Science with Neutrons

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Quantum Condensed Matter Division
Neutron Sciences

The Neutron Lifecycle Lecture Series
July 7, 2016

ORNL is the home of two powerful neutron sources

High Flux Isotope Reactor (HFIR)
Intense steady-state neutron flux and a high-brightness cold neutron source

Spallation Neutron Source (SNS)
World's most powerful accelerator-based neutron source

U.S. Department of Energy user facilities

OAK RIDGE National Laboratory | HIGH FLUX ISOTOPE REACTOR | SPALLATION NEUTRON SOURCE

ORNL is the home of two powerful neutron sources

Why do we go to so much trouble to produce so many neutrons?

Neutrons are a powerful probe to study materials!!

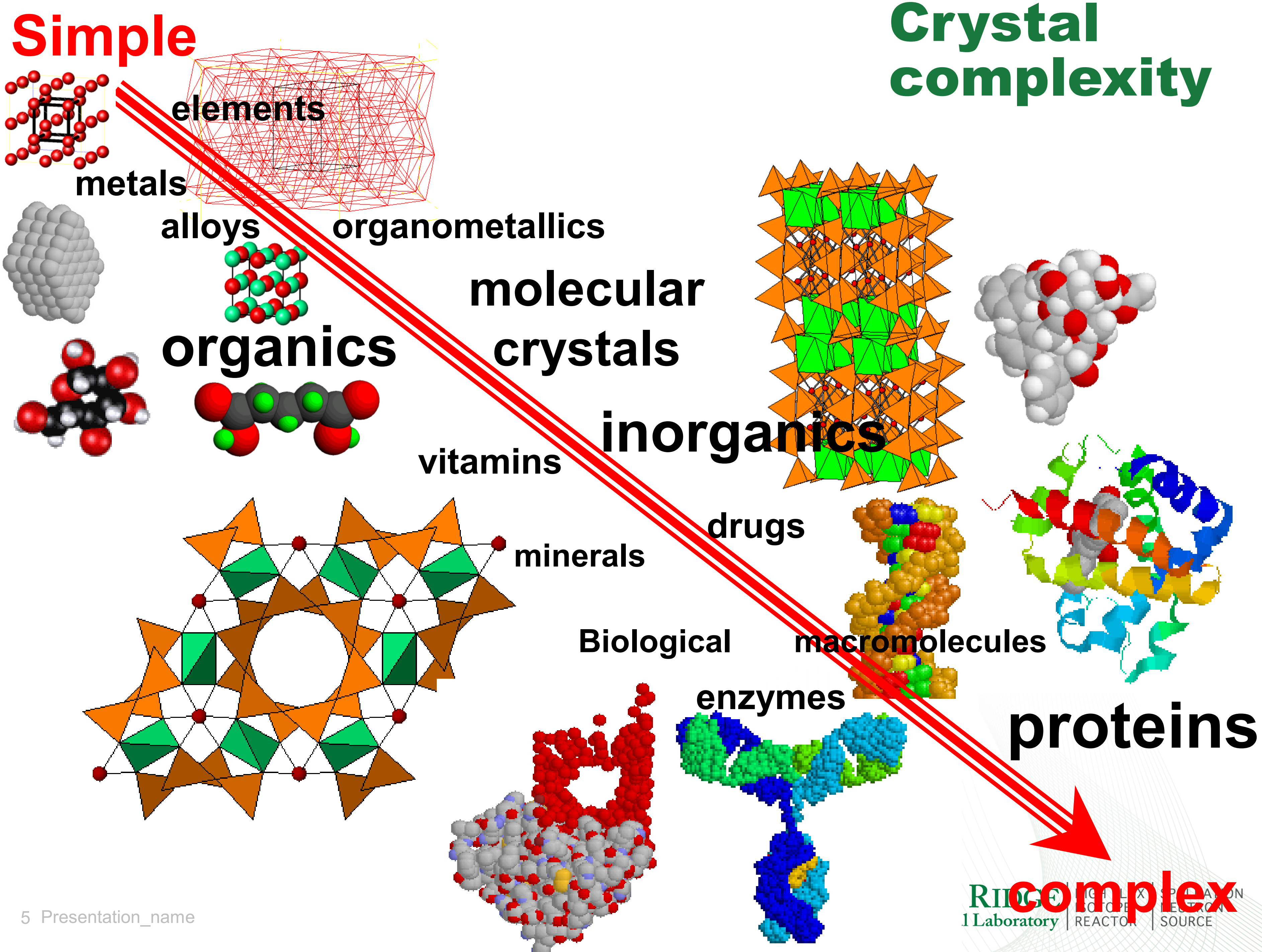
Why do we care about materials?

Material properties are largely determined by the atomic arrangement.

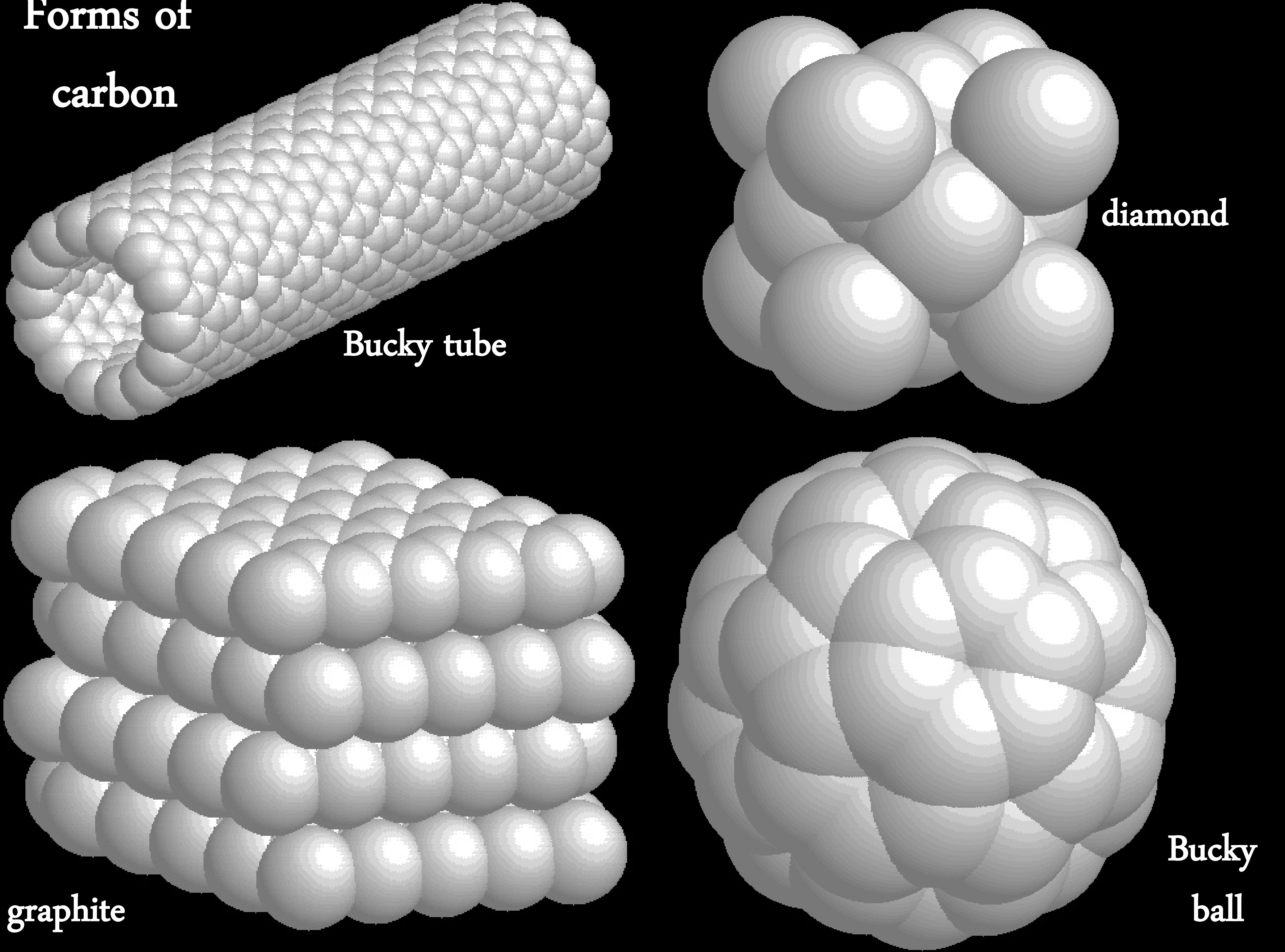
Understanding the atomic interactions in materials gives us clues about fundamental microscopic processes

Biological materials,
Engineering materials,
Quantum Materials, etc.

