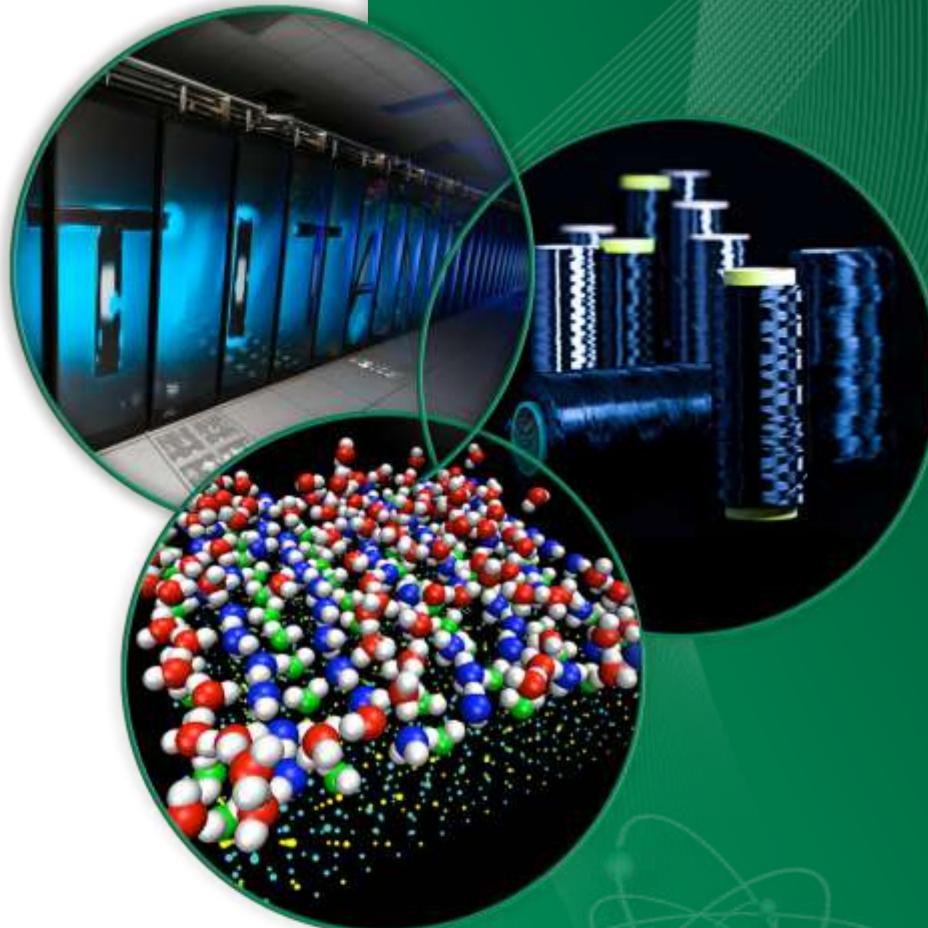
# The Early Development of Neutron Diffraction:

## Science in the Wings of the Manhattan Project



Presented to the

**Bragg Symposium: Celebrating 100 years of X-Ray Crystallography**


**Thomas E. Mason**
Director


Adelaide, SA, Australia
December 6, 2012

# Neutron scattering: The pre-reactor period

1930 | 1932 | 1934 | 1936 | 1938 | 1940

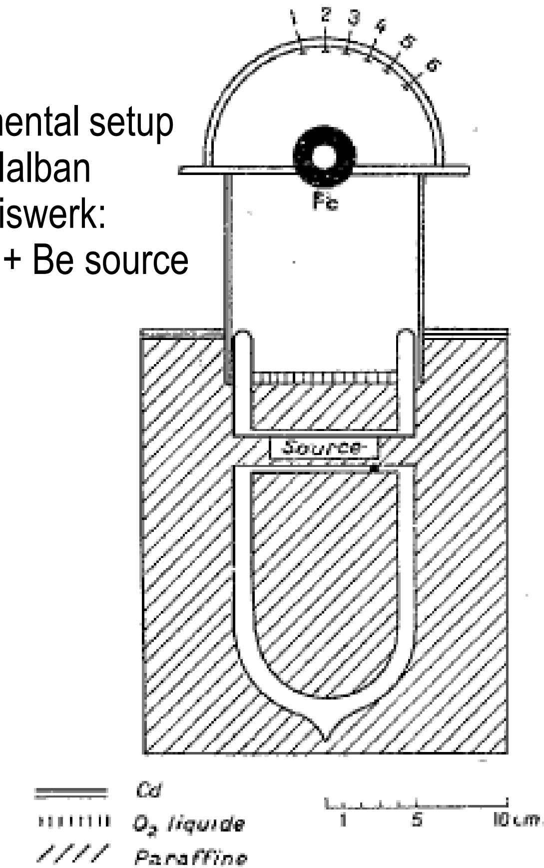
Chadwick's discovery of the neutron

Fermi's observation of neutron thermalization

Demonstrations of coherent neutron diffraction (Bragg scattering by crystal lattice planes)

