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THE LPTR SINGLE-CRYSTAL
NEUTRON SPECTROMETER AND
REACTOR SPECTRUM MEASUREMENTS

Livermore, California

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ABSTRACT

A single-crystal neutron spectrometer was designed and built for use at the Livermore Pool Type Reactor. The instrument is suitable for reactor spectrum measurements, neutron diffraction work, and certain neutron cross-section measurements. Among the novel design features are: neutron collimators coated with an organic compound that eliminates total reflection of neutrons from their surfaces, and a crystal orienter that is designed to provide a means of circumventing the problem of parasitic reflections.

Design considerations such as instrument resolution, collimator design, shielding, and component precision are discussed. The calibration and alignment of the spectrometer and its individual components are described in detail.

The problems of measuring reactor spectra with a single-crystal neutron spectrometer are considered individually. The equations commonly employed in the calculation of crystal reflectivity are shown to be deficient and appropriately modified equations are presented. A reactor spectrum measurement was made with the instrument and these modified equations were employed in the analysis of the data. The spectrum was measured from 0.5 to 4.0 Å (0.327 to 0.0051 eV) using the (111) planes of a lead single crystal. The effective neutron temperature was found to be $361 \pm 7^\circ\text{K}$, which is $51 \pm 7^\circ\text{K}$ above the average moderator temperature of 310°K . The experimental results are compared with a theoretical Maxwellian distribution and the differences are discussed.

I. DESIGN OF A SINGLE-CRYSTAL SPECTROMETER

Introduction

The LPTR Single-Crystal Neutron Spectrometer is a variable-energy instrument designed primarily to offer experimental versatility. Although many single-crystal neutron spectrometers and diffractometers have been built,¹⁻¹⁷ individual design requirements are seldom the same for any two instruments. The nature of the experiments to be conducted, the type of reactor being used, and the spatial limitations at a particular beam facility are but a few of the design considerations that prevent standardization. In this section we will discuss the over-all and specific design characteristics of this instrument.

Spectrometer Shielding

The Livermore Pool Type Reactor (LPTR) is a light-water moderated reactor operated at 2 MW (thermal). For this type of reactor, shielding problems are more severe than those encountered with graphite or heavy-water moderated reactors of comparable power. As will become evident, shielding considerations played a major role in determining the size and configuration of many spectrometer components.

An over-all view of the spectrometer, located at the B-1 beam tube facility of the LPTR, is shown in Fig. 1. It was necessary to place the main axis of the instrument 101 in. from the reactor face because of access requirements for an adjacent experimental facility on one side of B-1 and a reactor fission chamber on the other side. For the same reason, the beam tube extension and part of the peripheral shielding around the main axis were designed so that they could be readily moved aside.

As shown in Fig. 1, a 6-in. lead plug is situated at the beam tube inlet to decrease the gamma intensity. A series of converging paraffin and polyethylene collimators within the beam tube serve to reduce the fast neutron leakage by limiting the neutron beam to the solid angle subtended by the primary Soller collimator. The beam tube extension is an aluminum tube surrounded by approximately 2 in. of lead and 12 in. of high-density Masonite and serves to conduct the beam from the beam tube outlet at the reactor face to the inlet of the primary Soller collimator.

Fig. 1. Over-all view of spectrometer.

A cutaway drawing of the spectrometer is shown in Fig. 2. The main shielding tub, counterbalanced spectrometer arm, and shielded BF_3 detector, totaling almost 3-1/2 tons, rotate together. The angular range is 90° clockwise and 15° counterclockwise with respect to the direct beam, a total of 105° .

The cylindrical tub and peripheral shielding are constructed of Permalin, a high-density wood laminate. In addition to having greater strength and dimensional stability, shielding experiments showed the fast-neutron attenuation characteristics of Permalin to be superior to those of high-density Masonite. The tub, 44 in. in diameter and 42 in. high, was fabricated by the supplier in three sections with stepped mating surfaces. The individual sections were so constructed that there are no straight-through paths in the shielding. The central cavity, 14 in. in diameter and 16 in. high, houses the monochromating crystal and a device for remotely orienting the crystal. The cavity is surrounded by a cast-lead liner which is 4 in. thick over the 105° arc that can intercept the direct beam, and 2 in. thick elsewhere.

The primary Soller collimator, held stationary in the fixed peripheral shielding segment, extends to the edge of the inner cavity through a 105° pie-shaped opening in the back of the main shielding tub. It is this opening which necessitates the peripheral shielding.

It was found necessary to surround the spectrometer with additional shielding, in the form of water tanks, in order to reduce radiation levels below tolerance. Hence, access to the spectrometer is limited while the shutter is open. This proves to be no problem, however, since all normal operations are carried out and displayed remotely.

Shielding of the main BF_3 detector is provided by a 14-in.-diam. aluminum can filled with borated paraffin (30% boron by weight).

Resolution and Beam Collimation

The diffraction of neutrons by a single crystal is governed by the Bragg equation

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

(1)

Fig. 2. Detail of main spectrometer axis.

where n is the order of the reflection, λ is the neutron wavelength, d is the lattice spacing of the diffracting planes, and θ is the angle between the incident (as well as diffracted) beam and the diffracting planes.

We note that an uncertainty in the Bragg angle results in an uncertainty in the neutron wavelength. The energy resolution $\Delta E/E$ is obtained by differentiating Eq. (1) with respect to θ and making use of the relation¹⁸ $\lambda = 0.2860/\sqrt{E}$, where λ is in angstrom units and E is in electron volts. Hence

$$\Delta E/E = 13.99 d \sqrt{E} \cos \theta \Delta \theta, \quad (2)$$

where d is in angstrom units and E in electron volts.

Thus for a given energy and set of Bragg planes, the energy resolution of the spectrometer depends upon the uncertainty in the Bragg angle $\Delta \theta$. For identical primary and secondary collimators of horizontal angular divergence $\alpha = d/\ell$, where d is the width and ℓ is the length of the collimator channel. Sailor *et al.*⁶ have shown that $\Delta \theta = \alpha/\sqrt{2}$. The vertical angular divergence of the beam has a small but usually negligible effect on the energy resolution.¹⁹

However, it should be noted that the relation $\Delta \theta = \alpha/\sqrt{2}$ is derived for a monochromating crystal in which secondary extinction is negligible. This means that the crystal is thin enough that reflectivity is still proportional to crystal thickness. In practice, neutron monochromators are effectively of infinite thickness; that is, little or no further increase in crystal reflectivity is obtainable with further increase in crystal thickness. In this situation, secondary extinction is essentially complete rather than being negligible. As will be shown in Section III $\Delta \theta$ is a complicated function which includes the effects of both the horizontal angular divergence of the beam and the mosaic spread of the crystal. The functional form of the distribution which determines $\Delta \theta$ is given by the integrand of Eq. (20) or Eq. (21), depending upon whether the crystal is being used in reflection or transmission.

Effect of Total Reflection on Resolution

The horizontal angular divergence of the beam, α , is identical to the geometrical angular divergence of the collimator only in the absence of total

reflection. Hence, whether $\Delta\theta$ is taken as proportional to a , or calculated from the more complicated expression mentioned above, the energy resolution is also affected by total reflection.

The critical angle for the total reflection of neutrons is given by

$$\theta_c = \lambda (Na/\pi)^{1/2}, \quad (3)$$

where λ is the neutron wavelength, N is the number of scattering centers per cm^3 , and a is the bound coherent scattering amplitude of the reflecting surface. If more than one nuclide is present, a becomes the algebraic sum of the constituent nuclides, weighted according to their relative abundance.

To a first approximation for substances with positive values of Na , all neutrons having angles of incidence less than the critical angle will be totally reflected. Most structural materials possess positive values of Na ; for example, at a wavelength of 3 Å, critical angles for aluminum and a common stainless steel (type 304) are approximately 8 and 16 min respectively.²⁰

For multislit collimators, the slant thickness a neutron must travel in order to exit through a channel different from the one it entered is usually large enough that even weakly absorbing materials provide adequate attenuation. However, heavy absorption has relatively little effect on the critical angle and, hence, reflectivity of a surface. Thus the practice of coating collimator surfaces with cadmium accomplishes little. In fact, the reflectivity of cadmium is comparable to that of aluminum.²⁰

When $\theta_c \ll d/l$, $a = d/l$. However, when θ_c approaches or exceeds d/l , a increases and hence the energy resolution decreases. Calculation of $\Delta\theta$ then becomes quite complicated, in spite of simplifying assumptions,¹¹ including that of negligible secondary extinction mentioned above.

