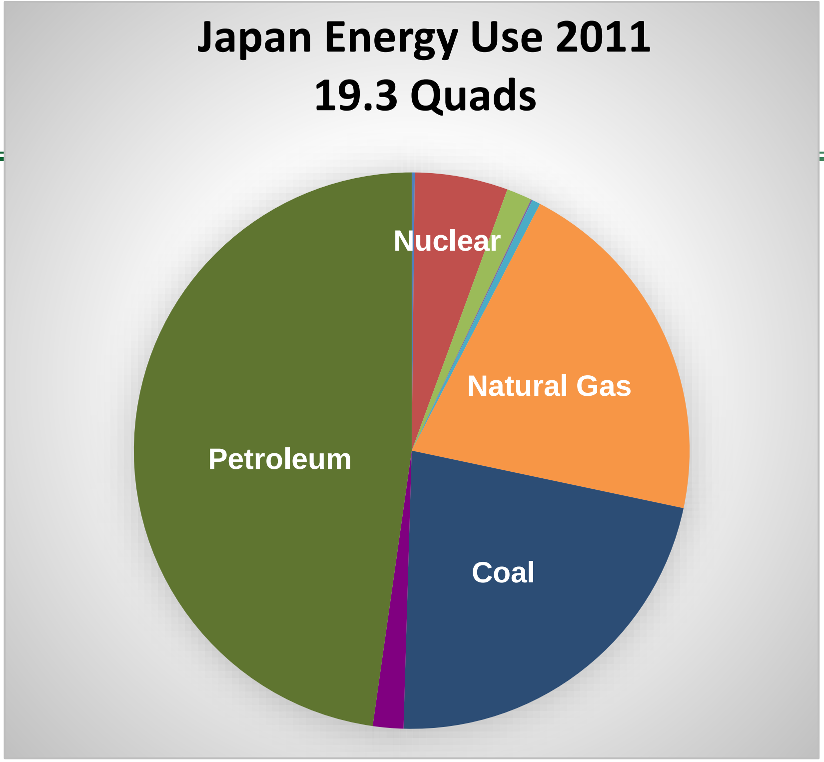






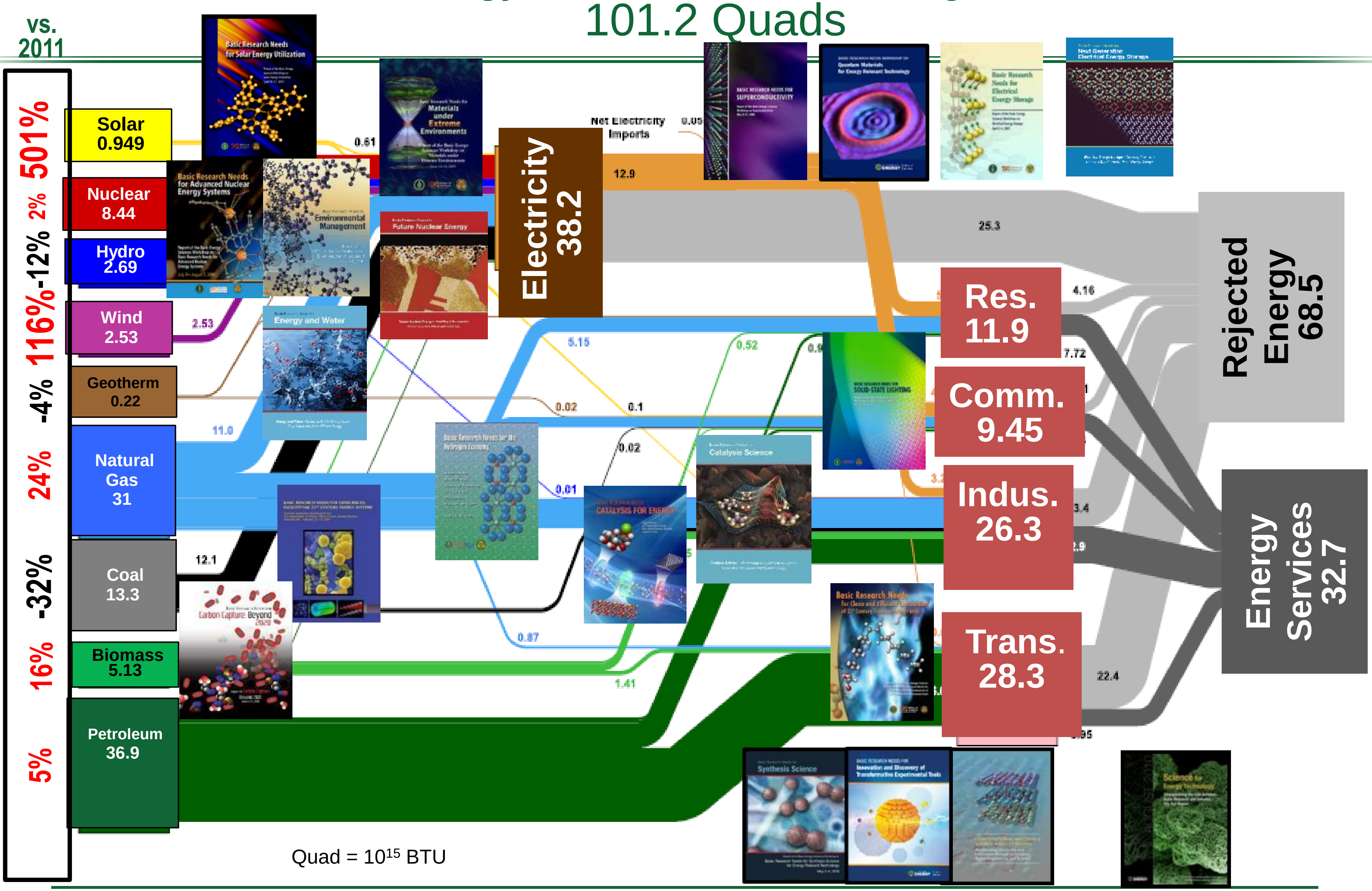





| | US | Japan |
|---|---|---|
| Solar | 0.16% | 0.18% |
| Nuclear | 8.50% | 5.42% |
| Hydro | 3.16% | 1.48% |
| Wind | 1.20% | 0.08% |
| Geothermal | 0.23% | 0.49% |
| Natural Gas | 25.62% | 20.68% |
| Coal | 20.27% | 22.15% |
| Biomass | 4.54% | 1.77% |
| Petroleum | 36.32% | 47.76% |

# U.S. Energy Production and Usage in 2018
# 101.2 Quads

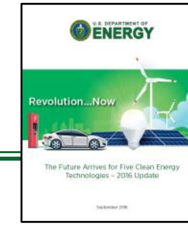





Energy Flow Charts Source:
https://flowcharts.llnl.gov/content/assets/docs/2018_United-States_Energy.pdf