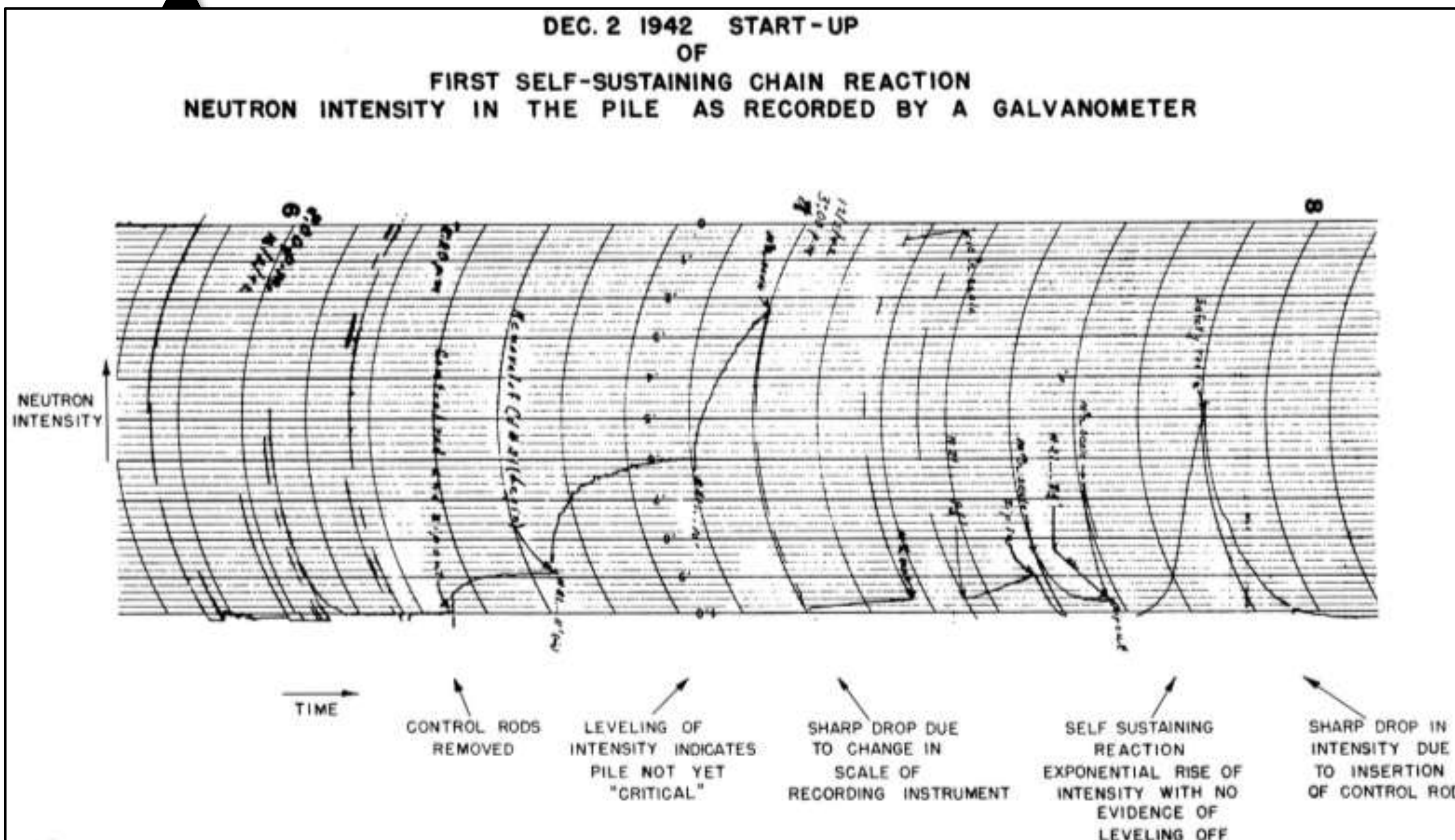
- Mitchell and Powers
- von Halban and Preiswerk

Experimental setup of von Halban and Preiswerk: 1 Ci Rn + Be source



# Successful operation of CP-1 marked the start of a new era

1942 1944 1946 1948 1950



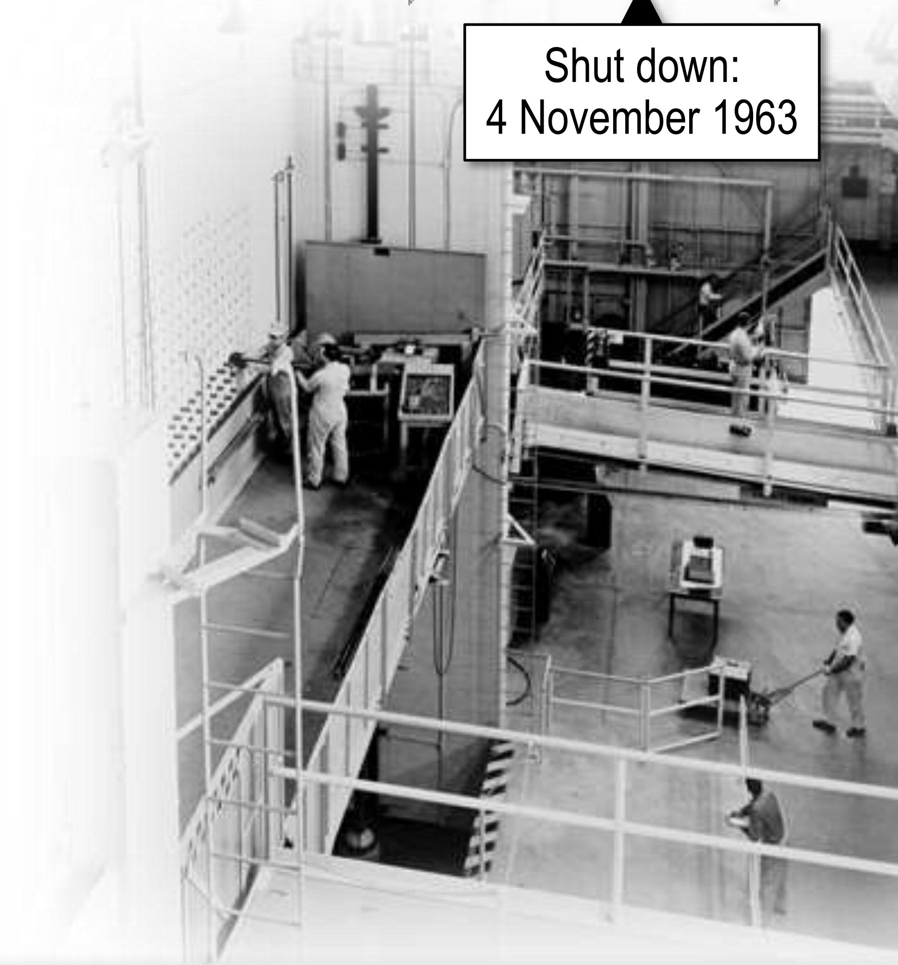
# Early nuclear reactors: The X-10 Pile (Graphite Reactor)