As described in detail elsewhere,²⁰ a commercially available organic acid has been found to have excellent total reflection suppression properties. The compound, tetradecanoic acid ($\text{C}_{14}\text{H}_{28}\text{O}_2$), also known as myristic acid, can be easily coated onto metal surfaces and produces a tough, tightly-adhering covering. All channel surfaces of the two Soller collimators to be described next were coated with this material. The 2-mil thickness employed has been shown to be adequate to completely mask the reflection properties of the base material.²⁰ The total reflection properties of the

tetradecanoic acid are such that we may assume $\alpha = d/l$ for all neutron energies of practical interest. The importance of this fact will become obvious in the section dealing with the calculation of crystal reflectivity.

Design of Soller Collimators

The design of neutron collimators is governed by the particular energy resolution requirements of the experiment. Accurate resonance analysis may require that the horizontal angular divergence (which will hereafter be referred to simply as the angular divergence) of the beam be on the order of a minute or two of arc.⁶ Collimators having angular divergences of less than one minute of arc have already been constructed.²¹ However, for most neutron diffraction work, reactor spectrum measurements, and many cross-section measurements, coarser collimation is tolerable and even desirable. Since work in all three areas was anticipated, a 10-min angular divergence was selected for the instrument. The actual angular divergence of the completed collimators was 9.47 min.

In order to obtain the required neutron intensity and still maintain the desired beam divergence, multichannel collimators are usually employed. Collimators of this type, originally used in x-ray work, are called Soller collimators.

An interesting problem arose in connection with the location of the main axis of the spectrometer. As mentioned in the section on spectrometer shielding, adjacent experiments and reactor fission chamber access requirements made it impossible to place the spectrometer at the reactor face. From this standpoint, locations farther from the reactor face were increasingly attractive, but increasingly unattractive from the standpoint of neutron intensity loss.

Now in order to compare alternatives in location of the primary axis, the maximum intensity attainable at each position is a desirable quantity to know. If the angular divergence, total collimator cross-sectional area, septum thickness, and source plane to collimator outlet are fixed, the number of collimating channels (and hence length of collimator) is still undetermined. There will be an optimum number of channels for which the total number of neutrons transmitted by the collimator is a maximum.

The procedure for calculating the neutron intensity emerging from a primary Soller collimator, neglecting total reflection, has been treated in detail by Szabó.²² The calculations are lengthy and there is no way of explicitly calculating the optimum number of collimating channels. However, subject to certain angular limitations, not usually encountered in practice, the equations governing the transmitted intensity reduce to a simpler form and the optimum number of collimating channels can be calculated explicitly.²³ Szabó²⁴ later showed that the effect of total reflection leaves the optimization conditions unchanged, although the magnitude of the transmitted intensity is increased. Since total reflection is absent from our collimators, this extension is of no concern to us except to note that the simplified equations for optimization²³ would still be valid if total reflection were present.

The calculations, outlined in Appendix A, are interesting for several reasons. The transmitted intensity decreases much less rapidly than the inverse square of the source plane to collimator outlet distance. As the distance is increased by about a factor of two from 93.5 to 187.5 in., the intensity is decreased by only a factor of 1.7.

The choice of collimator septum thickness was governed largely by structural considerations. To avoid having to tension the septa to obtain flatness, 1/64-in. precision-ground steel shim stock was used. Accurately milled slots, 15 mils deep, were used to hold the septa in place on the top and bottom plates of the collimator. The 2-mil coating of tetradecanoic acid on each side brought the total septum thickness to 19.6 mils. However, had we used 6-mil stock (10 mils total thickness with organic coating), the maximum intensity would have only been increased by about 9%.

Perhaps the most useful aspect is the comparison of the transmitted intensity for optimum and nonoptimum numbers of collimating channels. For our system parameters, the optimum number is thirteen, resulting in a collimator 35.6 in. long. Yet we can use considerably shorter collimators without much sacrifice in intensity. A collimator of approximately one-half optimum length, having 22 channels, transmits only 7% less intensity, while the 23.75-in. collimator actually constructed (18 channels) transmits only 3% less intensity than the optimized system. A diagram of one of the two identical Soller collimators is shown in Fig. 3.

Fig. 3. Detail of multislit (Soller) collimator.

2:1 Drive

As discussed earlier in connection with beam collimation, an uncertainty in angle results in an uncertainty in neutron energy. For the rotating components of the spectrometer, it was decided to limit this uncertainty to a modest fraction of the inherent energy resolution of the instrument. Hence the required angular precision was set somewhat arbitrarily at 0.01° .

Since such precision is difficult to obtain with a gear train, a stainless-steel belt and pulley system was chosen to maintain the required 2:1 angular ratio between the spectrometer arm and monochromating crystal axis. The linkage, shown in Fig. 4, is similar to that used by Hurst *et al.*⁴ at Chalk River. The belts are $1/2$ in. wide and 10 mils thick and are spring-tensioned to 30 lb. The pulleys were precision ground to $1/10$ mil in diameter. The effective radius of the pulleys was taken as the actual radius plus 40% of the belt thickness.⁴

The accuracy of the 2:1 drive was checked optically over the entire 105° arm rotation and the maximum deviation of the crystal axis from the required ratio was found to be 36.5 sec of arc. Although the precision of the optical instruments was about $1/10$ sec of arc, no backlash or slip in the linkage was detectable.

Eulerian Cradle

The mechanism for orienting the monochromating crystal is called an Eulerian cradle²⁵ since its three axes of rotation correspond to the three Eulerian axes.²⁶ The device, shown in detail in Fig. 5, is located in the central cavity of the main shielding tub, as shown in Fig. 2.

The vertical axis of rotation is called the "rock" axis since the crystal is rotated about this axis to produce a rocking curve (see Fig. 9). The top axis of the cradle is designated as the crystal "roll" axis and permits rotation of the crystal about a horizontal axis which is parallel to the reflecting planes. Hence, the orientation of the crystal face relative to this axis depends upon whether the crystal is being used in reflection or transmission. The middle axis is called the parasitic reflection axis (abbreviated as PR axis in Fig. 5). The reasons for having this additional axis will be detailed in a later section, but it suffices to mention here that this axis

Fig. 4. Detail of spectrometer arm and 2:1 drive.

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Fig. 5. Detail of Eulerian cradle.

permits rotation of the crystal about an axis perpendicular to the Bragg planes. That the PR axis must necessarily be the middle axis rather than the top axis is apparent after a moment's thought. The PR axis must always lie in the plane defined by the incident and diffracted beams. This is possible with the middle axis, no matter what the orientation of the top axis may be. The converse is not true, however.

Bodine 1.1-rpm gear motors were used for each of the three rotations. The worm and worm gear linkages are such that the rock axis rotates at approximately $2-1/2^\circ/\text{min}$ while the parasitic reflection and roll axes rotate at approximately half this speed. The angular ranges of rotation for the axes, defined by limit switches, are 190° , 23° , and 15° for the rock, parasitic reflection, and roll axes, respectively.

Ideally, the three axes should intersect at a point in space which remains fixed over the entire range of rotations for the three axes. The point should lie in the horizontal midplane of the crystal, being at the center or on the surface of the crystal depending upon whether the crystal is being used in transmission or reflection. However, the necessity of finite machining tolerances plus imperfections in the ball races and balls cause the point to wander a bit and the axes to exhibit a finite lack of coincidence. Precision measurements made by the Metrology Group here at the Laboratory showed the sum of these two effects to be larger than expected: as much as 0.040 in. for some positions. Hence parts were remachined and adjusted until the lack of coincidence was within 0.0005 in. over the entire angular range of rotation for the three axes.

The intersection of the three axes, dubbed the "magic spot" is about $1-1/2$ in. above the crystal holder table. Hence, monochromating crystals up to 3 in. in height can be used and still permit the horizontal midplane of the crystal to include the magic spot. The cradle calibration was carried out with a gage ball and dial indicators. The gage ball, centered on the magic spot, was left clamped to the cradle for subsequent use in the alignment and calibration of the spectrometer.

In addition to its three rotational motions, the Eulerian cradle has two linear translation motions called the X and Y traverses. These machine ways, shown in Fig. 5, are manually operated by threaded rods and are used primarily in obtaining concentricity between the rock axis of the cradle and the 2:1 axis of the spectrometer. Fifteen-turn discriminator dials attached to the end of each traverse rod aid in alignment procedures.

The crystal holder, not shown in Fig. 5, is counter-weighted so that the parasitic reflection and roll worms always remain in contact with the same surfaces of their worm gears, regardless of the sense of rotation.

