



HIGH FLUX
ISOTOPE
REACTOR

SPALLATION
NEUTRON
SOURCE

Lunch talk, March 30, 2026, 12:00-13:00

HFIR & SNS, a historical perspective

Bldg 8600, rm C-156

Barry Winn, HYSPEC instrument scientist

**Polarized Neutron Diffraction and Spectroscopy:
Applications to Quantum Materials**



U.S. DEPARTMENT OF
ENERGY

ORNL IS MANAGED BY UT-BATTELLE LLC
FOR THE US DEPARTMENT OF ENERGY



Chadwick discovers the neutron



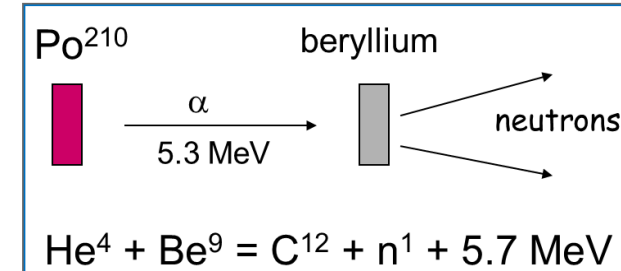
NATURE

[FEBRUARY 27, 1932

Possible Existence of a Neutron

It has been shown by Bothe and others that beryllium when bombarded by α -particles of polonium emits a radiation of great penetrating power, which has an absorption coefficient in lead of about 0.3 (cm.)^{-1} . Recently Mme. Curie-Joliot and M. Joliot found, when measuring the ionisation produced by this beryllium radiation in a vessel with a thin window, that the ionisation increased when matter containing hydrogen was placed in front of the window. The effect appeared to be due to the ejection of protons with velocities up to a maximum of nearly $3 \times 10^9 \text{ cm. per sec.}$ They suggested that the transference of energy to the proton was by a process similar to the Compton effect, and estimated that the beryllium radiation had a quantum energy of $50 \times 10^8 \text{ electron volts.}$

These results, and others I have obtained in the course of the work, are very difficult to explain on the assumption that the radiation from beryllium is a quantum radiation, if energy and momentum are to be conserved in the collisions. The difficulties disappear, however, if it be assumed that the radiation consists of particles of mass 1 and charge 0, or neutrons. The capture of the α -particle by the Be^9 nucleus may be supposed to result in the formation of a C^{12} nucleus and the emission of the neutron. From the energy relations of this process the velocity of the neutron emitted in the forward direction may well be about $3 \times 10^9 \text{ cm. per sec.}$

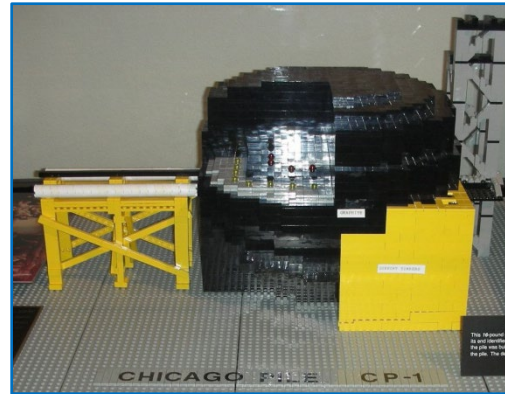
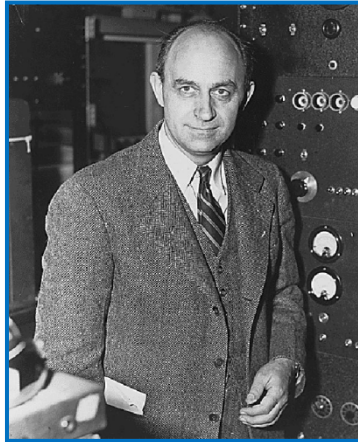


1935

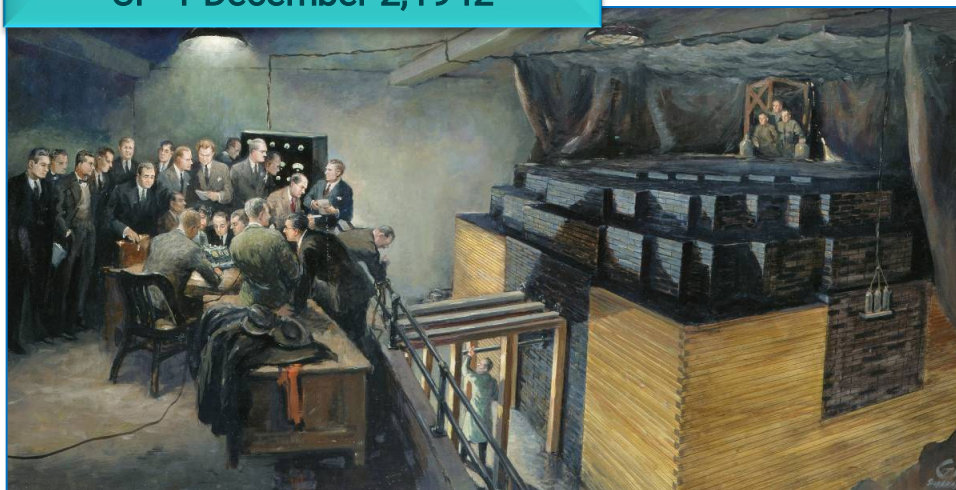
Nobel Prize
in Physics

The first chain reaction: CP-1

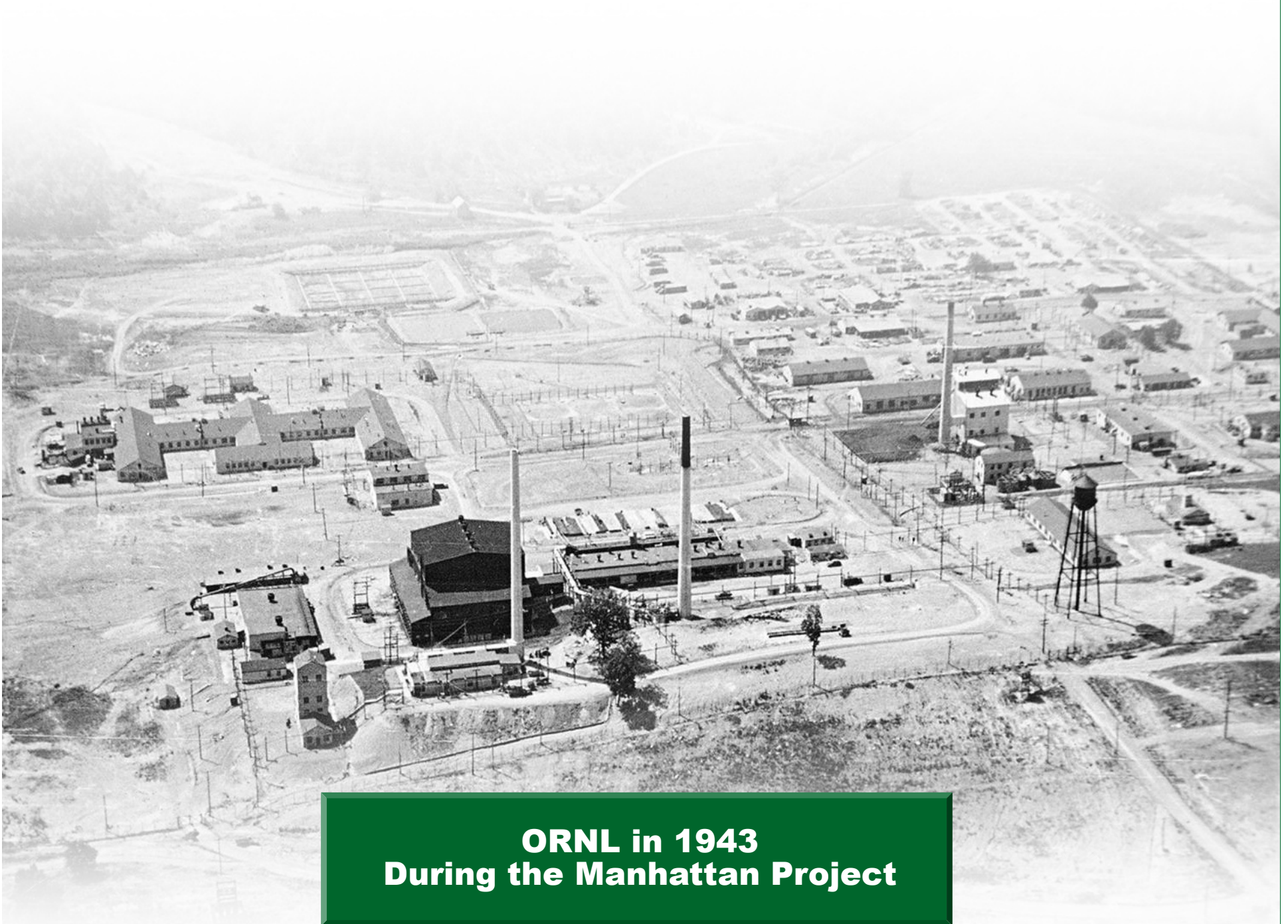
Enrico Fermi



CP-1 December 2, 1942

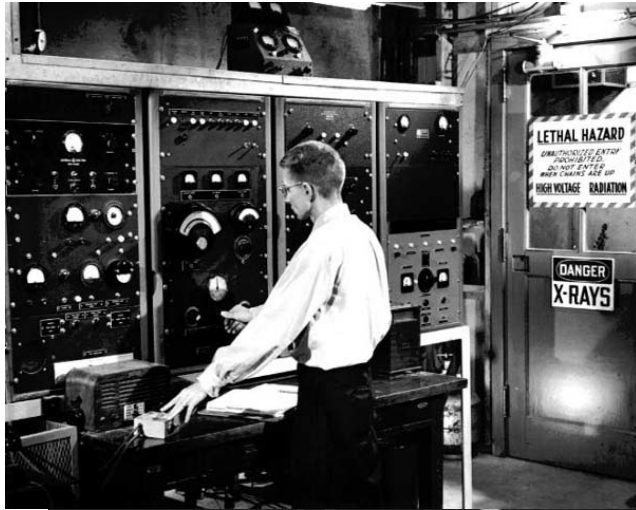


"Chicago Pile" #1



**ORNL in 1943
During the Manhattan Project**

X-10 Pile or Graphite Reactor



The graphite reactor at Clinton Laboratories went critical on Nov. 4, 1943, under the supervision of Enrico Fermi. It was dedicated as a national historic landmark in 1966.



Fun fact interlude

Secrets and Talking trash among scientists!

- The 'chance' of scattering, called a cross section, has a strange unit of area of 10^{-28} m^2 or 100 fm^2
- Nuclear physicists during WW-II chose a secret term for this unit
- Aligning for an experiment, we can talk trash with each other, using the old saying:
"Can't hit the broad side of a *barn*"



Can't hit the broad side of a *barn*!

Wollan's research proposal

Ernest Omar (Ernie) Wollan received his Ph.D. in 1929 at U. Chicago under Arthur Compton. He was present at the startup of CP-1, and X-10. Among other things he is the "father of health physics". Wollan moved to Oak Ridge in 1944.



202

This Document consists of 1 Pages and 0 Figures.
No. 1 of 2 Copies, Series A
DATE May 25, 1944

~~SECRET~~ CLINTON LABORATORIES

TO R. L. Doan DEPARTMENT
FROM E. O. Wollan DEPARTMENT
IN RE: Diffraction of neutrons

MAY 27 1944
R. L. DOAN

I would like to attempt to measure the diffraction of neutrons by single crystals. I have brought some equipment with me from Chicago, and Dr. Borst has shown me an opening in the pile at which this work could be done.

I would appreciate obtaining approval to go ahead with this experiment.

A problem assignment sheet for this work is enclosed.

E. O. Wollan

CLASSIFICATION CANCELLED
DATE OCT 17 1963
For The Atomic Energy Commission
H. R. Canine
Chief, Declassification Branch
RECEIVED
MAY 26 1944

EOW/o

December 1944 – rocking curves!