1940 1942 1944 1946 1948 1950 1952 1954 1956 1958 1960 1962 1964

First criticality:
4 November 1943

Shut down:
4 November 1963

| | |
|---|---|
| **Location** | Clinton Laboratories (later Oak Ridge National Laboratory), Tennessee |
| **Fuel** | Natural uranium |
| **Power** | 1,000 kW |
| **Moderator** | Graphite |
| **Coolant** | Air |
| **Flux** | $1 \times 10^{12}\ cm^{-2}\ s^{-1}$ |



# Early nuclear reactors:
# CP-3 (Chicago Pile 3)

1940 | 1942 | 1944 | 1946 | 1948 | 1950

First criticality:
15 May 1944

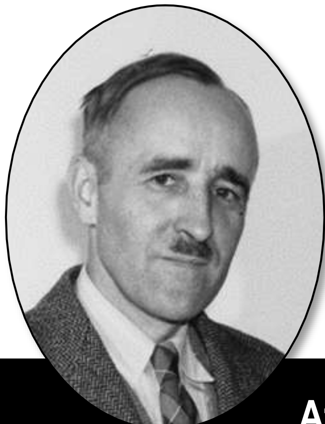
Shut down:
(later rebuilt as CP-3′)

| | |
|---|---|
| **Location** | Chicago Metallurgical Laboratory, Site A (later Argonne National Laboratory), Illinois |
| **Fuel** | Natural uranium |
| **Power** | 300 kW |
| **Moderator** | Heavy water |
| **Coolant** | Circulated moderator |
| **Flux** | $1 \times 10^{12}$ cm$^{-2}$ s$^{-1}$ |

Courtesy Argonne National Laboratory

# Early nuclear reactors: Neutron experimenters

At Clinton



Ernest O. Wollan

Lyle B. Borst
(Courtesy Brookhaven National Lab)

At Chicago

Enrico Fermi
(Courtesy Argonne National Lab)

Walter H. Zinn
(Courtesy Argonne National Lab)

# May 1944: Wollan formally proposed to measure neutron diffraction at X-10

[redacted] CLINTON LABORATORIES

To R. L. Doan
From E. O. Wollan
In re: Diffraction of neutrons

Department
Department

Date May 25, 1944

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

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

EOW/o

CLASSIFICATION CANCELLED
DATE
For The Atomic Energy Commission
Chief, Declassification Branch

RECEIVED
MAY 26 1944
R. L. DOAN

[redacted]

[redacted] contents in any manner to an unauthorized person is prohibited by law.



OAK RIDGE NATIONAL LABORATORY
MANAGED BY UT-BATTELLE FOR THE U.S. DEPARTMENT OF ENERGY

# Experiments by Wollan and Borst in summer 1944 were unsuccessful

May 31 July 8 July 12

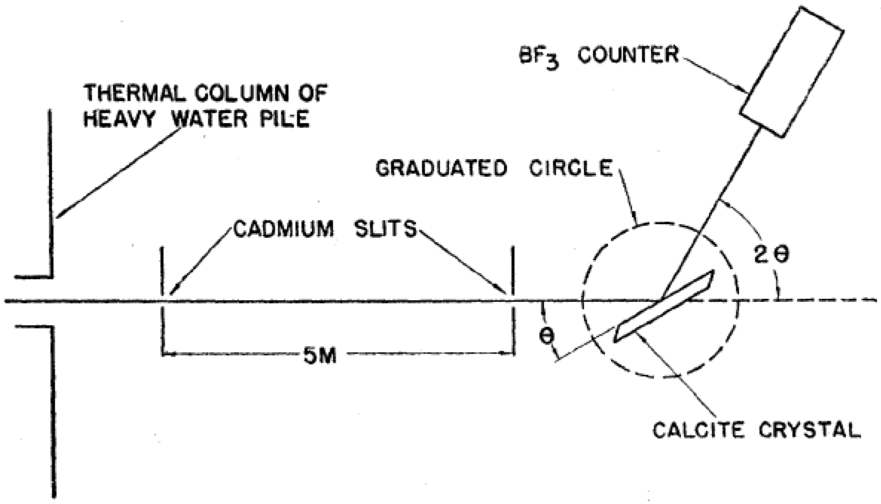
**Neutron Crystal Spectrometer - E. Wollan, L. Borst**

A neutron crystal spectrometer was brought down by E. O. Wollan. This was set up on the experimental face of the pile using as a beam the neutrons emerging from an open foil slot. The gypsum crystal used was 4 x 6 x 3/4" and apparently well-suited to the test. A $BF_3$ counter set up and used as a detector showed satisfactory amounts of scattered radiation. This radiation varied with angle but showed no sharp Laue maxima. The apparatus was removed until a diagnosis of the difficulty could be made.

307-X29P **Diffraction of neutrons.** The sample of calcite requested from Chicago was mounted in a neutron beam and photographic plates placed behind it to obtain a Laue diffraction pattern. These plates will be removed and developed on Monday.

307-X29P **Neutron diffraction.** No successful results were obtained from the photographic plates recently exposed to neutrons presumably diffracted from calcite. Arrangements are being made to use a $BF_3$ counter to detect neutrons diffracted from an epi-cadmium neutron beam. This beam emerges from hole 60 and is 3/4" square.