As indicated in the section on shielding, radiation levels adjacent to the spectrometer with the shutter open are too high to permit frequent access. Hence, remote operation and readout of the cradle motions were necessary. Vernitron Synchro-Torque transmitters and receivers were used to operate Veeder-Root registers at the console for each of the three axes. The transmitters were connected to the gear motor shafts by an antibacklash gear train, the gear ratios being such that one complete rotation of the transmitter-receiver system corresponded to a 1-min angular rotation of the corresponding cradle axis.

The accuracy of the three cradle readouts was determined optically by the Metrology Group and errors were found to be cumulative, but in general less than 1/2%. Hence, for measurements such as crystal rocking-curve widths which usually involve angular ranges of a degree or less, no corrections are needed. For larger angular intervals, tabulated corrections can be applied to yield values accurate within the 1-min least count of the readouts. Backlash was also found to be less than 1 min.

Arm Drive and Encoder

The rotating part of the spectrometer, weighing almost 3-1/2 tons, can be driven remotely or turned manually. For remote operation, a 5.7-rpm Bodine gear motor and reduction gear train can be engaged with a brass rack which is bent to a 22.500-in. radius. The arm is driven at approximately 1°/min. For large changes in angle, it is usually preferable to disengage the arm drive by turning the eccentric cam shown in Fig. 4 and effect the rotation manually. As shown in Fig. 2, a Theta Decittrak Encoder is spring-tensioned against a second brass rack of the same radius and remains engaged at all times. The encoder, which reads out in hundredths of a degree, was zeroed by optical means at the straight-through position of the arm. The encoder readout was then checked for accuracy and repeatability by the Metrology Group. The readout was found to deviate in continuous fashion from the actual arm angle by as much as 1/3 of a degree over the 105° angular range of the arm, due primarily to nonuniform tooth spacing on the brass rack. Hence, a correction chart was made up by noting

the encoder readout corresponding to the true arm angle for each integral degree over the angular range. Provided the same sense of rotation is observed, results are repeatable within the least count (0.01°) of the encoder.

The encoder output is also delivered to a Hewlett-Packard printer. The arm angle is printed out automatically with the count data from the arm and monitor BF_3 counters.

Neutron Detectors

The main detector is a Reuter-Stokes (RSN-45A) BF_3 tube with a diameter of 2 in. and an active length of 20 in. The BF_3 gas is enriched to 96% B^{10} and the tube is filled to a pressure of 70 cm. A thin aluminum can, 14 in. in diameter, filled with borated paraffin (30% boron by weight), surrounds the detector. The axis of the detector is parallel to the diffracted beam and the borated paraffin shield effectively eliminates detection of neutrons incident upon the detector from directions other than that of the diffracted beam.

Since the reactor power can fluctuate as much as 10% during the course of the operating week, the beam is monitored by a depleted ($\sim 9\% \text{B}^{10}$) BF_3 detector before reaching the monochromating crystal. Main detector counting times are determined by a preset monitor count, thus eliminating the effect of reactor power changes. The monitor is a Reuter-Stokes (RSN-137A) tube, 1/2 in. in diameter. This tube was found to have a much higher gamma sensitivity than claimed by the manufacturer. Although not directly in the beam, it was necessary to surround the tube with a 2-in. lead shield in order to obtain reproducible counting rates. The shielding configuration of both detectors is such that background count rates are independent of the operating condition at adjacent experiments.

The preamplifiers, amplifiers, scalars, and high-voltage supplies for the two detectors are all RIDL Designer Series transistorized units. However, both preamplifiers and amplifiers had to undergo major modification since the over-all gain of the system was woefully inadequate for use with BF_3 detectors.

Both six-decade scalars are connected through a RIDL programmer to a Hewlett-Packard printer. When the monitor scalar reaches the preset count, the preset count, main detector count and arm angle are automatically printed out.

II. SPECTROMETER CALIBRATION AND ALIGNMENT

Introduction

Thus far we have included pertinent calibration procedures along with the general description of components such as the 2:1 drive, Eulerian cradle, and arm drive. We now turn to the problem of positioning the spectrometer and properly aligning the components relative to each other. The precision optical measurements were carried out by the Metrology Group.

Spectrometer Alignment Requirements

Referring to Figs. 4 and 5, it can be seen that there are actually three separate vertical axes of the spectrometer: the main axis of the spectrometer, defined by the main thrust and radial bearings, the half-speed axis, and the vertical axis of the cradle.

Assuming that we do have an Eulerian cradle "magic spot" as defined earlier, we can now state the spectrometer alignment requirements:

1. The centerlines of the two Soller collimators (in the straight-through position) should be coincident and intersect the neutron source plane at its center.
2. The three vertical axes of the spectrometer should be coincident.
3. The horizontal midplanes of the collimators should be coplanar and oriented such that they intersect the vertical axes perpendicularly at the magic spot of the Eulerian cradle.

In passing, it should be noted that the apparent simplicity of the above requirements is misleading. For example, "coincidence" of axes really requires two separate measurements: parallelism and concentricity. Also, measurements were complicated by the fact that rotation of the main axis is limited to 105° , and the half-speed axis to half that interval. Further, unless the main axis of spectrometer is perpendicular to its base, the main spindle will precess about the true axis as the spectrometer rotates. Hence, it will be impossible to fulfill requirements 1 and 3 at more than one arm position.

The use of "horizontal" and "vertical" above is not in the absolute sense. The beam port is circular and the series of converging collimators bringing the neutron beam to the primary collimator inlet is slightly oversize.

Hence, the main axis of the spectrometer need only be within a few degrees of vertical. This is to be contrasted to parallelism requirements on the order of minutes to be discussed below.

Calibration and Alignment Procedure

First, a theodolite was positioned and leveled so that the intersection of the cross hairs defined the centerline of the beam port and neutron source plane. The spectrometer base was then positioned so that the main axis was parallel to the vertical cross hair of the theodolite. Next, the Eulerian cradle was bolted to the half-speed axis and, by means of three vertical adjusting screws, the height of the cradle was adjusted so that the gage ball representing the magic spot was centered on the theodolite cross hairs. The vertical axis of the cradle and the half-speed axis were then brought into parallelism by means of the three adjusting screws. With the aid of precision optical instruments, adjustments were continued until the two axes were within 7 sec of parallelism over the entire angular range. Using the magic spot gage ball and a dial indicator, the two axes were brought into concentricity using the X and Y traverses, which are located below the vertical axis of the cradle (see Fig. 5).

Next, parallelism of the main axis and half-speed axis were checked optically and found to be poorer than expected, the lack of parallelism being as much as 4 min 25 sec of arc over the angular range of the arm. By means of shims placed under the main thrust bearing the two axes were forced into closer parallelism. The final result was a maximum lack of parallelism of 1 min 27 sec.

The concentricity of all three axes was within 0.006 in. measured perpendicular to the arm and 0.012 in. measured parallel to the arm. Although these numbers reflect the effects of both concentricity and parallelism, they are actually within the limits ascribable to the measured lack of parallelism alone. In any event, they are small enough, compared to the monochromating crystal and Soller collimator dimensions, to be negligible.

The primary and secondary Soller collimators were then fitted with cross hairs at both ends and installed. The primary collimator was adjusted in the fixed peripheral shielding block (see Fig. 2) until the center septum and horizontal cross hairs were coincident with those of the previously

positioned theodolite. The spectrometer was rotated to the straight-through position and the procedure repeated for the secondary collimator, installed on the spectrometer arm.

Particularly for reactor spectrum measurements, it is important that the horizontal midplanes of the two collimators remain coplanar over the entire angular range of the spectrometer. Otherwise, the transmission function of the collimation system changes as the collimators become skewed relative to each other. Hence, after the collimators had been aligned in the straight-through position, their alignment was checked at the maximum clockwise arm angle of 90° . This was accomplished by moving the theodolite to the 90° position and positioning it so that the cross hairs of the secondary collimator on the spectrometer arm and those of the theodolite were aligned. Then a mirror was placed atop the Eulerian cradle at an angle of 45° and the reflection of the cross hairs and center septum of the primary collimator was observed through the secondary collimator by adjusting the theodolite focus. The collimator vanes were found to be out of parallel in the vertical plane by about 4 min. Since the vertical channel cross section is a rectangle 0.0654 by 1.470 in., its "angular divergence" in this direction is approximately 153 min. Hence, this amount of skewing was considered quite tolerable.

Finally, the arm BF_3 shield was aligned so that the axis of the detector would be coincident with the centerline of the secondary collimator.

III. MEASUREMENT OF REACTOR SPECTRA WITH A SINGLE-CRYSTAL NEUTRON SPECTROMETER

Introduction

In order to measure the spectral distribution of neutrons emerging from a reactor, three parameters are needed: crystal reflectivity, spectrometer resolution, and detector efficiency. Calculation of these quantities is complicated by order contamination and parasitic reflections. However, for the sake of clarity, we will defer discussion of these aspects to later paragraphs.

