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IDO-17022
October 1964

PRELIMINARY STUDIES FOR A
CURVED-CRYSTAL NEUTRON SPECTROMETER

R. R. Spencer, L. G. Miller and M. S. Moore

PHILLIPS
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NATIONAL REACTOR TESTING STATION
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IDO-17022
AEC Research and Development Report
Physics
TID-4500 (34th Ed.)
Issued: October 1964

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Idaho Operations Office

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ABSTRACT

Preliminary studies of the diffraction properties of both plane and curved crystals of various materials have been carried out, pursuant to their application as monochromators in a curved-crystal neutron spectrometer. Most promising of the crystals studied were a flat germanium crystal, a curved NaCl crystal, and a curved beryllium crystal. More detailed studies will be required in order to evaluate the possibility of using these crystals in such a spectrometer.

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PRELIMINARY STUDIES FOR A CURVED-CRYSTAL NEUTRON SPECTROMETER

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PRELIMINARY STUDIES FOR A CURVED-CRYSTAL NEUTRON SPECTROMETER

by

R. R. Spencer, L. G. Miller and M. S. Moore

I. Introduction

The curved-crystal spectrometer, suggested and developed to a precision instrument by DuMond^(1,2), has been used for many years as a device for analysis of gamma and X-ray spectra. As a neutron spectrometer, the curved-crystal monochromator holds the same attractive feature: its use enables one to focus the incident neutrons which fulfill the Bragg condition, and thus to obtain a monochromatic neutron beam which can be orders of magnitude more intense than if a flat crystal were used.*

Recent proposals for a curved-crystal neutron spectrometer at the MTR were based on the assumption that a suitable curved crystal can be obtained. For a neutron monochromating crystal, beryllium is the optimum material. Its coherent scattering length is large and its atomic density is higher than any other material of which large single crystals can be grown. Thus, the number of scattering centers in a diffracting plane is maximal, and the spacing of such planes is small. These two effects lead to the production of higher energy and higher intensity neutron beams at a given Bragg angle.

The obvious superiority of beryllium as a neutron monochromator does not rule out the possibility of using other materials, however.

*See Appendix A for a detailed comparison of the present MTR spectrometer, utilizing a plane crystal, and a proposed curved crystal spectrometer at the MTR.

The technological problems of producing and bending a suitable beryllium crystal are formidable and may even be impossibly difficult. However, the bending of crystals of quartz, germanium, and sodium chloride has been successfully done, and these crystals should be considered as possible second choices for neutron monochromators, should beryllium prove to be unsuitable.

It should be pointed out that two other possibilities exist for obtaining a focussed beam from a beryllium monochromator. The first technique, used successfully by Pattenden⁽³⁾, consists of brazing a plane beryllium crystal to an aluminum backing and slicing the beryllium into small aligned crystals, which can be arranged (by bending the backing to the appropriate curvature) into the form of an approximate curved crystal. The disadvantage of this approach is that the beam loses both intensity and resolution compared to that obtained from a true curved crystal.

The second approach involves the deliberate or accidental introduction of impurities in the melt which cause strains as the crystal grows, creating a situation somewhat analogous to the imposition of an external stress to cause bending. At the MTR, there are beryllium crystals grown from a melt which was known to be somewhat impure. These crystals appear to show some curvature which may have been caused by the presence of inhomogeneous impurities as the crystal solidified from the melt. Although the curvature appears to be non-uniform, there is a strong focussing effect from these crystals, and it may be that they can be used in a spectrometer of the type proposed.

The present report is divided into three sections. The first section describes the unsuccessful results of a research and development

subcontract, with Nuclear Metals, Incorporated, of Concord, Massachusetts, to produce a curved beryllium crystal of a given orientation. The second section describes an experimental study of the properties of germanium and sodium chloride single crystals and the bending of the latter. The third section gives the results of a preliminary study on existing beryllium crystals, which shows that these crystals do exhibit curvature and consequent focussing properties.

II. Studies of a Zone-Refined Beryllium Crystal for Possible Use With a Curved-Crystal Spectrometer

Nuclear Metals, Incorporated (NMI), the only available company with knowledge and experience in the technology of beryllium crystal growing, was engaged to attempt the growing of an oriented beryllium crystal suitable for experiments in bending. The orientation desired was the "c" axis of the crystal parallel to the axis of the approximately 1-1/4" diameter x 3" long cylinder. This orientation would allow use of the very highly reflecting (002) planes as the monochromator in the Cauchois spectrometer. Six zone refining passes were made for purification of the crystal and at least four further passes were made with crystal seeding in an attempt to achieve the desired orientation. It was found that beryllium exhibits a marked tendency to crystallize with the "c" axis perpendicular to the cylinder axis (direction of zone refining). The final zone pass resulted in an orientation of the "c" axis at 70° to the cylinder axis and an "a" axis at $21-1/2^{\circ}$ to the cylinder axis. Figure 1 shows the approximate orientation of the crystal with respect to the cylinder as was determined by X-ray diffraction. From this figure it was determined that the only planes which are suitably