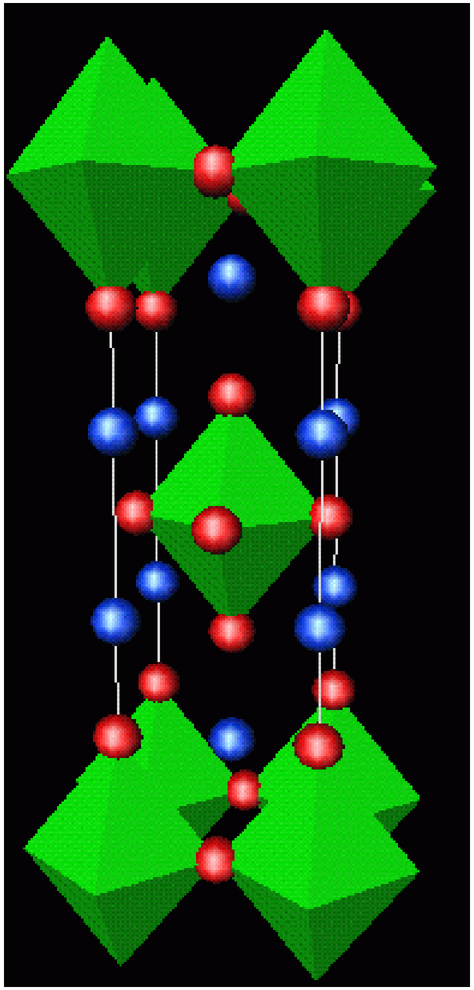
Crystal complexity



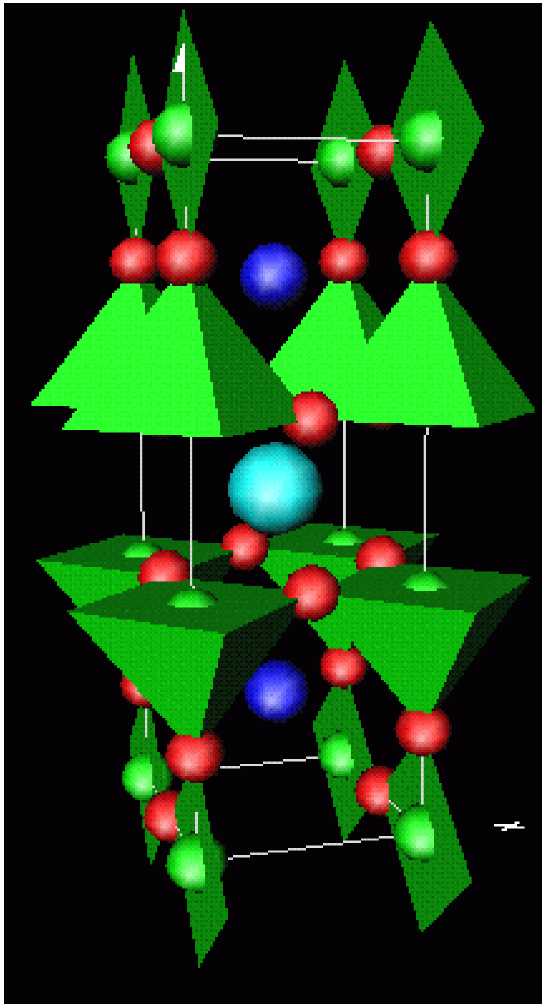
Forms of carbon



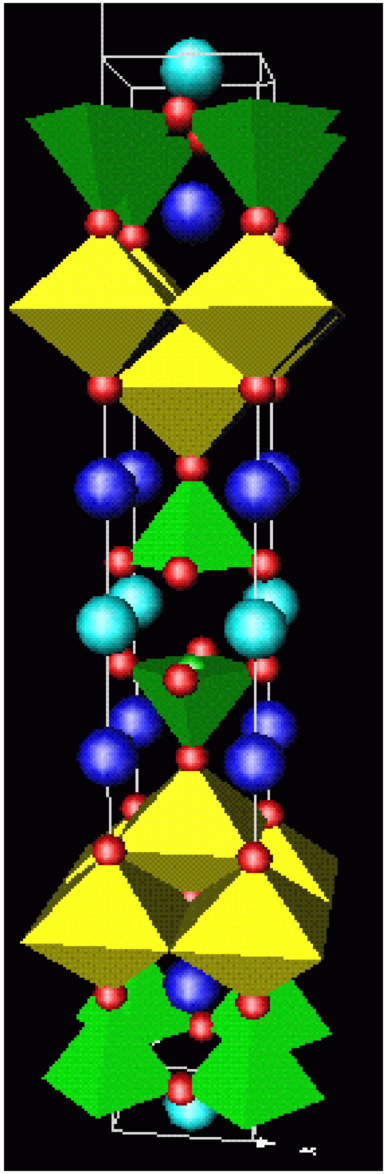
Cuprate Superconductors



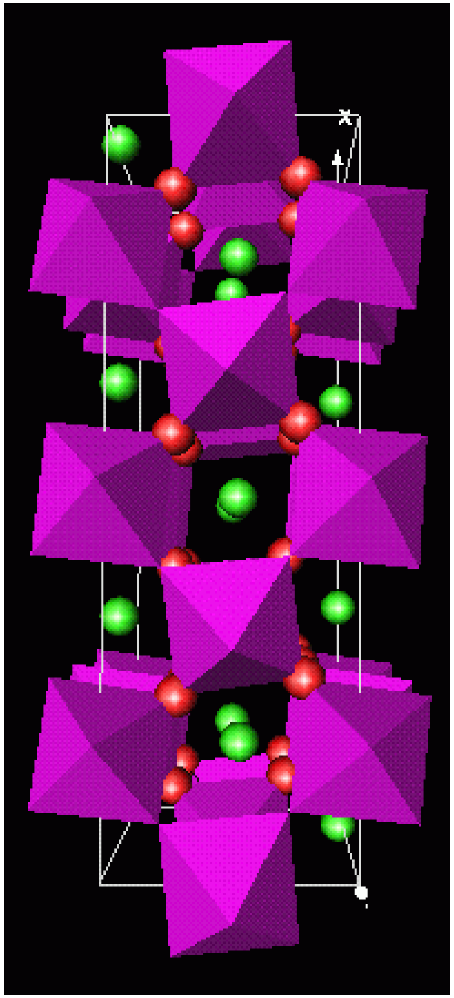
$(La,Ba)_2CuO_4$
40K

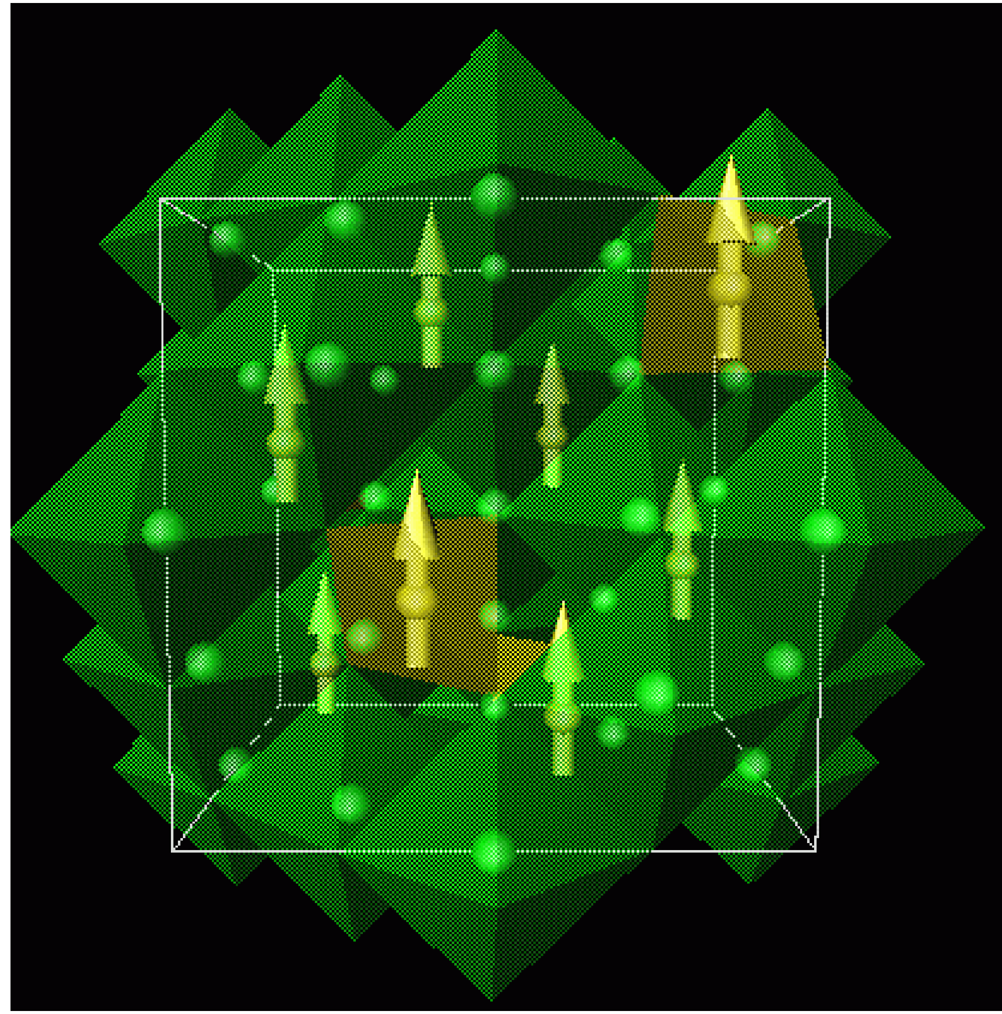


$YBa_2Cu_3O_7$
92K



$HgBa_2Ca_2Cu_3O_8$
122K

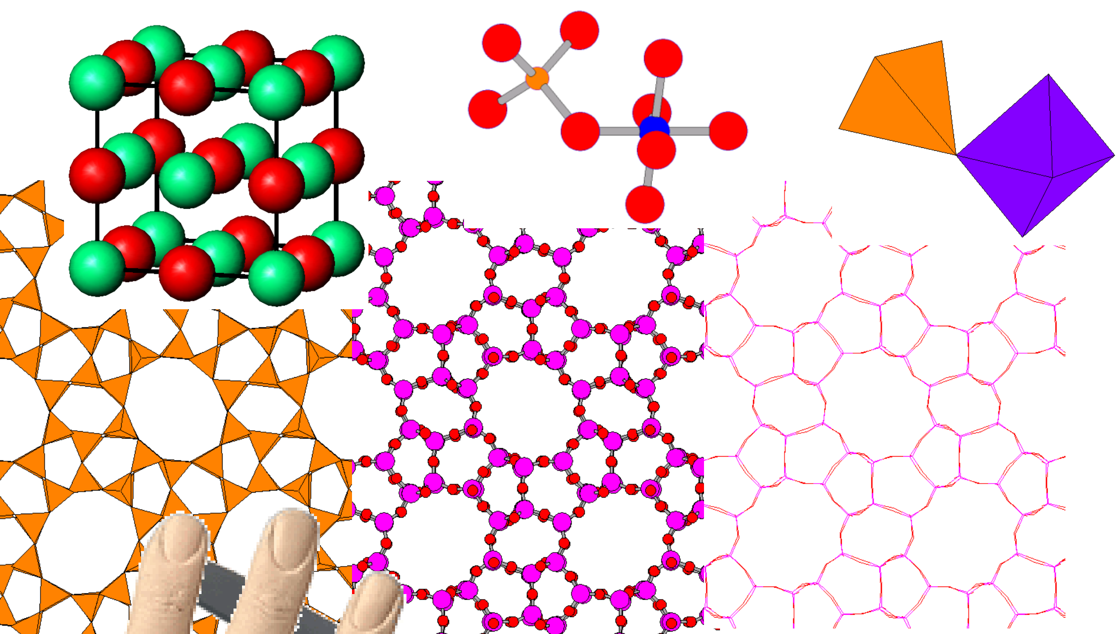


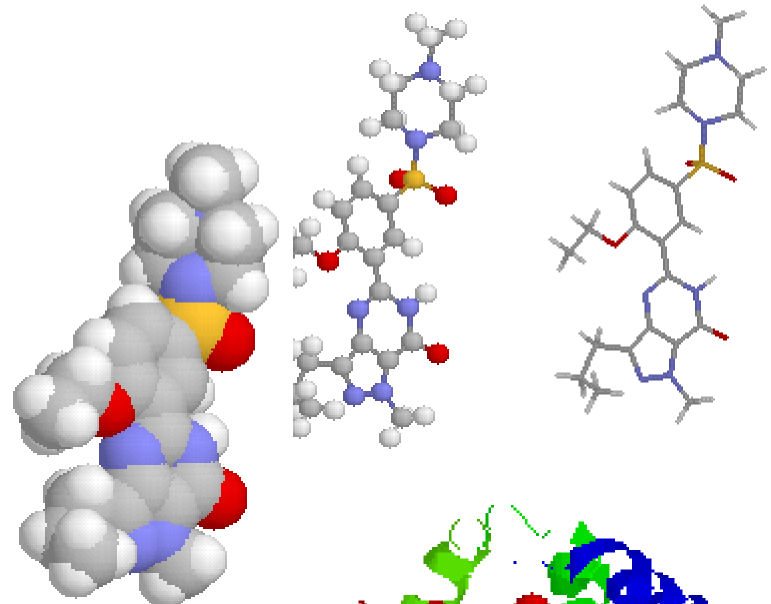


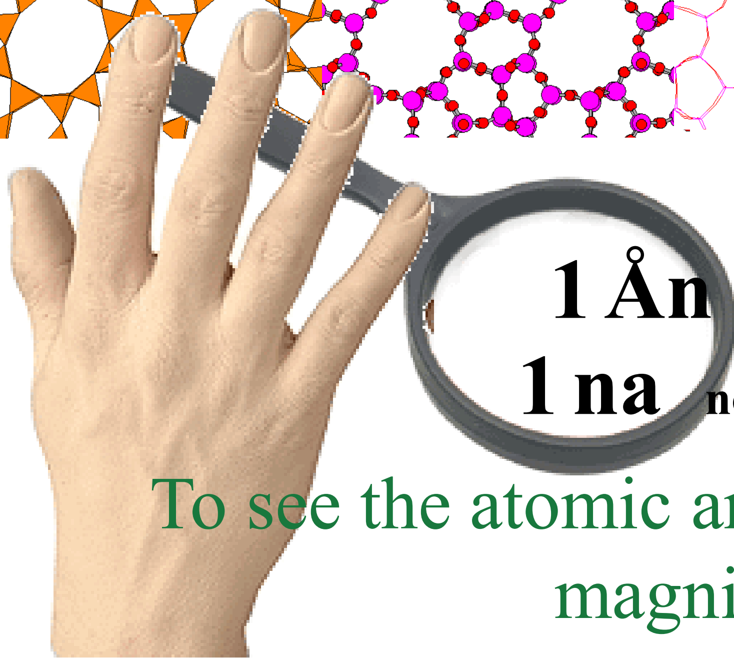
$(La,Ca)MnO_3$ perovksites
Colossal magnetoresistance
Magnetic storage devices

AB_2O_4 spinels
Ferrimagnets
MRI constrast agents

Visualizing crystal and molecular structures, cartoons of an invisible world



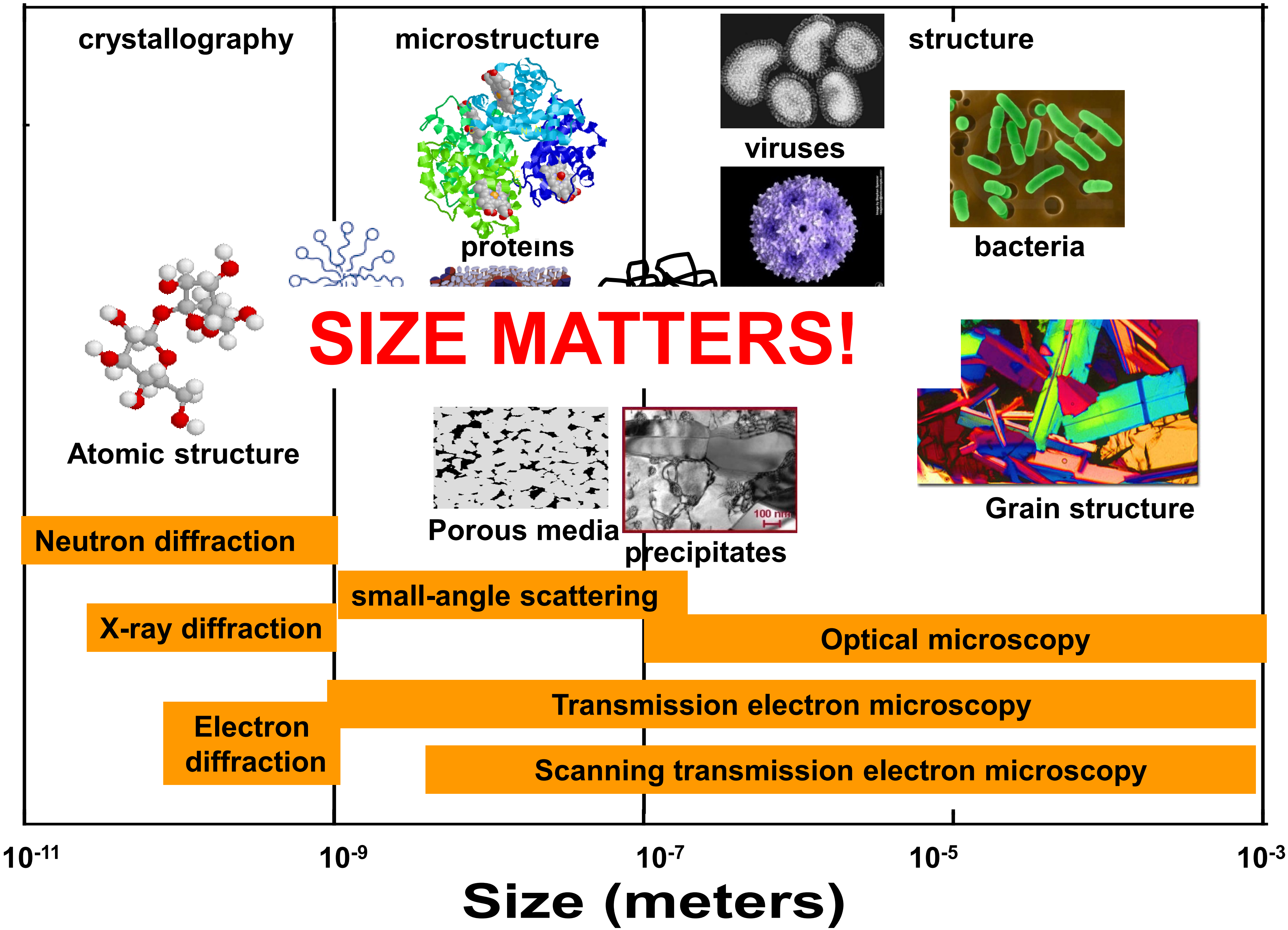




1 Ångstrom = 0.1 nanometer

1 nanometer = 0.000000001 meter

To see the atomic arrangement in a material, we must magnify 10,000,000 times



crystallography
microstructure
structure
viruses
bacteria
proteins
SIZE MATTERS!
Atomic structure
Porous media
precipitates
100 nm
Grain structure
Neutron diffraction
small-angle scattering
X-ray diffraction
Optical microscopy
Transmission electron microscopy
Electron diffraction
Scanning transmission electron microscopy
10^{-11}
10^{-9}
10^{-7}
10^{-5}
10^{-3}
Size (meters)

Visualizing atoms and molecules requires special probes

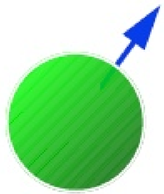
- X-rays
- Electrons
- Neutrons

Neutrons have unique properties that make them specially suitable to determine

- **Where the atoms are**
- **What do the atoms do**

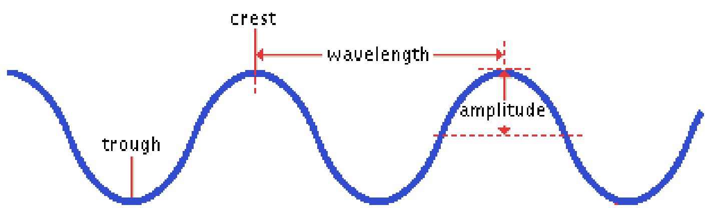
Neutrons

They are particles



Mass	$m = 1.67 \times 10^{-27}$ kg
Charge:	0
Spin:	1/2
Magnetic Moment	$\mu = -1.9\mu_N$

They also behave like "waves"



Wavelength λ (de Broglie)

Momentum $k=2\pi/\lambda$

Energy$=\hbar^2k^2/2m$

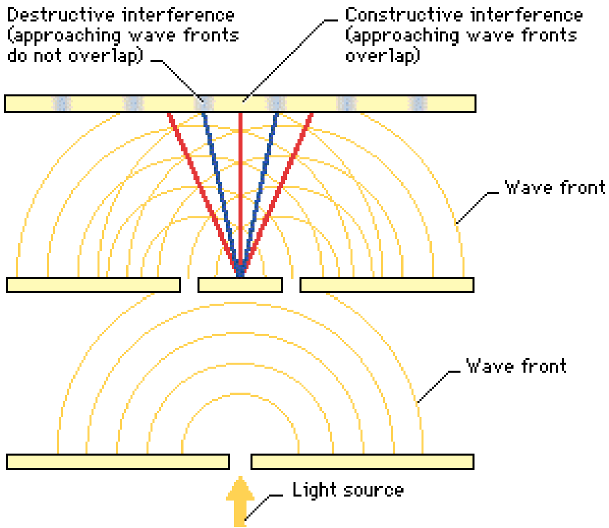
WAVE-PARTICLE DUALITY

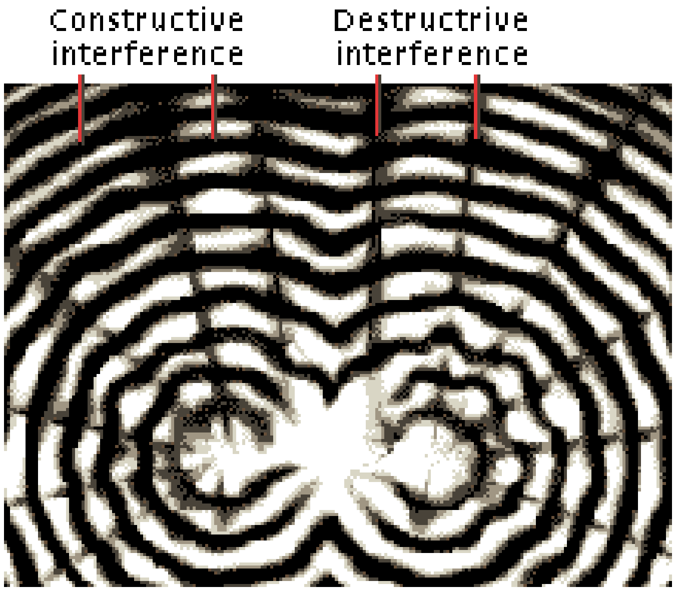
Louis-Victor Pierre Raymond de Broglie

Nobel Prize 1929 "for his discovery of the wave nature of electrons"

Wave Interference

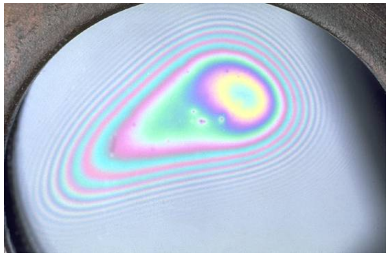
Waves can interfere constructively or destructively





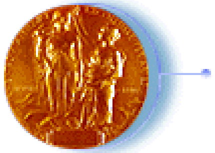
Interference patterns depend on the **wavelength** and the **distances between the sources**

Newton rings



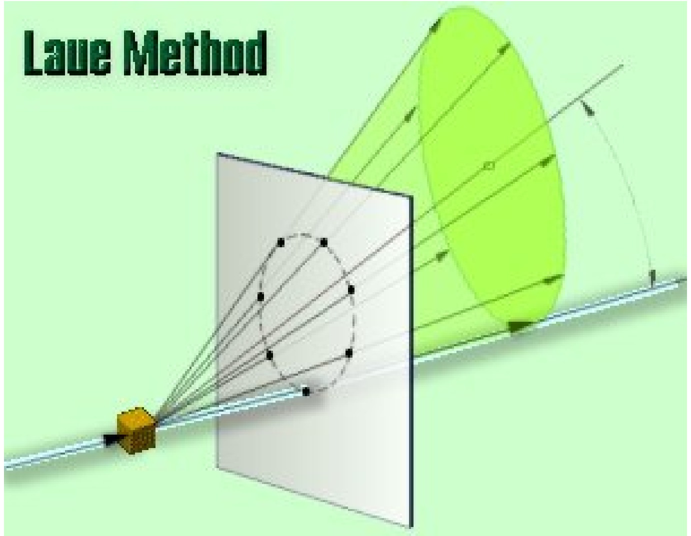
X-RAY DIFFRACTION

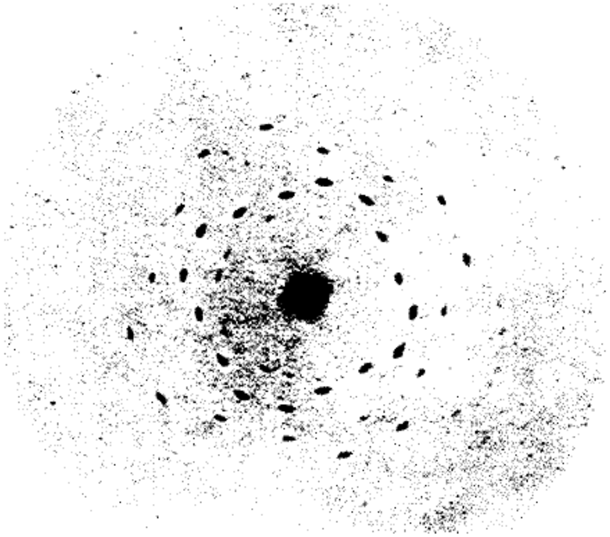
Nobel Prize in Physics 1914



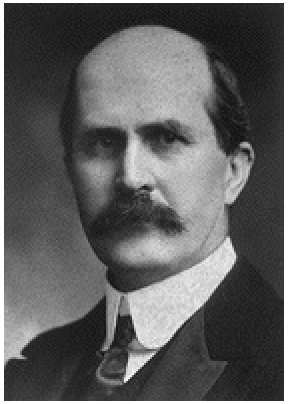
"for his discovery of the diffraction of X-rays by crystals"