Unfortunately, the problem of calculating crystal reflectivity has been treated neither completely nor correctly to date. Hence, we will spend some time outlining the derivation of the requisite equations. While the following considers only the symmetrical reflection case (reflecting planes parallel to crystal face) it applies equally to the transmission case. What we are going to do, briefly, is as follows: First, we will derive the integrated reflectivity of a crystal rotated through its Bragg position in a beam of monoenergetic neutrons with the detector remaining stationary, taking into account both the mosaic spread of the crystal and the finite angular divergence of the beam. Then we will show that this result bears a simple relationship to the integrated reflectivity of a crystal fixed at its Bragg position in a beam of white radiation, the detector again fixed. The latter, of course, corresponds to the conditions under which the reactor spectrum measurement is made at each neutron energy.

Before proceeding, it is perhaps worthwhile to digress for a moment to the nature of crystals used as neutron monochromators. Far from being true single crystals, they are actually composed of "mosaic blocks" on the order of 5000 Å across which, although individually perfect, are separated from each other by dislocations and hence not in perfect alignment. The usual assumption is that the "tilt" of these mosaic blocks relative to their mean orientation is given by a normal distribution whose standard deviation is η . Hence, the probability of finding a mosaic block with a tilt between ϵ and $\epsilon + d\epsilon$ of the mean is given by

$$W(\epsilon) = (1/\eta \sqrt{2\pi}) \exp - (\epsilon^2/2\eta^2) \quad (4)$$

where $W(\epsilon)$ has been normalized to unity. η is usually termed the "mosaic spread" of the crystal. The full width at half-maximum of this distribution is $\beta = 2\eta\sqrt{2\ln 2}$. Interestingly enough, the mosaic spread is relatable to the dislocation density and size of the crystal.²⁷ Suitable monochromating crystals generally have mosaic spreads on the order of 10 to 20 min of arc. The fact that metal single crystals with this degree of imperfection are easily grown is actually quite fortuitous. If a crystal were completely perfect, it would reflect neutrons over an angular range of only a few seconds of arc. The number of neutrons available for reflection in such a narrow angular range would be prohibitively small for most diffraction experiments. In fact, some crystals are too "perfect" to be used as neutron monochromators. Quartz is one such crystal.

If the individual mosaic blocks are assumed small enough that primary extinction can be ignored, which is usually the case for neutron monochromators, then the mosaic crystal is termed "ideally imperfect."²⁸

Incident and Diffracted Beam Power for Monoenergetic Parallel Beam

Since the mosaic blocks have a random orientation, there are no phase relationships to worry about. Hence, we can deal directly with the power in the incident and diffracted beams. The coupled differential equations describing the behavior of the incident beam power $\mathcal{P}_0$ and diffracted beam power $\mathcal{P}_H$ in traversing a layer dt of a crystal of total thickness t_0 , have been derived by Zachariasen.²⁹ We have taken the liberty of recasting them in the more familiar notation of Bacon and Lowde.²⁸

$$d\mathcal{P}_0(t) = -\mu\mathcal{P}_0(t)dt/\gamma_0 - QW(\Delta)\mathcal{P}_0(t)dt/\gamma_0 + QW(\Delta)\mathcal{P}_H(t)dt/\gamma_0, \quad (5)$$

$$d\mathcal{P}_H(t) = +\mu\mathcal{P}_H(t)dt/\gamma_0 + QW(\Delta)\mathcal{P}_H(t)dt/\gamma_0 - QW(\Delta)\mathcal{P}_0(t)dt/\gamma_0, \quad (6)$$

where μ is the effective linear absorption coefficient and γ_0 is the sine of the mean Bragg angle. $W(\Delta)$ is defined by

$$W(\Delta) = (1/\eta\sqrt{2\pi})\exp(-\Delta^2/2\eta^2) \quad (7)$$

and Δ now refers to angular difference between crystal face and its mean Bragg position. The quantity Q is given by

$$Q = \lambda^3 N_c^2 F^2 / \sin 2\theta, \quad (8)$$

where λ is the neutron wavelength, N_c is the number of unit cells per unit volume, F is the structure factor of a unit cell (including the Debye-Waller factor), and θ is the Bragg angle. We will have considerably more to say about some of the terms in Eqs. (5) and (6), but it suffices to note here that they apply only to the case of a monochromatic, parallel beam of neutrons incident upon a crystal whose angular orientation relative to its mean Bragg position is Δ . The first two terms on the right-hand side represent the power lost through "absorption" and diffraction, respectively. The third term represents the power gained through diffraction of the complementary beam.

Effect of Finite Angular Divergence of Beam

If a collimator (or an individual channel of a Soller collimator) of length l and width d had perfectly smooth and parallel sides, its transmission function would be triangular in shape with a full width at half-maximum $\alpha = d/l$. As proposed by Sailor *et al.*,⁶ we can adequately describe the transmission function of an actual collimator by a normal distribution having the same full width at half-maximum as the triangular function. The transmission function is then given by

$$I(\phi_1) = (1/\omega \sqrt{2\pi}) \exp(-\phi_1^2/2\omega^2), \quad (9)$$

where $\omega = \alpha/2\sqrt{2 \ln 2}$ and ϕ_1 is the angle between a given neutron direction and the collimator centerline. Similarly, we can define a transmission function for the second collimator, assumed identical by

$$I(\phi_2) = (1/\omega \sqrt{2\pi}) \exp(-\phi_2^2/2\omega^2), \quad (10)$$

where ϕ_2 is the angle between a given direction and the centerline of the second collimator. Equations (9) and (10) have been normalized to unity.

Now we have a beam of monoenergetic neutrons with the secondary collimator having its centerline at twice the Bragg angle with respect to the centerline of the primary collimator. The beam is incident upon a crystal of mosaic spread η , turned an amount Δ from its mean Bragg position and we wish to determine the effect of the finite angular divergence α on the terms

$W(\Delta)$ in Eqs. (5) and (6). For the monoenergetic case, we must have $\phi_1 = (\epsilon + \Delta)$ and $\phi_2 = -(\epsilon + \Delta)$. Integrating over all values of ϵ , we end up with the same distribution function as Sailer et al.⁶ obtained for the case of a white beam of neutrons, namely

$$W(\Delta) = \left[2\pi(\omega^2/2 + \eta^2) \right]^{-1/2} \exp \left[-\Delta^2 / 2(\omega^2/2 + \eta^2) \right]. \quad (11)$$

When the angular divergence $\alpha (= 2\omega \sqrt{2 \ln 2})$ goes to zero, Eq. (11) does indeed reduce to the parallel beam distribution given by Eq. (7).

As indicated above, the standard deviation $\sigma = (\omega^2/2 + \eta^2)^{1/2}$ of this distribution is the same as that observed by rocking a crystal through its Bragg position in a beam of white radiation. Hence, the quantity $(\omega^2/2 + \eta^2)^{1/2}$ can be experimentally determined quite easily.

Effective Absorption Coefficient

The bound coherent scattering cross section σ_c and the bound incoherent scattering cross section σ_i are fundamental properties of a single nucleus. The bound scattering cross section σ_s is the sum of these two terms (σ_s is listed as $\sigma_{fa} (A + 1)^2 / A^2$ in BNL 325, where σ_{fa} is the free-atom scattering cross section). Nonzero spin and the presence of more than one isotope are primarily responsible for the incoherent term. For a zero-spin, monoisotopic element $\sigma_s = \sigma_c$. However, it is important to realize that these quantities represent that part of the total bound cross section available for coherent or incoherent scattering. We noted in an earlier section on total reflection that the index of refraction (and hence the critical angle) depends solely on these quantities and is independent, except through density differences, of whether the specimen is a single crystal, polycrystalline, or a liquid.

The situation is entirely different for the scattering and diffraction of neutrons by an assembly of atoms. The scattering cross section depends not only on the physical arrangement of the atoms, but on the atomic weight of the scatterer and the ratio of its characteristic Debye temperature to its absolute temperature. The total macroscopic scattering cross section E_{TOTAL} can be divided into four parts as follows:³⁰

$E_{el}(S)$, the elastic coherent,
 $E_{el}(s)$, the elastic incoherent,
 $E_{inel}(S)$, the inelastic coherent,
 $E_{inel}(s)$, the inelastic incoherent.