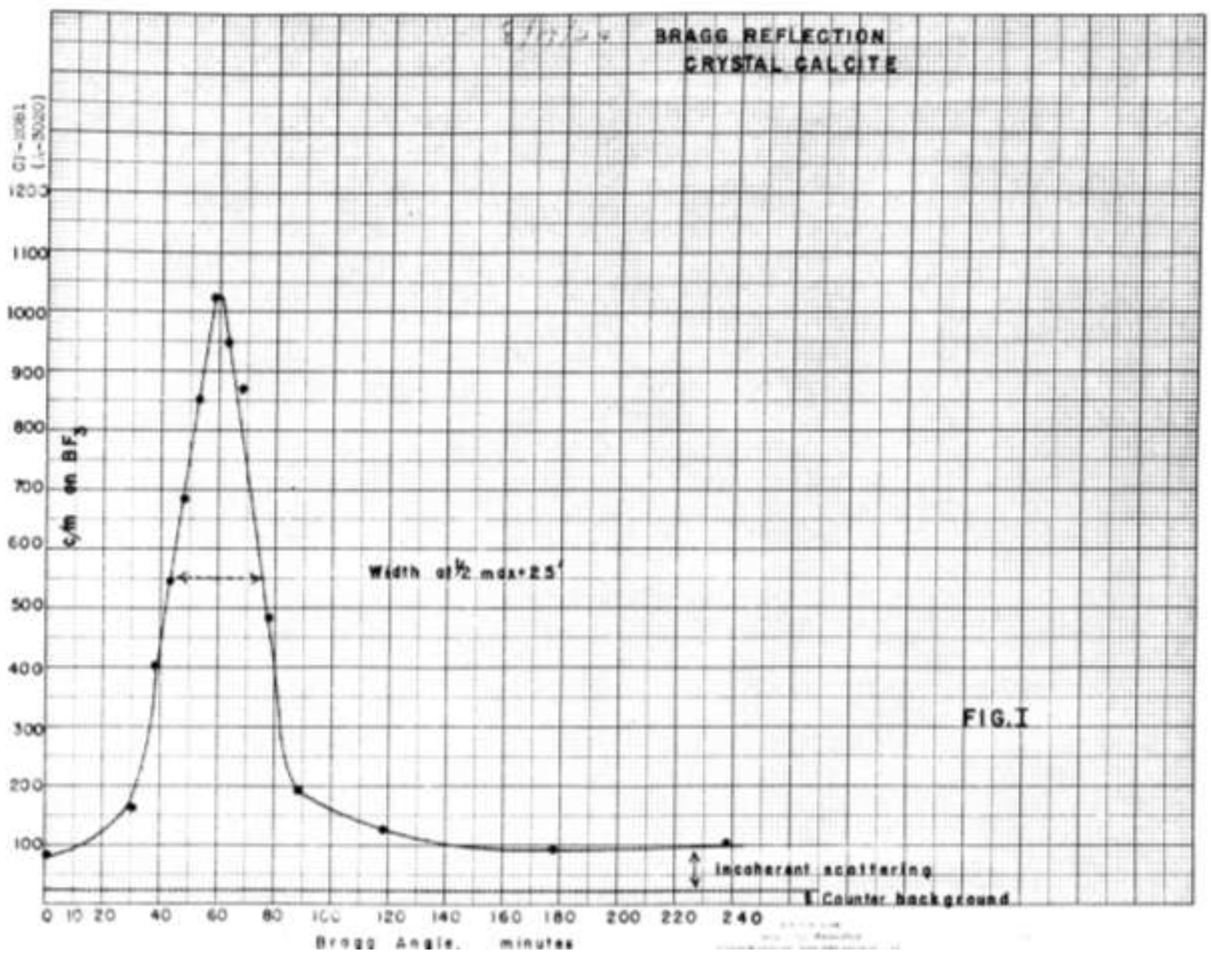
# July 1944: Fermi and Zinn initiated neutron diffraction experiments on CP-3



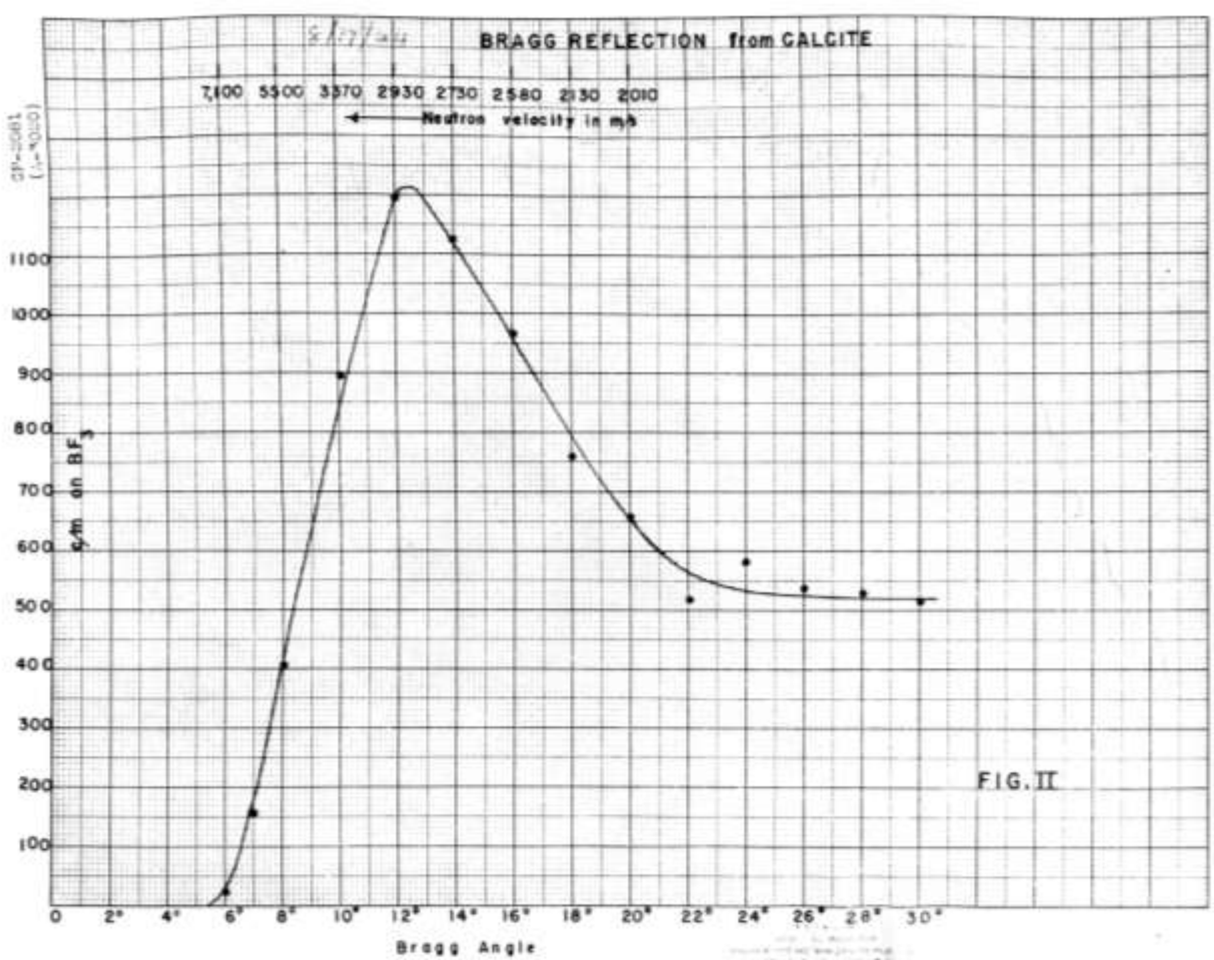

First experimental setup at CP-3
[W. H. Zinn, *Phys. Rev.* **71,** 752–757 (1946)]