Max Von Laue

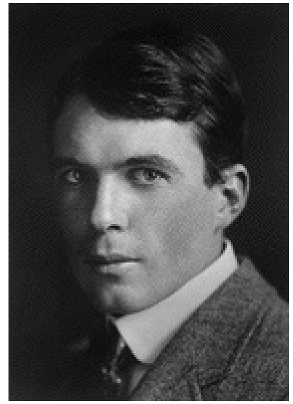




BRAGG DIFFRACTION LAW

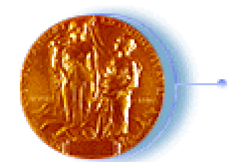


Sir William Henry Bragg

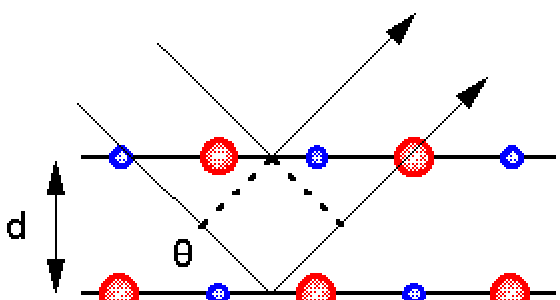


Sir William Lawrence Bragg

Nobel Prize in Physics 1915



$$n\ \lambda = 2d\ \sin\Theta$$



path difference=2dsinθ

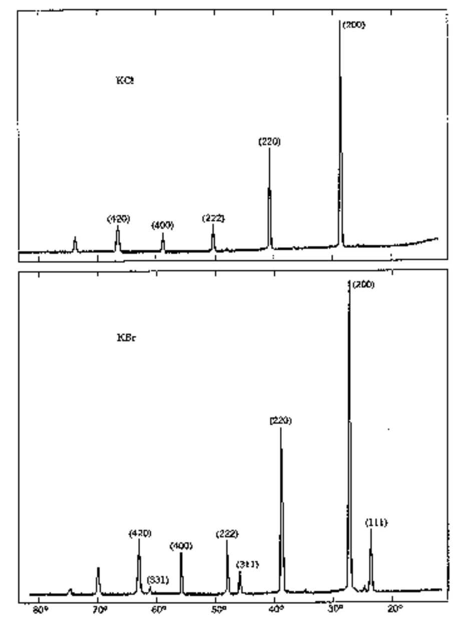


Figure 25 Comparison of x-ray reflections from KCl and KBr powders. In KCl the numbers of electrons of K^- and Cl^- ions are equal. The scattering amplitudes $f(K^+)$ and $f(Cl^-)$ are almost exactly equal, so that the crystal looks to x-rays as if it were a monatomic simple cubic lattice of lattice constant $a/2$. Only even integers occur in the reflection indices when these are based on a cubic lattice of lattice constant a. In KBr the form factor of Br^- is quite different than that of K^+, and all reflections of the fcc lattice are present. (Courtesy of Robert van Nordstrand.)

The beginnings of neutron diffraction

- Neutrons behave like waves like x-rays.
- In the 1940's Wollan and Shull applied the ideas of x-ray diffraction to neutrons (Oak Ridge Graphite Reactor)

Shull and Wollan at Oak Ridge Graphite Reactor

The Physical Review, Vol. 75 No 8, 830-841, April 15, 1948

The Diffraction of Neutrons by Crystalline Powders

E. O. Wollan and C. G. Shull

Oak Ridge National Laboratory, Oak Ridge, Tennessee

(Received April 15, 1948)

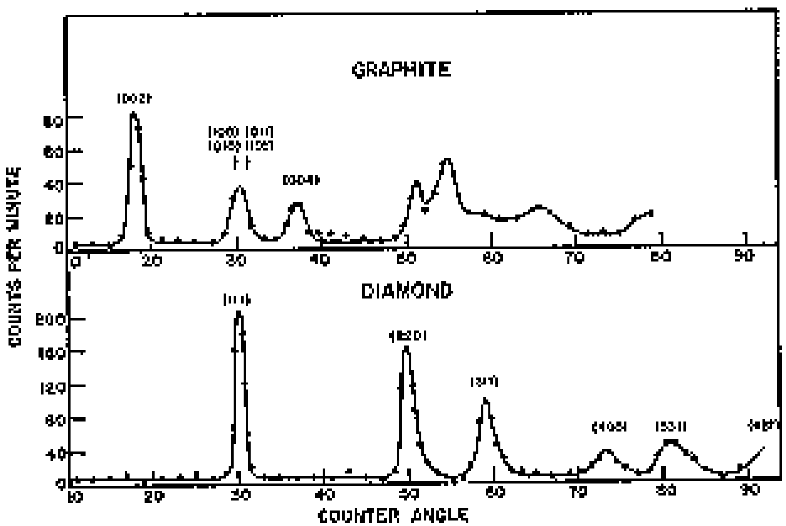


FIG. 3. Powder diffraction patterns for diamond and graphite. The major part of the diffuse scattering in these patterns arises from multiple scattering in the samples.

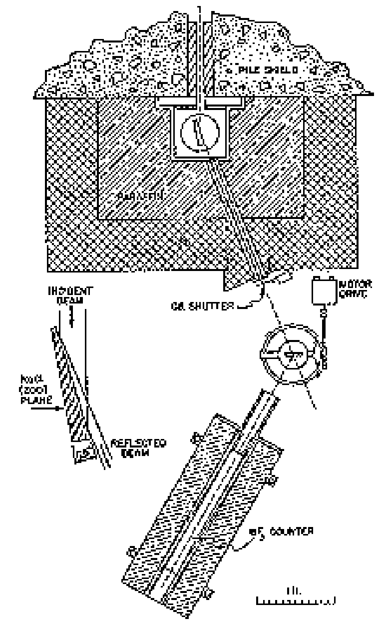


FIG. 1. Arrangement of apparatus, showing the monochromating crystal (detailed in left center) collimating slits, shielding, second spectrometer with location of powder specimen and counter.

Diffraction of neutrons by an antiferromagnetic lattice

- Neutrons have a magnetic spin and also interact with the electronic spins of materials.
- Neutron diffraction can be used to “see” a magnetic superstructure.

Detection of antiferromagnetism by neutron diffraction

C. G. Shull and J. S. Smart
Phys. Rev. **76**, 1256 (1949)

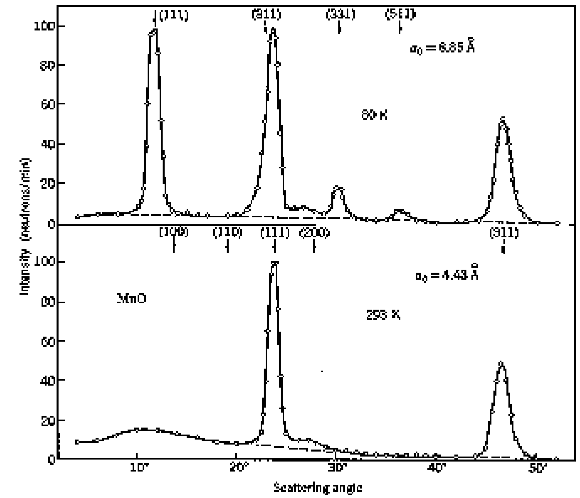


Figure 20 Neutron diffraction patterns for MnO below and above the spin-ordering temperature of 120 K, after C. G. Shull, W. A. Strauser, and E. O. Wollan, Phys. Rev. 83, 333 (1951). The reflection indices are based on an 8.85 Å cell at 80 K and on a 4.43 Å cell at 293 K. At the higher temperature the Mn^{2+} ions are still magnetic, but they are no longer ordered.

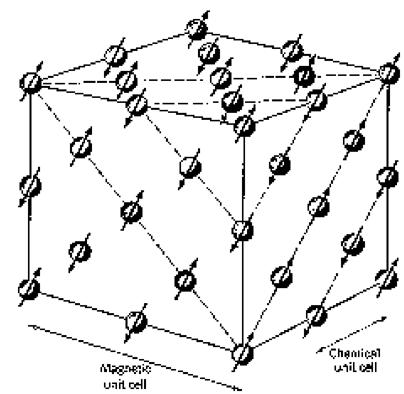
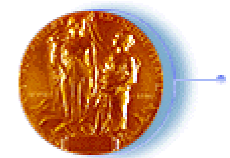


Figure 21 Ordered arrangements of spins of the Mn^{2+} ions in manganese oxide, MnO, as determined by neutron diffraction. The O^{2-} ions are not shown.

ANTIFERROMAGNETISM

Louis Eugène Félix Néel

Nobel Prize in Physics 1970



322 1970 LOUIS NÉEL

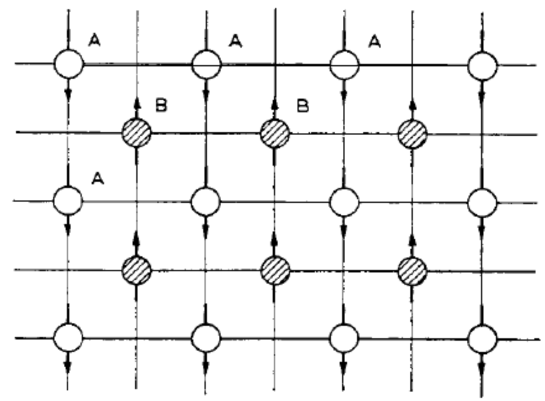


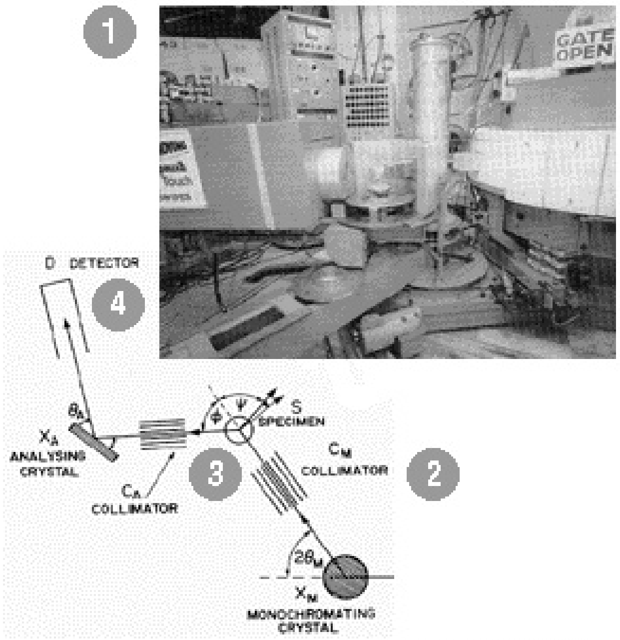
Fig. 1. Resolution of a plane lattice into two sub-lattices magnetized in antiparallel.

"for fundamental work and discoveries concerning antiferromagnetism and ferrimagnetism which have led to important applications in solid state physics".

The beginnings of neutron scattering

Bertram Brockhouse,
Chalk River, Canada,
1950's

First triple axis spectrometer to study the dynamics (vibrations of the atoms)



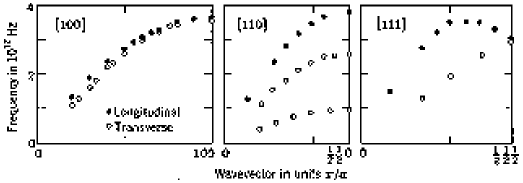


Figure 11 The dispersion curves of sodium for phonons propagating in the [001], [110], and [111] directions at 90 K, as determined by inelastic scattering of neutrons. [Woods, Brockhouse, March and Bowers, Proc. Phys. Soc. London 79, pt. 2, 440 (1962).]

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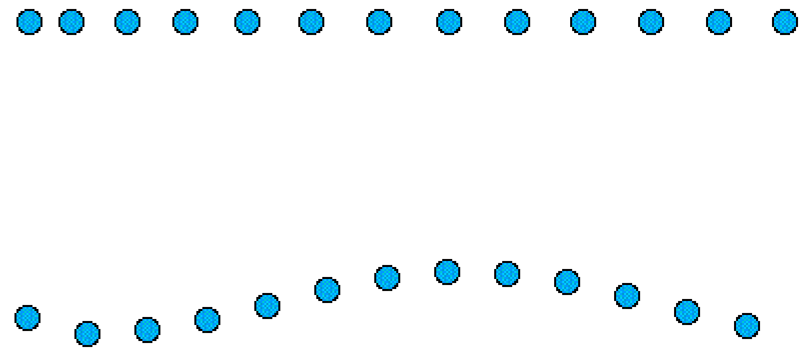

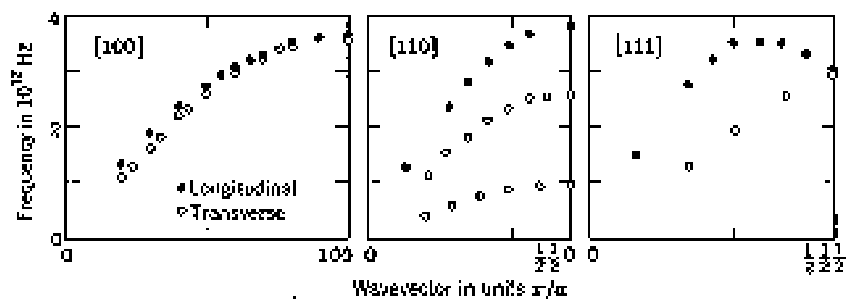

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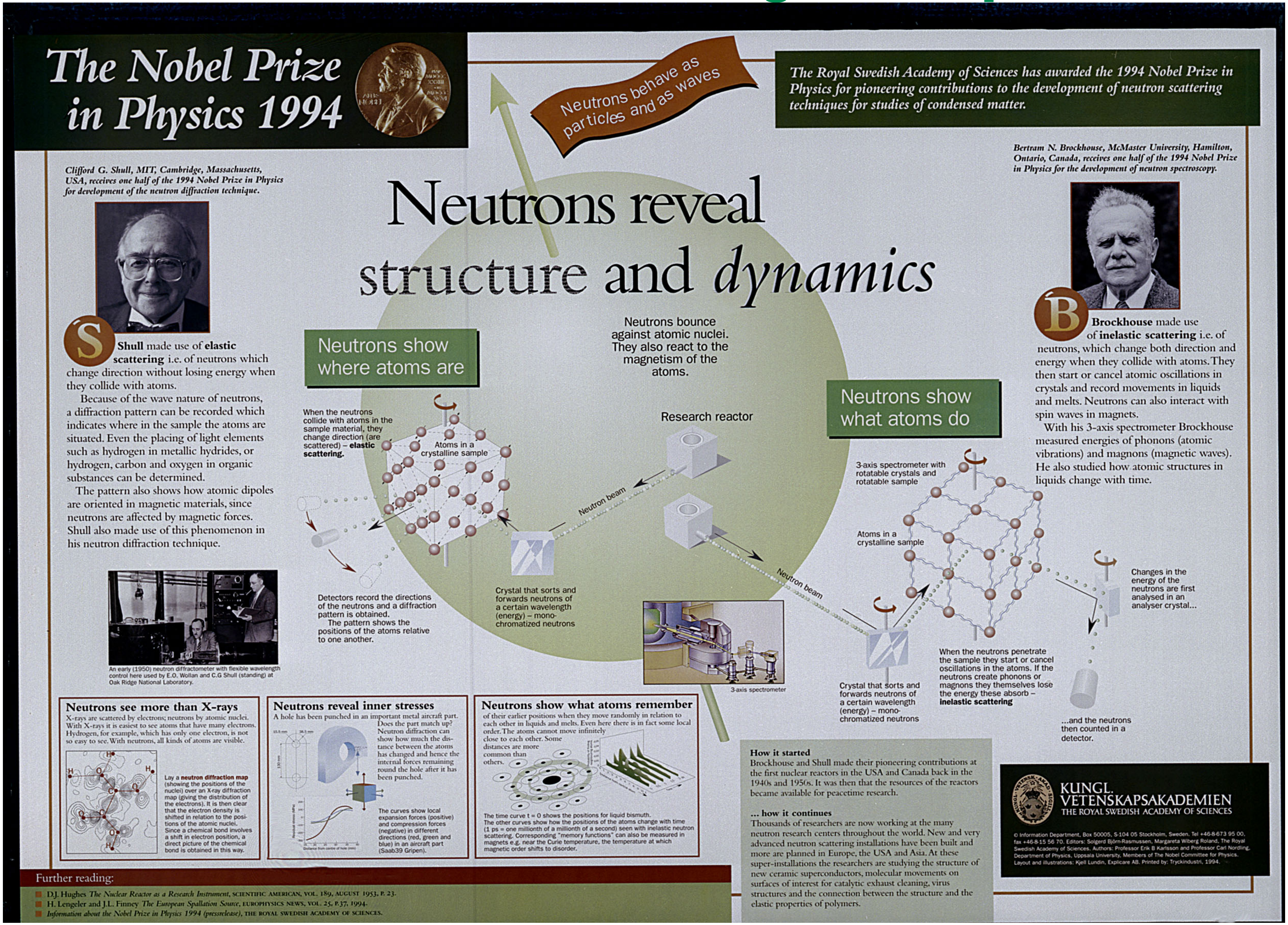
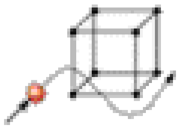
First triple axis spectrometer to study the dynamics (vibrations of the atoms)