Unfortunately, no simple relationships exist between the various macroscopic scattering cross sections and their single bound nucleus counterparts σ_c and σ_i . For example, no elastic coherent scattering is possible in a polycrystalline specimen at wavelengths longer than twice the largest lattice spacing. However, since all crystalline orientations are possible, neutrons having wavelengths shorter than this cutoff wavelength will eventually reach a part of the specimen for which the Bragg conditions are fulfilled and Bragg scattering will occur. Hence, if $E_{el}(S)$ is a sizable fraction of E_{TOTAL} at the cutoff wavelength, neutrons of lower energy will be preferentially transmitted by a polycrystalline specimen. In fact, such polycrystalline filters are widely used where long-wavelength beams are required.

For a single crystal, however, all crystalline orientations are not possible, and in general, the Bragg conditions will not be met by any of the mosaic blocks unless the crystal is close to its mean Bragg position. And even then, only those mosaic blocks within a second or two of arc of precisely fulfilling the Bragg conditions will give rise to Bragg scattering. Hence, unlike the polycrystalline case, the total cross section for a single crystal does not rise abruptly at the cutoff energy, but instead remains the same. Its rise to essentially the free-atom cross section at several eV is due to the increase in the $E_{inel}(S)$ with increasing energy. Hence, we have another type of filter through which thermal neutrons are transmitted preferentially to epithermal neutrons. That the total cross section for single crystals does behave in this manner, and that such filters are practical, is well established.³¹⁻³³

Let us now return to Eqs. (5) and (6) and consider the first term on the right-hand side of both equations. The effective absorption coefficient should include all interactions other than Bragg scattering that would take the neutron out of the incident or diffracted beam. The removal of neutrons from one beam by Bragg scattering into the complementary beam is taken care of by the second and third terms on the right-hand side of the equations. Hence, the effective absorption cross section can be written as

$$\mu = N\sigma_a + E_{inel}(S) + E_{el}(s) + E_{inel}(s). \quad (12)$$

Although both $E_{el}(s)$ and $E_{inel}(s)$ are strongly energy dependent, their sum is relatively constant.³⁴ Over the energy range 0.001 to 1.0 eV, a reasonable approximation is

$$N\sigma_i = E_{el}(s) + E_{inel}(s). \quad (13)$$

When used in Eq. (12), the approximation is quite good because σ_i is usually very small for suitable monochromating crystals, being zero within experimental uncertainties for Be, Al, Zn, and Pb. For Cu and Ge, it is small compared to the absorption cross section at the low energy end and is overshadowed by $E_{inel}(S)$ at the high energy end.

In the Placzek "incoherent approximation,"^{35, 36} which has been shown to be accurate within a few percent over the energy range 0.001 to 1.0 eV, provided the Debye temperature is properly chosen,³⁴ we can represent $E_{inel}(S)$ by

$$E_{inel}(S) = (\sigma_c/\sigma_i) E_{inel}(s) \quad (14)$$

or

$$E_{inel}(S) = (\sigma_c/\sigma_i) ([E_{el}(s) + E_{inel}(s)] - E_{el}(s)). \quad (15)$$

Using Eq. (13) and the expression for $E_{el}(s)$ ³⁰ we can write

$$E_{inel}(S) = N\sigma_c [1 - (1 - e^{-\tau})/\tau], \quad (16)$$

where

$$\tau = (24 Em/k \theta M) [(\phi(x)/x) + 1/4]. \quad (17)$$

In Eq. (17), E is the neutron energy, m is the mass of the neutron, k is Boltzmann's constant, θ is the Debye temperature of the crystal, M is the mass of the crystal atom, and $x = \theta/T$, T being the absolute temperature of the crystal. $\phi(x)$ is given by $(1/x) \int_0^x z dz / (e^z - 1)$ and over the range of x of interest to us can be approximated by the following expansion³⁷

$$\phi(x) = 1 - x/4 + x^2/36 - x^4/3600 + x^6/211680 + \dots \quad (18)$$

Hence Eq. (12) becomes

$$\mu = N \left\{ \sigma_a + \sigma_i + \sigma_c \left[1 - (1 - e^{-T})/\tau \right] \right\}. \quad (19)$$

Now for some reason the effective absorption coefficient used by Holm³⁸ and perpetuated by others^{39,40} was the total macroscopic cross section. In fact, only Taylor⁴¹ seems to have appreciated the physical significance of μ . In Fig. 6 the results of Eq. (19) are plotted for a lead single crystal over the wavelength range 0.1 to 5.5 Å (8.2 to 0.0027 eV). For comparison, the total macroscopic cross section is also plotted. The two quantities are seen to differ by as much as a factor of ten at longer wavelengths.

Integrated Reflectivity of Crystal Rotated in Monoenergetic,
Diverging Beam of Neutrons

By supplying the two appropriate boundary conditions, i. e., that the incident beam intensity at the face of the crystal $\mathcal{P}_0(0)$ is known and that the diffracted beam intensity at the back face of the crystal $\mathcal{P}_H(t_0)$ is zero, we can solve Eqs. (5) and (6). In particular, we are interested in the behavior of $\mathcal{P}_H(0)/\mathcal{P}_0(0)$, the ratio of diffracted to incident beam intensity at the face of the crystal as a function of Δ . The integral of this ratio over all values of Δ is called the integrated reflectivity of an ideally imperfect crystal in reflection and is given by²⁸

$$\mathcal{R}^0 = \int_{-\infty}^{\infty} a d\Delta / \left\{ (1+a) + (1+2a)^{1/2} \coth \left[A(1+2a)^{1/2} \right] \right\} \quad (20)$$

where $a = QW(\Delta)/\mu$ and $A = \mu t_0/\gamma_0$. $W(\Delta)$, given by Eq. (11), now includes the effect of both the mosaic spread η and the angular divergence α of primary and secondary collimators, assumed identical. The effective absorption coefficient μ is determined from Eq. (19). All remaining terms have been defined earlier in this section.

For the sake of completeness, we also include the result for the integrated reflectivity of an ideally imperfect crystal in transmission²⁸

$$\mathcal{R}^0 = \int_{-\infty}^{\infty} \sinh(Aa) \exp \left[-A(1+a) \right] d\Delta, \quad (21)$$

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Fig. 6. Comparison of total macroscopic cross section and effective absorption coefficient [as given by Eq. (19)] for a lead single crystal at 300°K.

where the definition of all terms is unchanged and all the foregoing comments apply without reservation.

Although the integrand in Eq. (21) can be expanded in a power series which is integrable term-by-term,³⁸ the integral in Eq. (20) does not have an analytically expressible solution. A computer program, to be discussed more fully later, was written for the IBM 7090 to perform the integration numerically.

In Fig. 7, the integrated reflectivity for a lead single crystal in reflection is plotted for both values of μ shown in Fig. 6. As might be expected, the two curves converge at short wavelengths, but for longer wavelengths, $\mathcal{R}^0$ calculated in the Placzek incoherent approximation lies as much as 30% above the curve calculated using $\mu = \Sigma_{\text{TOTAL}}$. That the discrepancy is not larger, considering the disparity in the μ 's, arises from the relative dominance of the second term on the right-hand side of Eqs. (5) and (6). For a comparable beryllium single crystal, the divergence is as much as 50%. However, since an order of magnitude difference in the two μ 's for lead produces a significantly smaller difference in the value of $\mathcal{R}^0$, the uncertainties inherent in the use of Eq. (19) become negligible. The effects of using the two values of μ in unfolding reactor spectra will be treated more fully in the section on experimental results.

Integrated Reflectivity of Stationary Crystal in a White, Diverging Beam of Neutrons

The integrated reflectivity $\mathcal{R}^0$ as defined by Eq. (20) or (21) has a simple physical interpretation. It is the angular range over which reflection can be considered complete. In other words, the width, in radians, of a rectangular function of unit height that encloses the same area as the actual bell-shaped function $\mathcal{P}_H(0)/\mathcal{P}_0(0)$, plotted as a function of Δ .

However, an angular interval $\Delta\theta$ is related to a wavelength interval $\Delta\lambda$ by the derivative of the Bragg equation

$$\Delta\lambda = 2d \cos \theta \Delta\theta / n. \quad (22)$$

Hence, rotating the crystal through its Bragg position in a monoenergetic beam of neutrons is comparable to leaving the crystal stationary at its Bragg position in a beam of white radiation. (The interval $\Delta\lambda$, on the order

Fig. 7. Integrated reflectivity of (111) planes of a lead single crystal in reflection. $t_0 = 0.686$ cm, $T = 300^\circ\text{K}$, and rocking curve width at half-maximum = 16.5 min.

of 0.01 Å, is small enough that the spectral distribution can be considered constant within a given interval.) The integrated reflectivity for this case, designated $\mathcal{R}^\lambda$, is simply⁴²

$$\mathcal{R}^\lambda = \mathcal{R}^\theta (2d \cos \theta / n) \quad (23)$$

Extending the analogy made above for $\mathcal{R}^\theta$, the value of $\mathcal{R}^\lambda$ can be thought of as the range of wavelengths over which reflection is complete. $\mathcal{R}^\lambda$ is therefore the product of the crystal reflectivity (the efficiency of reflection at a given wavelength) and the instrument resolution function (the wavelength interval over which reflection occurs), two of the three parameters mentioned in the introductory paragraph of this section.