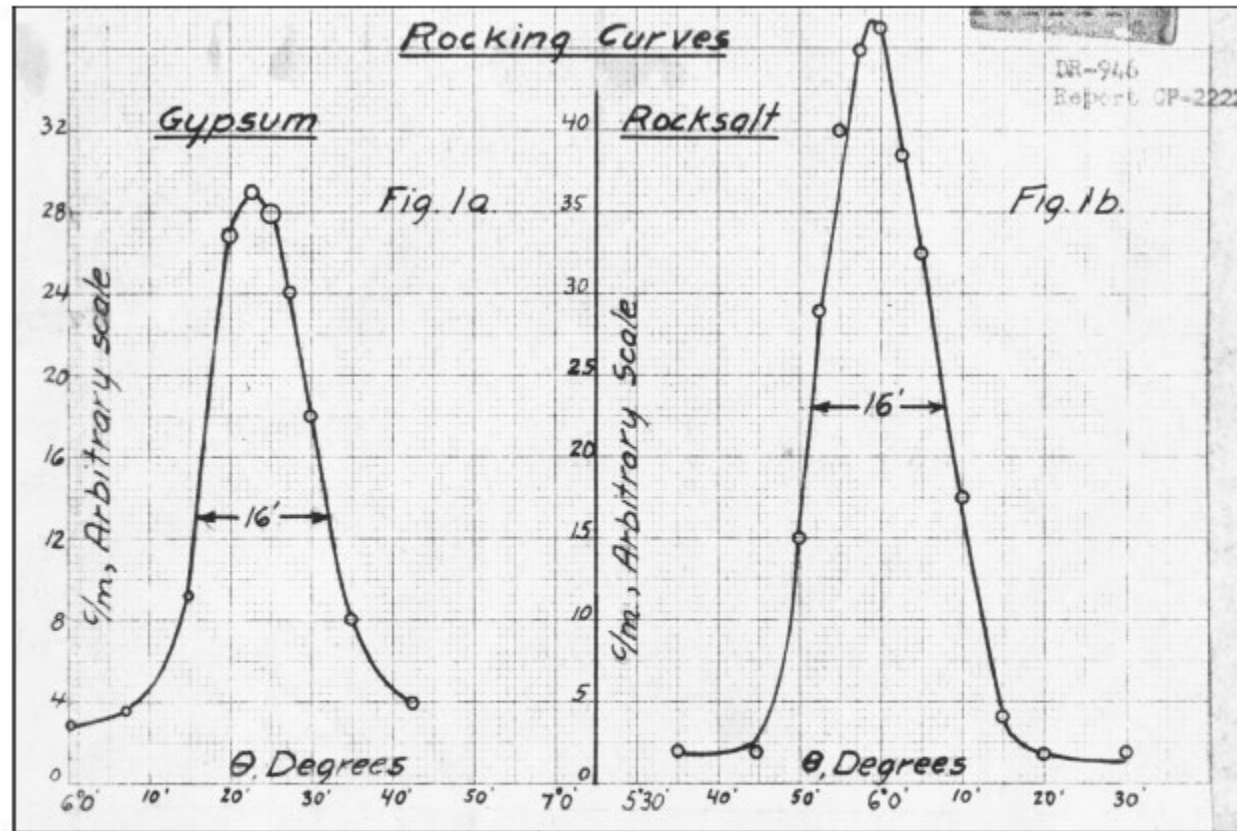


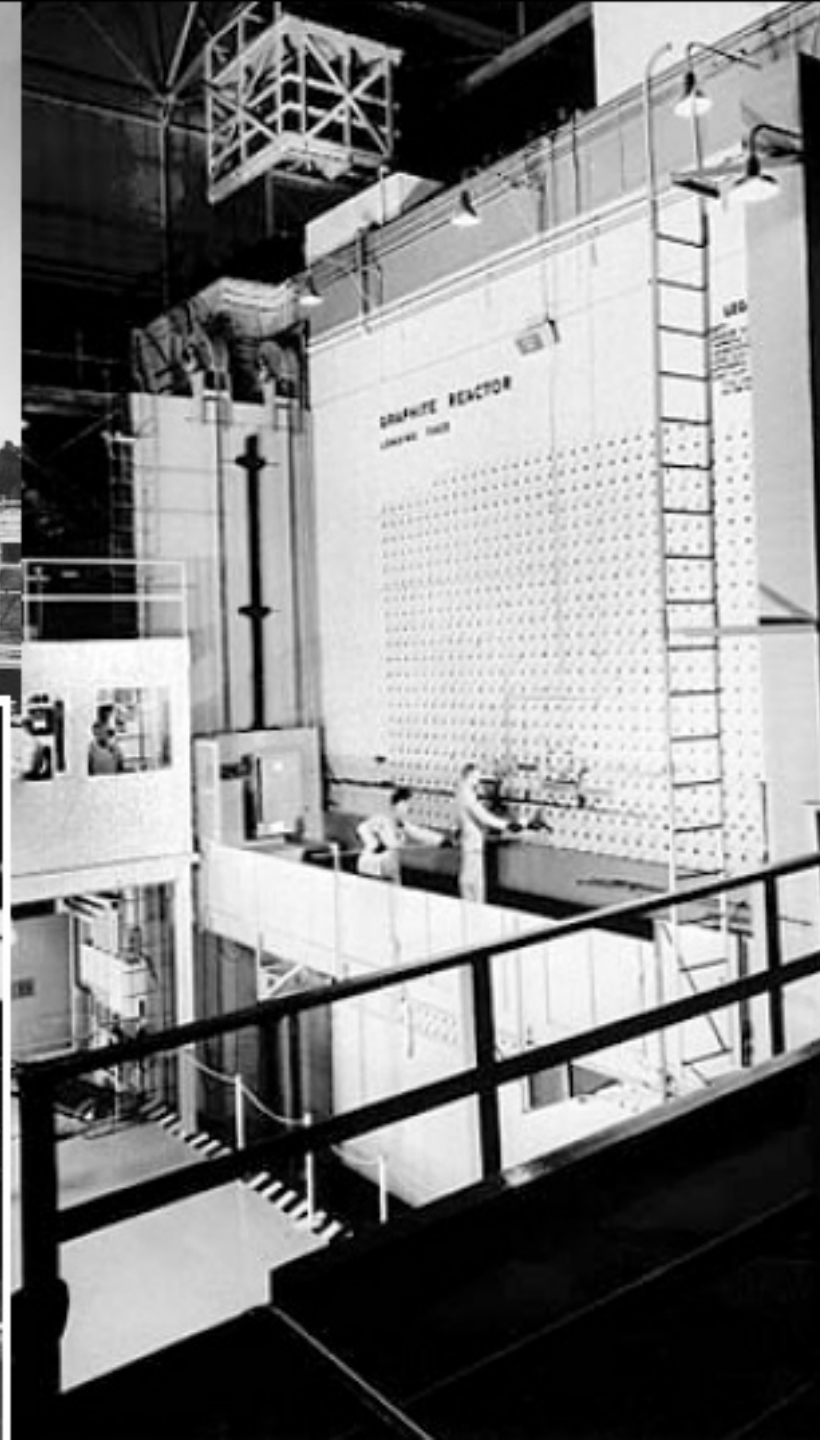
Fig. 4. Hand-plotted rocking curves for Bragg scattering from single crystals at the X-10 pile, obtained by Wollan & Borst in December 1944 with improved equipment installed on 2 December.

Nobel prizes and Neutron Scattering

Always the bridesmaid, never the bride (until 1994)

New set of magnetic Bragg peaks observed on MnO

ORNL Graphite Reactor
1943-1963



Detection of Antiferromagnetism by Neutron Diffraction*

C. G. SHULL

Oak Ridge National Laboratory, Oak Ridge, Tennessee

AND

J. SAMUEL SMART

Naval Ordnance Laboratory, White Oak, Silver Spring, Maryland

August 29, 1949



Shull & Ernie Wollan 1949

Neel's model confirmed

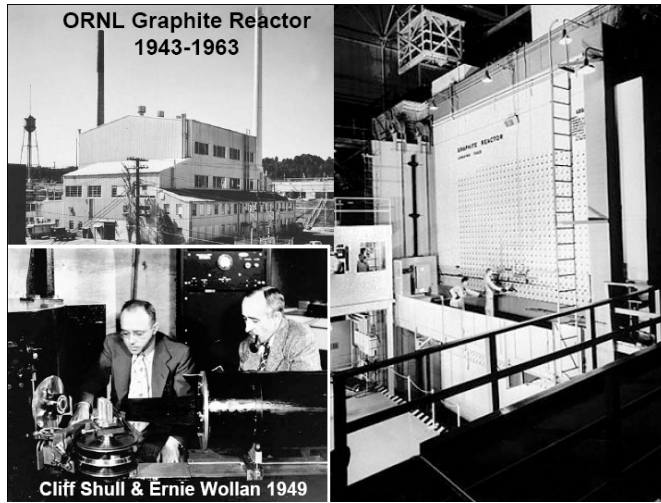


Half of the Nobel Prize in Physics 1970 was awarded to Louis Neel "for fundamental work and discoveries concerning antiferromagnetism and ferrimagnetism which have led to important applications in solid state physics".



**Nobel Prize
in Physics**

- ✓ The first direct evidence of antiferromagnetism was produced in determining the magnetic structure of MnO - 1949



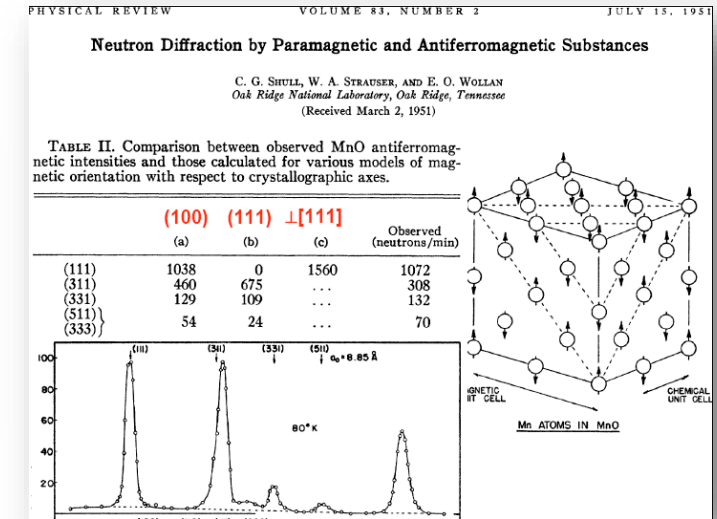
Detection of Antiferromagnetism by Neutron Diffraction*

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August 29, 1949



- ✓ the first magnetic form-factor data were obtained by measuring the paramagnetic scattering by Mn compounds (1951)
- ✓ the production of polarized neutrons by Bragg reflection from ferromagnets was demonstrated (1951)

H-D scattering and structure of ice

Shull and Wollan established the rules for scattering lengths of H and D (below left), and used this to prove that Pauling's model was the correct description for the structure of water (right).

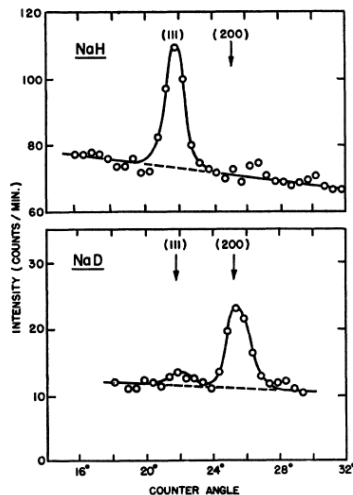


FIG. 1. Powder diffraction patterns over the (111) and (200) peaks of NaH and NaD. The diffuse scattering for NaH is seen to be much larger than that for NaD.

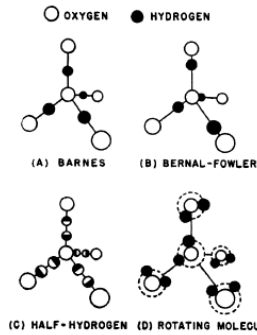


FIG. 2. Schematic diagram of four structural models which are used in calculating pattern intensities for various proposed structures of ice.

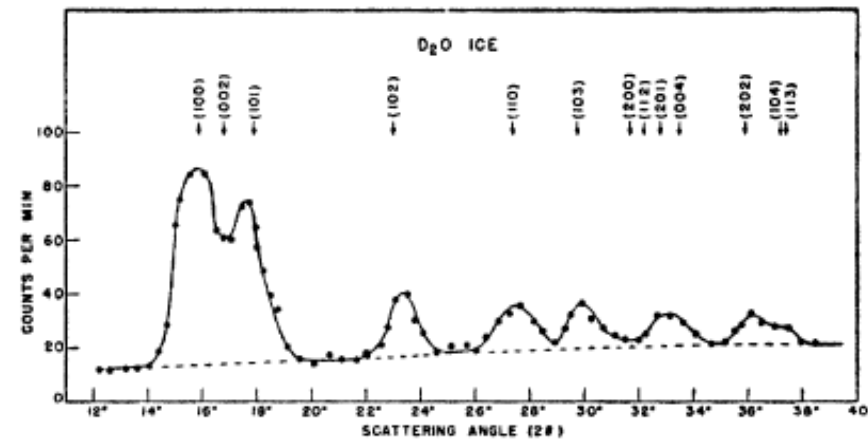
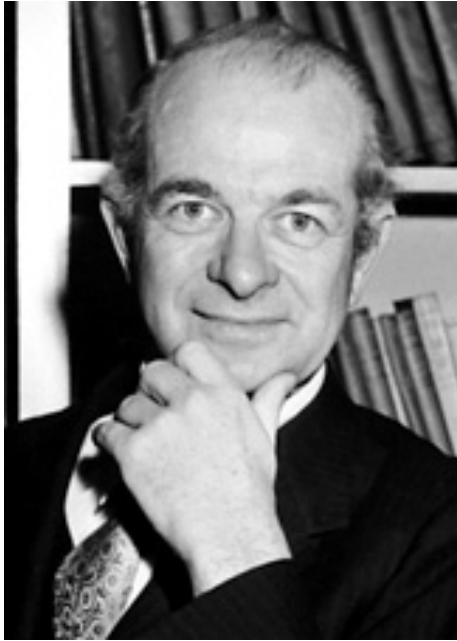


FIG. 1. Neutron diffraction powder pattern of heavy ice (D_2O) taken at $-90^\circ C$ with neutrons of wave-length 1.06Å.