"The crystal, having the dimensions 6 cm. x 12 cm., was mounted on the table of a spectrometer with two divided circles. One circle measured the position of the crystal and the other the position of an arm which carried the $BF_3$ counters. The two circles could be geared together so that the counter always moved through twice the angle of the crystal. The distance from the crystal to the counter was 135 cm."

# August 1944: “Typical crystal rocking curve” reported by Zinn



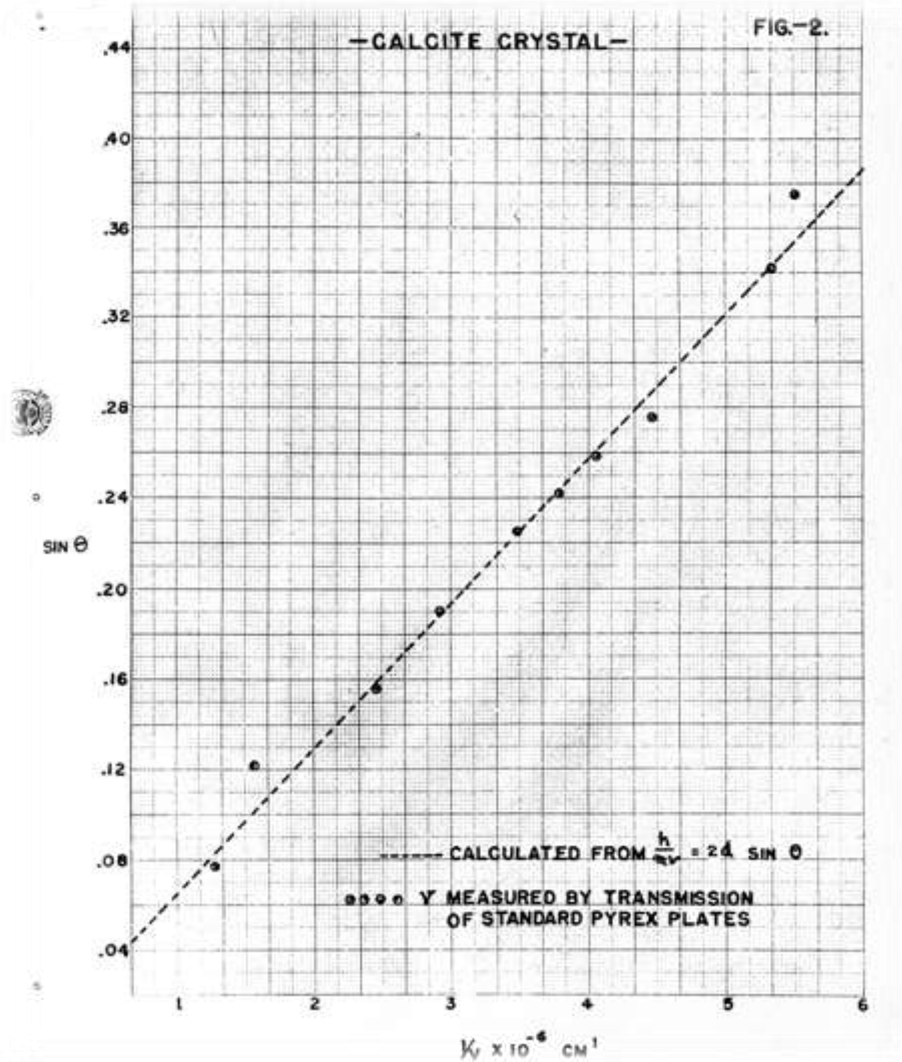
# August 1944: Diffraction measurement reported by Zinn



# Zinn continued his experiments with William Sturm

"The Bragg reflection of thermal neutrons from a crystal has been further investigated from the viewpoint of determining its usefulness as a neutron spectrometer.  It has been felt that the intensity available was sufficient to permit examinations of samples of rather small cross-sectional area."

– Metallurgical Project progress report, November 25, 1944



# Theoretical investigations of neutron diffraction were initiated at Chicago

- 1944: Classified reports acknowledged experiments only by Zinn at Argonne and Borst at Clinton
  - Goldberger and Seitz, *The Diffraction of Neutrons by Crystals I,* CP-2419, November 25, 1944 (declassified February 1, 1954)
  - Seitz and Goldberger, *The Diffraction of Neutrons by Crystals II,* MDDC-1036, December 20, 1944 (declassified June 26, 1947)
- 1947: Attribution expanded to include Fermi at Argonne and Wollan at Clinton

PHYSICAL REVIEW VOLUME 71, NUMBER 5 MARCH 1, 1947

**Theory of the Refraction and the Diffraction of Neutrons by Crystals**

M. L. GOLDBERGER, *Institute for Nuclear Studies, University of Chicago, Chicago, Illinois*

AND

FREDERICK SEITZ, *Physics Department, Carnegie Institute of Technology, Pittsburgh, Pennsylvania*

(Received November 27, 1946)

The equations for the elastic scattering of neutrons by a single crystal, governing simple refraction, reflection, and Laue-Bragg scattering, are derived under the assumption that the scattering and absorption cross sections are independent of spin. A brief summary of the results is given in the final section of the paper.

### 1. INTRODUCTION

THE experiments on the scattering of neutrons by single crystals carried out at the Argonne Laboratories by Fermi and Zinn and at the Clinton Laboratories by Borst and Wollan have given conclusive evidence that an appreciable part of the scattered intensity is coherent. In fact, a substantial part of the scattering takes place in accordance with the Laue-Bragg equations. The purpose of the present paper is to investigate the theoretical background for the scattering somewhat more fully than has been done previously[1] and to summarize the results in a form that may be of use in the course of the development of the experimental work.