Detector Efficiency

If $\phi(\lambda)$ is the neutron flux per unit wavelength interval, then per unit area, the number of neutrons reaching the detector each second is $\phi(\lambda)\mathcal{R}^\lambda$. If the detector efficiency is $\epsilon(\lambda)$, then the experimentally observed count rate $C(\lambda)$ is

$$C(\lambda) = \phi(\lambda)\mathcal{R}^\lambda\epsilon(\lambda). \quad (24)$$

For a neutron beam incident axially upon an enriched, high-pressure BF_3 tube, the detector efficiency is given by⁴³

$$\epsilon = \exp(-n_1\sigma_1t_1 + n_2\sigma_2t_2) \left[1 - \exp(-n_2\sigma_2t_3) \right], \quad (25)$$

where $n_1\sigma_1$ is the macroscopic absorption cross section of the window material, t_1 is the window thickness, n_2 is the number of B^{10} atoms/cm³, σ_2 is the cross section of B^{10} , t_2 is the length of the inactive region at the window end, and t_3 is the active length of the detector.

The detector employed in the experiments to be described was a 2-in.-diam Reuter-Stokes RSN 45A, filled to a pressure of 70 cm, enriched to 96% B^{10} , and having $t_1 = 0.055$ in. of aluminum, $t_2 = 1-1/4$ in., and $t_3 = 20$ in. In this case $n_1\sigma_1t_1$ is negligible compared to $n_2\sigma_2t_2$.

Using a value of 3838 barns⁴⁴ for σ_2 at 0.0253 eV, the detector efficiency as a function of wavelength is given by

$$\epsilon(\lambda) = \exp(-0.161 \lambda) \left[1 - \exp(-2.57 \lambda) \right]. \quad (26)$$

A plot of detector efficiency from 0.001 to 10 eV is shown in Fig. 8. The effect of the inactive length is seen to dominate the detector performance at low energies.

Order Contamination

We now turn to one of the factors complicating reactor spectrum measurements made with a single-crystal spectrometer. The Bragg equation predicts that for conditions such that $\lambda = 2d \sin \theta$, neutrons of wavelength $\lambda/2$, $\lambda/3$, $\lambda/4$, etc., will also be reflected. Order contamination is particularly severe at energies below the peak of the thermal spectrum since the flux at $\lambda/2$ or $\lambda/3$ may be comparable or even considerably larger than the flux at the primary wavelength λ . Hence, at longer wavelengths, additional terms must be added to the right-hand side of Eq. (24) for the higher-order contributions to the count rate. The measured count rate $C(\lambda)$ thus becomes

$$C(\lambda) = \sum_{n=1}^m \phi(\lambda/n) \mathcal{R}^{\lambda n} \epsilon(\lambda/n), \quad (27)$$

where the value of m used for the upper limit of the summation will depend on the part of the spectrum being measured and the accuracy desired.

Fortunately, several mechanisms help to suppress the higher-order reflections relative to the first-order reflection. The integrated reflectivity $\mathcal{R}^{\theta}$ is reduced in three ways. First, as can be seen from Fig. 6, the effective absorption coefficient tends to discriminate against shorter wavelength neutrons relative to those of longer wavelength (the operating principle of single-crystal neutron filters). The second and third reductions are effected through the factor Q , defined by Eq. (8), which contains two wavelength-dependent quantities. Because of the λ^3 factor, higher-order values of Q will be reduced by a factor of n^3 . Further reduction of Q is accomplished through the structure factor F . This quantity contains the factor $\exp(-W)$ where

$$W = (1.5 h^2 n^2 / M k \theta d^2) \left[(\phi(x)/x) + 1/4 \right] \quad (28)$$

is the Debye-Waller factor. All symbols, except Planck's constant h and the spacing of the diffracting planes d were defined earlier for Eq. (17). And finally, as seen in Eq. (23), an additional factor of $1/n$ is introduced in the

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Fig. 8. Main detector efficiency versus neutron energy.

transformation of $\mathcal{R}^0$ to $\mathcal{R}^\lambda$. It should be pointed out that for higher orders of a given primary wavelength, $\sin 2\theta$ in Eq. (8) does not change.

As mentioned earlier, $\mathcal{R}^0$ defined by Eq. (20), must be integrated numerically. A general program was written for the IBM 7090 to perform calculation of the product $\mathcal{R}^{0n} (2d \cos \theta/n) \epsilon(\lambda/n) = \mathcal{R}^{\lambda n} \epsilon(\lambda/n)$ for the first five orders ($n = 1$ to 5). Computations are carried out for primary wavelengths from 0.1 Å to the Bragg cutoff appropriate to the diffracting planes being used, in steps of 0.1 Å. By supplying the requisite input data, the program can be used for any monochromating crystal, as well as any set of diffracting planes, crystal thickness, mosaic spread, and collimator divergence. The effective absorption coefficient μ is calculated using Eq. (19).

Parasitic and Double-Bragg Reflections

A parasitic reflection occurs when a second set of diffracting planes also simultaneously satisfies the Bragg conditions for a given wavelength. Since these parasitic planes are not parallel to the primary planes, the reflected intensity in the direction of the primary diffracted beam is decreased. In terms of the reciprocal lattice, the above situation means that two reciprocal lattice points lie on the sphere of reflection.⁴⁵ However, this is just the requirement for double-Bragg reflection, which can either decrease or increase the reflected intensity in the direction of the primary diffracted beam.⁴⁶

Parasitic and double-Bragg reflections have received considerable attention during the past few years⁴⁶⁻⁵² because of their complicating effects on reactor spectrum and precision cross-section measurements. In the case of reactor spectra, the parasitic reflections are so numerous that over wide intervals there are no spectral points unperturbed by one or more of these competing reflections. Knowing the orientation of all three crystallographic axes of the monochromating crystal, relative to the neutron beam, the Bragg angles at which these parasitic reflection will occur are predictable. But there is no feasible method of calculating the magnitude of their perturbing influence.

The problem is further complicated because the occurrence and magnitude of these parasitic reflections will be different for different orders

of reflection. Hence, for precision cross-section and fission parameter measurements at low energies, order corrections must be determined experimentally for each data point.

As mentioned in an earlier section, the monochromating crystal holder, called an Eulerian cradle, was provided with an extra degree of rotational freedom to allow rotation of the crystal about an axis perpendicular to the primary reflecting planes. As will be shown in the next section, this provides a means of mechanically circumventing the problem of parasitic and double-Bragg reflections in reactor spectrum measurements.

IV. MEASUREMENT OF THE LPTR REACTOR SPECTRUM

Description of Monochromating Crystal

The spectrum measurement to be described was made with a lead single crystal measuring $2 \times 9 \times 0.27$ in. with the (111) planes parallel to the large face. The crystal was grown by the Atomic Energy Division of American-Standard. To check the identity of these planes a 3-in. polycrystalline Be filter was placed in front of the secondary collimator (see Fig. 2). The spacing* of the lead (111) planes is $2.8580 \text{ \AA}$ and the Be cutoff* is at $3.9587 \text{ \AA}$. Using $\lambda = 3.9587 \text{ \AA}$ and $d = 2.8580 \text{ \AA}$ in Eq. (1), the Be cutoff should be observed at a Bragg angle of $43^\circ 50'$. The lead crystal was rotated through this angle by rotating the arm through 2θ . The diffracted beam intensity transmitted by the Be filter was measured and the center of the break was observed at a Bragg angle of $43^\circ 53'$. Hence, the planes were verified as being (111).

The crystal was used in reflection rather than transmission for the following reason. One of the 2×9 in. faces evidenced considerable surface irregularity. By reflecting off the "good" face, irregularities at the opposite face would be much less important than if the crystal were used in transmission.

A rocking curve of the crystal in reflection (taken with the arm stationary and the crystal rotated through its Bragg position) is shown in Fig. 9. The smaller peak to the left of the main peak indicates that there is a small portion of the crystal misaligned by about 40 min with respect to the rest of the crystal. However, because of its relative size and separation from the main peak, its presence was not felt to be of importance.