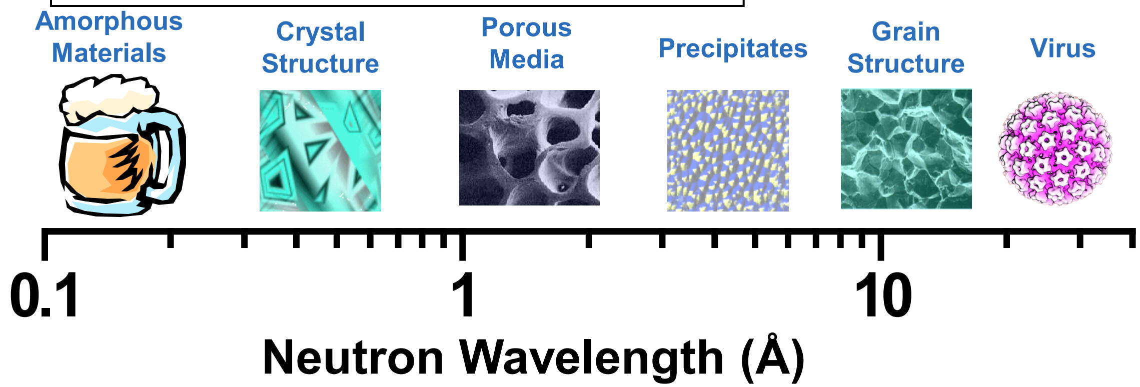
1994 Nobel Prize in Physics for the development of the neutron scattering technique



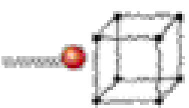
Match the probe with the system

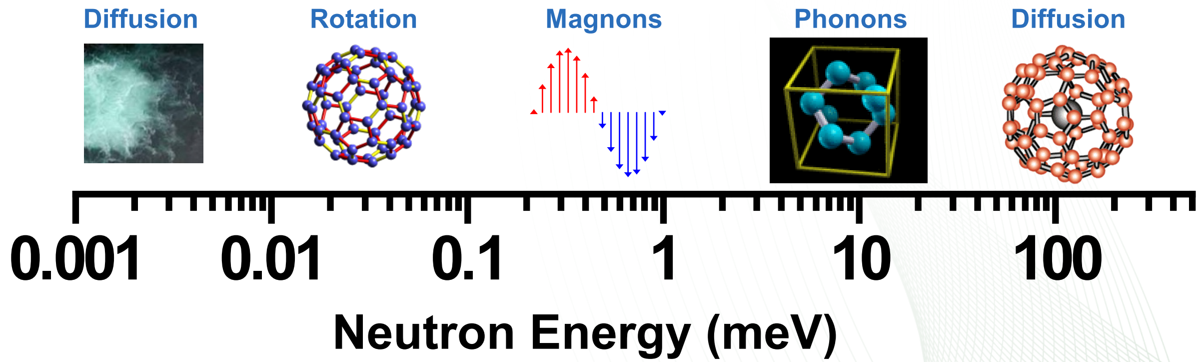
$$q = \frac{4\pi \sin(\theta)}{\lambda} = \frac{2\pi}{d}$$





Neutron **WAVELENGTHS** are similar to atomic scale dimensions





The **ENERGY** of thermal neutrons is on the same order as many atomic scale dynamical processes

Science with Neutrons

- **Neutrons show where atoms are**
- Neutrons show where the nuclei and the electron spins are:
  - Ordering of the nuclei
  - Ordering of the electron spins
- **Neutrons show what atoms do**
- Neutrons show what the nuclei and the electron spins do:
  - Lattice vibrations-phonons
  - Spin excitations-magnons, etc

Science with Neutrons

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  - Ordering of the electron spins

Elastic scattering -- Diffraction

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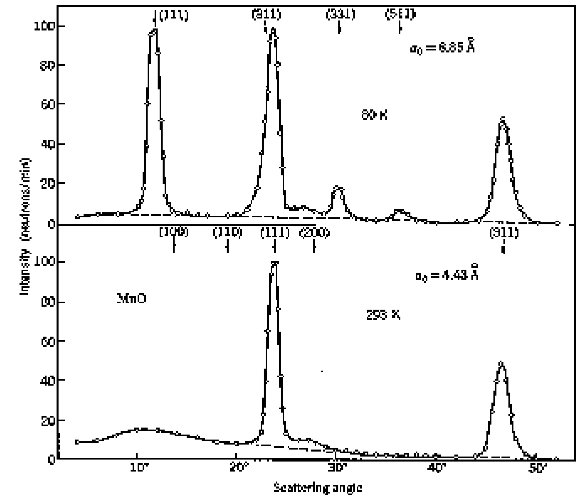

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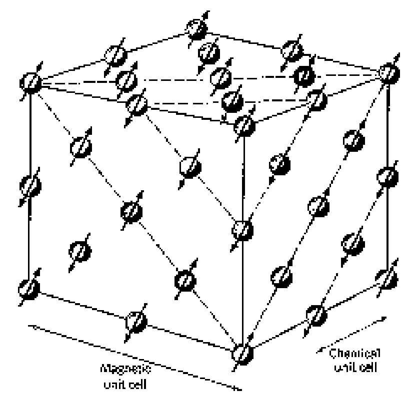

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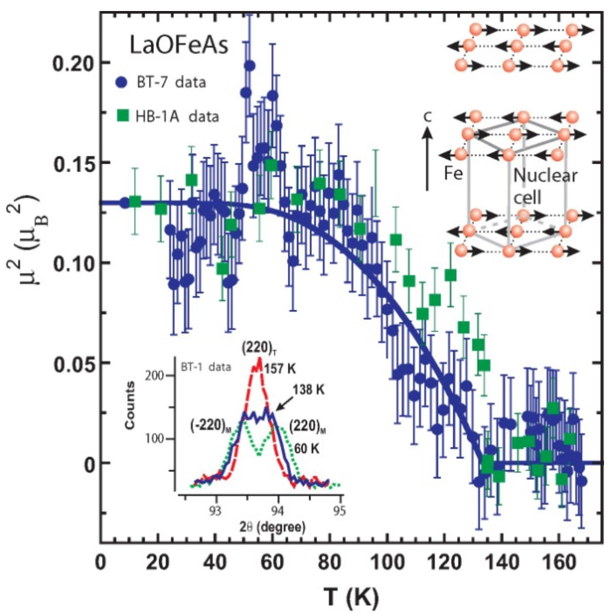
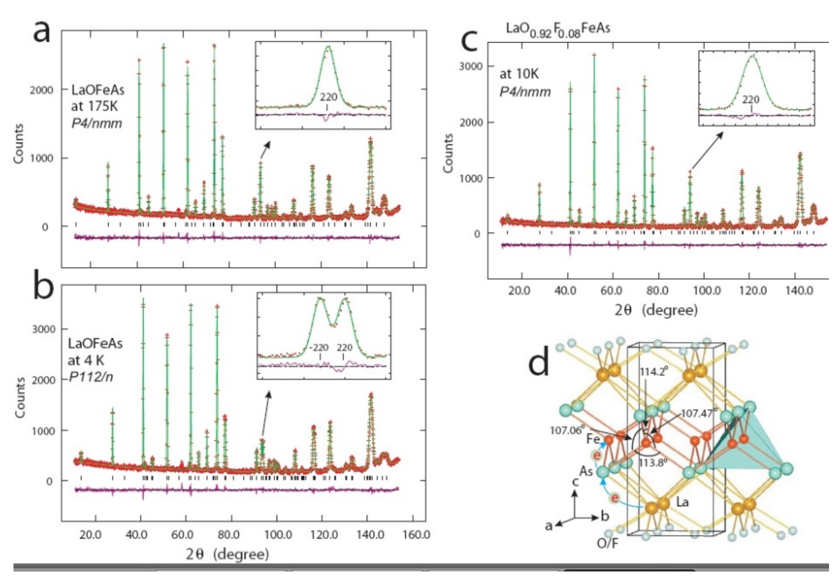
doi:10.1038/nature07057

LETTERS

Magnetic order close to superconductivity in the iron-based layered $LaO_{1-x}F_xFeAs$ systems

Clarina de la Cruz[1,2], Q. Huang[3], J. W. Lynn[3], Jiying Li[3,4], W. Ratcliff II[3], J. L. Zarestky[5], H. A. Mook[2], G. F. Chen[6], J. L. Luo[6], N. L. Wang[6] & Pengcheng Dai[1,2]





Structural and magnetic phase transitions in $Na_{1-\delta}FeAs$

Shiliang Li,[1,2] Clarina de la Cruz,[2,3] Q. Huang,[4] G. F. Chen,[5,1] T.-L. Xia,[5] J. L. Luo,[1] N. L. Wang,[1] and Pengcheng Dai[2,3,1]
[1]Institute of Physics, China; [2]The University of Tennessee; [3]Oak Ridge National Laboratory; [4]NIST Center for Neutron Research; [5]Remin University, China

We use neutron scattering to study the spin and lattice structures of single crystal and powder samples of $Na_{1-\delta}FeAs$ (T_c=23 K). Upon cooling from room temperature, the system goes through a series of phase transitions: first changing the crystal symmetry from tetragonal to orthorhombic at 49 K, then ordering antiferromagnetically with a spin structure similar to that of LaFeAsO and a small moment ($0.09\pm0.04\mu_B$), and finally becoming superconducting below about 23 K. These results confirm that antiferromagnetic order is ubiquitous for the parent compounds of the iron arsenide superconductors and suggest that the separated structural and magnetic phase-transition temperatures are due to the reduction in the c-axis exchange coupling of the system.

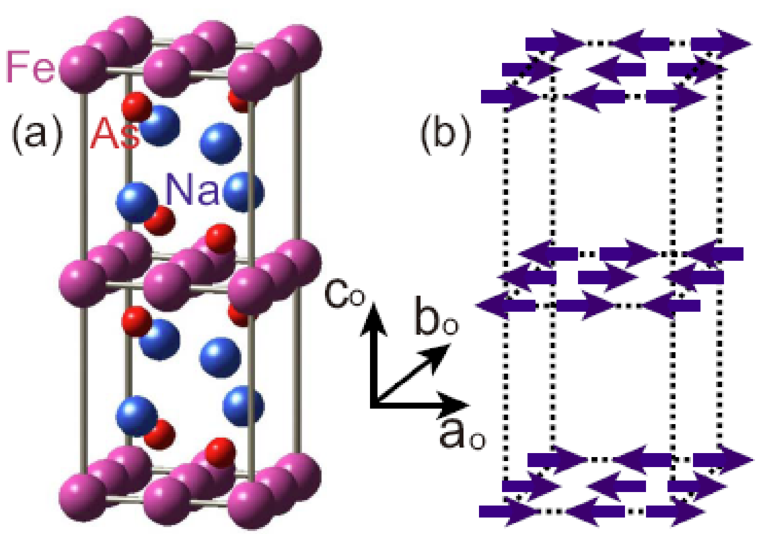


FIG. 1. (a)Nuclear and (b) magnetic structures of the ideal NaFeAs. (a) includes two orthorhombic nuclear unit cells for comparison with the magnetic unit cell in (b).

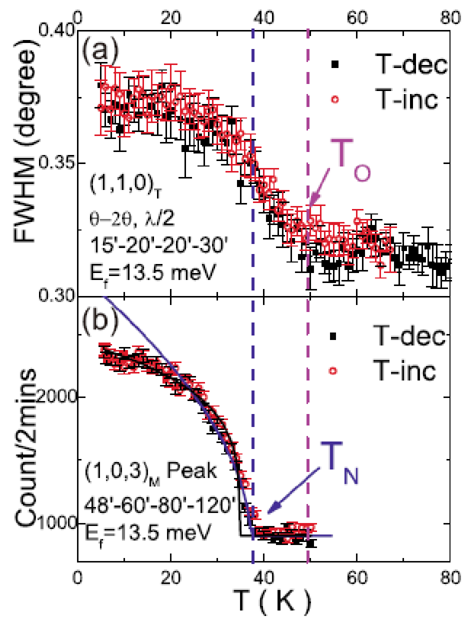


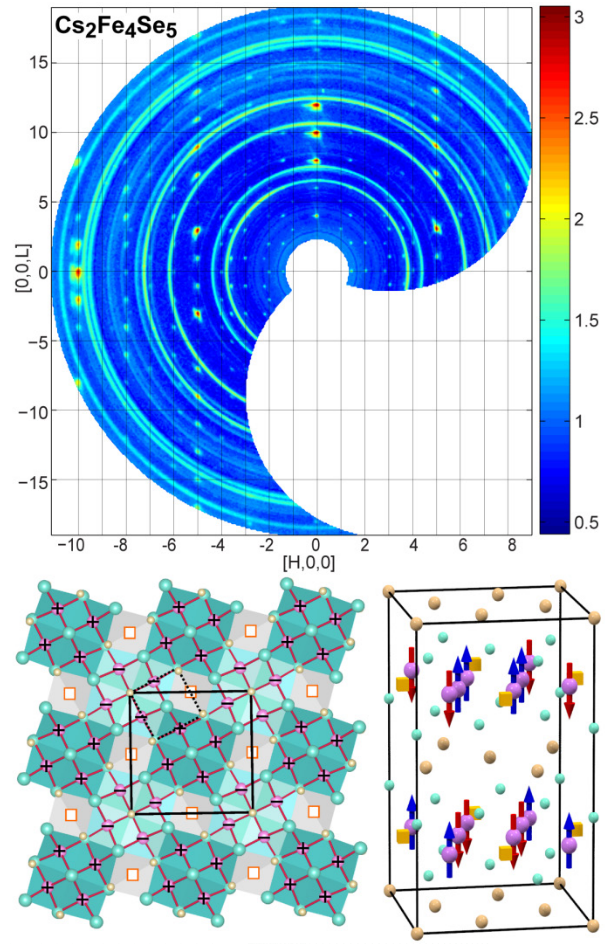
FIG. 2. (a) T dependence of the FWHM of θ-2θ scan at the nuclear peak $(1,1,0)_T$ position using λ/2 scattering by removing the PG filter. (b) T dependence of the peak intensity at the AF peak $(1,0,3)_M$ position suggesting a Néel temperature of 39 K. Data from HB-1 at HFIR.

Neutron scattering identifies new family of Fe-based superconductors with Tc above 30 K

- **A new family of Fe-based superconductors, $A_xFe_{2-y}Se_2$ [A: Rb, Cs, (TiRb) and (Ti, K)], has been discovered with a Tc above 30 K.**
- **Single-crystal neutron diffraction experiments on the Wide-Angle Neutron Diffractometer and the Four-Circle Diffractometer at HFIR revealed common crystalline and magnetic structures.**
- **The crystalline structure forms an expanded $\sqrt{5} \times \sqrt{5} \times 1$ super-cell with the Fe_1 site almost empty and the Fe_2 site fully occupied because of ~20% Fe vacancies.**
- **The magnetic moments carried by Fe ions develop a block checkerboard antiferromagnetic order at around 470–570 K.**
- **The large-moment antiferromagnetic orders and the strong interplay between superconducting and magnetic orders differ distinctly from the dynamics of the Fe square-lattice-based families of iron superconductors previously discovered.**

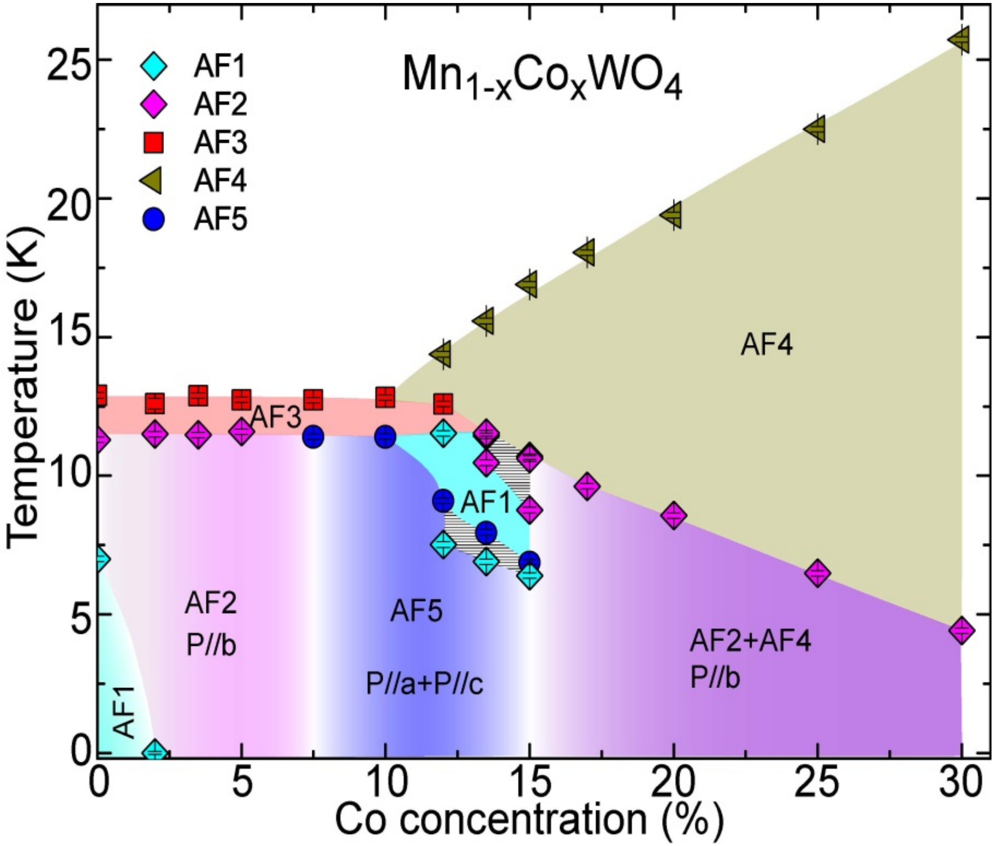
"Common crystalline and magnetic structure of superconducting $A_2Fe_4Se_5$ (A = K, Rb, Cs, Ti) single crystals measured using neutron diffraction," F. Ye, et al. S. Chi, W. Bao, X. F. Wang, J. J. Ying, X. H. Chen, H. D. Wang, C. H. Dong, and M. Fang, ***Phys. Rev. Lett.* 107, 137003 (2011)**.