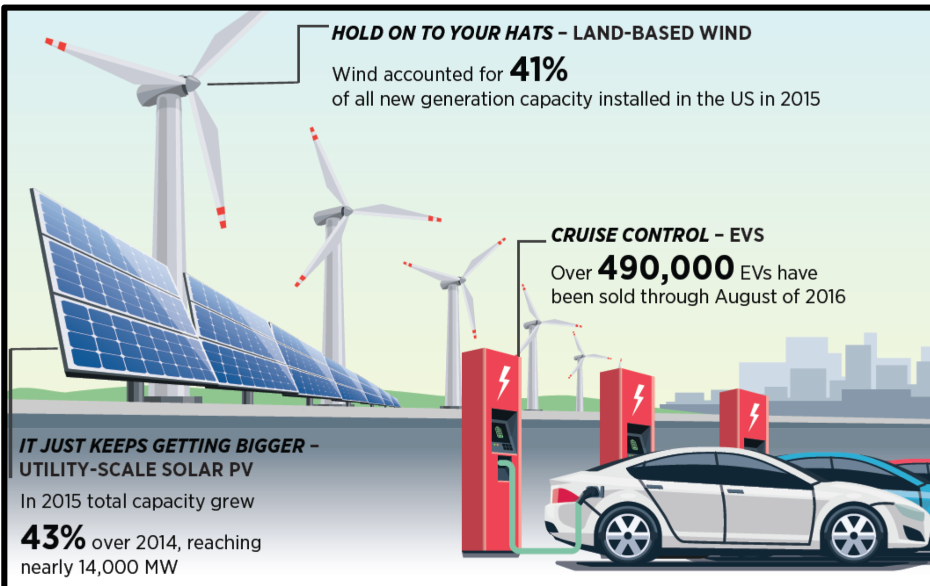
# DOE Report: Revolution…Now 2016

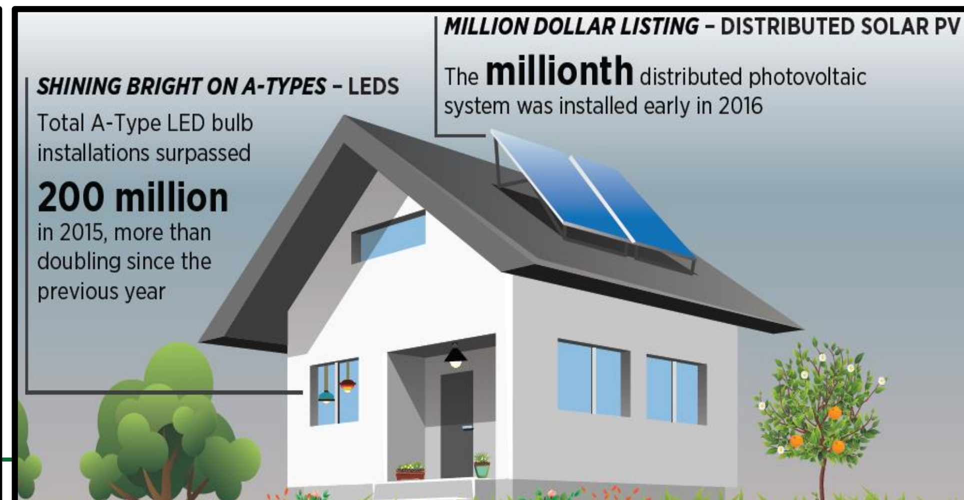
## Accelerating Clean Energy Deployment




### Major Findings

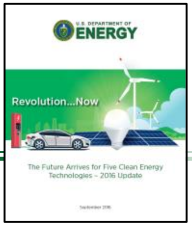
- Wind and solar PV accounted for over 2/3 of all new electricity generating capacity installed in the U.S. in 2015
- Land-based wind accounted for 41 percent of all new capacity brought online in the United States in 2015
- Utility-scale PV generated enough electricity to power more than 2 million homes in 2015. It also represented 15 percent of all newly installed electricity generation capacity in 2015.
- More than 1 million distributed PV systems have been installed on American homes and businesses.
- Total installations of A-type LED bulbs exceeded 200 million through 2015 - growing 160 percent over 2014.
- Total sales of electric vehicles has soared closer to the half million mark with 490,000 EVs on the road as of August 2016.