The full width at half-maximum of the rocking curve in Fig. 9, corrected for background, is seen to be 16.5 min . As shown by Sailor et al.,⁶ the rocking curve for a mosaic single crystal in a beam of white radiation can be approximated by a normal distribution with a standard deviation $\sigma = (\omega^2/2 + \eta^2)^{1/2}$ where η is the mosaic spread of the crystal as defined by Eq. (4) and $\omega = \alpha/2 \sqrt{2 \ln 2}$, α being the horizontal angular divergence of the neutron collimators. For a normal distribution the full width at half-

* Computed from "best values" of lattice parameters given in Ref. 53.

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Fig. 9. Rocking curve for lead crystal in reflection. Peak counting rate is approximately $1.6 \times 10^6/\text{min}$.

maximum = $2\sigma \sqrt{2 \ln 2}$. Hence, we have experimentally determined the quantity $(\omega^2/2 + \eta^2)^{1/2}$ which enters directly into calculation of the integrated reflectivity $\mathcal{R}^0$ as given by Eqs. (20) and (11). Since $\alpha = 9.47$ min, we can use the above information to determine the mosaic spread η of the crystal, which turns out to 6.4 min.

Effects of Parasitic and Double-Bragg Reflections

In Fig. 10 the observed count rate for neutrons reflected from the (111) planes of a lead crystal is plotted as a function of neutron wavelength. Since counts were taken for a preset number of monitor counts, the data are unaffected by changes in reactor power. The jagged appearance of the curve is due to the parasitic and double-Bragg reflections previously described. From this plot, it is difficult to ascertain whether any of the data points in the 1.0 to 2.0 Å interval are free from their perturbing influence.

Use of the Eulerian Cradle

The Eulerian cradle, shown in Fig. 2 and in greater detail in Fig. 5, was described at length in an earlier section. The rock axis, which is independent of the 2:1 drive, is used to rotate the crystal through its Bragg position about a vertical axis. The roll axis, which is the topmost axis of the cradle, permits rotation of the crystal about a horizontal axis parallel to the Bragg planes. Hence, the orientation of these two axes that produces a maximum diffracted intensity is used to locate the crystal in its Bragg position for the collection of data.

However, the parasitic planes which produce the jagged count-rate spectrum in Fig. 10 are not parallel to the primary Bragg planes. Hence, if we rotate the crystal about an axis perpendicular to these planes, the Bragg angles at which given parasitic reflections occur will change, as will their relative intensity. As will be shown shortly, this fact permits us to obtain enough points free of parasitic reflections to determine the unperturbed count-rate spectrum with reasonable accuracy.

The middle axis of the cradle, called the PR (parasitic reflection) axis is designed to enable such a rotation. However, if the crystal is to remain in its Bragg position for different orientations of the PR axis, then obviously the Bragg planes must be aligned perpendicular to this axis. The

Fig. 10. Effect of parasitic and double-Bragg reflections on count-rate spectrum using a lead (111) crystal in reflection. Data uncorrected for background.

alignment is carried out experimentally as follows: The aluminum crystal holder (not shown in Fig. 5) in which the crystal is rigidly held, is screwed atop the Eulerian cradle and can be adjusted relative to the crystal-holder table. First, rocking curves are taken for two different angular position of the PR axis (4° apart in this case). If the peaks of the rocking curves occur at different positions of the rock axis, then the primary Bragg planes are not perpendicular to the PR axis. Hence adjustment of the crystal holder relative to the crystal-holder table must be continued. Such a procedure was followed until the peak positions of the two rocking curves differed by less than 1 min of arc.

Determination of Unperturbed Count-Rate Spectrum

In Fig. 11 sections of three spectral curves are shown for a wavelength interval near the peak of the spectrum. The orientation of the PR axis differed by 2° for each curve. Considered individually, considerable freedom could be exercised in drawing the parasitic reflection-free curve for each. However, from the composite plot, the unperturbed spectrum can be drawn with considerably more confidence. As might be expected, there are wavelengths at which none of the data points are free from the effects of parasitic reflections. The size of the data points is indicative of the statistical uncertainty.

The unperturbed count-rate spectrum, as determined by the above procedure, is shown in Fig. 12 for the wavelength interval 0.5 to $4.0 \text{ \AA}$. The background was determined by turning the crystal 100 min from its Bragg position about the vertical rock axis, and averaged about 2% of the peak intensity.

Spectrum Unfolding

The spectrum unfolding proceeds in straightforward fashion by the application of Eq. (27) to the unperturbed count-rate data. For the sake of discussion, we can rewrite Eq. (27) as

$$C(\lambda) = \sum_{n=1}^m C_n(\lambda), \quad (29)$$

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Fig. 11. Detail of count-rate spectrum for three different orientations of parasitic reflection axis. Data uncorrected for background.

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Fig. 12. Count-rate spectrum for lead (111) crystal in reflection. Corrected for background and effects of parasitic and double-Bragg reflections.

where

$$C_n(\lambda) = \phi(\lambda/n) \mathcal{R}^{\lambda n} \epsilon(\lambda/n). \quad (30)$$

Starting at the short-wavelength end, to a very good approximation $C(\lambda) = C_1(\lambda)$. That is, higher-order contributions to the count rate are negligible. In fact, even at 1.0 Å, $C_2(\lambda)$ is only 1/2% of the observed count-rate and does not exceed 1% of $C(\lambda)$ until a wavelength of 1.2 Å is reached. However, for computational purposes, higher-order terms were taken into account whenever they exceeded 1/2% of the measured $C(\lambda)$. Hence, second-order contributions were included from 1.0 Å onward, third-order from 1.7 Å, and fourth-order from 3.0 Å onward.

The resultant plot of neutron flux per unit wavelength interval versus wavelength is shown in Fig. 13. Since we have been comparing the integrated reflectivity $\mathcal{R}^0$ calculated with μ as given by Eq. (19) to that obtained with $\mu = \Sigma_{\text{TOTAL}}$ (see Fig. 7) it is of interest to see how this difference affects the spectrum unfolded from the experimental data. Hence, in Fig. 14 is plotted the spectrum unfolded in the same manner and from the same count-rate data as in Fig. 13, but with $\mu = \Sigma_{\text{TOTAL}}$. Comparison of these results, as well as a discussion of other spectral details will be made in the following section.

Discussion of Results

A flux distribution can be described in three ways: the flux per unit energy, velocity, or wavelength interval. For a Maxwellian distribution having a characteristic temperature $T(^{\circ}\text{K})$, these distributions have the form

$$\phi(E) = \mathcal{N}(E/kT)^2 [\exp(-E/kT)]/E \quad (31)$$

$$\phi(v) = 2\mathcal{N}(E/kT)^2 [\exp(-E/kT)]/v \quad (32)$$

$$\phi(\lambda) = 2\mathcal{N}(E/kT)^2 [\exp(-E/kT)]/\lambda \quad (33)$$

where $\mathcal{N}$ is the total neutron flux of all energies. Since E , v , and λ are not linearly related, the most probable (peak) fluxes for the three distributions occur at different energies. These are shown in Table I.

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Fig. 13. Reactor spectrum unfolded from experimental data using Eq. (27) with μ given by Eq. (19).

Fig. 14. Reactor spectrum unfolded with $\mu = \Sigma_{\text{TOTAL}}$. Compare with Fig. 13.

Table I. Energy of most probable flux for a Maxwellian distribution.

Flux distribution	Energy of most probable (peak) flux
$\phi(E)$	kT
$\phi(v)$	$(3/2) kT$
$\phi(\lambda)$	$(5/2) kT$

Although one would not expect the in-pile reactor spectrum (let alone one measured exterior to the core) to exhibit true Maxwellian behavior, it is still of interest to see how closely the two compare. Hence, in Figs. 13 and 14, the appropriate Maxwellian distribution, as given by Eq. (33), is also plotted with a temperature and normalizing constant chosen such that the most probable flux is the same as that for the experimentally measured distribution.

Returning to Fig. 7 for a moment, we note that the slope of $\mathcal{R}^0$ in the region around $1.0 \text{ \AA}$ differs for the two methods of calculating μ , the effective absorption coefficient. Hence, it is not surprising that the spectra in Figs. 13 and 14 exhibit different peak wavelengths. Now the experimental uncertainty in the count-rate data from which these spectra were unfolded leads to an estimated maximum uncertainty of $0.01 \text{ \AA}$ in the wavelength of the peak fluxes. Hence, the most probable wavelength of $1.025 \pm 0.01 \text{ \AA}$ in Fig. 13 corresponds to an effective neutron temperature of $361 \pm 7^\circ\text{K}$, while the most probable wavelength $1.075 \pm 0.01 \text{ \AA}$ in Fig. 14 corresponds to an effective neutron temperature of $328 \pm 7^\circ\text{K}$.