Figure 1 - Orientation of crystal axes of the zone-refined beryllium crystal.

near the proper orientation with respect to the cylinder axis are a set of (222)'s and a set of (121)'s. The crystal was scanned in the MTR crystal spectrometer neutron beam in order to determine something of its mosaic. Figure 2 shows a rocking curve of the beam from a set of planes tentatively identified as (110); this is typical of other Bragg reflections which were observed. This rocking curve shows that the crystal has considerable multiple sub crystals and a quite large resulting angular spread (approximately $2\frac{1}{2}^{\circ}$ in Bragg angle). This is prohibitively large for the sort of resolution desired with the proposed curved-crystal facility. It should be pointed out that this rocking curve was taken with the neutron beam traversing the full thickness of the crystal (at least $1\frac{1}{4}$ ") and there exists a possibility that the multiple crystal effects are entirely in a surface layer. It is planned that further studies using a technique mentioned by Knowles⁽⁴⁾ will be carried out in order to determine whether the interior portion of the crystal might have the required narrow mosaic (10-15 minutes of arc).

III. Studies of Germanium and Sodium Chloride Crystals for Possible Use With a Curved-Crystal Spectrometer

A. Germanium

Recently, investigators have succeeded in constructing bent germanium crystal spectrometers for use in gamma-ray spectroscopy⁽⁵⁾. Since the neutron coherent scattering length for germanium is even larger than for beryllium there may be some set of planes in crystals of germanium suitable for a bent crystal neutron spectrometer. A 1" x 2" x 3" germanium crystal was available at this laboratory and a preliminary neutron diffraction scan was taken. Figure 3 shows the rocking

Figure 2 - Neutron rocking curve for the 110 planes of the zone-refined beryllium crystal.

Figure 3 - Neutron rocking curve for the 331 planes in Germanium

curve for a set of (331) planes at a neutron energy of approximately 0.08 eV. The (331) planes have a spacing of 1.298 Å, which is reasonable to achieve high energies. The width of this diffraction peak is about that of the coarse collimator used (15') and hence reveals nothing of the mosaic width of the crystal. In general, germanium crystals exhibit quite narrow mosaic widths (a typical width is 20 seconds of angular divergence), which are considered to be too narrow to yield adequately intense neutron beams. Consequently, even though the present preliminary results show that measurable intensities can be obtained, further studies on germanium as a monochromator for a curved-crystal spectrometer are not planned unless the alternate approaches described below prove fruitless.

B. Sodium Chloride

Sodium chloride crystals may be bent quite easily⁽⁶⁾. In the present study, a set of bending blocks was constructed of aluminum by milling a 2 meter radius-of-curvature concave cut in an aluminum block and a corresponding convex cut in a second block. A preliminary attempt at bending was carried out on a rather poor crystal approximately 3/16" thick by immersing the crystal sandwiched between the two blocks in a saturated salt solution and loading the sandwich until bending was achieved. Rocking curves, taken in a parallel beam with 15 minute divergence (very coarse collimation), showed no significant change before and after bending. Beam profiles of the (200) Bragg reflected neutrons at various distances from the crystal showed that some focussing was attained (defocussing of the beam was observed when the crystal was rotated 180°).

A second considerably better crystal has been cut and milled to approximately 1/4" thickness for an attempt at bending and for more detailed studies.

IV. Studies of a Beryllium Crystal Showing Curvature Without External Stress

For use with the MTR crystal spectrometer, there are three beryllium single crystals available, with the specifications shown in Table I. All three are crystals cut from a single large crystal grown by NMI prior to 1956, before they applied the zone-refining technique to beryllium. A typical rocking curve is shown in Figure 4. This curve was taken under the same conditions as for the zone-refined beryllium crystal previously discussed and should be compared with Figure 2.

For low-resolution high-intensity applications, the MTR crystal spectrometer is most often used with the crystal designated in Table I as "Mark I", with the a-axis vertical. In this orientation, there are four sets of planes which give neutron beams of comparable high intensity: the so-called "reflection planes" with Miller indices (002), the (100) planes at 90° to these, and two sets of (101) planes at $\pm 67^\circ$ to the (002) planes.