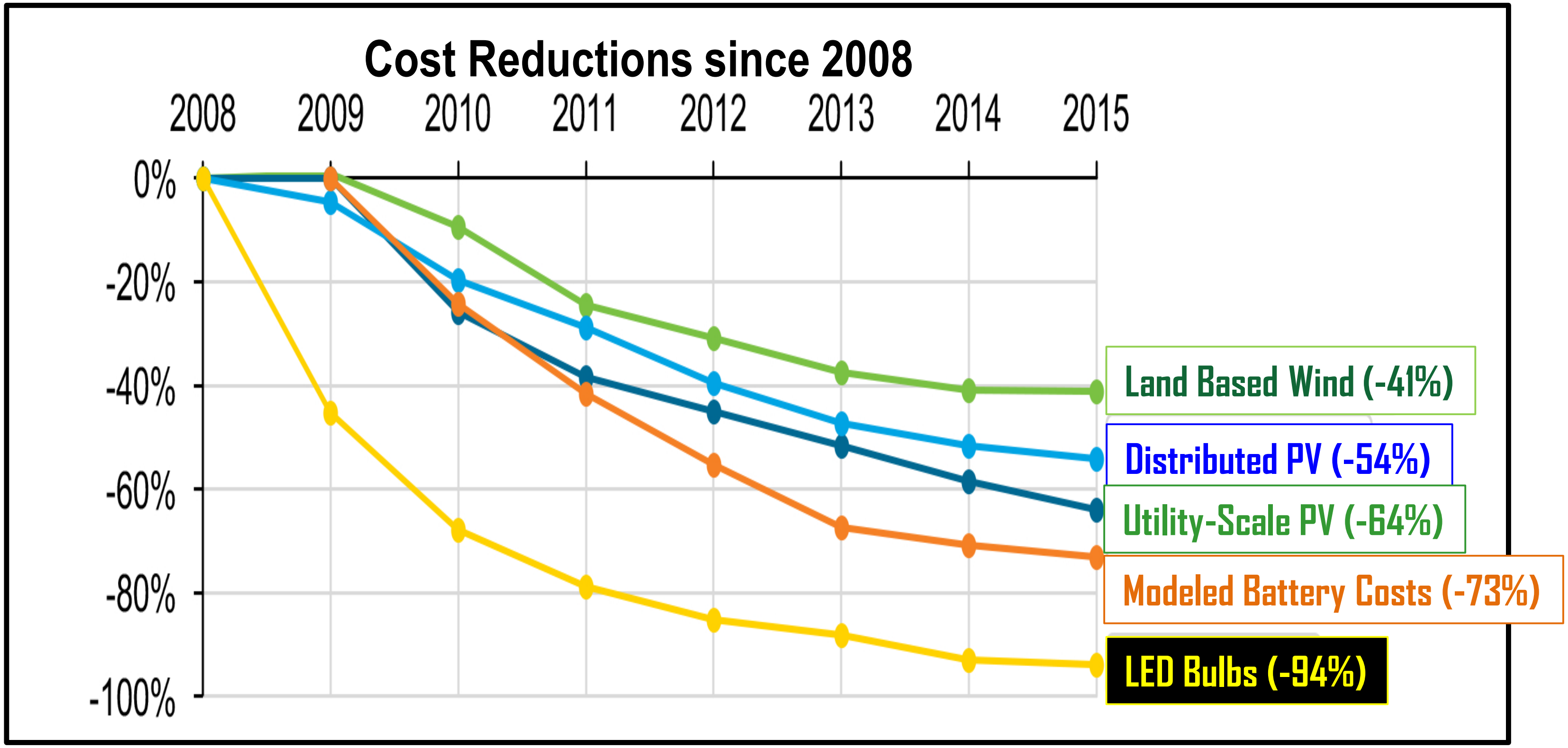




http://energy.gov/revolution-now

# How Low Can They Go?

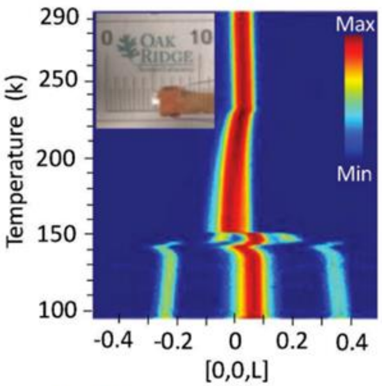


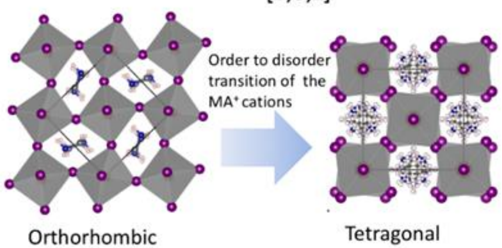

Thanks to decades of strategic investments by DOE, energy technologies have rapidly decreased in cost since 2008.




Notes: Land based wind costs are derived from levelized cost of energy from representative wind sites. Distributed PV cost is average residential installed cost. Utility-Scale PV cost is the median installed cost. Modeled battery costs are at high-volume production of battery systems, derived from DOE/UIS Advanced Battery Consortium PHEV Battery development projects. LED bulb costs are cost per lumen for A-type bulbs. See full report for full citations and details.

# Neutrons Reveal Role of Cation Ordering in Hybrid Perovskites for More Efficient Solar Cells




Neutron diffraction revealed a structural phase transition at low temperatures in novel hybrid perovskite materials used for solar cells that correlates with an anomalous increase in photoluminescence. The inset shows the $CH_3NH_3PbBr_3$ single crystal grown at CNMS.

**Work was performed at SNS's TOPAZ instrument, a DOE Office of Science User Facility.**

## Scientific Achievement

**Neutron diffraction coupled with Density Functional Theory (DFT) calculations revealed how temperature-induced disorder of caged organic cations induce a structural transformation in organic-inorganic hybrid perovskites (OIHPs) - materials that have an inorganic cage with organic materials inside - which significantly affects their optoelectronic properties.**