- C. G. Shull, E. O. Wollan, G. A. Morton, and W. L. Davidson, *Neutron diffraction studies of NaH and NaD*, Phys. Rev. **73**, 842-847 (1948).
- E. O. Wollan, W. L. Davidson, and C. G. Shull, *Neutron diffraction study of the structure of ice*, Phys. Rev. **75**, 1348-1352 (1949).

Pauling's model confirmed



The Nobel Prize in Chemistry 1954 was awarded to Linus Pauling *"for his research into the nature of the chemical bond and its application to the elucidation of the structure of complex substances"*.



Nobel Prize
in
Chemistry

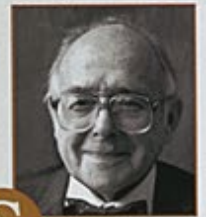
The Nobel Prize in Physics 1994



Neutrons behave as particles and as waves

The Royal Swedish Academy of Sciences has awarded the 1994 Nobel Prize in Physics for pioneering contributions to the development of neutron scattering techniques for studies of condensed matter.

Clifford G. Shull, MIT, Cambridge, Massachusetts, USA, receives one half of the 1994 Nobel Prize in Physics for development of the neutron diffraction technique.



S Shull made use of **elastic scattering** i.e. of neutrons which change direction without losing energy when they collide with atoms.

Because of the wave nature of neutrons, a diffraction pattern can be recorded which indicates where in the sample the atoms are situated. Even the placing of light elements such as hydrogen in metallic hydrides, or hydrogen, carbon and oxygen in organic substances can be determined.

The pattern also shows how atomic dipoles are oriented in magnetic materials, since neutrons are affected by magnetic forces. Shull also made use of this phenomenon in his neutron diffraction technique.



An early (1950) neutron diffractometer with flexible wavelength control here used by E.O. Wollan and C.G. Shull (standing) at Oak Ridge National Laboratory.

Neutrons see more than X-rays

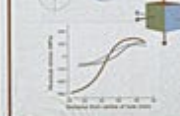
X-rays are scattered by electronic neutrons by atomic nuclei. With X-rays it is easiest to see atoms that have many electrons. Hydrogen, for example, which has only one electron, is not so easy to see. With neutrons, all kinds of atoms are visible.



Lay a neutron diffraction map (showing the positions of the nuclei) over an X-ray diffraction map (giving the distribution of the electrons). It is then clear that the electron density is shifted in relation to the positions of the atomic nuclei. Since a chemical bond involves a shift in electron position, a direct picture of the chemical bond is obtained in this way.

Neutrons reveal inner stresses

A hole has been punched in an important metal aircraft part. Does the part match up? Neutron diffraction can show how much the distance between the atoms has changed and hence the internal forces remaining round the hole after it has been punched.



The curves show local expansion forces (positive) and compression forces (negative) in different directions (red, green and blue) in an aircraft part (David Griener).

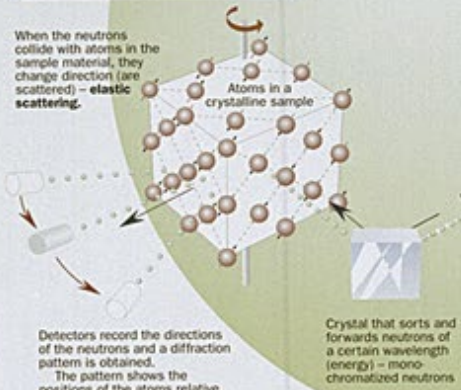
Neutrons show what atoms remember

of their earlier positions when they move randomly in relation to each other in liquids and solids. Even here there is in fact some local order. The atoms cannot move infinitely close to each other. Some distances are more common than others.



The time curve $t = 0$ shows the positions for liquid bromine. The other curves show how the positions of the atoms change with time (1 ps = one millionth of a millionth of a second) seen with inelastic neutron scattering. Corresponding "memory functions" can also be measured in magnets e.g. near the Curie temperature, the temperature at which magnetic order shifts to disorder.

Neutrons show where atoms are

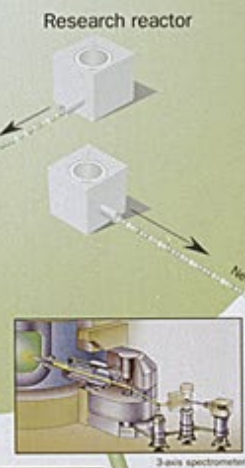


When the neutrons collide with atoms in the sample material, they change direction (are scattered) - **elastic scattering**.

Detectors record the directions of the neutrons and a diffraction pattern is obtained. The pattern shows the positions of the atoms relative to one another.

Crystal that sorts and forwards neutrons of a certain wavelength (energy) - **monochromatized neutrons**

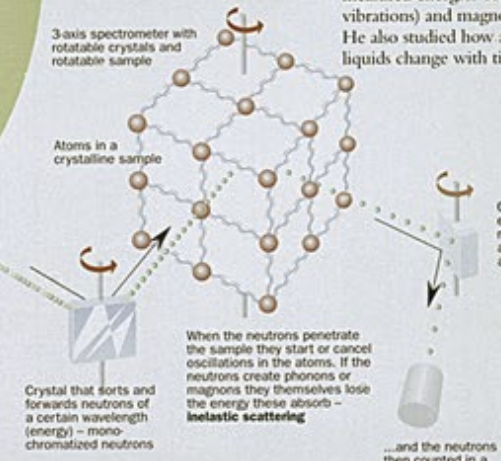
Neutrons bounce against atomic nuclei. They also react to the magnetism of the atoms.



Research reactor

3-axis spectrometer

Neutrons show what atoms do



3-axis spectrometer with rotatable crystals and rotatable sample

Atoms in a crystalline sample

Crystal that sorts and forwards neutrons of a certain wavelength (energy) - **monochromatized neutrons**

When the neutrons penetrate the sample they start or cancel oscillations in the atoms. If the neutrons create phonons or magnons they themselves lose the energy these absorb - **inelastic scattering**

Changes in the energy of the neutrons are first analysed in an analyser crystal...

...and the neutrons then counted in a detector.

Bertram N. Brockhouse, McMaster University, Hamilton, Ontario, Canada, receives one half of the 1994 Nobel Prize in Physics for the development of neutron spectroscopy.



B Brockhouse made use of **inelastic scattering** i.e. of neutrons, which change both direction and energy when they collide with atoms. They then start or cancel atomic oscillations in crystals and record movements in liquids and melts. Neutrons can also interact with spin waves in magnets.

With his 3-axis spectrometer Brockhouse measured energies of phonons (atomic vibrations) and magnons (magnetic waves). He also studied how atomic structures in liquids change with time.

How it started
Brockhouse and Shull made their pioneering contributions at the first nuclear reactors in the USA and Canada back in the 1940s and 1950s. It was then that the resources of the reactors became available for peacetime research.

... how it continues
Thousands of researchers are now working at the many neutron research centers throughout the world. New and very advanced neutron scattering installations have been built and more are planned in Europe, the USA and Asia. At these super-installations the researchers are studying the structure of new ceramic superconductors, molecular movements on surfaces of interest for catalytic, exhaust cleaning, virus structures and the connection between the structure and the elastic properties of polymers.



Information Department, Box 50005, S-104 05 Stockholm, Sweden. Tel: +46-8-673 95 00. Fax: +46-8-673 95 25. Editors: Bengt-Åke Rosenqvist, Margareta Åberg-Rosend, The Royal Swedish Academy of Sciences, Århus; Professor Erik B. Kramers and Professor Carl Norling, Department of Physics, Uppsala University, Members of the Nobel Committee for Physics. Layout and illustrations: Kjell Lindén, Expresso AB. Printed by Trycknord, 1994.

- Further reading:
- D.J. Hughes *The Nuclear Reactor as a Research Instrument*, SCIENTIFIC AMERICAN, VOL. 156, AUGUST 1957, P. 23
 - H. Lengeler and J.L. Finney *The European Spallation Source*, EUROPHYSICS NEWS, VOL. 25, P.32, 1994.
 - Information about the Nobel Prize in Physics 1994 (<http://www.nobel.se/physics/1994/>), THE ROYAL SWEDISH ACADEMY OF SCIENCES.

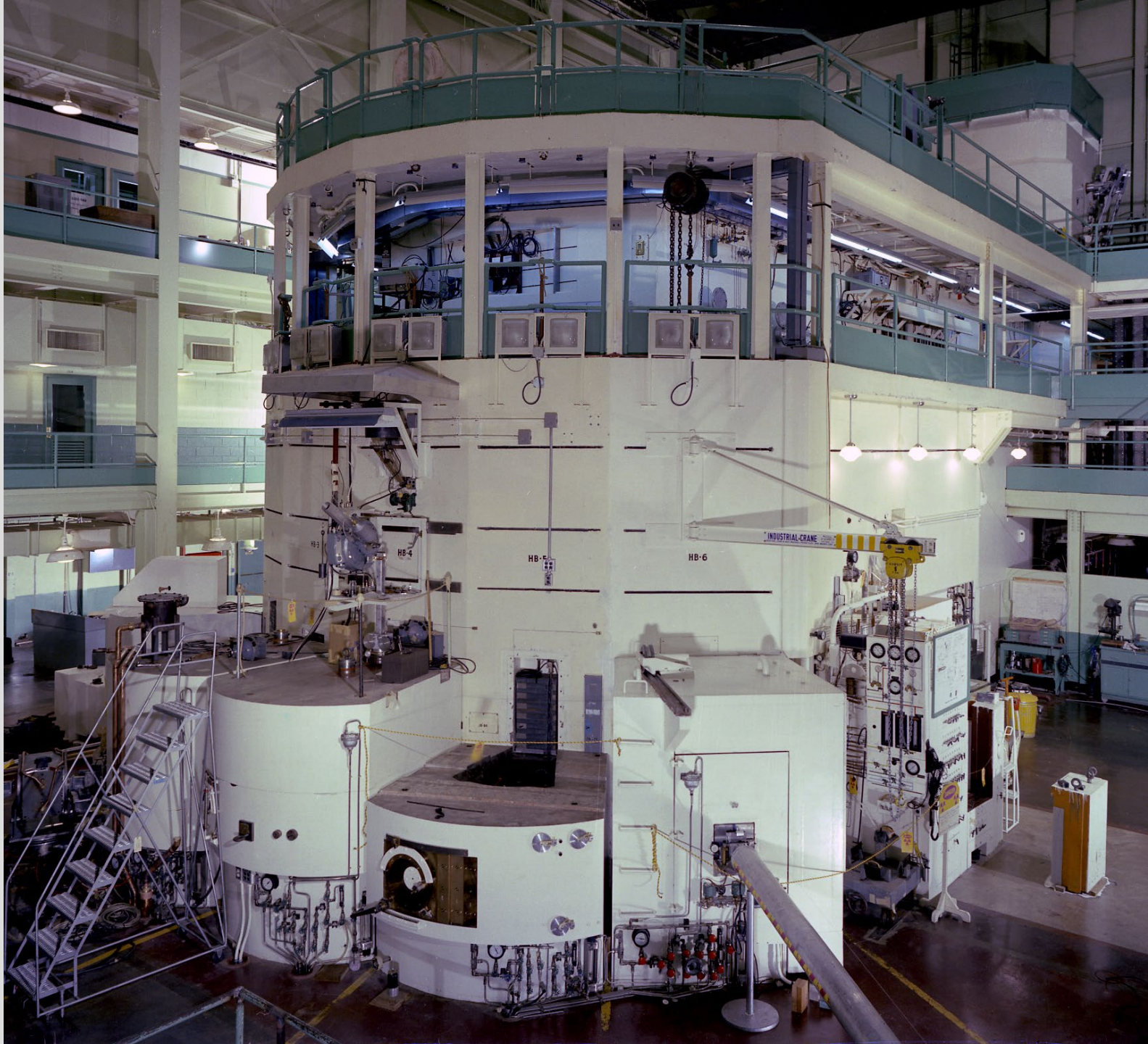
Ramping up Reactor-based Neutron Scattering