With a two-inch-diameter lithium glass as a neutron detector, it was noted that the neutron intensity at a given Bragg angle appeared to change in a regular fashion, depending upon which of four possible (101) orientations was used (two sets of (101) planes, and front and back reflections from each). Probing the beam with a small BF_3 tube indicated that the beam area was different for different orientations

TABLE I

Beryllium Crystal Specifications

Crystal	Mark I	Mark II	Mark III
Physical Description	Rectangular Parallelopiped	Rectangular Parallelopiped	Truncated Wedge
Height	5 3/4 "	6 7/16"	7 3/16"
Width	1 3/4 "	1 3/4 "	1 3/16" to 1 3/4"
Thickness	5/16"	3/8 "	3/8 " to 1"
Radiation History	Continuous Use Since 1957	Intermittent Use Since 1958	None of any Consequence

Figure 4 - Neutron rocking curve for the 100 planes of the melt-grown beryllium crystal.

of the crystal! For certain orientations, the crystal appeared to be focussing the beam in the vertical dimension.

In order to study this phenomenon in greater detail, beam profiles were obtained with a Polaroid neutron camera. (The neutron camera contains a lithium-loaded zinc-sulfide screen adjacent to a Polaroid film pack, which can be used to give a full size neutron beam profile.) A punched cadmium sheet over the end of the collimator was used to obtain two parallel neutron beams spaced vertically 1.5 inches apart. Ten of these beam profiles, which are representative of the behavior observed, are shown in Figure 5. It is evident that a focussing effect does occur.

It was conjectured that the crystal may have had strains induced by radiation damage, since this particular beryllium crystal (Mark I) has been in more or less continuous use in the MTR crystal spectrometer since 1957 and shows some cracks or splitting along the edges. Consequently, the same experiment was performed on Mark II, which has had only very occasional use and does not show any visible evidence of radiation damage. The same results were obtained: focussing occurs for the same orientations as for Mark I. It can thus be concluded that the strains were introduced in the growing of the large crystal from which these were cut, possibly by impurities in the melt.

Both crystals were investigated for physical curvature (curvature which might have been induced after the crystals were machined). No curvature of this nature could be discerned. If any exists, it is less than 0.05%.

The possibility was also investigated that the crystals are twinned instead of curved, i.e., that each crystal consists of two crystals of very nearly the same orientation, fortuitously inclined at just the

100
Planes
At
143°

100
Planes
At
323°

Distance
From
Crystal

24 Inches

43 Inches

61 Inches

79 Inches

98 Inches

Figure 5 - Beam profile of two round beams spaced vertically 1 1/2 inches apart.

correct angle to show a pseudo-curvature (somewhat analogous to the pseudo-bent crystal made by Pattenden⁽³⁾). It was demonstrated that the crystal is curved and not twinned by allowing three beams to strike it instead of two. The beam profiles showed convergence of all three beams, as well as convergence of the individual beam spots.

In order to determine the radius of curvature of this crystal, it is necessary to remove the effects of neutron beam (collimator) divergence, which is a large effect in the vertical dimension on the MTR spectrometer. Beam profiles were obtained from all the major planes, the (002), (100), and (101), at all crystal orientations for which the Bragg condition is fulfilled at a given angle while keeping the "a"-axis vertical, and for various distances from the crystal. The data are summarized in Figure 6, which is a polar plot of the distances between centers of the beam spots (which at the crystal was approximately 1.5 inches). The beam divergence can be clearly distinguished from the focussing effect, since if there were no collimator divergence, the circular loci would all have a radius of 1.5 inches. Several determinations of the radius of curvature were made for different locations of beams incident on the crystal - the results showed a radius of curvature of 80 ± 20 inches. Some evidence was found that the curvature of the crystal is not uniform, although a quantitative determination of this effect requires more careful study.

It was recognized that those (101) planes which show such a large focussing effect in the vertical dimension cannot be used as the monochromating planes in a curved crystal spectrometer, because the Bragg condition is not the same at either end of the crystal as it is in the center. Consequently, one more investigation was carried out, using

Figure 6 - Polar plot of beam spacings for planes studied.

reflections from the (110) planes which are perpendicular to that a-axis which was originally vertical. The crystal was rotated 90° , and two vertical parallel beams were allowed to strike it. The profiles taken at various distances from the crystal are shown in Figure 7. The focussing effect persists; in fact, the slit profiles have actually crossed in the last of the figures shown.

Although the crystal designated as Mark III was not investigated in the present study, it too was a part of the larger crystal from which all three pieces were cut, and there is every reason to believe that its behavior is the same. It is planned, if further studies show that these beryllium crystals can be used in a curved crystal spectrometer, that Mark III will be cut in an appropriate manner to be used. Studies are planned which will answer the following questions:

- (1) What are the focussing (or defocussing) effects in the other crystal dimension?
- (2) How markedly does the curvature change from one part of the crystal to another?
- (3) Can a convergent collimator (see Appendix A) be designed in such a way as to fulfill the Bragg condition for all parts of the crystal, even though the curvature changes?