## Significance and Impact

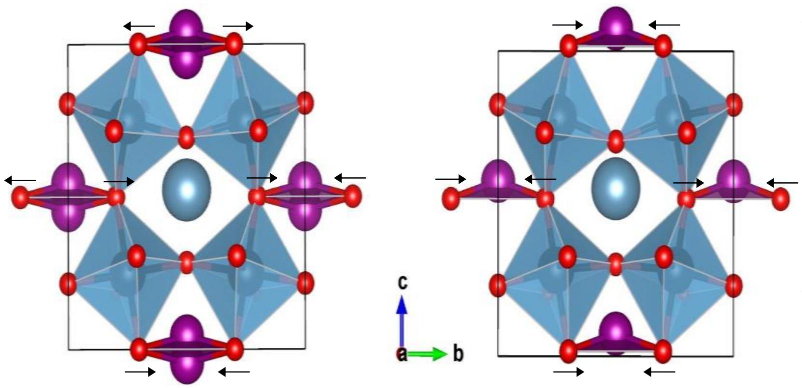
**Understanding how organic cation ordering affects the structure and optoelectronic properties in OIHPs enables the design and synthesis of new OIHP materials for solar cells with higher power conversion efficiency.**

## Research Details

- Real-time and in situ neutron and X-ray diffraction studies show the order to disorder transformation of $MA^+$ cations in OIHPs is responsible for anomalous photoluminescence (PL) measured with increasing temperature.
- DFT calculations explained the anomalous increase in PL intensity by the reduction of defect density and dielectric screening caused by increasing disorder of the caged $MA^+$ cations in OIHPs.


B. Yang, W. Ming, M.-H. Du, J. K. Keum, A. A. Puretzky, C. M. Rouleau, J. Huang, D. B. Geohegan, X. Wang, and K. Xiao, *Adv. Materials* (2018).

# New Mechanism for Ferroelectricity in Double Perovskites




T > $T_c$ T < $T_c$

*At high temp (above 650K) $Ca_{2-x}Mn_xTi_2O_6$ loses ferroelectric properties (left). At low temp (below 650K, which includes room temp) the ferroelectric phase remains (right). This is important because it means that this new thinner ferroelectric material is more likely to work at room temperature than conventional materials like $BaTiO_3$ and $PbTiO_3$.*

**Work performed at the ORNL Spallation Neutron Source's POWGEN instrument, a DOE Office of Science User Facility.**

Z. Li, Y. Cho, X. Li, X. Li, A. Aimi, Y. Inaguma, J.A. Alonso, M.T. Fernandez-Diaz, J. Yan, M.C. Downer, G. Henkelman, J.B. Goodenough, and J. Zhou, *JACS*, (2018).

## Scientific Achievement

**Neutron diffraction data coupled with first-principles calculations revealed the origin of ferroelectricity in the double perovskite $CaMnTi_2O_6$. This discovery made it possible to develop a new method to mass produce $Ca_{2-x}Mn_xTi_2O_6$ (a new ferroelectric material).**

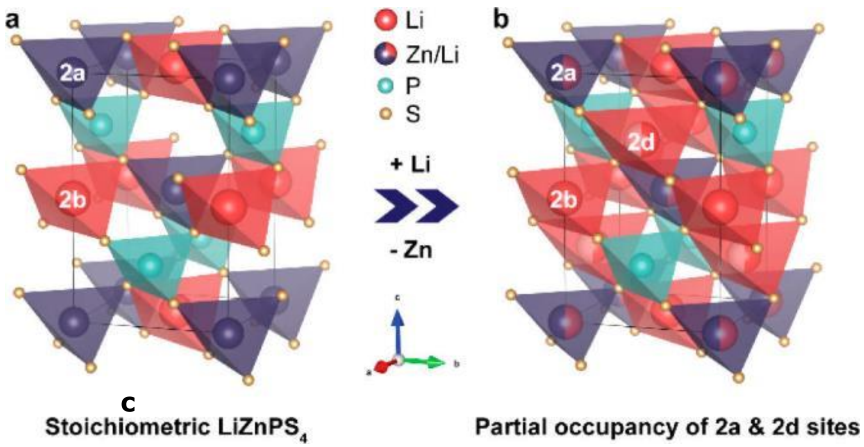
## Significance and Impact

**Materials lose their ferroelectric properties when the material is too thin. The ferroelectricity caused by an order-disorder transition has the potential to overcome this critical thickness problem in all proper ferroelectric materials. This coupled with the new production method may make it possible to mass produce new ferroelectric materials and greatly simplify the structure of microelectronic devices like smart phones which would make them more efficient and less expensive to produce.**

## Research Details

— Neutron powder diffraction was performed at temperature range 2K-700K.
— Rietveld refinement results provide the detailed information of the structural transition at $T_C$ and the interplay between magnetic ordering and the ferroelectric displacement below $T_N$.
— Origin of ferroelectricity is where a polar structure is caused by the order-disorder transition at the coplanar $Mn^{2+}$ sites, rather than the displacement-type in conventional ferroelectric materials such as $BaTiO_3$ and $PbTiO_3$