The neutron scattering emphasis in the 1960's was at another reactor

Beam room of the Oak Ridge Research Reactor, ORR, at Oak Ridge National Laboratory, December 29, 1960

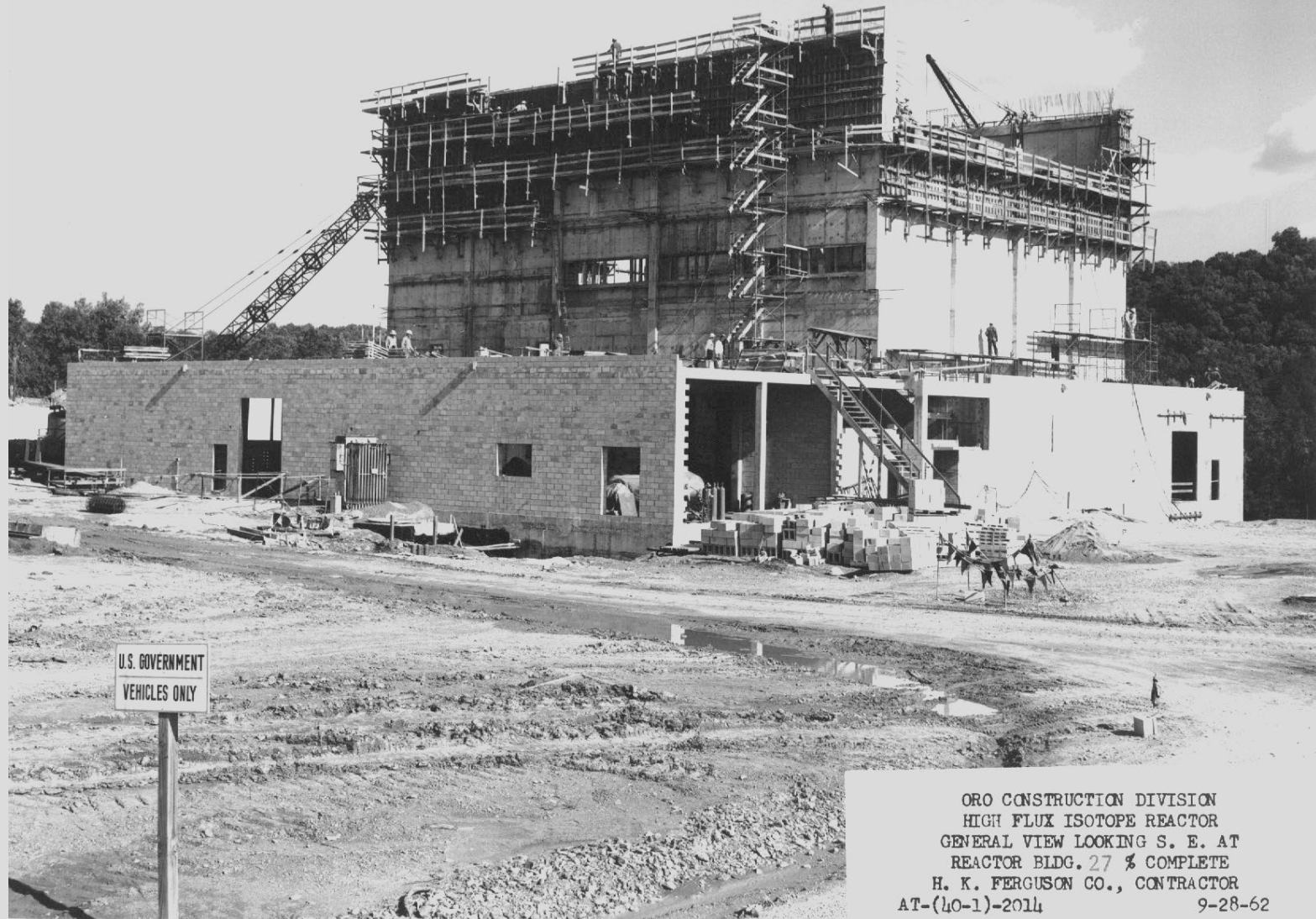


Oak Ridge Research Reactor (ORR), January 1960



A research reactor for isotope production, irradiation studies (& a bit-o-scattering)

High Flux Isotope Reactor (HFIR). General view looking southeast at Reactor building, 27 percent complete. September 28, 1962.



ORO CONSTRUCTION DIVISION
HIGH FLUX ISOTOPE REACTOR
GENERAL VIEW LOOKING S. E. AT
REACTOR BLDG. 27 % COMPLETE
H. K. FERGUSON CO., CONTRACTOR
AT-(10-1)-2014 9-28-62

Herb Mook, HB-3, Beam Room,
HFIR, December 13, 1982

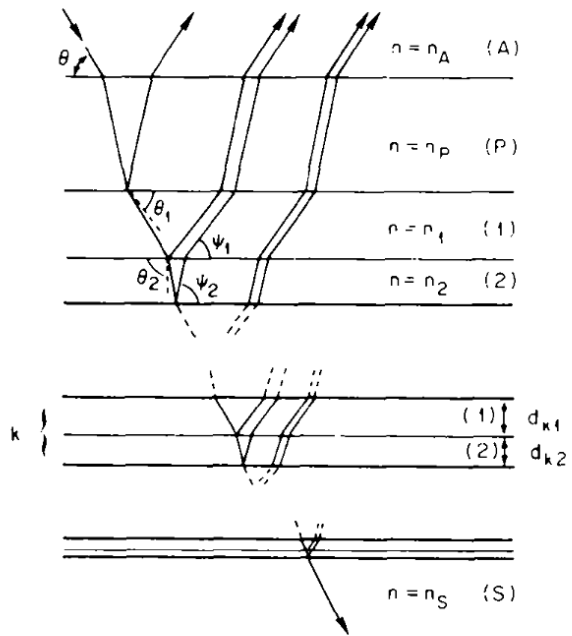


Fig. 1. Geometry of a general multi-bilayer structure composed of $2N$ alternating thin films of generic materials 1 and 2, having respective refractive indices n_1 and n_2 , and thicknesses d_{k1} and d_{k2} in the k th bilayer ($k = 1, \dots, N$). The structure is assumed to lie on an infinitely thick substrate of material S (refractive index n_S), and to be immersed in an atmosphere $A(n_A)$ from which it may be protected by the presence of a coating $P(n_P)$. All angles have been greatly exaggerated for clarity.

J. Appl. Cryst. (1989). 22, 35-41

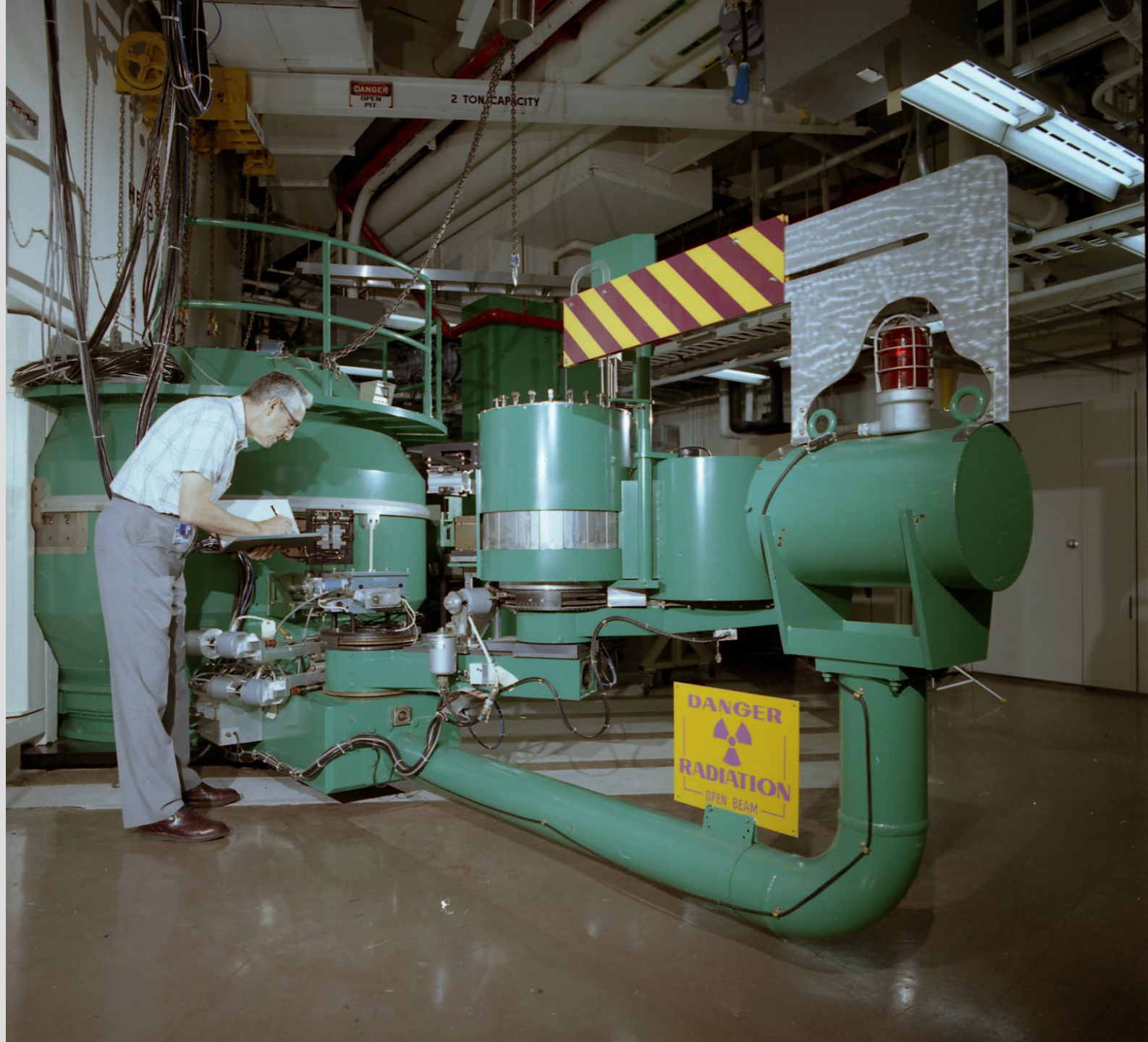
Discrete Thin-Film Multilayer Design for X-ray and Neutron Supermirrors

BY JOHN B. HAYTER AND H. A. MOOK

Solid State Division, Oak Ridge National Laboratory, PO Box 2008, Oak Ridge, TN 37831-6031, USA

Ralph Moon (1929-2004)

HB3, HFIR



Triple Axis Polarization Analysis

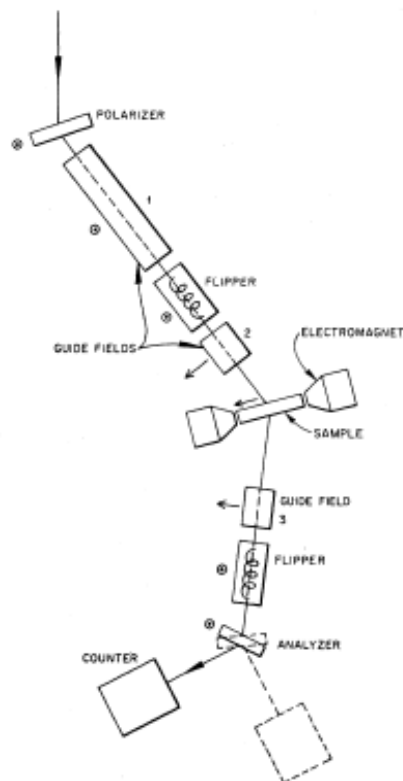


FIG. 1. Experimental arrangement. Arrows adjacent to the guide fields show the direction of the magnetic field sensed by the neutrons.

In a seminal paper in 1969, Ralph Moon and colleagues invented the method of neutron polarization analysis, opening up new fields for investigation by neutron scattering. A schematic diagram of the original apparatus is shown at left. Below, right, are measurements of spin-flip scattering in magnetic excitations.