Neutron experiments: HB-1A, HB-2C (WAND)

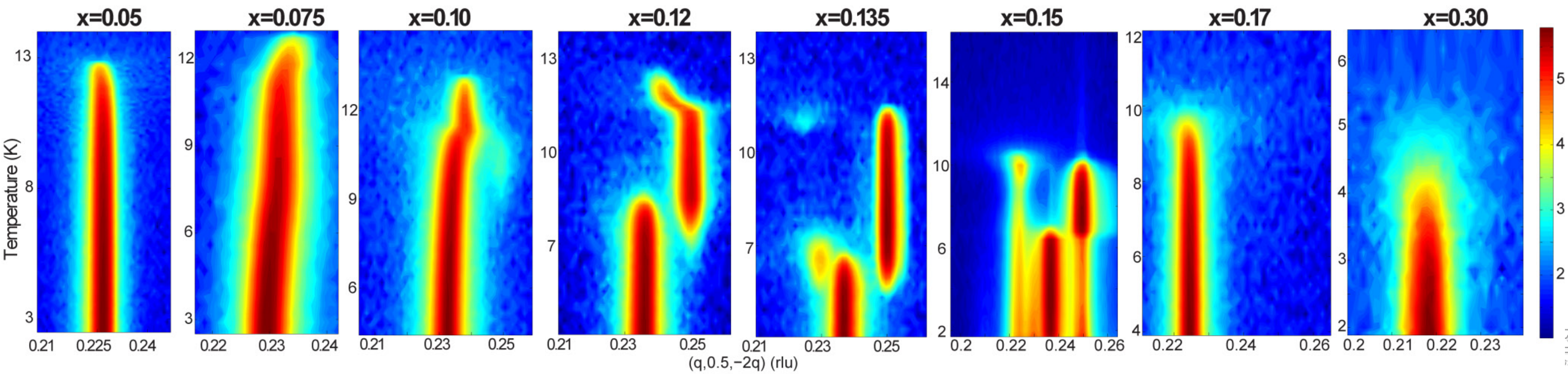


Top: Single-crystal neutron diffraction pattern on $Cs_2Fe_4Se_5$. The weak peaks indicate the formation of a super-cell structure below the structural and magnetic transition temperatures. Bottom: Crystalline (left) and magnetic (right) structures of superconducting $A_2Fe_4Se_5$ derived from neutron diffraction measurements.

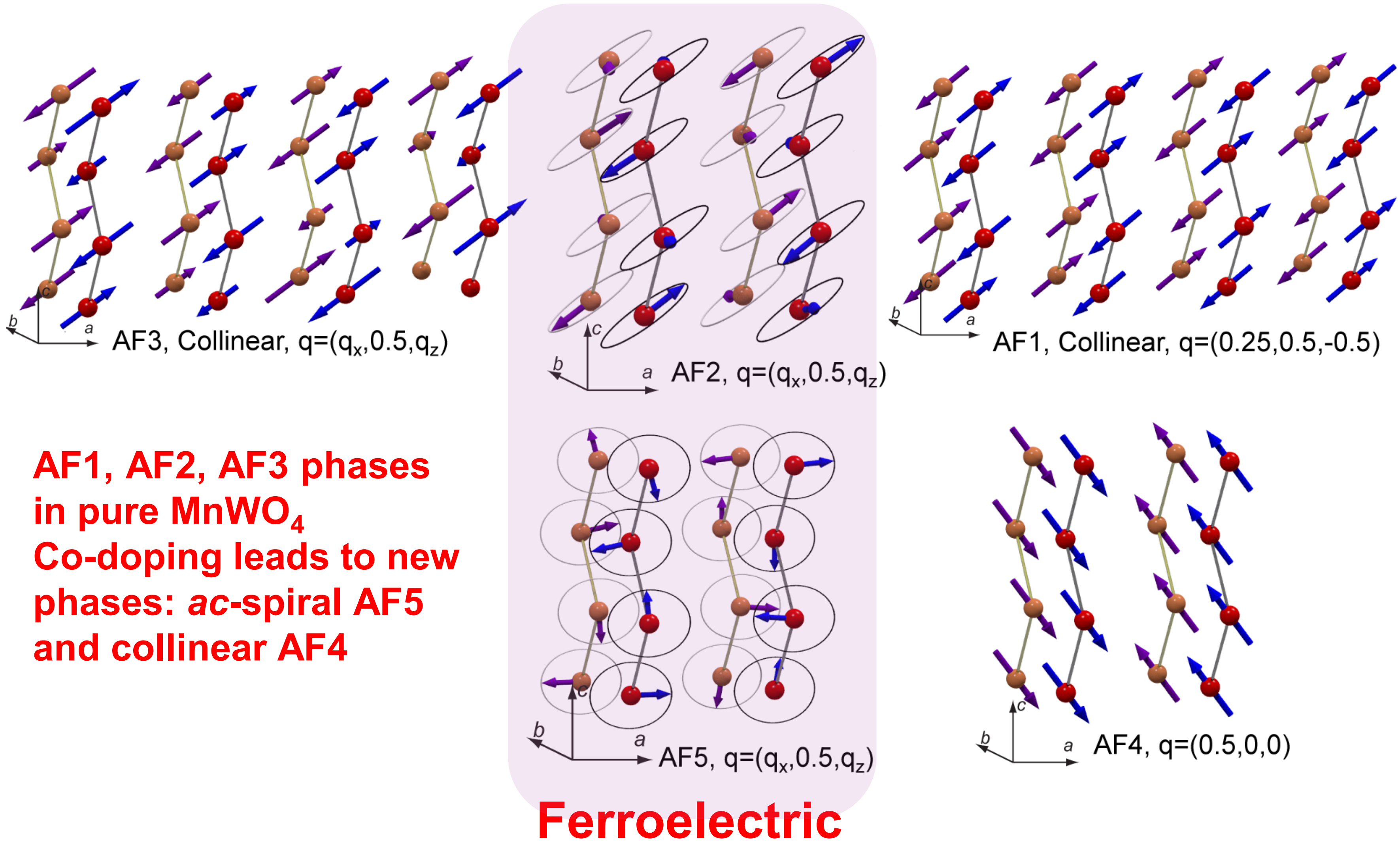
$MnWO_4$: Co substitutions lead to rich phase diagrams



Ye et al.
Phys. Rev. B 86, 094429 (2012)



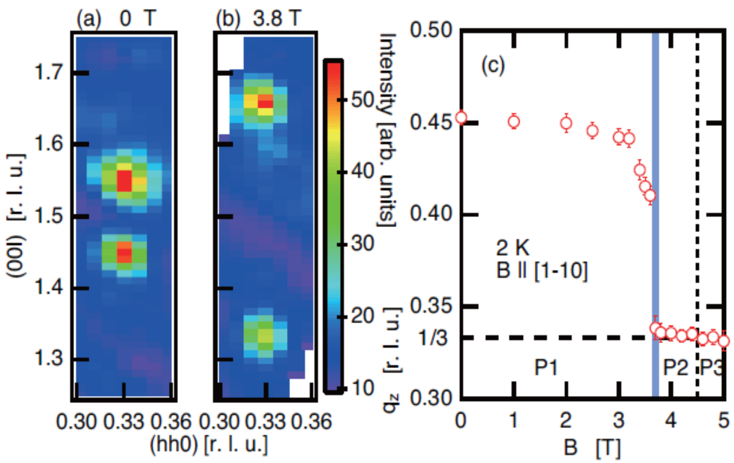
Various competing magnetic orders in doped $MnWO_4$

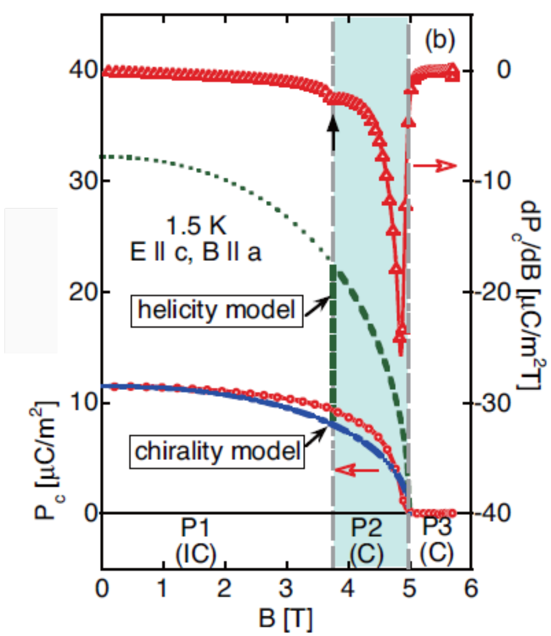


- **AF1, AF2, AF3 phases in pure $MnWO_4$**
- **Co-doping leads to new phases: *ac*-spiral AF5 and collinear AF4**

Ye et al. Phys. Rev. B 86, 094429 (2012)

Spin-Chirality-Driven Ferroelectricity in a Triangular Lattice Antiferromagnet



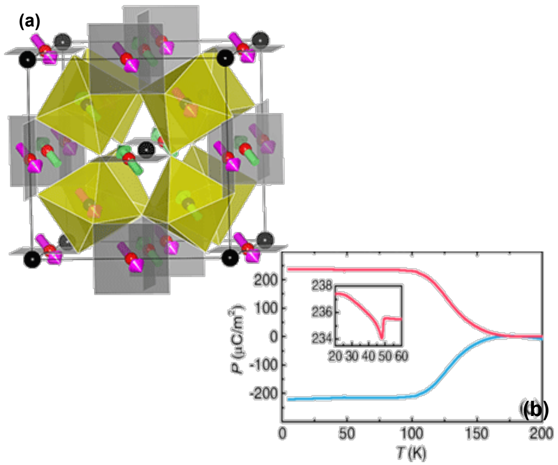


- $RbFe(MoO_4)_2$ is a triangular lattice AF multiferroic below T_N=3.9K.
- The $Fe3^+$ ions form a 120° spin structure for which none of the existing microscopic models predict electric polarization
- Neutron scattering measurements reveal that when a magnetic field is applied along the c-axis there is a change in the propagation wavevector of the helicity at 3.7T from a incommensurate to a commensurate value (see top figure)
- Surprisingly at this magnetic field there is no change in the electric polarization (red open circles in bottom figure) like it would be expected in a spin current model (broken green line).
- This result suggest a new mechanism for multiferroicity in which the electric polarization is induced by spin chirality (blue line).

H. Mitamura, R. Watanuki, K. Kaneko, N. Onozaki, Y. Amou, S. Kittaka, R. Kobayashi, Y. Shimura, I. Yamamoto, K. Suzuki, S. Chi, and T. Sakakibara, "Spin-Chirality-Driven Ferroelectricity on a Perfect Triangular Lattice Antiferromagnet". *Phys. Rev. Lett* **113,**147202 (2014)

Research performed at the HFIR WAND under the US-Japan Cooperative Program in neutron Scattering

New multiferroic material with a surprisingly highly symmetric crystal structure



Neutron diffraction reveals cubic nuclear structure of $LaMn_3Cr_4O_{12}$ hosting the G-type AFM structure of the Cr and Mn sublattice with spin orientation along the [111] direction that lacks an inversion center. The magnetism occurs concurrently with the onset of ferroelectric polarization and is enhanced by the application of a magnetic field.

Work was performed at the ORNL High Flux Isotope Reactor's HB-2A high resolution powder diffraction instrument, which is a DOE Office of Science user facility.

Scientific Achievement

$LaMn_3Cr_4O_{12}$ is a new multiferroic material that has a cubic structure that presents a new mechanism of multiferroicity.

Significance and Impact

Multiferroics are materials with correlated magnetic and electric subsystems, making them viable for spintronic and other magnetoelectric applications. Cubic crystals have an inversion center, so they are not expected to be multiferroic. In $LaMn_3Cr_4O_{12}$, the magnetization within the cubic material forms a spin pattern that is asymmetric. Thus, a unique mechanism via spin-orbit coupling between the two independent magnetic sites can induce ferroelectricity.

Research Details

– Neutron powder diffraction determine the temperature dependent crystal and magnetic structure.

X. Wang, Y. Chai, L. Zhou, H. Cao, C. dela Cruz, J. Yang, J. Dai, Y. Yin, Z. Yuan, S. Zhang, R. Yu, M. Azuma, Y. Shimakawa, H. Zhang, S. Dong, Y. Sun, C. Jin, and Y. Long. *Physical Review Letters*. 115 (2015): 087601.

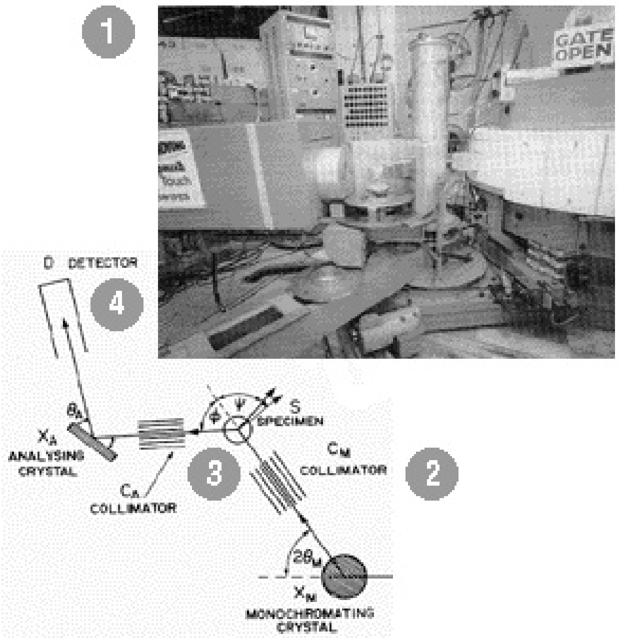
Science with Neutrons

- **Neutrons show what atoms do**
- Neutrons show what the nuclei and the electron spins do:
  - Lattice vibrations-phonons
  - Spin excitations-magnons, etc

Inelastic scattering

The beginnings of neutron scattering

Bertram Brockhouse, Chalk River, Canada, 1950's



First triple axis spectrometer to study the dynamics (vibrations of the atoms)

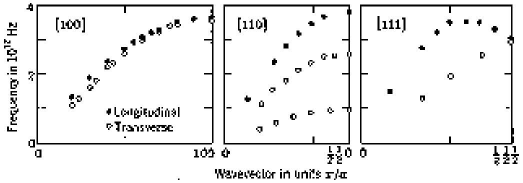
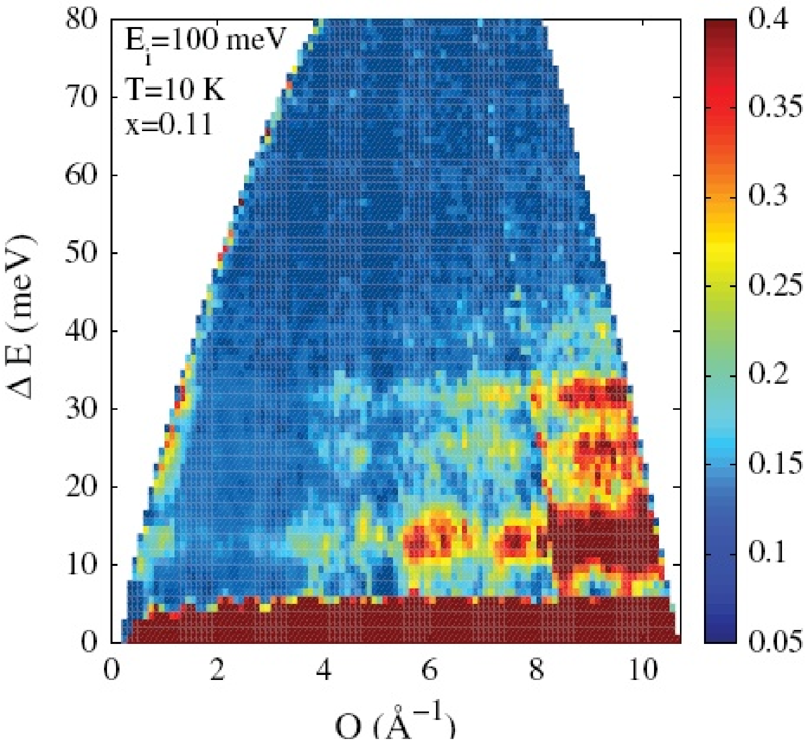
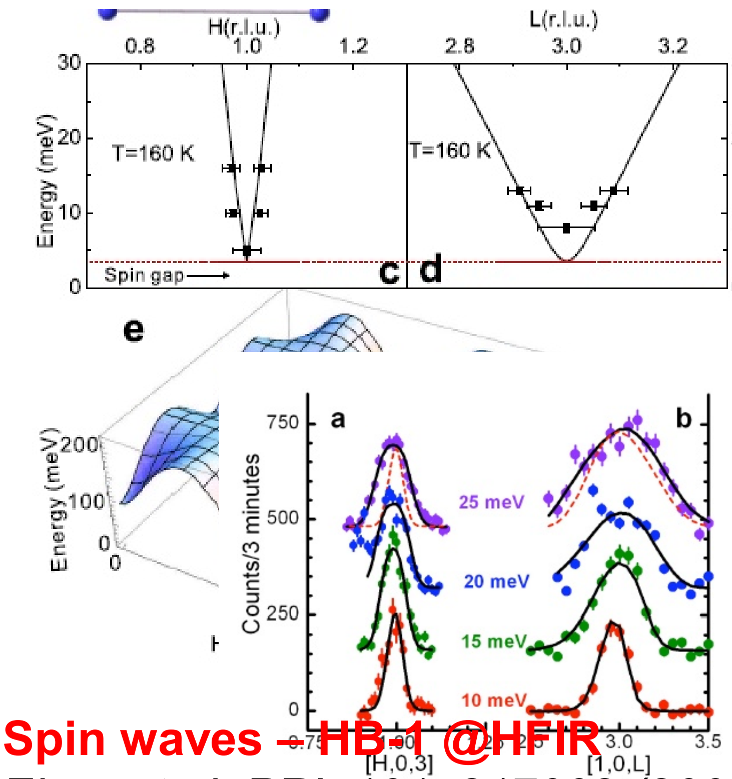


Figure 11 The dispersion curves of sodium for phonons propagating in the [001], [110], and [111] directions at 90 K, as determined by inelastic scattering of neutrons. [Woods, Brockhouse, March and Bowers, Proc. Phys. Soc. London 79, pt. 2, 440 (1962).]