# New Pathway to Develop Safer and More Efficient Batteries




*a) Stoichiometric $LiZnPS_4$, b) partial occupancy of the 2a and interstitial 2d Li site in $Li_{1+2x}Zn_{1-x}PS_4$ leading to increase in ionic conductivity. Understanding the complex relationship between lithium site occupation and ionic conductivity provides a significant step forward towards realizing rechargeable all solid state battery technologies.*

**Work performed at the ORNL Spallation Neutron Source's POWGEN instrument, a DOE Office of Science User Facility.**

K. Kaup, F. Lalère, A. Huq, A. Shyamsunder, T. Adermann, P. Hartmann and L. F. Nazar, Chem. Mat., (2018).

## Scientific Achievement

**Neutron powder diffraction and synchrotron X-ray data were used to provide a detailed structural characterization of the $Li_{1+2x}Zn_{1-x}PS_4$ solid solution. Neutrons are sensitive to lighter elements like lithium which made it possible to find the position and occupancy of interstitial Li sites that play a key role in achieving fast lithium ion conduction.**

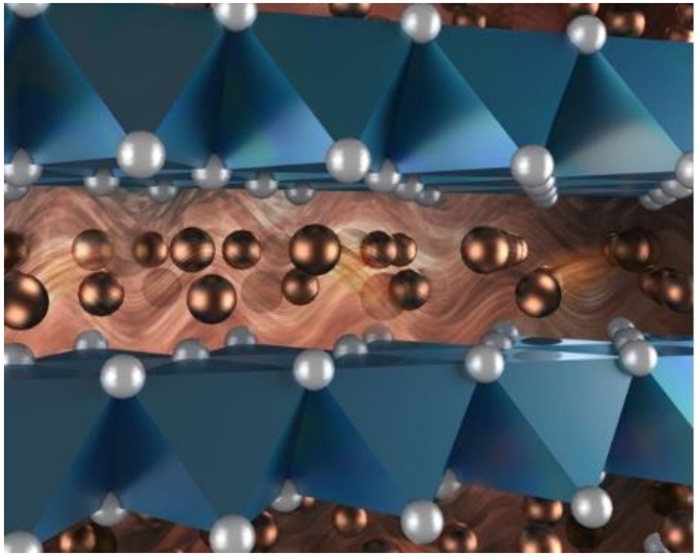
## Significance and Impact

**This work validates previous theoretical Density functional theory and ab initio molecular dynamics calculations on this structure, and indicate its potential as an improved lithium ion conductor in an all solid state battery (ASSB). This technology has the potential to increase the maximum stored energy in solid state batteries, while also making them safer (less explosive) and extending their useful life.**

## Research Details

- The partial occupancy of Li in the 2a and 2d site leads to a substantial increase in lithium ion conduction from $10^{-8}$ S/cm to $10^{-4}$ S/cm.
- A deeper understanding of the mechanisms driving the structure property relationships in the next generation of battery materials is required to commercially realize the technology.

# Ultra-low Thermal Conductivity for Thermoelectrics and Batteries



Artist representation of Cu ions hopping (brown balls) in superionic $CuCrSe_2$. ORNL/Jill Hemman

Work used CNCS & ARCS spectrometers at the SNS (ORNL), HERIX spectrometer at APS sector 30 (ANL), and computational resources at NERSC and OLCF.

J. L. Niedziela*, D. Bansal*, A. May, J. Ding, T. Lanigan-Atkins, G. Ehlers, D.L. Abernathy, A. Said and O. Delaire. Selective Breakdown of Phonon Quasiparticles Across the Superionic Transition in $CuCrSe_2$, Nature Physics, (2018). DOI:https://doi.org/10.1038/s41567-018-0298-2

## Scientific Achievement

**For the first time, the evolution of atomic dynamics across a superionic transition has been fully elucidated, by resolving both phonon vibrations and atomic diffusion in $CuCrSe_2$.**

## Significance and Impact

**This discovery provides a new route to design solid-state electrolytes for safer batteries and also to improve thermoelectric devices by suppressing thermal conductivity.**