Measurements at other reactors indicate that the effective temperature of leakage neutrons is on the order of $50\text{-}150^\circ\text{C}$ above the moderator temperature.⁵⁴ Since the average moderator temperature of the LPTR is 310°K , the spectrum obtained by using $\mu = \Sigma_{\text{TOTAL}}$ gives a temperature difference of only 18°C , while that obtained using μ as given by Eq. (19) yields a temperature difference of 51°C , a value within the observed limits.

As may be seen in Fig. 13, the measured spectrum lies above the Maxwellian distribution at both the short- and long-wavelength ends. The presence of a fission source and the resultant slowing-down spectrum is

responsible for the difference at the short-wavelength end. The disparity at the long-wavelength end is less readily explained. In fact, it can be argued that the measured leakage spectrum should actually fall slightly below the theoretical Maxwellian distribution.⁵⁴ However, in Fig. 1, we note that the beam must pass through a 6-in. lead plug, used to reduce the direct-beam gamma intensity. Now the wavelength interval of 4.0 to 0.5 Å corresponds to an energy interval of 0.0051 to 0.327 eV. Reference to BNL 325 reveals that the total cross section of lead rises steadily from 2.5 barns at 0.0051 eV (4.0 Å) to essentially the free-atom cross section of 11 barns at 0.82 eV (1.0 Å). Hence, the lead plug scatters 4 Å neutrons preferentially to 1 Å, by about a factor of 70 for a 6-in. plug. However, this ratio would only be observable in so-called "narrow-beam" geometry. Since the beam geometry is fairly broad, the filtering action would be less pronounced. The measured spectrum at 4 Å is a factor of 4 greater (relative to 1 Å) than the theoretical Maxwellian distribution. Hence, qualitatively, the long-wavelength behavior of the measured spectrum is plausible. The complexity of the beam geometry makes quantitative comparison difficult. The chief utility of Fig. 13 is that it describes the spectrum available for experimental use at the B-1 beam tube facility.

A final point worth mentioning is that although the measured spectrum at long wavelengths is greater in intensity than that predicted by a Maxwellian, the slope beyond about 3.5 Å is actually steeper than that of a Maxwellian. This is in agreement with leakage spectrum measurements made at the LPTR for wavelengths of 4 Å and greater, using a slotted-cylinder mechanical velocity selector.⁵⁵

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APPENDIX A SOLLER COLLIMATOR CALCULATIONS

Introduction

The elevation and end views of a Soller collimator having its centerline coincident with that of a square reactor channel are shown in Fig. 15. The symbols used in Fig. A-1 and in the equations of this section are those used by Szabó¹⁹ and are defined as follows.

- α the horizontal angular divergence of the collimator,
- β the vertical angular divergence of the collimator,
- L the distance from the neutron source plane to the collimator outlet,
- e the width and height of the reactor channel of square cross section,
- ℓ the total length of the collimator,
- h the total internal height of the collimator,
- s the total internal width of the collimator,
- n the number of collimating channels,
- d the internal width of the collimating channels,
- v the thickness of the channel septa.

The last four symbols are related by the expression $s = nd + (n - 1)v$. Since the angular range of interest here is of the order of a degree or less, the neutron flux at the source plane can be considered isotropic and the collimator angular divergences defined by $\alpha = d/\ell$ and $\beta = h/\ell$. The septa are assumed "black" to neutrons and although total reflection is neglected, Szabó has shown³¹ that its inclusion does not alter the conditions for optimization.

Provided that

$$\alpha \leq [(1/2)(e - s) + d] / L, \quad (A-1)$$

$$\beta \geq (e + h)/2L, \quad (A-2)$$

the optimum number of channels n_{opt} , is given by²¹

$$n_{\text{opt}} = \frac{b + (b^2 + ac)^{1/2}}{a}, \quad (A-3)$$

where

$$a = vW(WY^2 - \alpha LX^2),$$

$$b = \alpha Z/W,$$

$$c = Z^2(\alpha LX^2 - vY^2),$$

Fig. A-1. Elevation (at left) and end views of Soller collimator.

$$W = (aL + v),$$

$$X = (e - h),$$

$$Y = (e + h),$$

$$Z = (s + v).$$

Since n is not a continuous variable, the value obtained from (A-3) is rounded off to the nearest integer. Using this integral value of n , the corresponding maximum intensity is given by

$$I_{\max} = \frac{dn_{\text{opt}}^a}{4} \left(\frac{Y^2}{L} + \frac{X^2}{L - l} \right). \quad (\text{A-4})$$

Equation (A-4) is also valid for calculating the intensity with non-optimum values of n , as long as the conditions of Eqs. (A-1) and (A-2) are met.

The transmitted intensity I is defined¹⁹ as the ratio of the total number of neutrons transmitted per second by the collimator to the neutron flux per unit solid angle at the source plane. Hence, the total number of neutrons leaving the collimator per second is given by $I\phi/4\pi$.

Calculations

Although the above calculational procedure is intended for a beam port of square cross section, with a little care it can be applied to other shapes. For example, the beam port at which the spectrometer is located has a circular cross section 5.56 in. in diameter at the source plane. However, because of the narrow horizontal angular divergence of the collimator (less than 10 min), it can "see" only about 1.2 in. to either side of the vertical centerline of the circular source plane. Hence, as far as the collimator is concerned, the source plane could just as well be a square measuring 5.56 in. on a side. The discrepancy between the two areas is about 3% for the worst case considered. On this basis, the source plane dimension was taken to be the same as the beam port diameter.

Collimator Design

Adjacent experiments plus access requirements for a nearby reactor fission chamber made it difficult to locate the main axis of the spectrometer at the reactor face ($L = 93.5$ in.). Since spatial limitations decreased with increasing distance from the reactor face, it was of interest to determine what the price would be at each position in terms of intensity loss. The

fixed system parameters in this case were $a = 9.47$ in., $v = 0.0196$ in., $e = 5.56$ in., $h = 1.47$ in., and $s = 1.5$ in.. The values of L chosen for comparison were 93.5, 150, and 187.5 in.. Conditions (A-1) and (A-2) are met and the maximum intensity attainable at each position, using Eqs. (A-3) and (A-4) is shown in Table A-1.

Table A-1. Variation of n_{opt} , ℓ_{opt} , and I_{max} with source to collimator outlet distance L .

$L(\text{in.})$	n_{opt}	ℓ_{opt}	$I_{\text{max}}(\text{in}^2) \times 10^4$
93.5	19	22.11	2.35
150	15	29.92	1.62
187.5	13	35.61	1.35

It is interesting to note that the maximum intensity decreases much less rapidly than predicted by the simple inverse square law. The reason is apparent if one considers a given collimator transmitting neutrons from a source plane of infinite extent. The transmitted intensity will be independent of the source plane to collimator outlet distance. Because of the narrow horizontal angular acceptance of the collimator, it cannot "see" the sides of the source plane, while the much larger vertical angular acceptance permits the collimator to "see" that the source plane is of finite vertical extent. Hence, the transmitted intensity would be expected to vary less strongly with distance than for a point source.

The effect of septum thickness on the transmitted intensity is of interest because the complexity of collimator fabrication increases markedly for thin septa.⁴ For the $L = 187.5$ in. position, the maximum intensity for septum thicknesses of 0.005, 0.010, and 0.0196 in. are shown in Table A-2.

Table A-2. Variation of n_{opt} , ℓ_{opt} , and I_{max} with septum thickness v at $L = 187.5$ in.

$v(\text{in.})$	n_{opt}	ℓ_{opt}	$I_{\text{max}}(\text{in}^2) \times 10^4$
0.005	24	21.11	1.57
0.010	18	27.03	1.47
0.0196	13	35.61	1.35

Finally, it is worthwhile to ascertain just how sensitive the transmitted intensity is to deviations from the optimum configuration. For $L = 187.5$ and $v = 0.0196$ in., we have calculated the transmitted intensity for values of n ranging from 7 to 22. In all cases, conditions (A-1) and (A-2) are satisfied, so that Eq. (A-4) can be employed. The results are shown in Table A-3 and are plotted in Fig. A-2.

Table A-3. Variation of transmitted intensity with number and length of collimating channels for $L = 187.5$ and $v = 0.0196$ in.

n	$l(\text{in.})$	$I (\text{in}^2) \times 10^4$
7	72.21	1.14
8	62.32	1.23
10	48.44	1.32
12	39.15	1.34
13	35.61	1.35
14	32.55	1.34
16	27.60	1.33
18	23.74	1.31
20	20.66	1.28
22	18.12	1.25

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Fig. A-2. Transmitted intensity versus number of collimating channels for $L = 187.5$ in. and $v = 0.0196$ in.

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