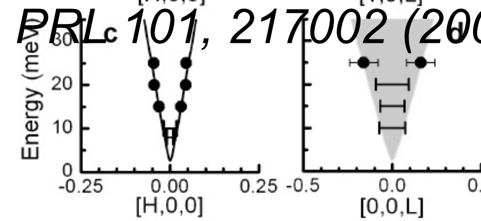
Complementarity of SNS and HFIR: Iron arsenide superconductors





Spin waves – HB-1 @HFIR

Zhao et al. PRL 101, 217002 (2008)



Phonon DOS - ARCS @SNS

Christianson et al.,

PRL 101, 157004 (2008)

Spin waves – HB-3 @HFIR

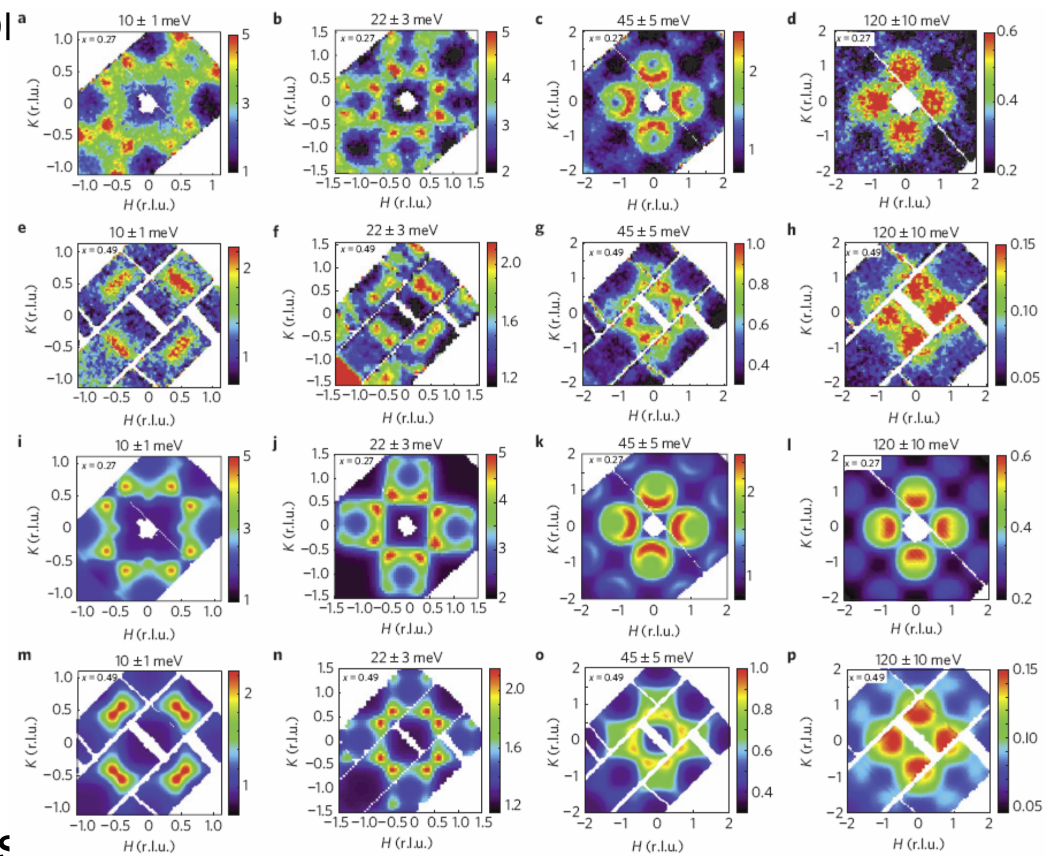
McQueeney et al. PRL 101, 227205 (2008)

Oak Ridge National Laboratory | HIGH FLUX ISOTOPE REACTOR | SPALLATION NEUTRON SOURCE

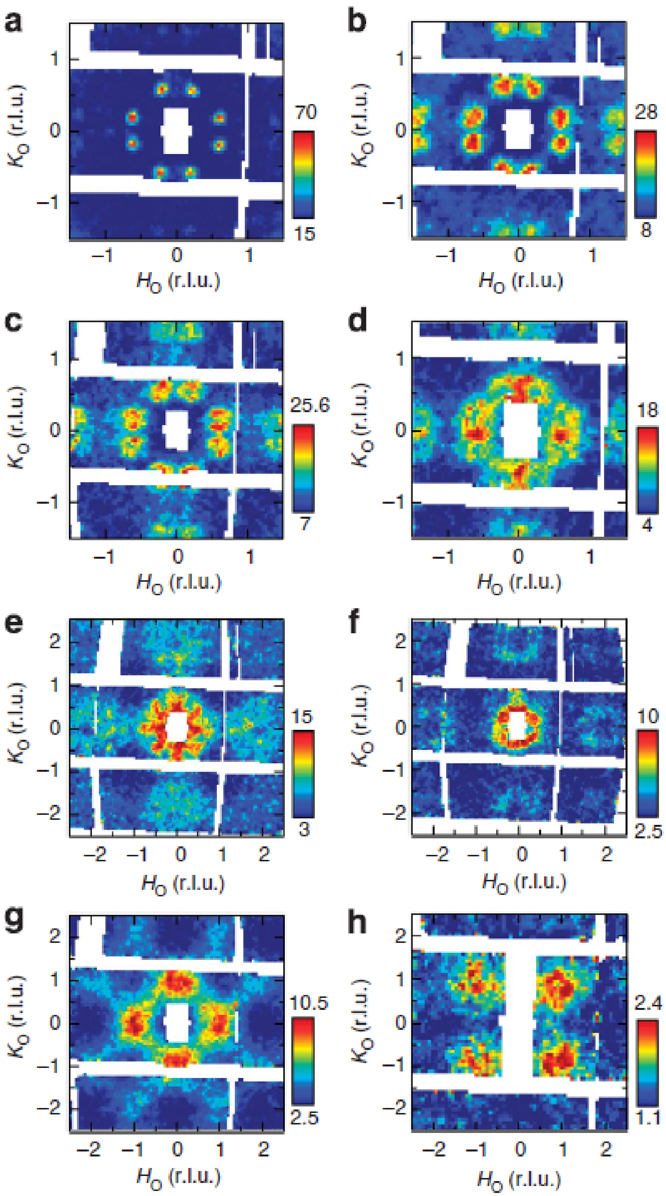
Evolution of spin excitations into the superconducting state in $FeTe_{1-x}Se_x$

M.D. Lumsden, A.D. Christianson *et al.*, Nature Physics: doi:10.1038/nphys1512

- Inelastic neutron scattering studies of the magnetic excitation spectrum in crystals of $Fe_{1+y}Te_{1-x}Se_x$ with x=0.27 (not superconducting) and x=0.49 (superconducting).
- Measurements indicate 2d incommensurate magnetic excitations extending up to high energies (> 250 meV).
- Excitations are 4-fold symmetric about the square lattice (π, π) point and are characterized by the same wavevector as the spectrum of the high-T_C cuprates showing strong similarities in the magnetism of the 2 families of high-T_C superconductors.



Spin waves in insulating Rb0.89Fe1.58Se2



Spin waves and magnetic exchange interactions in insulating $Rb_{0.89}Fe_{1.58}Se_2$

Miaoyin Wang, Chen Fang, Dao-Xin Yao, GuoTai Tan, Leland W. Harriger, Yu Song, Tucker Netherton, Chenglin Zhang, Meng Wang, Matthew B. Stone, Wei Tian, Jiangping Hu, and Pengcheng Dai

Nature Communications 2, 580 (2011).

- $A_yFe_{1.6+x}Se_2$ (A = K, Rb, Cs) superconductors are isostructural with iron pnictides. However, the undoped members are insulators, forming a block AFM order with a Néel temperature of ~500 K.
- the spin waves of the insulating antiferromagnet $Rb_{0.89}Fe_{1.58}Se_2$ can be accurately described by a local moment Heisenberg Hamiltonian
- Fitting reveals a next-near-neighbor interaction which is similar to $(Ba,Ca,Sr)Fe_2As_2$ and $Fe_{1.05}Te$

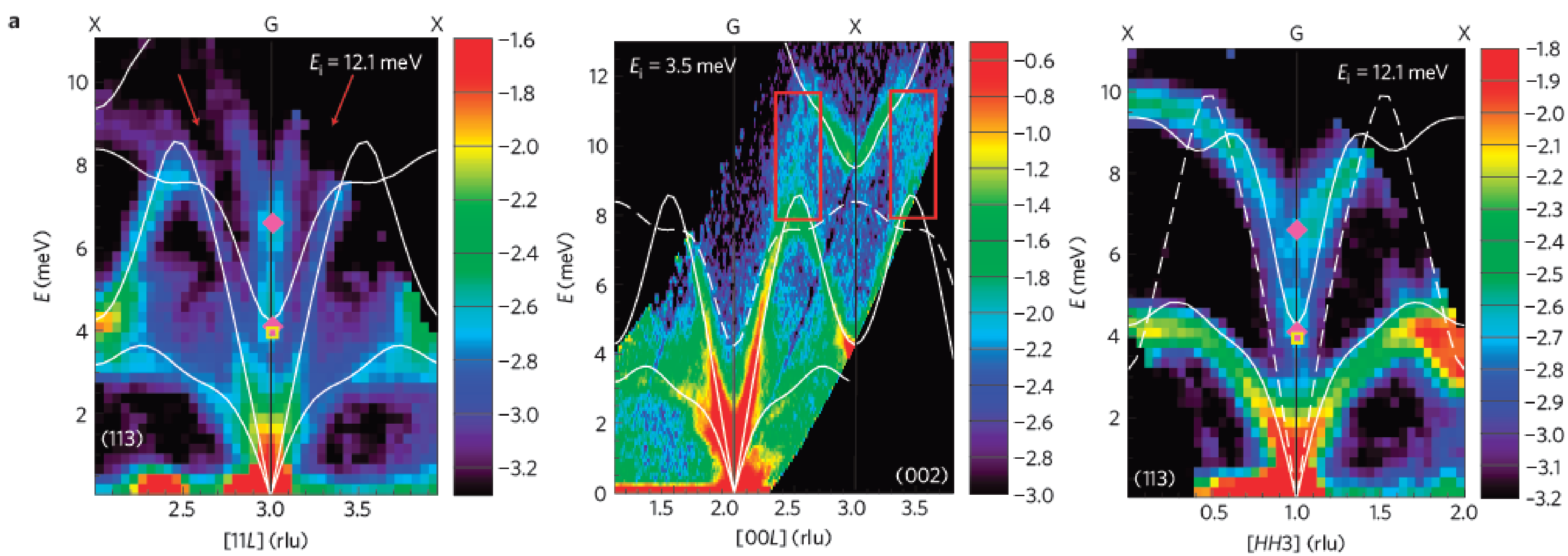
Giant Anharmonic Effects in Thermoelectrics

Giant anharmonic phonon scattering in PbTe

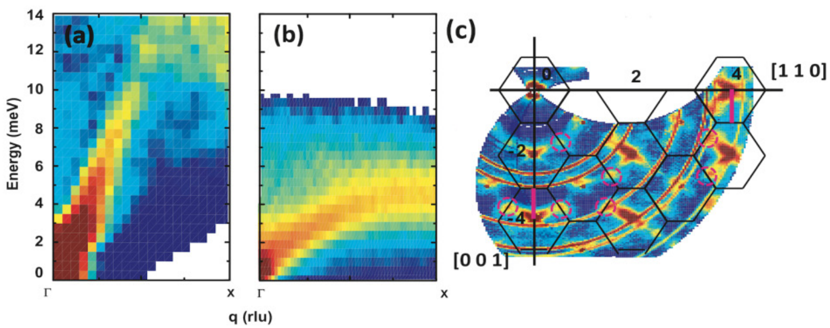
O. Delaire, J. Ma, K. Marty, A. F. May, M. A. McGuire, M-H. Du, D. J. Singh, A. Podlesnyak, G. Ehlers, M. D. Lumsden and B. C. Sales

Nature Materials **10**, 614 (2011).

- PbTe is one of leading thermoelectric materials due to low thermal conductivity.
- Combination of INS and first-principle calculations of phonons strong anharmonic coupling between the ferroelectric TO mode and the LA modes.
- This coupling is likely to play a central role in explaining the low thermal conductivity.



Nanoscale Insights into Glass-Like Thermal Transport Offer a Breakthrough in Thermoelectric Efficiency



Phonon energies vs. phonon momentum transfer. The broadening of the phonon modes indicates large phonon scattering rates and low thermal conductivity (a, b). Map of phonon scattering across 3 nm domains in $AgSbTe_2$ from ARCS measurements (c).

J. Ma, O. Delaire, A. F. May, C. E. Carlton, M. A. McGuire, L. H. VanBebber, D. L. Abernathy, G. Ehlers, T. Hong, A. Huq, W. Tian, V. M. Keppens, Y. Shao-Horn, and B. C. Sales, *Nature Nanotechnology* **8** ,445 (2013). A highlight of this paper appears in Austin J. Minnich, “Naturally glassy crystals,” *Nature Nanotechnology* **8**, 392-393 (2013). doi:10.1038/nnano.2013.106

Work was performed at the SNS CNCS, POWGEN, and ARCS instruments and at HFIR’s HB-1A, HB-3, and CTAX triple-axis spectrometers.

Scientific Achievement

The origin of the extremely low glass-like thermal conductivity in $AgSbTe_2$ was elucidated using neutron scattering and electron microscopy. Neutron scattering is the optimal technique to systematically probe phonon dispersions and line widths throughout reciprocal space.

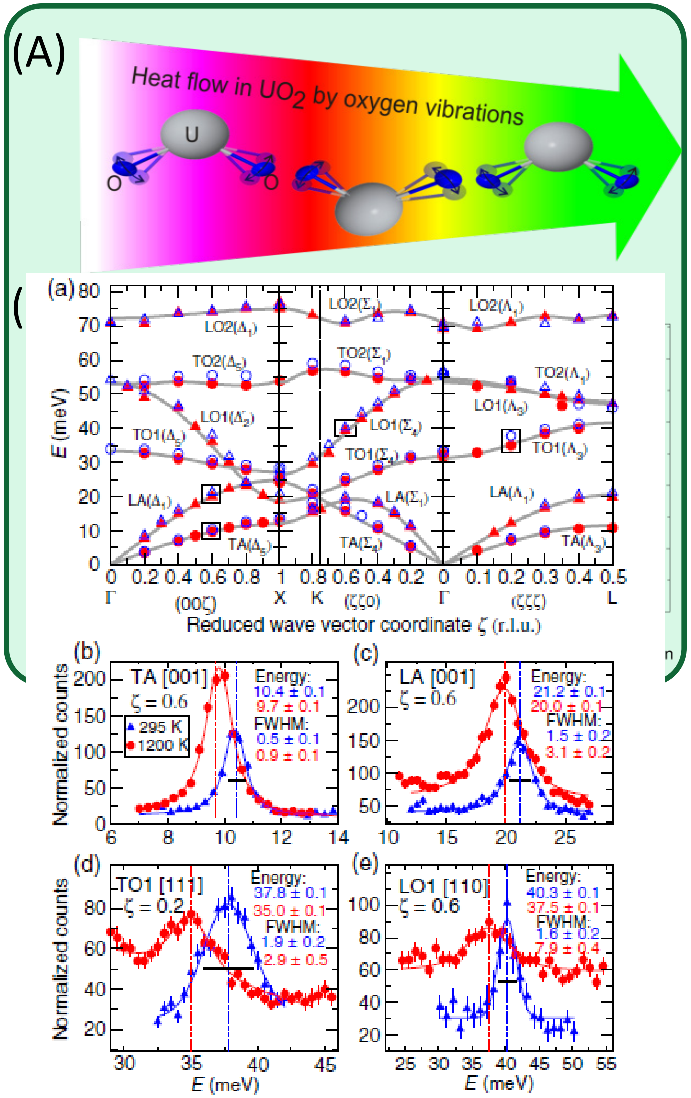
Significance and Impact

This work identifies a new mechanism by which the thermal conductivity of crystals can be manipulated and could prove useful in applications such as thermoelectric energy conversion and phase-change memory devices.