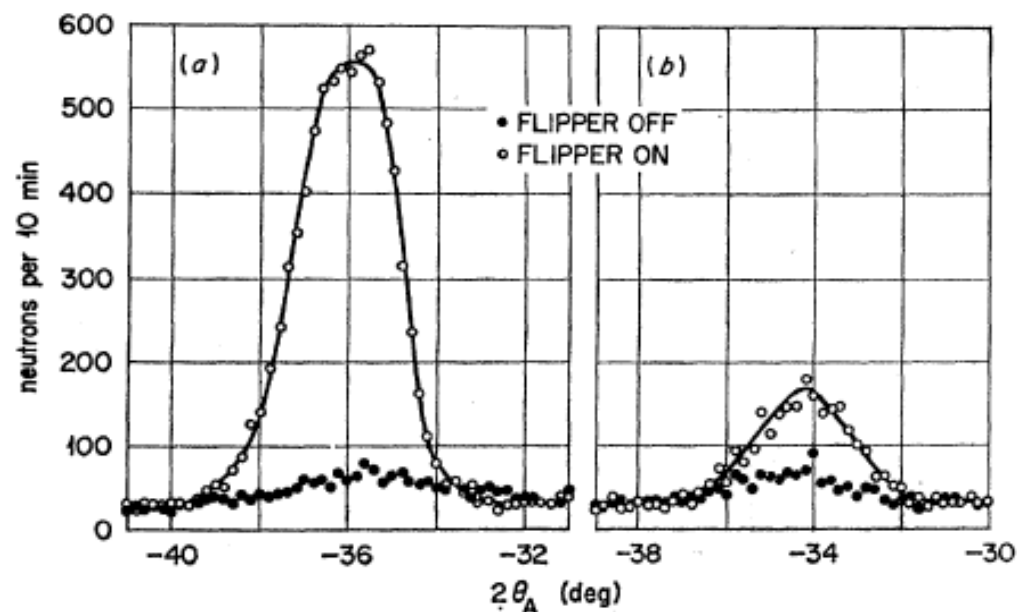


FIG. 9. Polarization reversal in magnon scattering from Li ferrite. In (a), $\hat{K} \cdot \hat{P}_0 = 1$, and in (b), $\hat{K} \cdot \hat{P}_0 = 0$. The small peak in the flipper-off data is due to nuclear disorder scattering.

R. M. Moon, T. Riste, and W. C. Koehler, *Polarization Analysis of Thermal-Neutron Scattering*, Phys. Rev. **181**, 920-931 (1969). [345]

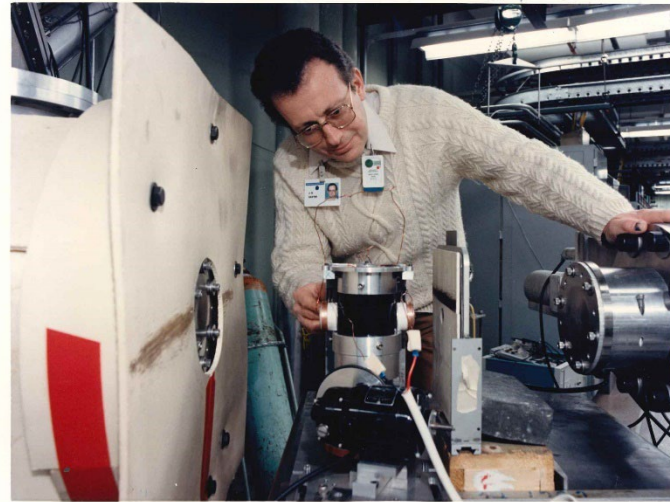
Beyond the solid state 'bubble'

Wally Koehler tried to expand the neutron science beyond the traditional scope of Solid State Physics



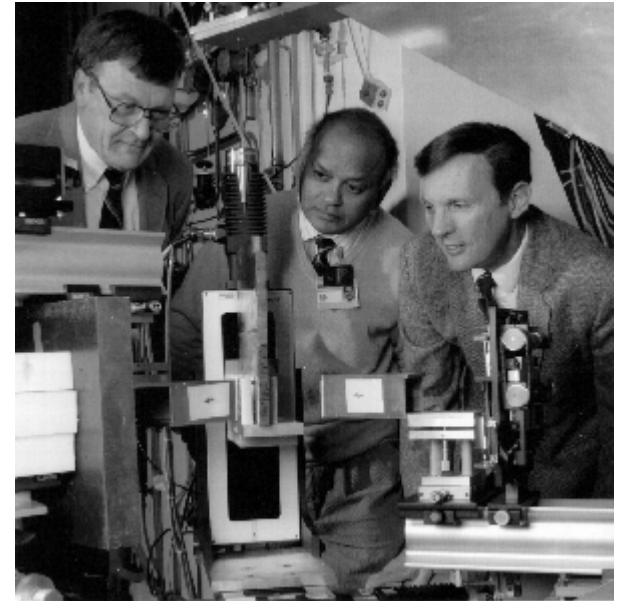
George Wignall

- First SANS User Facility in the US
- Polymers



John Hayter

- Ferrofluids
- Colloids
- Neutron optics



Steve Spooner & Cam Hubbard

- Metallurgy using SANS
- Residual stress

George Wignall was one of the pioneers in the use of H-D labeling in SANS

Colloid & Polymer Sci. 252, 749–752 (1974)



*Institut für Festkörperforschung of the Kernforschungsanlage Jülich
and from the Corporate Laboratory of the Imperial Chemical Industries Limited, Runcorn, Cheshire, United Kingdom*

Neutron small-angle scattering by mixtures of H and D-tagged molecules of polystyrene and polyethylene

J. Schelten, G. D. Wignall, D. G. H. Ballard, and W. Schmatz

With 6 figures and 2 tables

(Received May 29, 1974)

The initiative for the use of the H-D contrast technique in SANS from polymers to "color" individual molecules and make them visible in the condensed state was taken independently By G. Wignall (ICI/Julich) and Julia Higgins in France (Saclay/ILL)

Macromolecules

Vol. 7, No. 6, November–December 1974

Conformation of Polymer Chain in the Bulk 863

Conformation of Polymer Chain in the Bulk

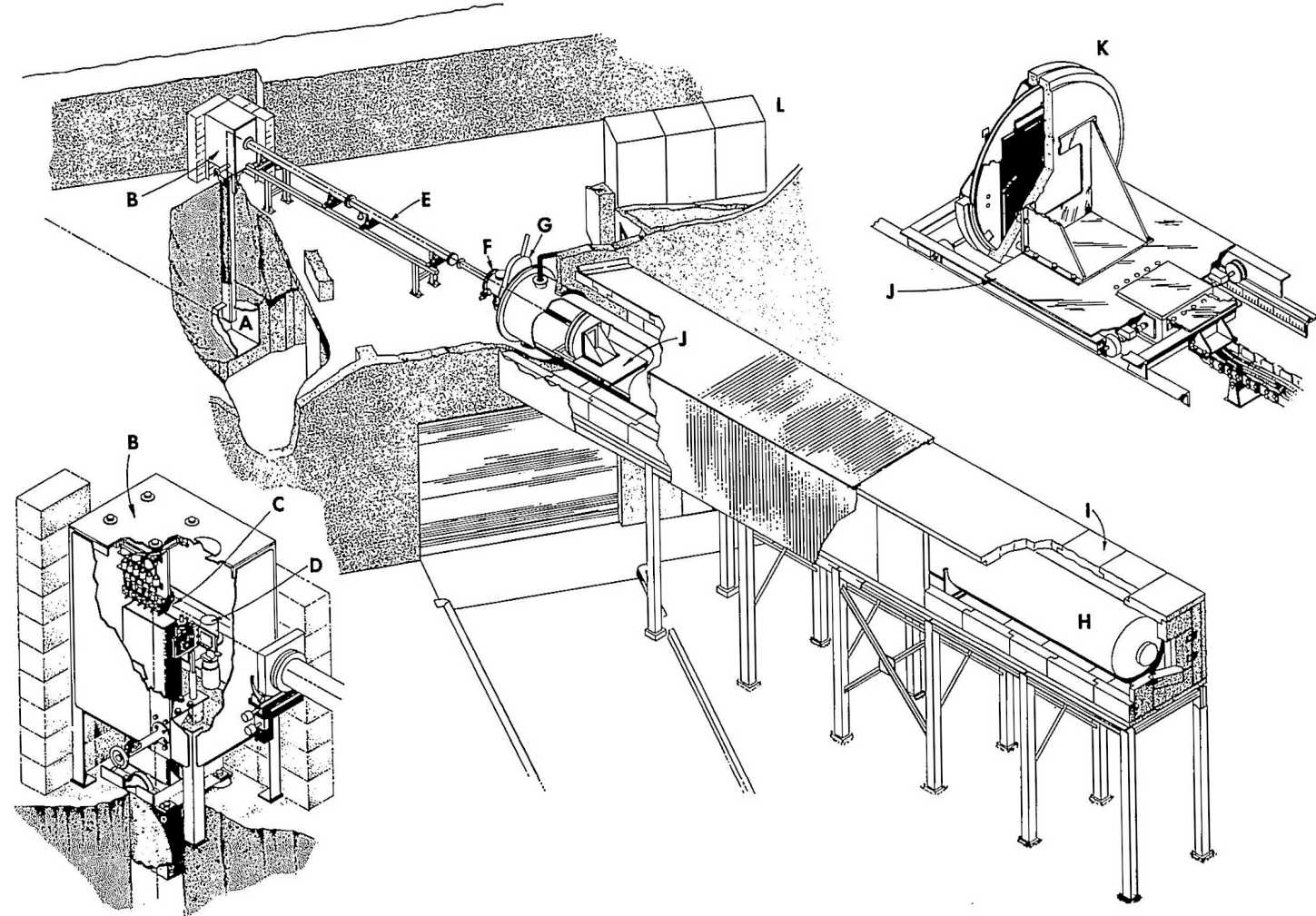
J. P. Cotton,^{1a} D. Decker,^{1b} H. Benoit,^{1b} B. Farnoux,^{1a} J. Higgins,^{1c} G. Jannink,^{1a} R. Ober,^{1d} C. Picot,^{1b} and J. des Cloizeaux^{1e}

Centre de Recherches sur les Macromolécules (CNRS), 67083 Strasbourg Cedex, France; Service de Physique du Solide et de Résonance Magnétique and Service de Physique Théorique, Centre d'Études Nucléaires de Saclay, 91190 Gif-sur-Yvette, France; Institut Max Von Laue-Paul Langevin, 38042 Grenoble, France; and Laboratoire de la Matière Condensée, Collège de France, 11, Place M. Berthelot, 75005 Paris, France. Received May 2, 1974



The 30m SANS Facility at HFIR

- Beam delivery by 6 pair PG crystals in a “periscope” fashion
- 4.75 angstrom , beryllium filter, no cold source
- 3.4×10^4 neutrons/(cm² s)
- Aperture up to 2 x 2 cm² separated by 10 m.
- Large 60 x 60 cm² movable PSD
- Vacuum flight-path of 20m on stilts stuck out of the HFIR building!



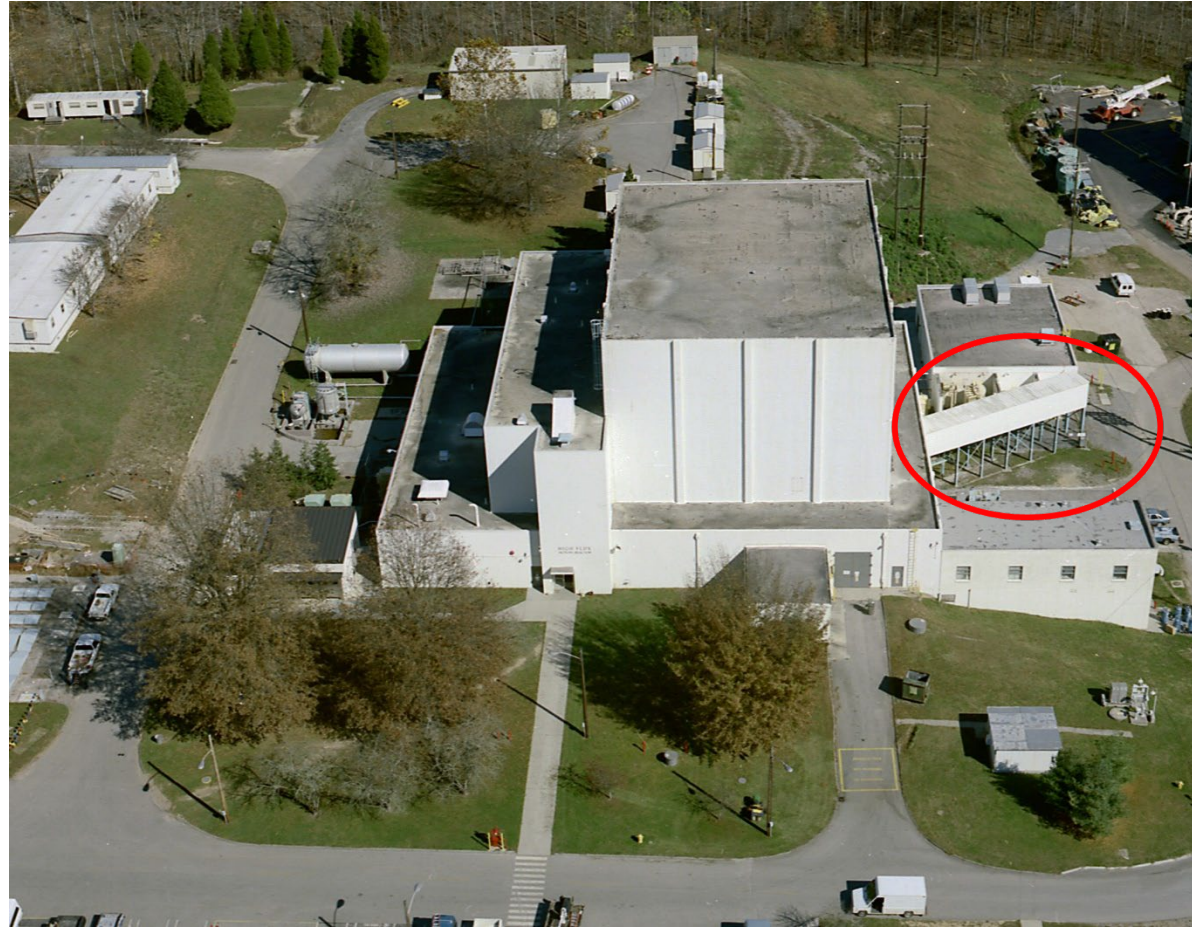
- | | | |
|---------------------------------------|--|--|
| (A) LOWER MONOCHROMATOR HOUSING | (F) SAMPLE CHAMBER | (K) 112-cm-diam TWO-DIMENSIONAL POSITION-SENSITIVE DETECTOR (64- BY 64-cm ACTIVE AREA) |
| (B) UPPER MONOCHROMATOR HOUSING | (G) 56-cm GATE VALVE | (L) DATA ACQUISITION SYSTEM |
| (C) GRAPHITE MONOCHROMATING CRYSTALS | (H) 20-m BY 152-cm-diam VACUUM FLIGHT PATH | |
| (D) COLD BERYLLIUM FILTER | (I) 28-cm WOOD SHIELDING | |
| (E) COLLIMATOR AND NEUTRON BEAM GUIDE | (J) DETECTOR CARRIAGE | |