110
Planes
At
172°

110
Planes
At
352°

Distance
From
Crystal

24 Inches

43 Inches

61 Inches

79 Inches

98 inches

Figure 7 - Beam profile for two vertical beams spaced $3/8$ inches apart.

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Appendix A

In order to make a meaningful comparison of the relative neutron intensity in the Bragg beam from a curved crystal spectrometer and from a conventional plane crystal spectrometer, it is necessary to consider the angular resolution conditions which exist. These are assumed to be determined by the properties of the collimation system and by the mosaic of the crystal; the latter is assumed to remain virtually unchanged by curving the crystal.

The resolution function of a parallel slit collimator cannot be directly compared to that of a convergent collimator, since the shapes of the resolution function are different. This is illustrated in Figure 8 for the case of two collimators which are idealized representations of the MTR plane and curved crystal spectrometers, respectively. In one dimension, the relative intensity distributions are the block functions shown. The idealized resolution functions are a triangle for the parallel collimator, and a trapezoid for the convergent collimator. Both resolution functions can be formed by the convolution of two "block" resolution functions, as shown. In the convolution the root-mean-square deviation can be shown to be given by

$$\rho^2 = \rho_1^2 + \rho_2^2,$$

where ρ_1 and ρ_2 are the RMS deviations of the block functions. Since the RMS deviation is preserved under convolution, it is proper to compare the resolution functions of two such collimators according to their respective RMS deviations. (It may be noted that the resolution function of each spectrometer is a convolution of the resolution functions of the collimator and of the crystal mosaic spread, in the

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Figure 8 - Comparison of Soller slit and convergent collimators. The neutron source area is assumed to be infinitely extended, and is represented by the solid line at the left of the figure. The neutron intensity at the detectors for the two collimators is represented as rectangular functions, where I_0 is the intensity which would obtain for a Soller slit collimator of length S which extended from source to detector. The resolution functions for the two collimators are a triangle and a trapezoid, both of full width $6\Delta/S$ at the base. The resolution function was calculated by integrating the angular divergence across the slit. This procedure is justified since functions shown can be formed by convolution of the two block functions given below each of the resolution functions.

idealized case chosen). By quadrature it is easily shown that the RMS deviation of the triangular function is given by $\frac{\Delta}{5}\sqrt{6}$. For the trapezoid it is $\frac{\Delta}{5}\sqrt{5/3}$. Thus, for equivalent resolution, the parallel slit collimator is permitted to have slits separated a distance $\frac{\Delta\sqrt{10}}{3}$, where Δ is the slit width of the convergent collimator.

The present MTR crystal spectrometer has a maximum beam area of 1 x 3 sq. inches; its plane crystal is illuminated by a source of the same area, located a distance of 20 feet from the crystal. The proposed curved-crystal spectrometer would consist of a crystal of area 7.25 sq. inches, located a distance of 12 feet from a source of area 135 sq. inches. It is assumed that the neutron flux of the two sources can be made the same.

With the curved-crystal spectrometer a collimator would be used which is 6 feet long and is constructed of infinitely absorbing foils of negligible thickness spaced 0.1 inch apart at the crystal, and 0.15 inch apart at the distance of maximum divergence. It is shown above that the resolution characteristics of such a collimator are equivalent to a Soller slit (parallel foil) collimator which is 4 feet long and is constructed of foils spaced $0.1\sqrt{10/3}$ inches apart. It can be easily shown that the number of neutrons transmitted through a Soller slit collimator is independent of the distance from the collimator to the source, provided the source is plane and infinitely extended. In this case, a single slot of the above converging collimator transmits a factor of $4/\sqrt{10}$ times as many neutrons as the equivalent parallel slot collimator. However, for the actual case considered, the source area is not infinitely extended, being 1 x 3 inches for the plane crystal spectrometer, and 9 x 15 inches for the curved crystal spectrometer.

For a single slot of the respective collimators, the source solid angle intercepted by the curved crystal is a factor of 3.8 higher than for the plane crystal.... Thus the number of neutrons passing through each slot is a factor of 4.8 higher for the converging collimator than for the parallel collimator. For the curved-crystal spectrometer, the converging collimator is composed of 50 slots; the existing spectrometer can utilize only 9.5 slots of the equivalent Soller-slit collimator, because of the smaller beam dimensions. If it is assumed that the crystal reflectivities are equivalent, the Bragg beam of the curved-crystal spectrometer will contain a factor of 25 more neutrons than that of the existing spectrometer, at the same resolution.

There is also a neutron intensity gain, as a result of the focussing properties of the curved crystal. The proposed spectrometer will focus the neutrons to a beam spot of dimensions 0.1" x 6". About 65% of the neutrons are contained in a spot of dimensions 0.1" x 2.5". The present spectrometer beam measures 1" x 3", so the expected neutron intensity of the proposed spectrometer is a factor of 200 higher than is presently available, at comparable resolutions.

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