The scattering of slow neutrons by crystals has been of considerable interest since the earliest days of neutron physics. Following Fermi's development[2] of a simplified or semi-empirical method of treating the interaction between neutron and nucleus, Wick[3] showed that one should expect highly crystalline media to scatter slow neutrons in a way that is very different from that expected for gases or completely amorphous materials because of the interference effects which occur. Wick's work has formed the basis for much of the theoretical work in this field. Following a very similar line of reasoning, Teller[4] pointed out that one should expect ortho- and parahydrogen to possess markedly different scattering cross sections because of interference, provided the large cross section for the scattering of slow neutrons by protons could be ascribed to a virtual singlet level of the type used in the Breit-Wigner formalism of resonance scattering. The subsequent experimental verification[5] of the quantitative predictions of Teller and Schwinger[4] on the basis of Teller's original suggestion demonstrated that one can expect to obtain a reasonable description of the scattering of slow neutrons by polyatomic systems with the use of wave mechanics and the Breit-Wigner formalism.

In the period following Wick's work, numerous investigators extended his treatment of the scattering of neutrons by crystals. Most prominent among these are the work of Pomerantschuk,[6] Van Vleck,[7] Halpern, Hamermesh and Johnson,[8] Seeger and Teller,[9] and Weinstock.[10] Pomerantschuk examined more carefully than Wick the influence of low temperatures upon the scattering cross section. Van Vleck investigated the scatter-

[1] This document is based on work performed under Contract No. W-35-058-eng-71 for the Manhattan Project. Part of the information covered in this document appeared in Report CP-2419, and a more complete survey will appear in volume IIIB, Division IV of the Manhattan Project Technical Series, as part of the contribution of the Clinton Laboratories. Some of the results derived in this report, particularly those for the index of refraction, were derived earlier by Fermi for simple cases. Fermi's measurements of the total reflection of thermal neutrons by graphite and subsequent measurements of Bragg scattering by Zinn and Borst furnished the incentive for much of the work described here. Professor W. E. Lamb has informed us that he investigated theoretical aspects of the problem of neutron refraction as early as 1940.

[2] E. Fermi, Ricerca Scientifica, **7**, Part 2, 13 (1936).

[3] G. C. Wick, Phys. Zeits. **38**, 403 (1937).

[4] J. Schwinger and E. Teller, Phys. Rev. **51**, 775 (1937); J. Schwinger and E. Teller, Phys. Rev. **52**, 286 (1937).

[5] J. Halpern, I. Estermann, and O. Stern, Phys. Rev. **52**, 142 (1937); L. W. Alvarez and K. S. Pitzer, Phys. Rev. **55**, 596 (1939).

[6] I. Pomerantschuk, Phys. Zeits. Sowjetunion **13**, 65 (1938).

[7] J. H. Van Vleck, Phys. Rev. **55**, 924 (1939).

[8] O. Halpern, M. Hamermesh, and M. H. Johnson, Phys. Rev. **59**, 981 (1941).

[9] R. J. Seeger and E. Teller, Phys. Rev. **62**, 37 (1942).

[10] R. Weinstock, Phys. Rev. **65**, 1 (1944).