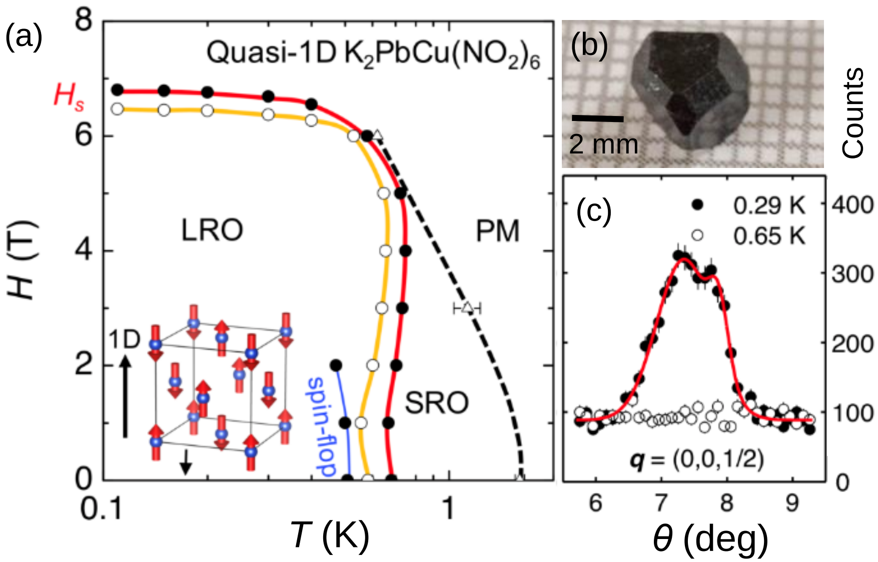
## Research Details

– Used inelastic neutron scattering, quasi-elastic neutron scattering, inelastic x-ray scattering, and first-principles simulations to track evolution of the lattice dynamics and ionic hopping across the superionic transition, when ionic conductivity becomes liquid-like.

– Research establishes that superionic diffusion of Cu ions arises with the breakdown of anharmonic low-energy optic phonon modes in the normal state.

– Strongly anharmonic lattice vibrations are also the origin of the very low thermal conductivity enabling high-efficiency thermoelectric conversion. Yet, the observation of persisting acoustic phonons in the superionic phase shows that an oppotunity exists to reduce the thermal conductivity even further.

# New Method to Design Highly-entangled Quantum Matter




(a) Applied magnetic field (H) vs temperature (T) phase diagram of quasi-1D $K_2PbCu(NO_2)_6$. The applied magnetic field suppresses the 3D long-range magnetic order between coupled chains and produces a highly-entangled state. (b) Single crystal used in this study. (b) Rocking scans through the magnetic Bragg peak from HB-1A both above and below the 3D ordering temperature in zero field. Collectively this data made it possible to determine the type of long-range magnetic order in this material.

**Work was performed at the HB-1A spectrometer at the High Flux Isotope Reactor, a DOE Office of Science User Facility.**

N. Blanc, J. Trinh, L. Dong, X. Bai, A.A. Aczel, M. Mourigal, L. Balents, T. Siegrist, and A.P. Ramirez, *Nature Physics* (2017).

## Scientific Achievement

**A neutron scattering and specific heat study found that a modest applied magnetic field of 6 Tesla can melt the 3D long-range magnetic order in $K_2PbCu(NO_2)_6$, which is comprised of weakly-coupled antiferromagnetic spin-1/2 chains, and produce a highly-entangled quantum state of matter near the quantum critical point.**

## Significance and Impact

**This work demonstrates a new approach for producing highly-entangled quantum matter which may make it possible to create highly frustrated quantum matter in a larger family of materials. This state makes electrons more sensitive to outside forces which in turn may enable the creation of qubits (the smallest bit of info in a quantum computer made of electrons that spin up (1s) and down (0s). The efficient creation of qubits could contribute to the development of a quantum computer.**

## Research Details

- Single crystals of $K_2PbCu(NO_2)_6$ were grown by Siegrist's group at Florida State University using a sol-gel technique.
- Bulk characterization measurements were performed at Georgia Tech and UC Santa Cruz by PIs Mourigal and Ramirez.
- The high flux and excellent signal-to-noise of the HB-1A spectrometer at the HFIR enabled this research team to detect weak magnetic Bragg peaks associated with the 3D ordered state of this material.