Fig. 22. 30-m Small-Angle Neutron Scattering Facility.

W. Koehler and G. D. Wignall built the first Small Angle Neutron Scattering (SANS) User Program in the US in 1980



The first NSF Center for Small Angle Neutron Scattering



Transition from collaborative research to User program

1970's - 2000's

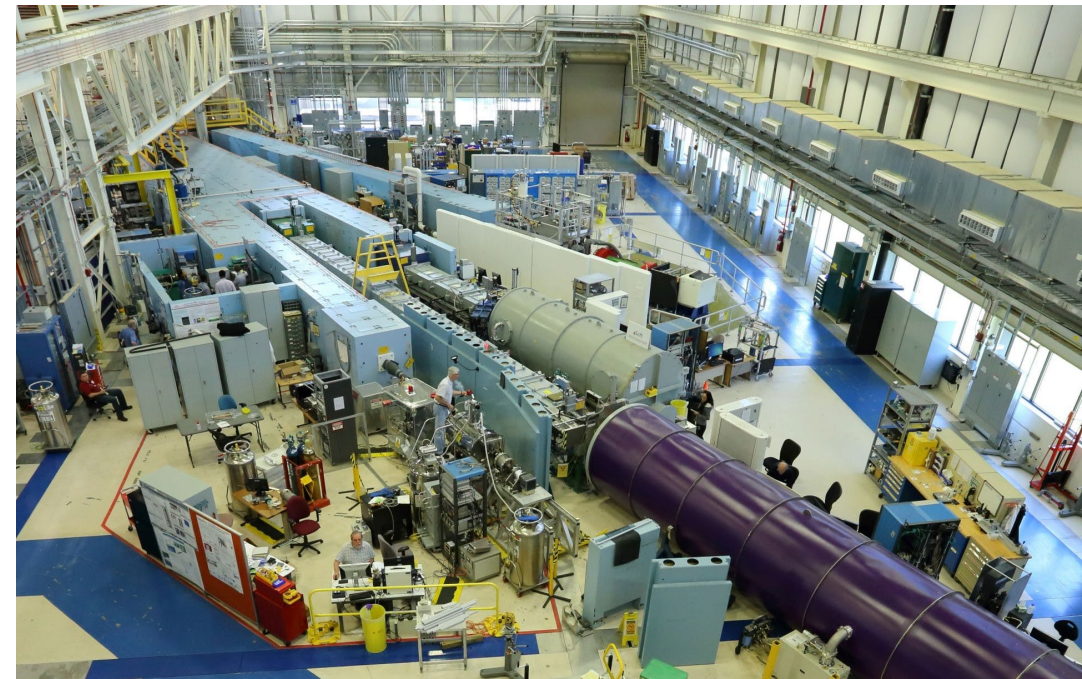
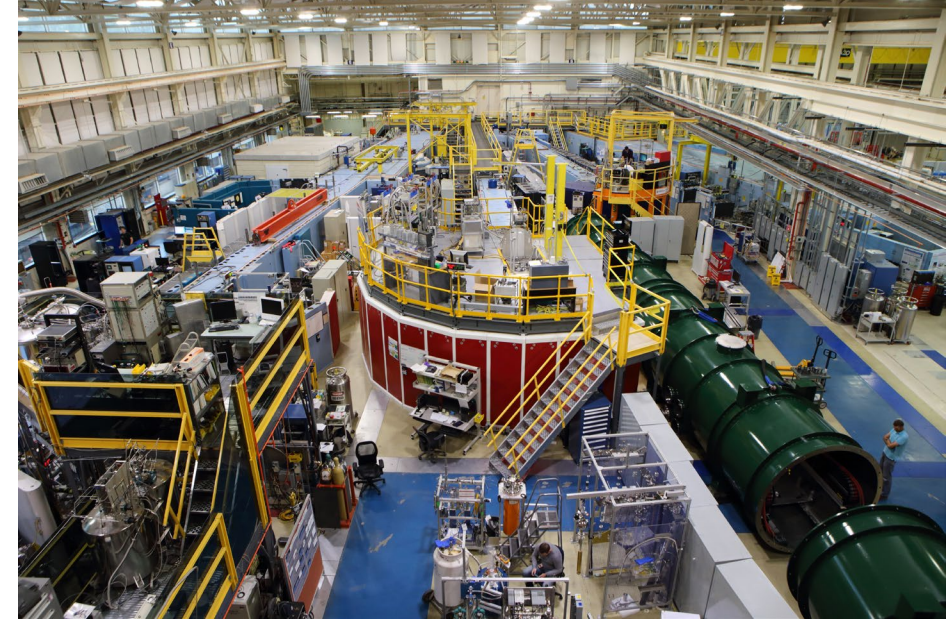
The National Center for Small Angle Scattering Research


NCSAR

- NCSAR conceived as a National User Facility for Small Angle Research
(A NOVEL IDEA IN THE US at the time!)
- Limited SANS availability in the US: Biology at BNL, metallurgy at ORR-ORNL
- Only ILL a truly User Facility. Julich and Saclay also focused programmatic efforts
- Proposed to NSF/DOE in 1977 to serve a larger community of University and Industry
- Approved in Jan1978 as a joint NSF/DOE program
 - New 30m SANS at HFIR
 - Existing 5m SANS at ORR
 - A SAXS facility
 - A double perfect crystal SANS at HFIR
- 30m SANS completed in mid 1980
- First publication in 1981


Sister reactors in the US and a major swing-and-miss

Department of
Commerce / NIST /
was NBSR /
now NCNR

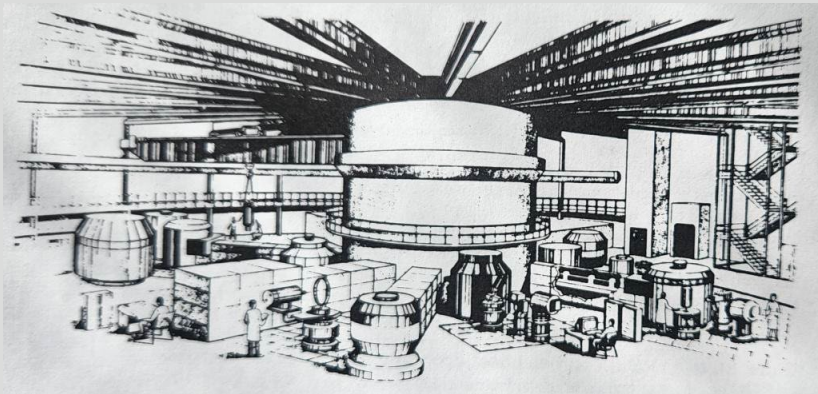




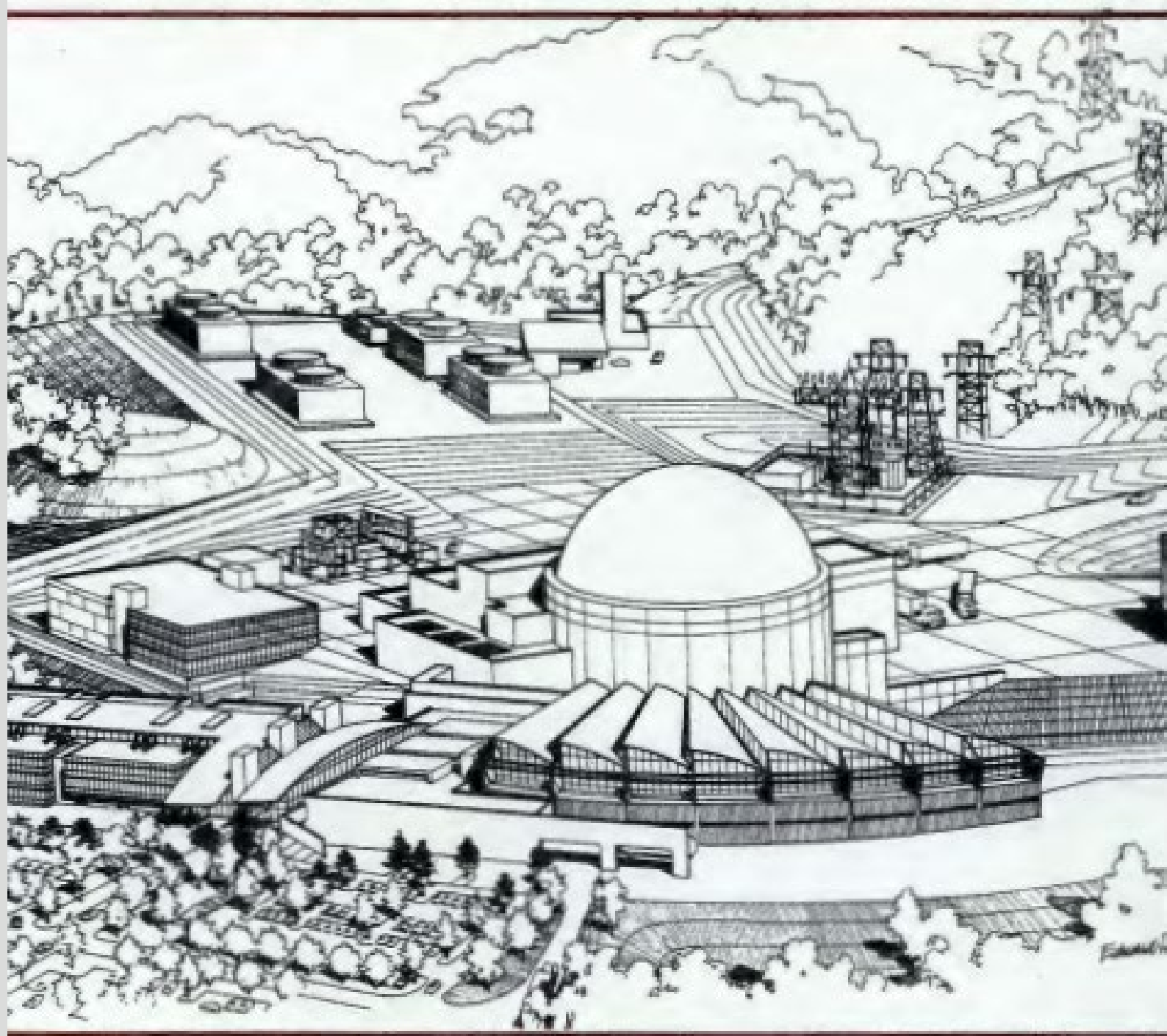
High Flux Beam Reactor (HFBR) Sister reactor at BNL (1965-1996)

The Leak 
**Politics,
Activists, and
Loss of Trust
at Brookhaven
National
Laboratory**
Robert P. Crease with Peter D. Bond