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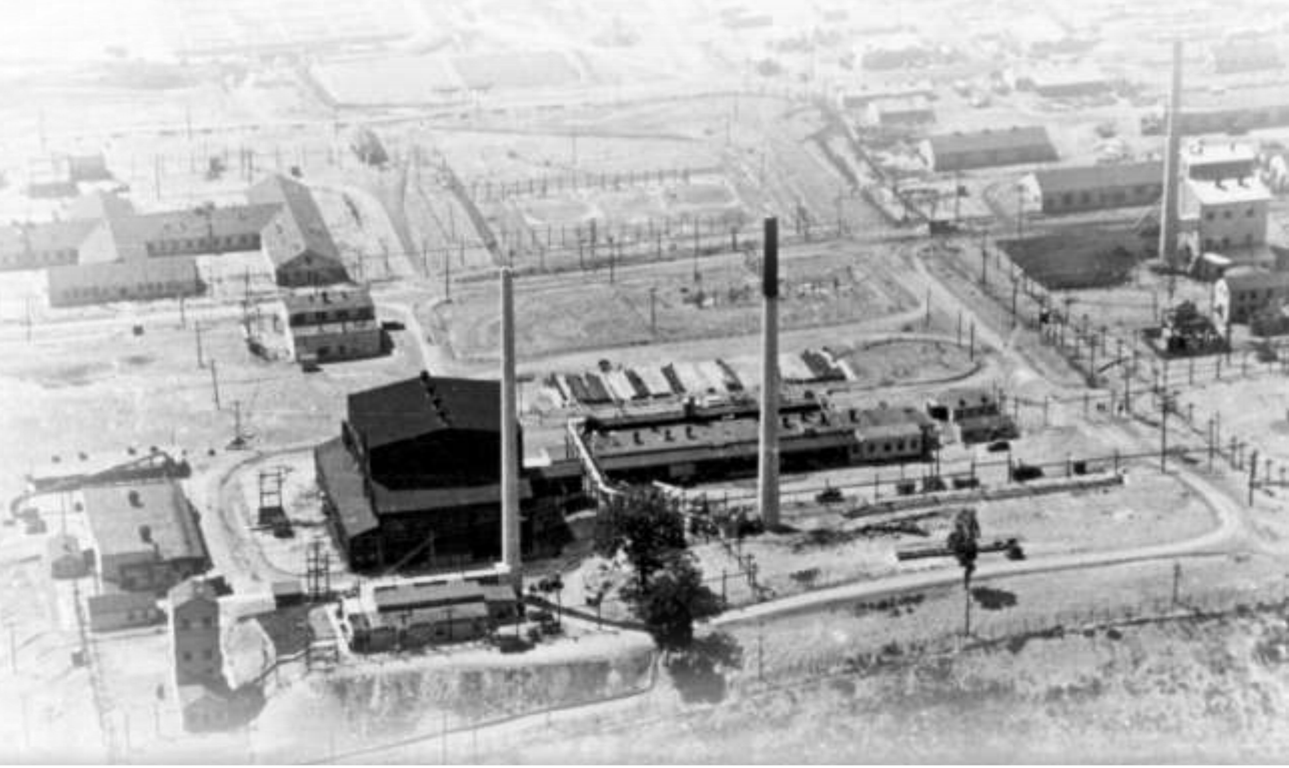
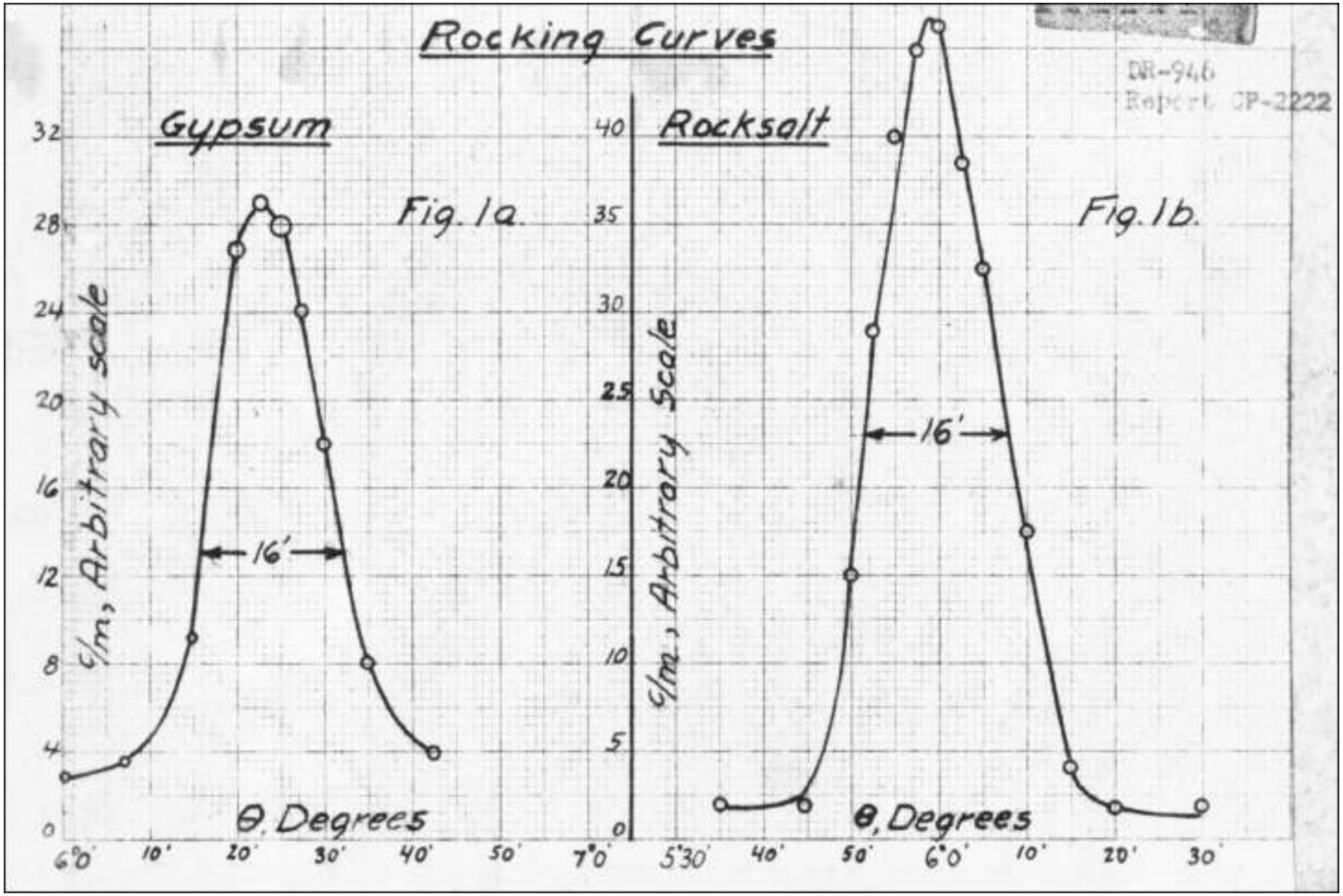
# December 1944: Wollan reported success at Clinton

| Upgraded equipment | Other improvements | Source of neutrons |
|---|---|---|
| • Diffraction of neutrons from gypsum and rock salt achieved using “much more satisfactory” equipment:<br>– X-ray spectrometer borrowed from University of Chicago Physics Department<br>– 30 cm long, 5 cm diam $BF_3$ proportional counter filled to ~20 cm Hg pressure and used end on to the diffracted beam<br>– Cadmium Soller slit placed in front of counter | • Shielding and “geometrical disposition” of the counter<br>• Thinner crystal slab | • Beam 1/4 in. wide by 3/4 in. high |



# Rock salt offered advantages for work "at energies up to 1 ev or more"



# January 1945: Wollan articulated the potential of neutron diffraction

In general, I believe it can be said that neutron diffraction constitutes a very useful and simple physical "tool" when used in conjunction with a pile, and this will be especially so when piles of greater flux are available.

210 This document consists of 2 pages and 0 figures. No. 1 of 5 copies, Series A

[redacted]

CLINTON LABORATORIES

1. R. L. Doan
2. A. H. Snell
3. E. O. Wollan
4. Reading File
5. Central File

DATE Jan. 13, 1945

TO R. L. Doan DEPARTMENT

FROM E. O. Wollan DEPARTMENT

IN RE: Program Relative to Diffraction of Neutrons by Crystals

CLASSIFICATION CANCELLED
Ted Davis 5-16-96
ADD signature Date
Single rereview of CCRP-declassified documents was authorized by DOE Office of Declassification memo of August 22, 1994.

The diffraction of neutrons by crystals has at least two apparent aspects, (1) its use as a "tool" in studying various physical constants as a function of neutron energy in the region from 0.001 ev to about 10 ev and (2) a study of the phenomenon of neutron diffraction itself.