# Neutron Scattering Facilities Worldwide:
## Reactors & Spallation Sources


US Neutron Scattering Facilities

European Neutron Scattering Facilities

Asian Neutron Scattering Facilities

Office of Science

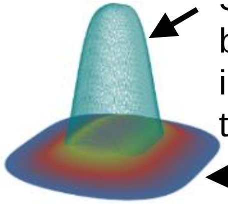
# SNS Upgrades at ORNL

**Goal:** To provide high neutron fluxes (Proton Power Upgrade, PPU) & cold neutrons with higher intensity and energy resolution (Second Target Station, STS)



## Project Deliverables:

- PPU raises the power delivered to the first target station to 2 MW
- PPU provides the capability to deliver 0.5 MW to the STS
- STS adds a new tungsten target, experimental hall, and 8 world-class instruments with room for 14 more instruments
- STS will be optimized for cold neutrons



STS provides a smaller beam footprint and 20x intensity compared to the first target station

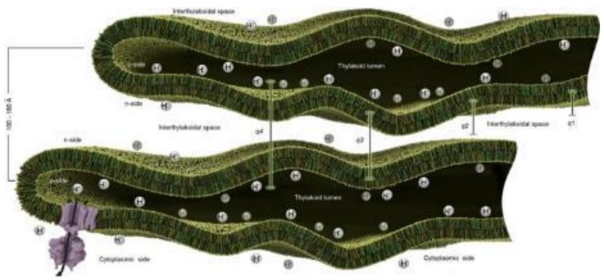
## PPU enables:

- Increased neutron flux to existing beamlines
- New experiments currently limited by low intensity
- A platform for providing high intensity neutrons to the STS

## STS enables:

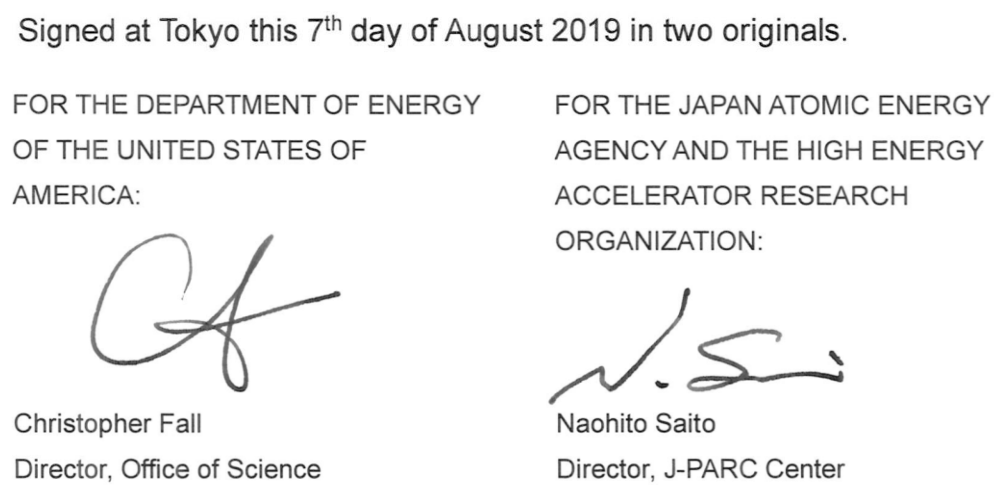
- 20-fold increase in the cold neutron flux through novel moderator designs
- 100-fold improvement in time resolution for real-time observation of dynamic phenomena
- Investigation of smaller samples

STS will provide higher resolution for more lifetime detail about soft matters and bio-related processes



# DOE-MEXT Neutron Science Project Arrangement

- For SNS and J-PARC to purse a collaborative research program which focuses on research and development of high power spallation neutron science and related technologies.


Signed at Tokyo this 7th day of August 2019 in two originals.

FOR THE DEPARTMENT OF ENERGY OF THE UNITED STATES OF AMERICA:

Christopher Fall
Director, Office of Science

FOR THE JAPAN ATOMIC ENERGY AGENCY AND THE HIGH ENERGY ACCELERATOR RESEARCH ORGANIZATION:

Naohito Saito
Director, J-PARC Center

# SNS / J-PARC possible future collaboration areas

## H$^-$ ion sources

Long life antennas needed for high reliability, and high current RF plasma volume source

## High power RF systems

High power and robust RF systems are a common area of concern for reliable operations

## Target lifetime extensions

Post-irradiation examination of targets to understand cavitation and rad-damage lifetime limitations

Robotic device to cut target vessel samples

## Charge exchange injection to rings

Robust stripper foils required for high power / long life operation

New

Used