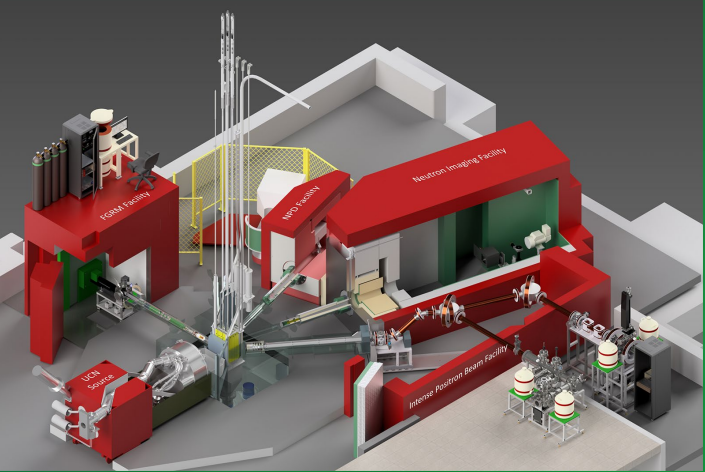
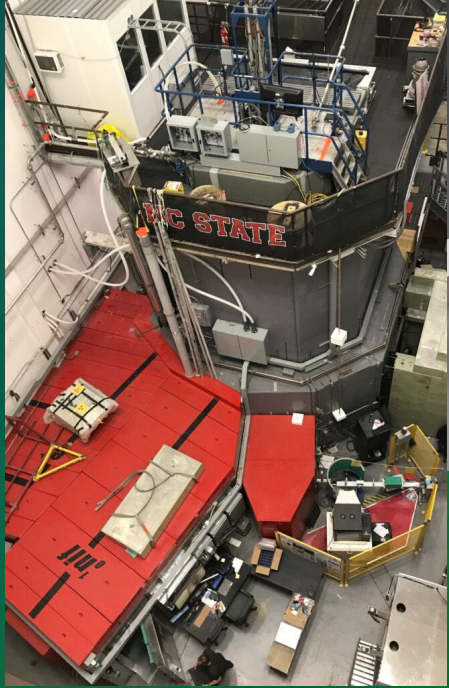
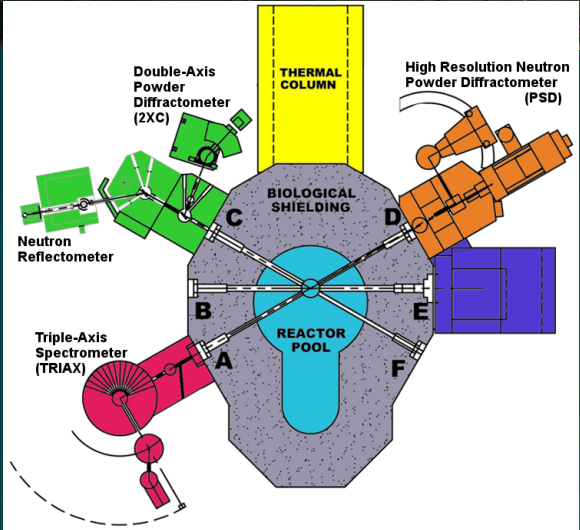
Proposed research reactor, Advanced Neutron Source, ORNL (1990-1996)



<https://www.ornl.gov/blog/advanced-neutron-source-projects-demise-resulted-anothers-rise>



Smaller, university-based research reactors in US with neutron scattering capabilities



**HFIR, Changing Be reflector, new cold source & guide hall,
with another round coming!**

A new cold source and guide hall for HFIR

Exterior of building for the Center for Neutron Scattering - Cold Guide Hall. Initial helium heater test at High Flux Isotope Reactor. HFIR's cold source has passed tests on its refrigeration system, clearing the way for researchers to begin receiving cold neutrons, September 29, 2006.

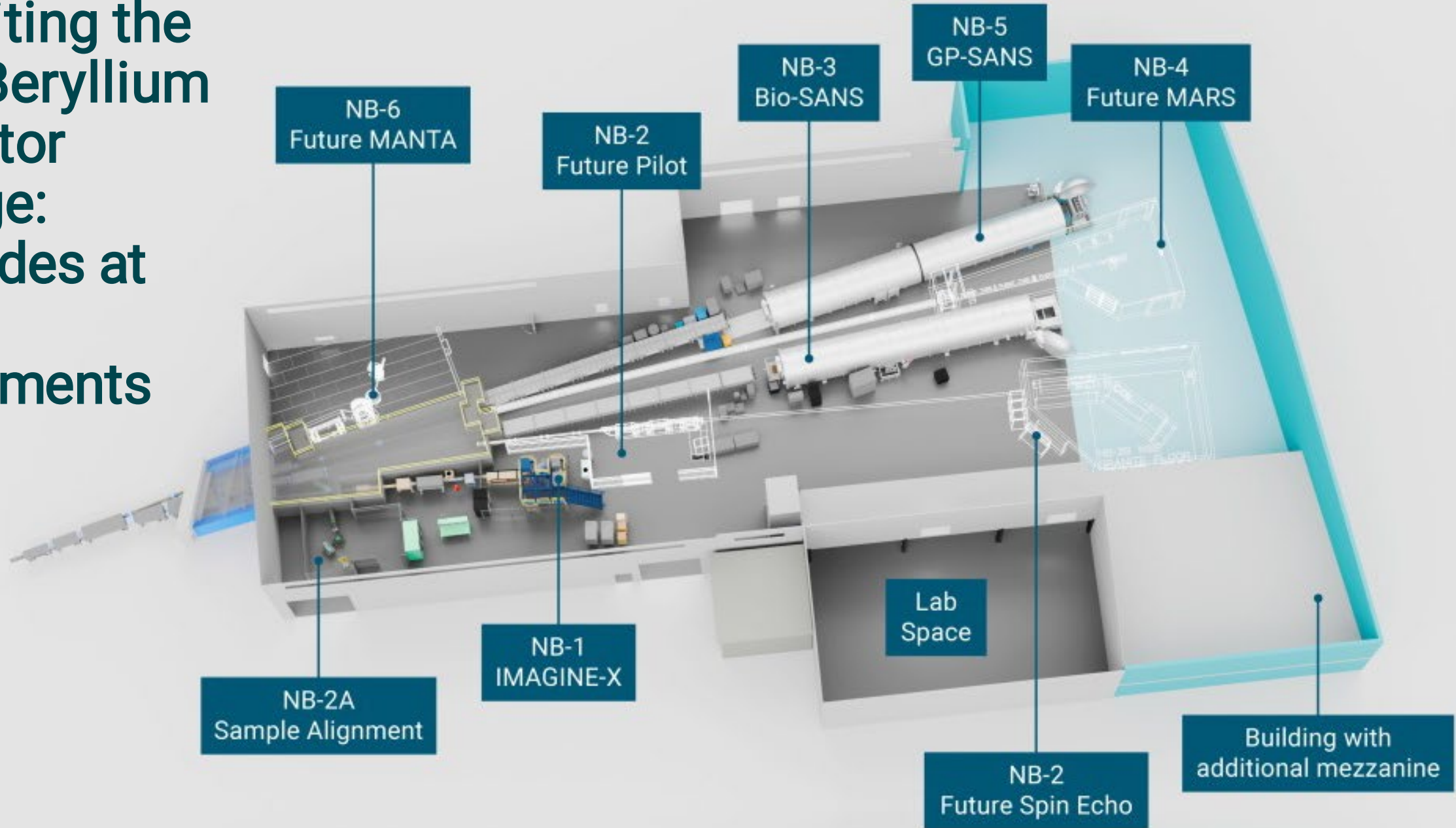


Setting up inside

Cold Guide Hall guide hall,
January 3, 2008.



Exploiting the next Beryllium reflector change: upgrades at HFIR instruments



Pulsed sources & the Spallation Neutron Source

Pulsing the steady state source at HFIR

Ferrite crystal ${}^7\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$

1,000 Gauss, pulsed

Both periodic and correlation

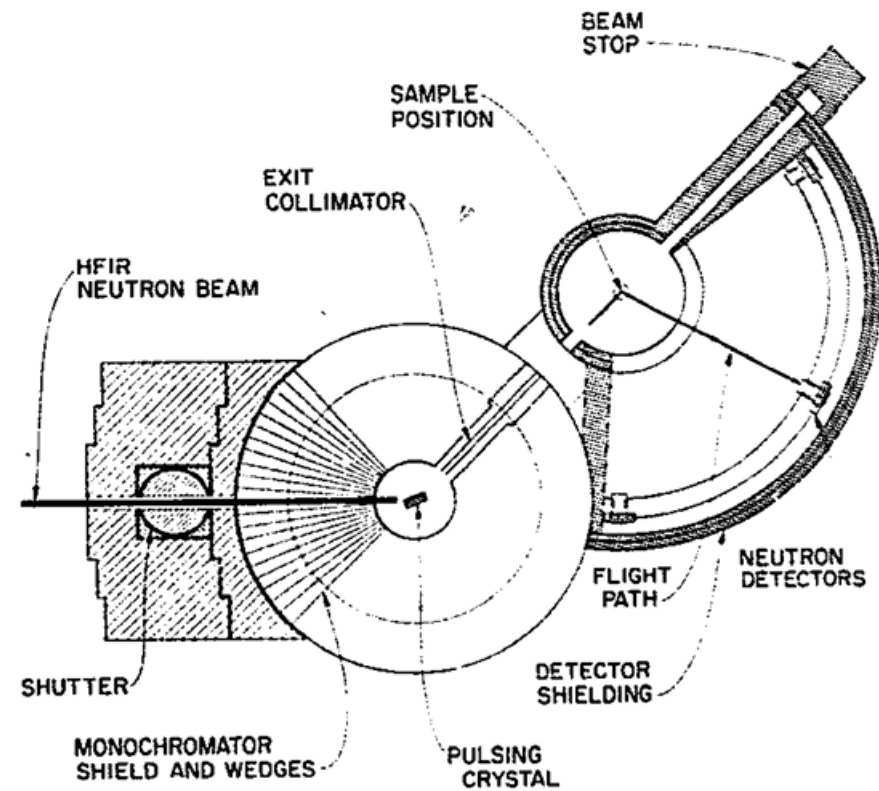


Fig. 4. Layout of the time-of-flight spectrometer installed at the HFIR.

NUCLEAR INSTRUMENTS AND METHODS 116 (1974) 205-216; © NORTH-HOLLAND PUBLISHING CO.

A MAGNETICALLY PULSED NEUTRON TIME-OF-FLIGHT SPECTROMETER FOR INELASTIC SCATTERING*

H. A. MOOK[†], F. W. SNODGRASS⁺ and D. D. BATES⁺

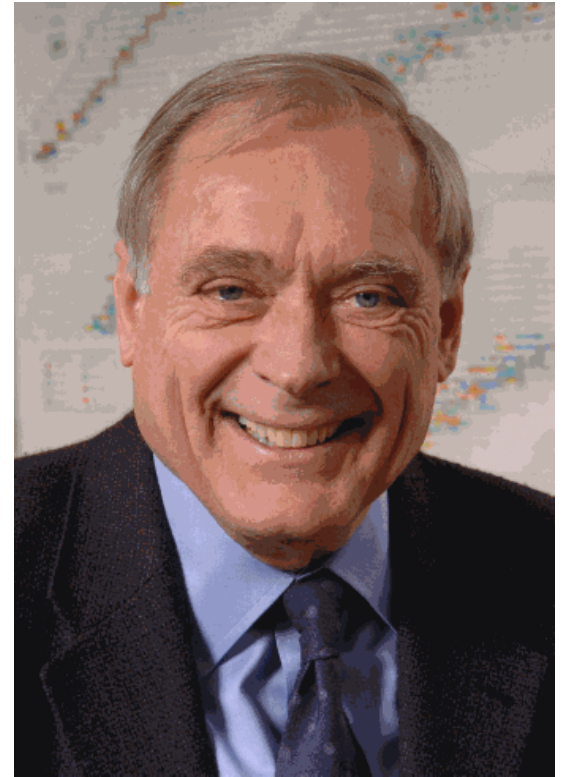
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830, U.S.A.