We have made a start on both of these aspects of the problem. Relative to the first we have made what we feel to be a careful study of the absorption by cadmium from 0.05 to 1.0 ev and have shown from this that the Breit-Wigner formula represents the experimental data within very close limits. Relative to the second aspect we have shown that both gypsum and rock salt give good Bragg reflections with neutrons.

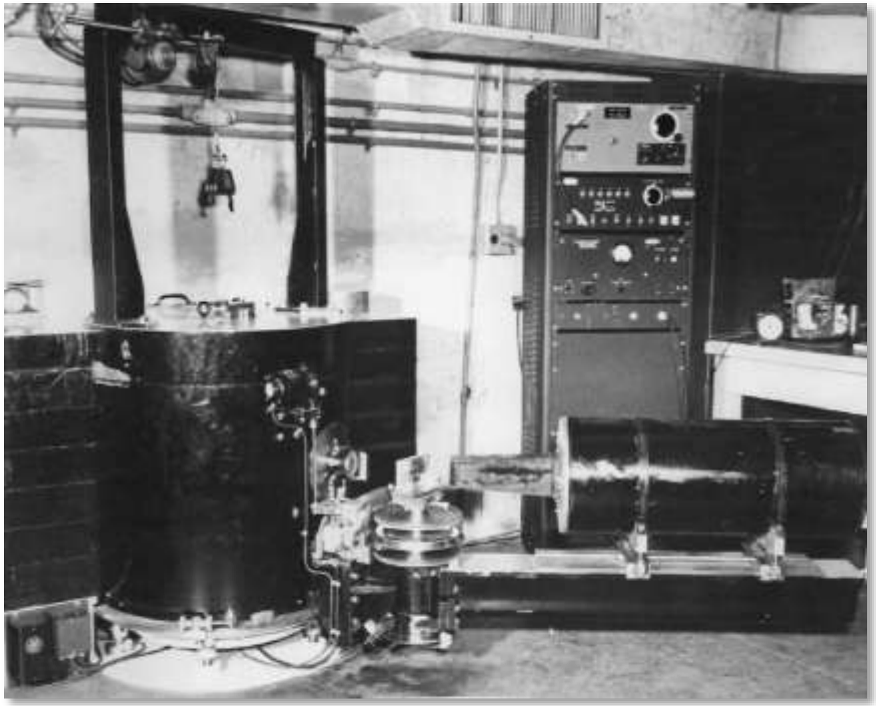
2. A study of the phenomenon of neutron diffraction as related to the type of crystal used is of interest although probably of less importance to the project. We have planned to divert a rather small per cent of our time in this direction. We have ordered a rock salt crystal which can be cut along a 111 plane so the coherent scattering by Na and Cl atoms can be independently determined. This is of importance in checking the diffraction theory as it relates to the role played by atoms of different spins and of different isotopic composition.

Classification Cancelled

In general, I believe it can be said that neutron diffraction constitutes a very useful and simple physical "tool" when used in conjunction with a pile and this will be especially so when piles of greater flux are available.

# Wollan continued his explorations of neutron diffraction on the X-10 pile

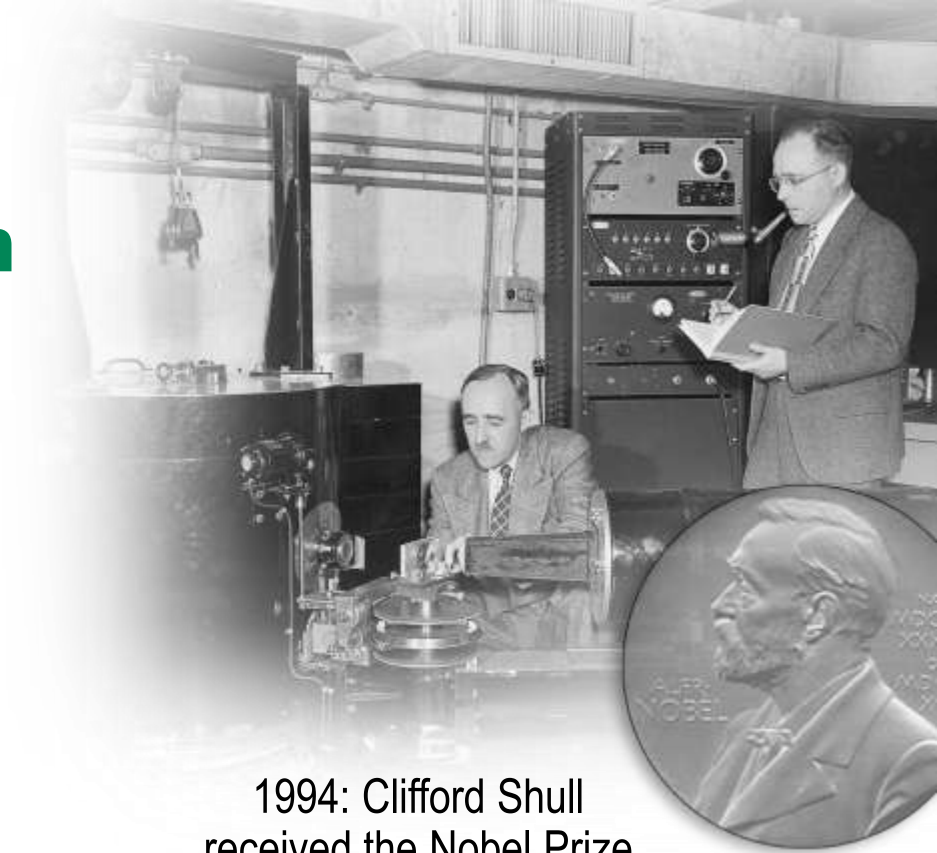
1946: Wollan's double-axis spectrometer, modified for use in powder diffraction studies



1949: Double-axis spectrometer constructed specifically for neutron use

# Wollan and Shull pioneered the field of neutron diffraction

- Development of techniques and instrumentation
- Application to previously insoluble problems in:
  - Crystallography and chemical binding
  - Nuclear studies
  - Magnetism



1994: Clifford Shull received the Nobel Prize in Physics for the development of neutron scattering techniques to analyze condensed matter

# Discussion