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

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December 6, 2012
Neutron scattering: The pre-reactor period

- 1930: Chadwick's discovery of the neutron
- 1932: Fermi's observation of neutron thermalization
- 1934: Demonstrations of coherent neutron diffraction (Bragg scattering by crystal lattice planes)
  - Mitchell and Powers
  - von Halban and Preiswerk
- 1936: Chadwick's discovery of the neutron
- 1938: Fermi's observation of neutron thermalization
- 1940: 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.
### Early nuclear reactors: The X-10 Pile (Graphite Reactor)

| Year | 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

| Property | Value |
|----------|-------|
| Fuel     | Natural uranium |
| Power    | 1,000 kW |
| Moderator| Graphite |
| Coolant  | Air |
| Flux     | $1 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$ |
Early nuclear reactors:
CP-3 (Chicago Pile 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
$1 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$

First criticality:
15 May 1944

Shut down:
(later rebuilt as CP-3')

1940 1942 1944 1946 1948 1950

Courtesy Argonne National Laboratory
Early nuclear reactors: Neutron experimenters

Ernest O. Wollan
(Courtesy Brookhaven National Lab)

Lyle B. Borst
(Courtesy Brookhaven National Lab)

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
Experiments by Wollan and Borst in summer 1944 were unsuccessful.

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 $\text{BF}_3$ 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 $\text{BF}_3$ 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

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

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1. This document is based on work performed under Contract No. W-7405-eng-14 for the Manhattan Project. Part of the information contained in this document appeared in Report CP-1749, and a more complete version will appear in volume III, Division IV of the Manhattan Project Technical Series, as part of the compilation of the Clinton Laboratory. Some of the results derived in this report, particularly those for the theory of reflection, were derived earlier by Fermi for single rays. Fermi's measurements of the total reflection of thermal neutrons by graphite and subsequent measurements of Bragg scattering by Zinn and Bethe furnished the incentive for much of the work described here. Professor W. E. Lamb has informed me that he investigated theoretical aspects of the problem of neutron reflection as early as 1940.

2. G. C. Wick, Phys. Ztschr. 88, 60 (1917).

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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: |
|   − X-ray spectrometer borrowed from University of Chicago Physics Department |
|   − 30 cm long, 5 cm diam BF$_3$ proportional counter filled to ~20 cm Hg pressure and used end on to the diffracted beam |
|   − Cadmium Soller slit placed in front of counter |
| • Shielding and “geometrical disposition” of the counter |
| • 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.

To R. L. Dean
From E. O. Wollan

In re: Program Relative to Diffraction of Neutrons by Crystals

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.
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