Received 26 November 1973

Pulsed spallation source, ZGS Intense Neutron Generator (ZING) preceded IPNS @ Argonne (1977-2008)

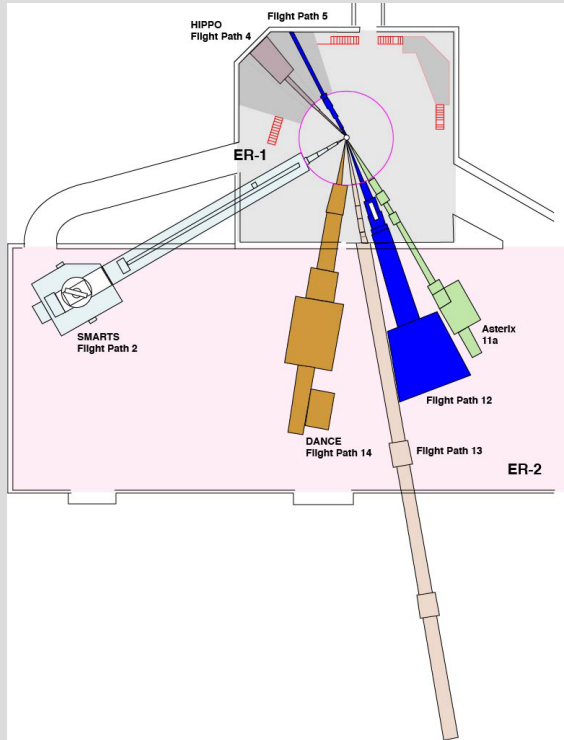


Jack Carpenter, (1936-2020)



ANL/HIST-5

Pulsed spallation source, Lujan @ LANSCE @ Los Alamos: User program (1977-1995-2012) *Partial user program continues*



Manuel Lujan Jr. Neutron Scattering Center

Low energy neutron source (LENS) @ U Indiana-Bloomington

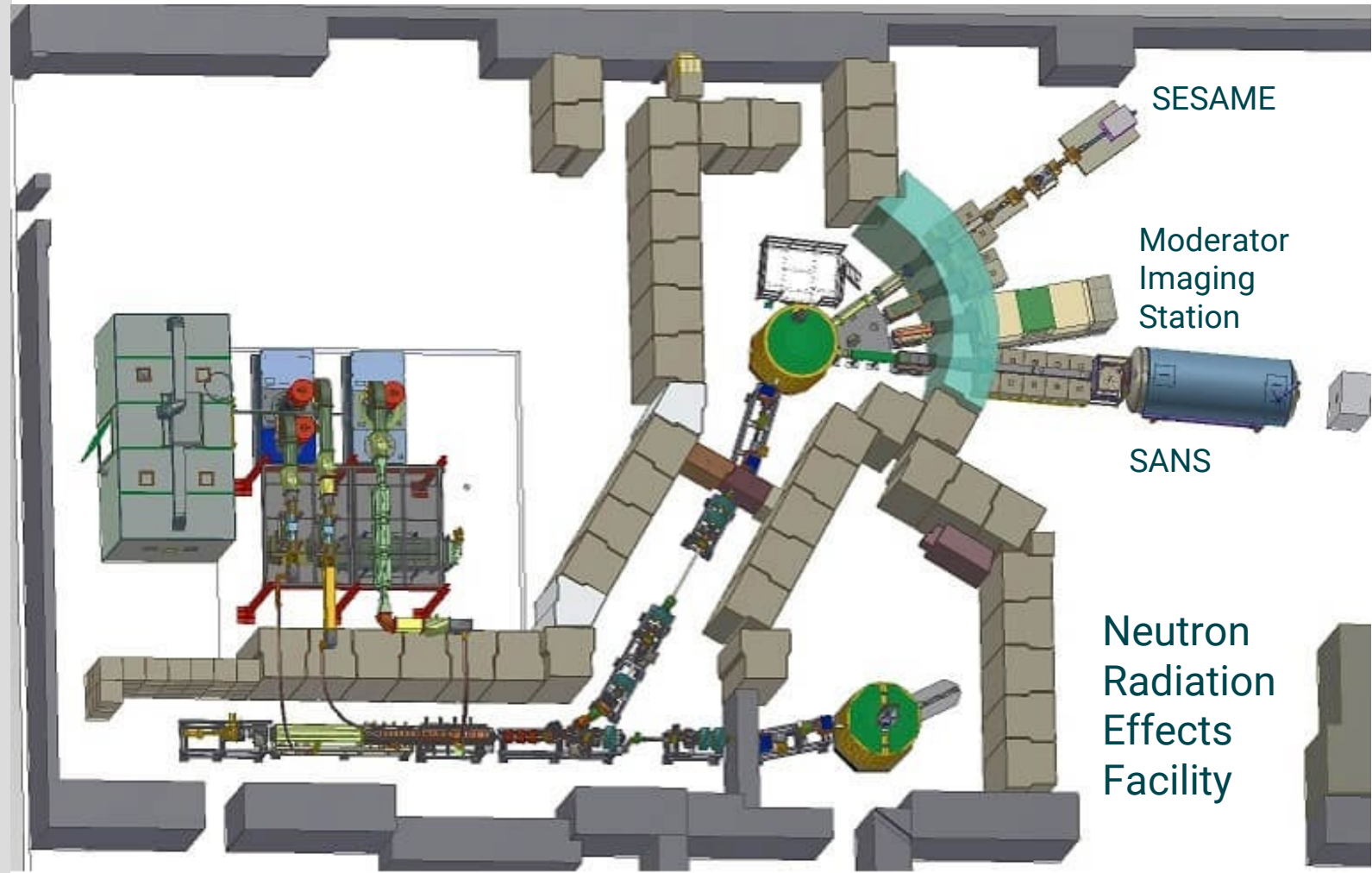


Roger Pynn

Difficult to keep open (may now be closed)

Moderator innovations for SNS tested here

Innovative optics for polarization analysis and Larmor techniques (enhance resolution in 'one direction')



Spallation Neutron Source

Spallation Neutron Source (SNS)
construction site, August 1, 2003

BASIS in foreground

Office complex in background



SNS celebration in the Central Control Room after the first neutron pulse hit the Target, April 28, 2006.



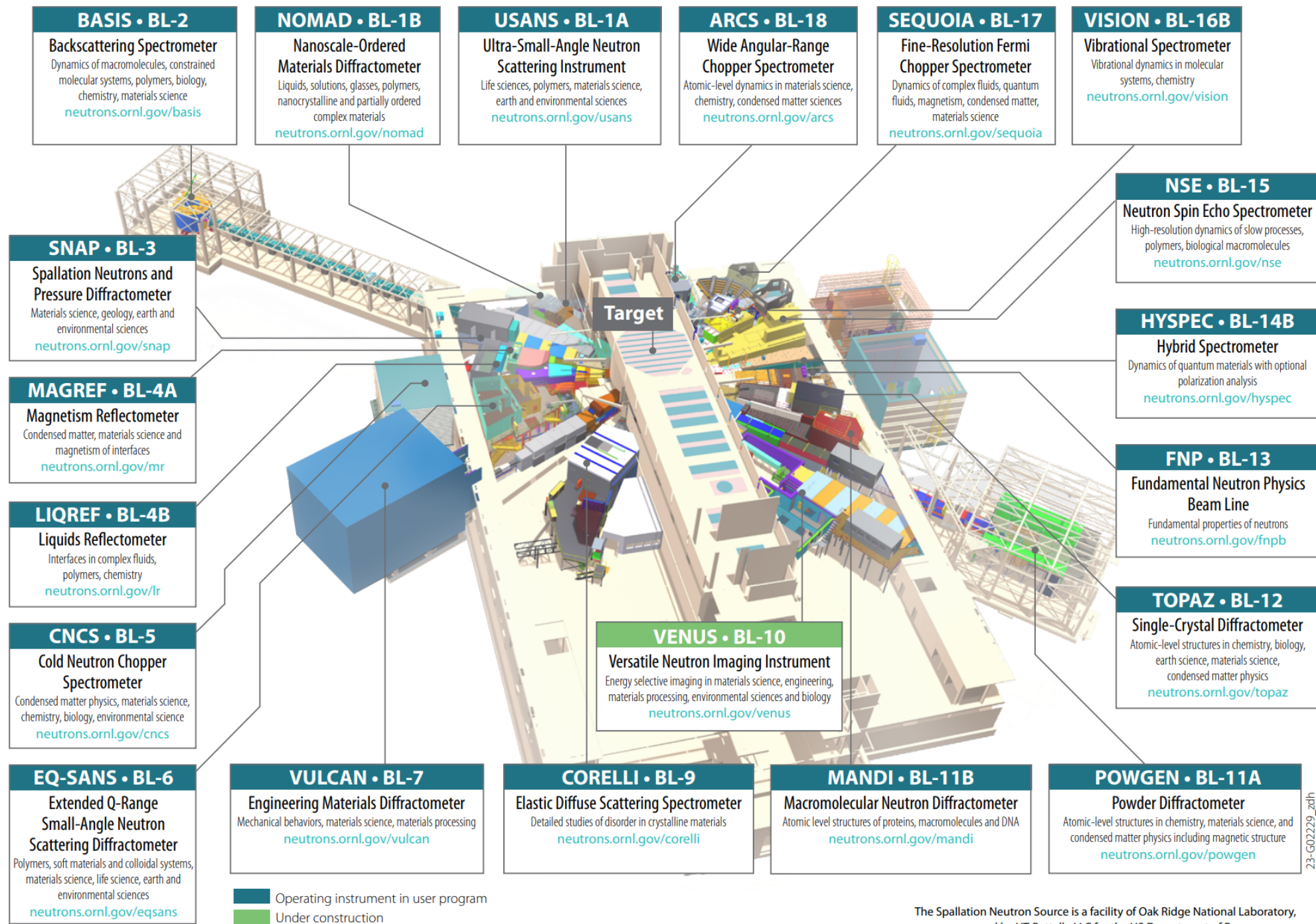
Gradual roll-out of instruments at the SNS

SNS capital project: ~4 instruments

A few 1-off's

SING (SNS Instruments Next Generation) ~5 instruments

SING II ~5 instruments



Joint Institute for Neutron Sciences, later Schull Wollan Center

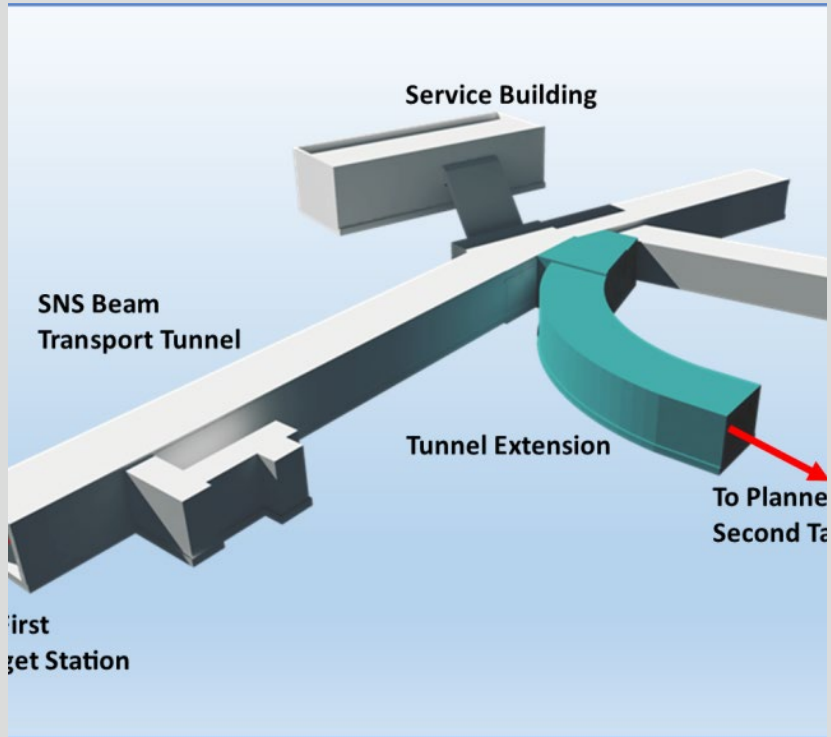
June 21, 2011



Proton Power Upgrade Project:

Increased signal & S:N (1.4 to 2 MW with better ortho-para mix),

& Paving the path to the Second Target Station



Science recap, ORNL neutron scattering

Some ORNL Neutron Scattering Accomplishments

- Measurement of the neutron scattering amplitudes of over 60 elements and isotopes.
- First direct evidence of antiferromagnetism.
- Confirmation of the Néel model of ferrimagnetism.
- First use of neutrons to determine the structure of hydrides.
- First observation of magnetic critical scattering.
- First measurements of 3d and 4f electronic form factors.
- First measurements of magnetic moment distributions in 3d alloys.
- Determination of the magnetic structures of rare-earth metals.

Some ORNL Neutron Scattering Accomplishments

- Development of the neutron polarization analysis technique
- Phonon anomalies in superconductors
- Magnetic excitations in 3d and 4f metals
- Measurement of Bose condensate in liquid helium
- Form factors of paramagnetic metals
- Phonons and magnetic excitations in high-Tc superconductors
- Structure & Excitations in Manganites
- Excitations in quantum magnets

Some ORNL Neutron Scattering Accomplishments

- Core-shell morphologies and film formation in polymer latexes.
- Compatibility, segregation, and interactions in polymer blends (alloys).
- Quantification of isotope effects and phase segregation in deuterium-labeled polymer mixtures.
- Order-disorder transitions in block copolymers and metal alloys.
- Structure and interactions in micelles, microemulsions, and ferrofluids.
- Flux-line lattices in high-T_c superconductors.

Now it's YOUR turn to add to our rich legacy!