Research Details

- $AgSbTe_2$’s ground-state degeneracy leads to forming of nanoscale domains that spontaneously scatter phonons, indicating a new avenue to nanoscale engineering.
- Transmission Electron Microscopy (TEM) and diffraction show that nanoscale domains are present.
- Inelastic neutron scattering shows large-phonon line widths, indicating scattering of phonon domain walls, leading to low thermal conductivity.

Neutrons Find Good Vibrations in Oxide Nuclear Fuel



Scientific Achievement

Neutron scattering measurements demonstrated the surprising result that highly anharmonic oxygen vibrations and low velocity uranium vibrations transport the largest amount of heat in uranium dioxide (UO_2).

Significance and Impact

First measurements of atomic-scale thermal transport for the strongly correlated nuclear fuel UO_2 reveal gaps in present simulations and help quantify interatomic forces in this system.

Research Details

- Existing theoretical analyses had concluded that 90% of the thermal transport in UO_2 is by the high-velocity longitudinal acoustic (LA) phonons dominated by uranium atom vibrations.
- Neutron scattering measurements have shown, conversely, that the pure oxygen longitudinal optical (LO1) phonons, and the transverse acoustic (TA) phonons, are strong heat carriers as well.
- New first-principles simulations correctly predict the relative importance of the TA and LO1 phonon modes, but anharmonic phonon-phonon scattering magnitudes are off by a factor of two.

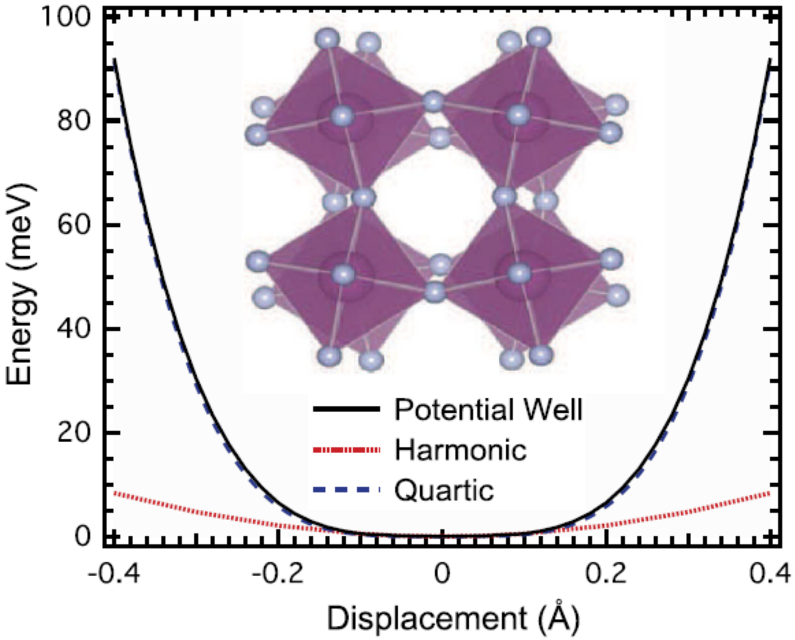
J. W. L. Pang, W. J. L. Buyers, A. Chernatynskiy, M. D. Lumsden, B. C. Larson, and S. R. Phillpot, ***Phys Rev Lett* 2013 *110*, 157401.**

Center for Materials Science of Nuclear Fuel

Structural Relationship between Negative Thermal Expansion and Quartic Anharmonicity of Cubic ScF_3

Chen W. Li, Xiaoli Tang, J. A. Muñoz, J. B. Keith, S. J. Tracy, D. L. Abernathy, and B. Fultz

Phys. Rev. Lett. 107, 195504 (2011).



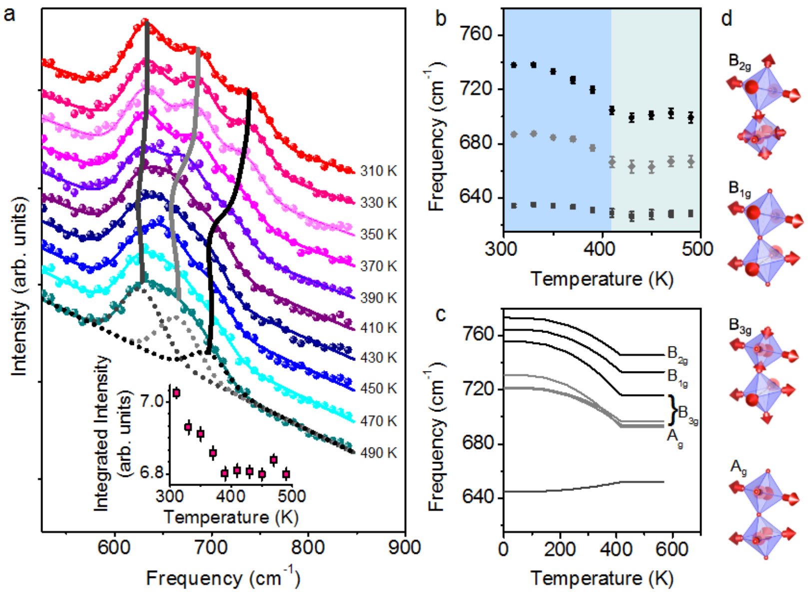
- Phonon density of states measurements combined with first principles calculations indicate that some of the modes act as quartic oscillators

- Quartic potential accounts for phonon stiffening with temperature and a significant part of the negative thermal expansion

“**News and Views: A fresh twist on shrinking materials**”, J. Paul Attfield, Nature **480**, 465 (2011)

“**Focus: New Vibration in Material That Shrinks When Heated**”, Michael Schirber, Physics **4**, 90 (2011)

5*d* ions revealed as key ingredient in creating record spin-phonon coupling



a-b Inelastic neutron scattering reveals a record phonon shift of 40 cm^{-1} coupled to the magnetic and metal-insulator transition in $NaOsO_3$. **c** Density functional theory calculations reproduce the results and reveal the modes responsible as all involving Os-O distortions, shown in **d**. The intrinsically extended Os-O orbital overlap allows magnetism to couple to the modes on an unprecedented scale and results in the record phonon shift.

The research conducted at ORNL's Spallation Neutron Source was sponsored by the Scientific User Facilities Division, Office of Basic Energy Sciences, US Department of Energy. Part of the research at ORNL was sponsored by the Scientific User Facilities Division and Materials Sciences and Engineering Division. Experiments were performed at the ORNL Spallation Neutron Source's ARCS, SEQUOIA and NOMAD instruments.

Scientific Achievement

$NaOsO_3$ found to host a spin-phonon transition with the largest ever measured phonon shift of 40 cm^{-1}.

Significance and Impact

Enhanced coupling of material properties offers new fundamental insights and routes to multifunctional devices. In this context 5*d* oxides provide new paradigms of cooperative interactions that drive novel emergent behavior. Here we studied $NaOsO_3$ that hosts a rare "Slater" metal-insulator transition driven by magnetic order. Inelastic neutron scattering used to access the collective excitations uncovered the largest ever recorded phonon shift at a spin-phonon transition. In combination with detailed theory the extended Os-O orbital overlap was found to be the mechanism driving the giant coupling. The results offer new routes to design materials with enhanced coupling, with 5*d* ions a key ingredient.

Research Details

– Record phonon shift measured with neutrons.
– Neutrons used to rule out a structural change.
– Theoretical calculations at ORNL uncovered the phonons and mechanism for enhanced magnitude.

Calder, S., J. H. Lee, M. B. Stone, M. D. Lumsden, J. C. Lang, M. Feygenson, Z. Zhao, J.-Q. Yan, Y. G. Shi, Y. S. Sun, Y. Tsujimoto, K. Yamaura, A. D. Christianson, Giant spin-phonon-electronic coupling in a 5d oxide. *Nature Communications* 6, 8916 (2015).

OAK RIDGE National Laboratory | HIGH FLUX ISOTOPE REACTOR | SPALLATION NEUTRON SOURCE

Magnetic ground state of multiferroic $CuFe_{1-x}Ga_xO_2$ revealed by inelastic neutron scattering

The triangle lattice $CuFeO_2$ is of great interest because of the magnetic frustration within each hexagonal plane. A multiferroic phase can be induced either by applying a magnetic field above about 7 T or by doping with nonmagnetic Al or Ga impurities, where the resulting multiferroic ground state has noncollinear spin configuration.

Using inelastic neutron scattering and theoretical modeling, the magnetic ground state of the multiferroic $CuFe_{1-x}Ga_xO_2$ was determined.

The magnetic spins of Fe^{3+} ions in the hexagonal plane form a complex noncollinear spin structure that gives rise to the electric polarization below the magnetic transitions.

The lattice distortion associated with displacement of oxygen atoms plays an important role in determining the magnetic ground state.

This complex ground state provides an alternative way to realize multiferroic coupling that might be found in many other frustrated magnets with rhombohedral or hexagonal symmetries.

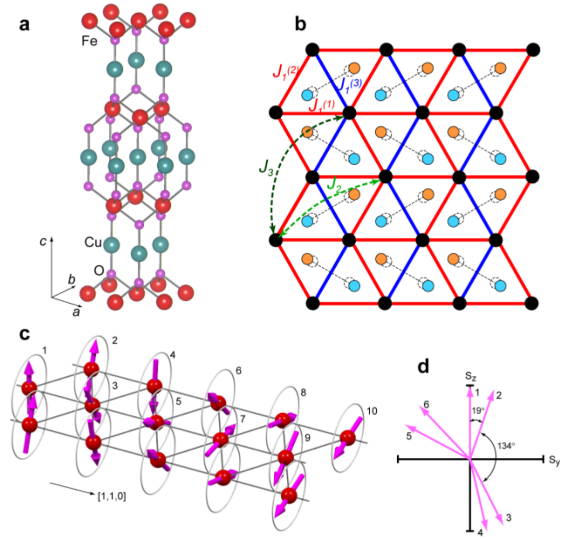


Figure 1.(a) Crystal structure of the triangle lattice $CuFeO_2$ (b) Two-dimensional hexagonal lattice structure of Fe^{3+} layer and distorted oxygen ions. (c) The complex noncollinear spin configuration at low temperature. (d) Projection of spin along [1,1,0] direction.

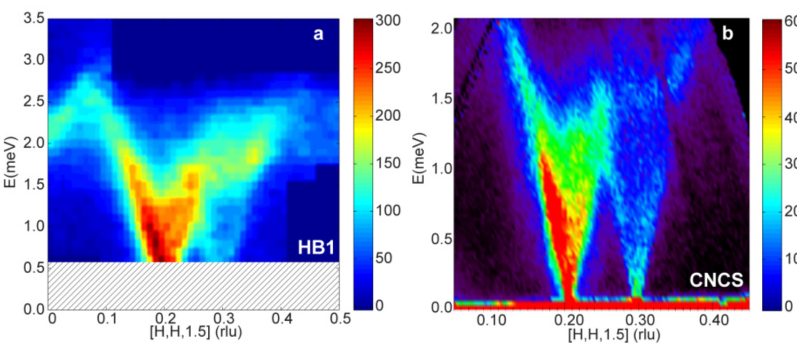
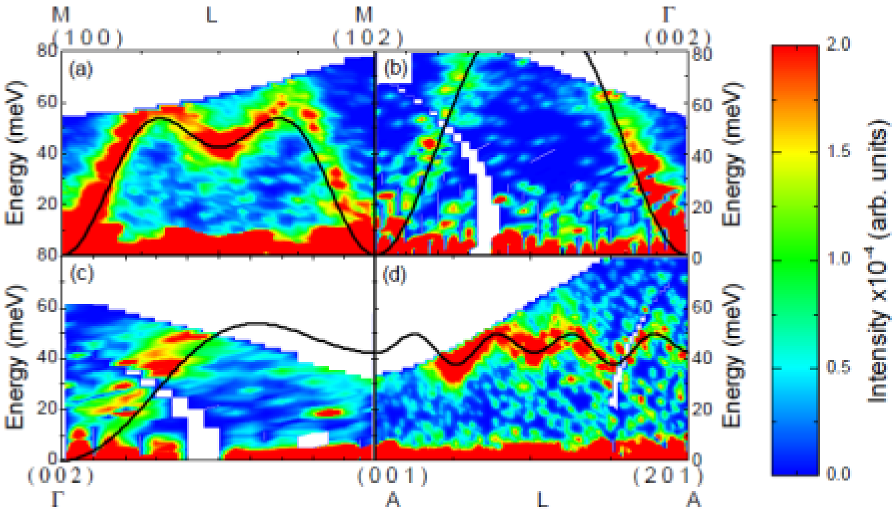


Figure 2. Inelastic neutron scattering spectra of 3.5% Ga-doped $CuFeO_2$ measured at HB1 at the HFIR (a) and at the CNCS at the SNS (b). The data match well with spin dynamics produced by the magnetic configuration shown in Fig. 1.

Contact: Feng Ye, 865-576-0931, yef1@ornl.gov
Funding Sources: DOE Office of Science–Division of Materials Science and Engineering and the Division of Scientific User Facilities, ORNL Laboratory-Directed Research and Development Program
Resources: Cold Neutron Chopper Spectrometer (CNCS) at the Spallation Neutron Source at ORNL, and HB1 Triple-Axis Spectrometer at High Flux Isotope Reactor at ORNL.

Long-range interactions lead to a high-temperature Mn-based ferromagnet



a-d The inelastic neutron scattering measurements on MnBi that were measured on the ARCS spectrometer. The solid line in a fit to the model, which required interactions up to 6th nearest neighbor, a distance of ~7.5 Å.

Table The Curie temperatures and moment sizes of various Mn-based binary ferromagnets are shown, along with the nearest-neighbor (NN) Mn-Mn distance. This suggests increased separation leads to a stronger ferromagnet, consistent with the exchange constants measured.

Compound	T_{Curie} (K)	Moment (μ_B)	NN Mn Dist. (Å)
MnBi	630	3.90	3.055
MnSb	587	3.55	2.895
MnAs	318	3.20	2.852
MnP	292	1.33	2.743

Work performed on the ARCS Spectrometer, BL-18 at ORNL's Spallation Neutron Source, a US DOE Office of Science User Facility.

Scientific Achievement

Measurements on the high-temperature ferromagnet MnBi suggest that long-ranged interactions stabilize the ground state.

Significance and Impact

The high-temperature ferromagnet MnBi has been proposed as a candidate to replace costly rare earth magnets for room temperature applications. Neutron scattering measurements were used to determine the magnetic exchanges in this system, which showed some unexpected trends. Firstly, the fits required exchanges out to 7.5 Å to reproduce the spin wave spectrum, suggesting long-ranged magnetic interactions. Secondly, the nearest-neighbor exchange is antiferromagnetic, and the ferromagnetic ground state is a result of the longer-range terms. This explains the unexpected trend of stronger magnetism in Mn*T* compounds as the Mn atoms are separated.

Research Details

– The spin waves were measured by TOF neutron scattering.

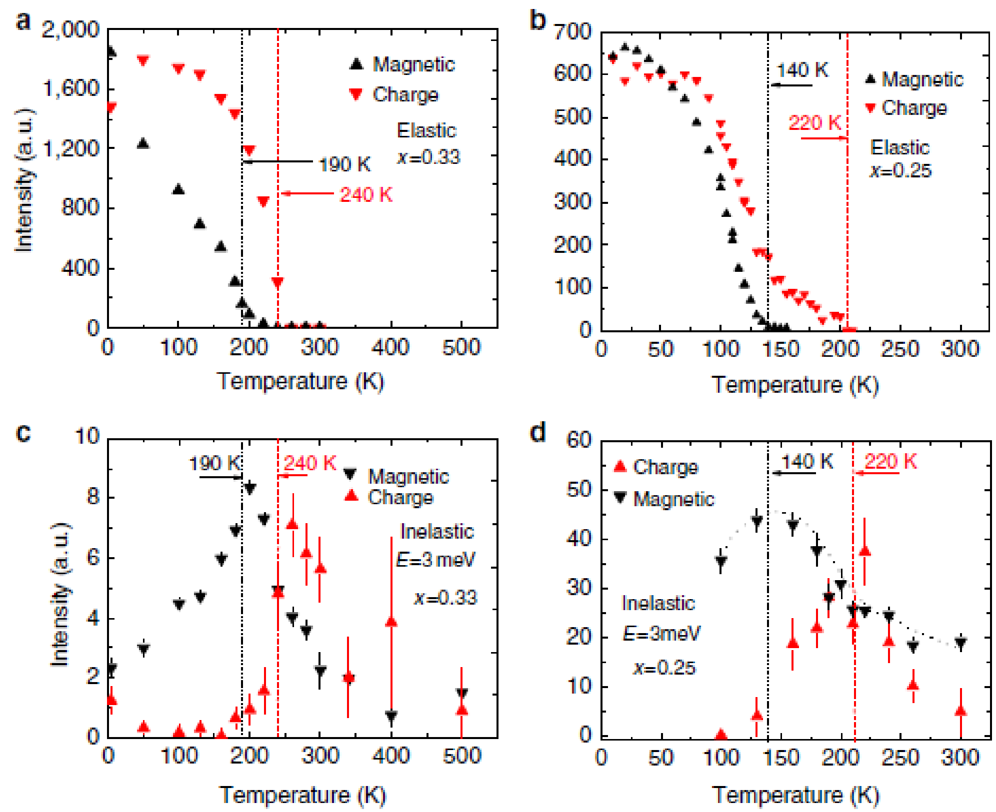
– The exchange parameters were determined by fitting to a Heisenberg model with 6 exchange interactions.

– First principles calculations of the exchange constants were in excellent agreement with the fits.

T.J. Williams, A.E. Taylor, A.D. Christianson, S.E. Hahn, R.S. Fishman, D.S. Parker, M.A. McGuire, B.C. Sales and M.D. Lumsden, Extended Magnetic Exchange Interactions in the High-Temperature Ferromagnet MnBi, *Applied Physics Letters* 108, 192403 (2016).

Direct observation of dynamic charge stripes in $La_{2-x}Sr_xNiO_4$

- In layered compounds the doped charge carriers can segregate into periodically spaced **charge stripes** separating narrow domains of antiferromagnetic order.
- There have been theoretical proposals of dynamically fluctuating stripes but direct spectroscopic evidence of these has been lacking until now.
- This is the first observation of dynamical charge stripes in $La_{2-x}Sr_xNiO_4$. The largest fluctuations are near the charge-stripe melting temperature.



S. Anissimova, D. Parshall, G. Gu, K. Marty, MD Lumsden, Songxue Chi, JA Fernandez-Baca, D. Abernathy, D. Lamago, J. Tranquada, and D. Reznik, "Direct observation of dynamic charge stripes in La2-xSrxNiO4", ***Nature Communications* 5, 3467 (2014)**

What about the future?

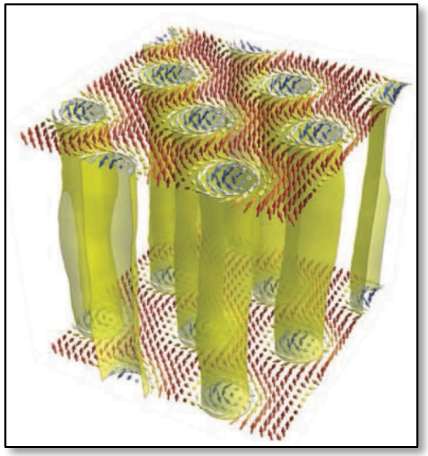
Quantum condensed matter: Moving into the mesoscale

Goal: Understanding materials response on the mesoscale

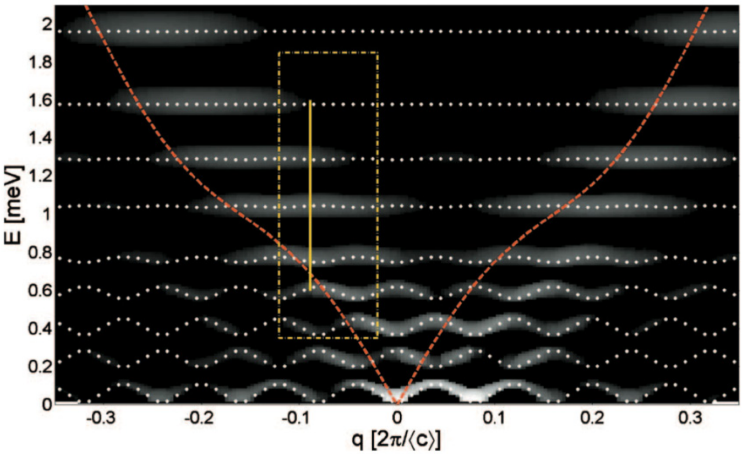
- Topological phases and excitations
- Dynamics in heterostructures/interfaces
- Quantum phases in extreme conditions

Capabilities required

- Higher brightness at long wavelengths
- Access to smaller energy scales (< 1 meV)
- High-field (40 T) and high-pressure (100 GPa) sample environments



Skyrmion lattice
Milde et al., *Science* (2013)



Dy/Y multilayers
Grunwald et al., *Phys. Rev. B* (2010)

OAK RIDGE National Laboratory | HIGH FLUX ISOTOPE REACTOR | SPALLATION NEUTRON SOURCE

Achieving High-Temperature Ferromagnetic Topological Insulators by Proximity Coupling

Scientific Achievement

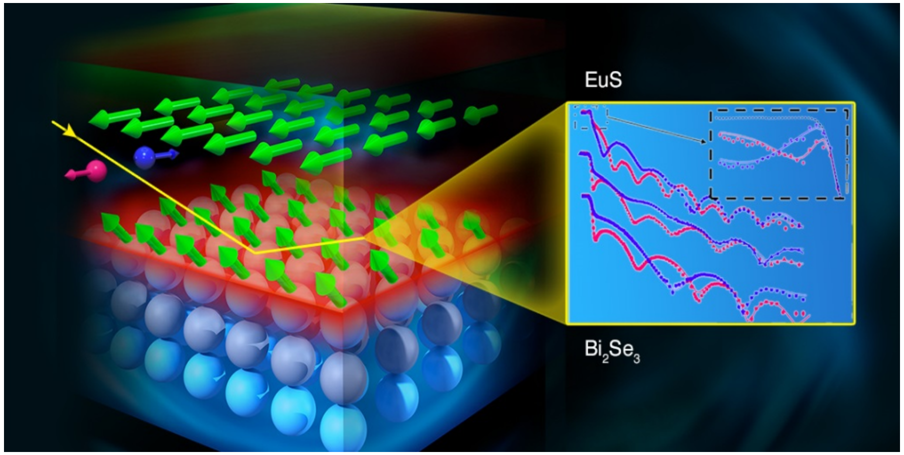
Using polarized neutron reflectometry (PNR), researchers have discovered magnetic moments in hybrid topological insulator (TI) materials at room temperature, hundreds of degrees Fahrenheit warmer than the sub-zero temperature where the properties are expected to occur.

Significance and Impact

TIs are insulating materials *in bulk* and display conducting *surface states* protected by time-reversal symmetry, wherein electron spins are locked to their momentum. Inducing ferromagnetic surface states in TIs are thought to enable the emergence of exotic phenomena such as interfacial magneto-electric coupling, and Majorana fermions. This discovery promises new opportunities for next-generation electronic and spintronic devices such as improved transistors and quantum computing technologies.

Research Details

- Proximity coupling enabled hybrid bilayer heterostructures of bismuth selenide (Bi_2Se_3) TIs combined with a europium sulfide (EuS) ferromagnetic insulator (FMI) are grown with atomically sharp interfaces and crystalline orientation by molecular beam epitaxy.
- The ferromagnetic state was directly observed in the top two quintuple layers (QL, where 1 QL ≈ 0.96 nm) of Bi_2Se_3 near the TI-FMI interface up to temperatures higher than 300 kelvins.



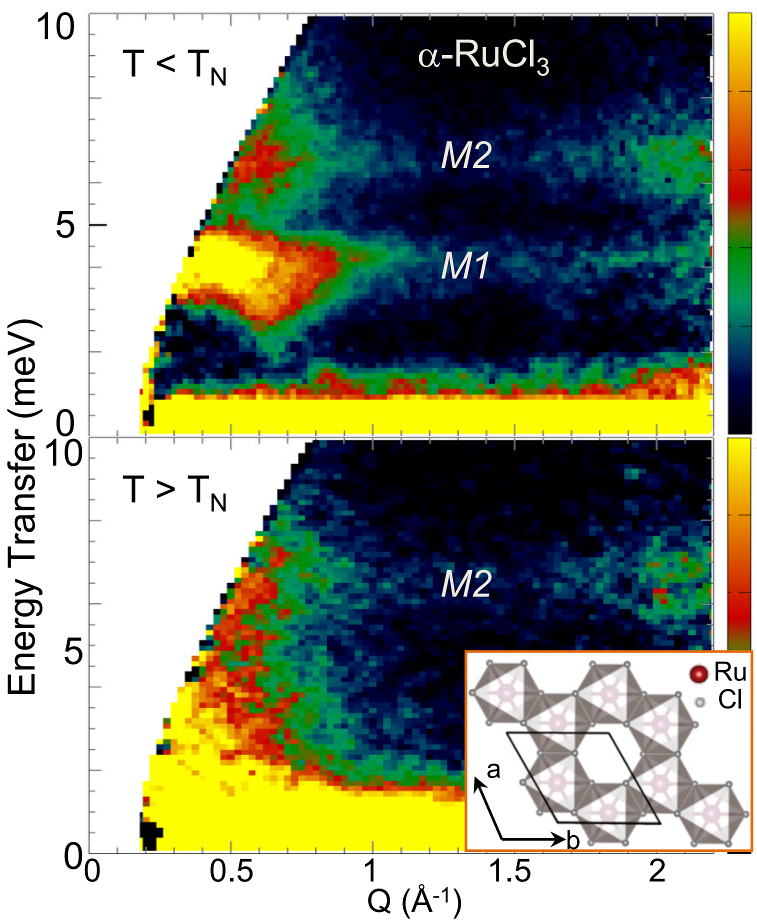
Schematic of the PNR experimental set up for Bi_2Se_3-EuS bilayer films and measured and fitted (solid lines) reflectivity curves for two neutron spin-polarization. The inset is an expanded view of the reflectivity below its critical edge that is sensitive to the distribution of the Eu ions due to the absorption cross section and the magnetic moment.

- PNR provides characterization of the depth profiles of the elemental nuclear density, the magnetization density, and is also particularly element-sensitive to Eu via the absorption density profile. This affords a very precise disentanglement of the intrinsic ferromagnetism of EuS, from its interfacial magnetism and the induced magnetization in Bi2Se3.

F. Katmis, V. Lauter, F. Nogueira, B. Assaf, M. Jamer, P. Wei, B. Satpati, J. Freeland, I. Eremin, D. Heiman, P. Jarillo-Herrero, and J. Moodera, *Nature*, **2016.**

Work was performed at ORNL's SNS Magnetism Reflectometer instrument, BL-4A, a DOE Office of Science User Facility.

Neutrons expose Kitaev quantum spin liquid excitations in α-$RuCl_3$



Magnetic excitations in α-$RuCl_3$ showing M1, the spin-wave mode from the ordered ground state, and thermally resilient M2 mode from itinerant Majorana Fermions. The crystal structure of α-$RuCl_3$, bottom right, exhibits a layered honeycomb arrangement of S=1/2 magnetic moments.

Scientific Achievement

Inelastic neutron scattering (INS) experiments on the layered honeycomb magnet α-$RuCl_3$ provide clear evidence for excitations related to a Kitaev quantum spin liquid (QSL). The measured response function cannot be explained by conventional spin waves but is consistent with scattering from itinerant Majorana fermions. Majorana fermions are theirs own antiparticles and are potential building blocks of future quantum computers.

Significance and Impact

Under appropriate circumstances, including strong spin-orbit coupling, S=1/2 spins on a honeycomb lattice can form a Kitaev QSL. INS measurements on graphene-like α-$RuCl_3$ yielded estimates of the magnetic interactions placing this material in proximity to the Kitaev QSL limit, and found evidence for an excitation continuum arising from Majorana fermions associated with the QSL state. This is a major step forward in identifying fractionalized excitations that someday could be useful for quantum information.

Research Details

- α-$RuCl_3$ samples were purified to 99.9% purity at ORNL.
- These samples were measured using SEQUOIA, the high resolution chopper spectrometer at SNS, and HB-1A triple-axis instrument at HFIR.

A. Banerjee, C.A. Bridges, J.-Q. Yan, A.A. Aczel, L. Li, M.B. Stone, G.E. Granroth, M.D. Lumsden, Y. Yiu, J. Knolle, S. Bhattacharjee, D.L. Kovrizhin, R. Moessner, D.A. Tennant, D.G. Mandrus and S.E. Nagler, *Nature Materials*, doi:10.1038/nmat4604 (2016).

Work performed at SEQUOIA (SNS) and HB-1A (HFIR), DOE Office of Science User Facilities.

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N. Peter Armitage
Nature Materials 15, 701–702 (2016)
doi:10.1038/nmat4667, Published online 22 June 2016

NATURE MATERIALS | Journal Cover: Inelastic neutron scattering characterization shows that α-$RuCl_3$ is close to an experimental realization of a Kitaev quantum spin liquid on a honeycomb lattice. The collective excitations provide evidence for deconfined Majorana fermions.
Image: Genevieve Martin and Arnab Banerjee, Oak Ridge National Laboratory
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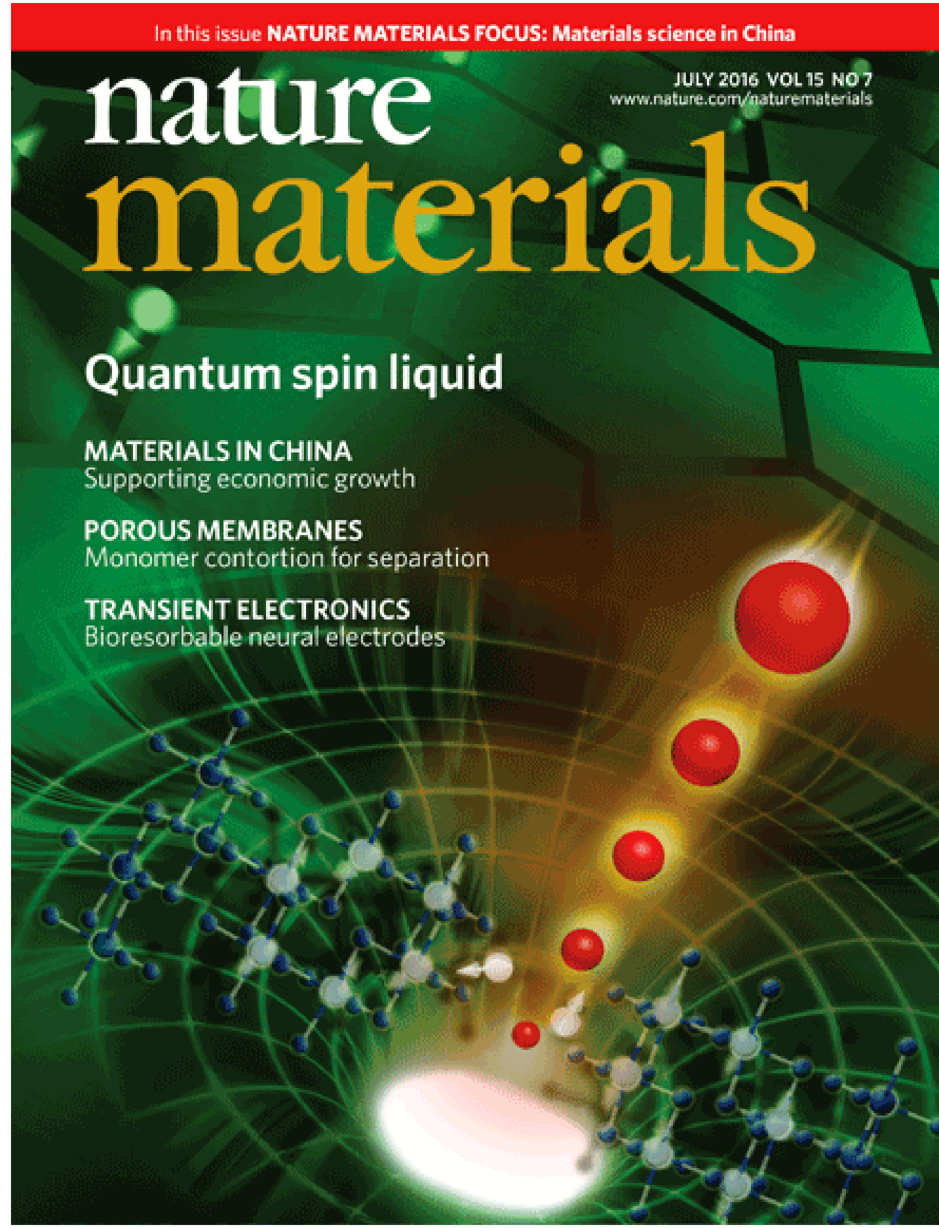

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Science with Neutrons

- **Neutrons show where atoms are**
- Neutrons show where the nuclei and the electron spins are:
  - Ordering of the nuclei
  - Ordering of the electron spins
- **Neutrons show what atoms do**
- Neutrons show what the nuclei and the electron spins do:
  - Lattice vibrations-phonons
  - Spin excitations-magnons, etc