Perfect Crystals and Imperfect Neutrons*

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Neutron diffraction studies of perfect, non-absorbing crystals have been used to explore aspects of dynamical diffraction theory. The Pendellösung fringe patterns obtained from Laue transmitting crystals with full-spectrum incident radiation have been used to determine crystal structure factors with high precision. It has been established that spherical wave theory is called for, and small crystal curvature must be allowed for, in the interpretation of the fringe patterns. Novel information about the characteristics of the neutron wave packet becomes available from the experimental studies.

Introduction

You may have been puzzled about the wording of my title, so I feel that I owe some words of explanation. As to the first part of my title, 'perfect crystals': if I had anticipated some months ago that our congress was to have scheduled so many sessions devoted to crystal topography with so many expert topographers in attendance, all of which and all of whom are dedicated to the sole proposition that there is no such thing as a perfect crystal, then I am sure I would have hesitated to have used this phraseology. Now I have no desire to run counter current to this tide, (particularly since the neutron experiments that I propose to discuss illustrate deviations from perfection), so I would ask you to enclose in mental quotation marks any references that I might make to perfect crystals. The second part of my title, 'imperfect neutrons', is more worthy of explanation. I was introduced to neutrons many years ago through areas of nuclear physics, and we were told then that neutrons were to be considered as one of the building blocks of atomic nuclei. Furthermore, we learned on the basis of very good experiments that this parcel of nuclear matter possessed dimensions of the order of one hundred thousandth of our Å unit, and when observed by itself, unclothed, it moved with a particular velocity not necessarily the same for different neutrons but always along straight lines unless acted upon by other atomic nuclei or by other field agencies. However, we diffractionists now know, on the basis of equally good experimental evidence, that we must endow the neutron particle with other characteristics, namely the mysterious de Broglie wave property, the wave structure with the characteristic de Broglie wavelength. This endowment presents complications and I outline some of these in Fig. 1.

Here I classify neutrons as being 'perfect' or 'imperfect'. The 'perfect' neutron, in the sense that I am using the term, implies propagation with a fixed propagation wave vector \( \mathbf{k} \), and with infinite dimension, or coherence length, associated with the de Broglie wave structure. If we think of the de Broglie wave structure as being of longitudinal extent \( L \), then we can define \( N \) as the characteristic number of de Broglie waves within this packet and the ideal or perfect neutron would be one where \( N \) was infinitely large. For such a travelling neutron an alternative description would be in terms of its wavelength band width \( \Delta \lambda \), and here \( \Delta \lambda / \lambda \) would be vanishingly small in the limit and its momentum uncertainty would also be vanishingly small. Such a neutron would propagate with completely defined transport properties, i.e. its propagation direction, its group velocity and its de Broglie wavelength would all be completely specified.

On the other hand my designation of an 'imperfect' neutron would be applicable to one in which the wave structure is to be considered as diverging and where the propagation direction is no longer defined, this representing one form of imperfection. For this case we are unable to define precisely the directional propagation of our entity. Another sort of imperfection that I illustrate in Fig. 1 is associated with a finite limitation of the wave packet. Unlike the 'perfect' case with an infinite number of de Broglie waves forming the wave packet, a finite value for \( N \) implies an uncertainty in the de Broglie wavelength to be associated with this moving packet and this means uncertainty in the propagation velocity. Lastly I show in the bottom of the figure another form of imperfection. If we somehow perform a coherent splitting of the travelling neutron wave packet into subpackets, then we have to think of partial neutrons as being in evidence and this is again classified as a state of imperfection. There exist other sorts of imperfection in addition to the three that I have listed and we would have no trouble in extending the list. My reason for illustrating these three types of imperfection is that there exists present experimentation which offers us some information about these states of imperfection, and that is really the theme of this lecture.

As diffractionists we are interested in diffraction by...
crystals, and in talking about perfect crystals we need of course dynamical diffraction theory. There are two forms of experimental approach to this that I shall be discussing, that characteristic of Laue geometry and that of Bragg geometry. In Fig. 2, I show the former of these cases. Here we have a parallel-faced crystal with diffracting planes oriented at right angles to these faces, and we envisage neutron radiation as approaching this crystal at an approach angle $\theta$ relative to the diffracting planes. If we define the incident position of this radiation by means of an entrance slit, slit $A$, then as soon as this radiation enters the crystal, there is a coherent splitting of the incident wave into four travelling wave fields, two in the forward direction and two in the general Bragg direction. These components are all coherent with respect to each other, and there is some modification in the magnitude of the propagating wave vectors. Hence we are in an ideal position to observe coherent interference effects as these wave fields propagate through the crystal. For the case of radiation approaching at exactly the Bragg angle, two of the wave fields can be combined to form one Bloch wave which propagates through the crystal directly along the Bragg planes, in other words perpendicularly to the entrance surface, and the other two wave fields can be combined to form a second Bloch travelling wave which also propagates along the net planes perpendicular to the entrance surface. These two Bloch waves possess slightly different wave vectors, and they can interfere with each other as they propagate through the crystal. Accordingly a beat pattern can be produced, the well known Pendellösung beating action. If one investigates what is released from the back side of the crystal, a very interesting intensity distribution is to be expected according to dynamical diffraction theory. First of all, the Bragg-reflected radiation that is released from the right-hand side of the crystal is no longer confined to a limited area as was the entrance beam, but is spread out, as indicated in Fig. 2, with a finite spatial width. This width depends on the Bragg angle and the thickness of the crystal and this fanning is sometimes referred to as the Borrmann fan within the crystal. What we are first interested in is the intensity distribution that describes the release of Bragg-reflected radiation from the crystal.

I might digress a moment and say a few words about the neutron case insofar as it compares with the X-ray case. X-ray dynamical diffraction theory has been explored for many years and is apparently very well at hand. Are there differences now between the characteristics of neutron dynamical diffraction theory and X-ray dynamical diffraction theory? The answer seems to be that there is very little difference. This has been looked at closely by a number of people including ourselves, and by present-day experimentation standards one is perfectly safe in transcribing X-ray dynamical diffraction theory over to the neutron case. However, the presence of processes (2) and (4) necessitates special handling, which

![Diagram of neutron classifications](image)

Fig. 1. Description of neutron classifications.

![Diagram of Bragg reflection](image)

Fig. 2. Diffraction by a crystal slice with Laue geometry.
in some cases can result in significant changes. In Fig. 2 four travelling waves are illustrated; if this were a crystal with magnetic atoms we would have to consider eight wave components with strong dependence upon the neutron polarization. Indeed neutron experimentalists have been searching for magnetic crystals of sufficient perfection to test some of these novelties. To date no usefully perfect crystals have been uncovered but they will undoubtedly be available in the future. In similar vein, it has been attractive to think of the development of a neutron interferometer system along lines developed so successfully and beautifully by Bonse & Hart (1965) for X-rays. Referring again to Fig. 2, energy propagation through the crystal along the symmetrical directions, ± ε, is shown, which is obtained for an incident ray slightly displaced from the exact Bragg angle. There are coherent components in the two Bragg beams emerging, separated, from the crystal, and it is the challenging task of the neutron experimentalist to get them back together and thereby demonstrate phase interference. Many interesting facets of neutron physics could be explored with such a system. It is also worth mentioning at this stage that most crystals, including silicon and germanium which we shall be considering shortly, are quite transparent to neutron radiation with little true absorption. Thus in dynamical diffraction all wave fields are fully excited in the crystal and only in exceptional cases does the Borrmann effect, anomalous transmission of some of the wave field components, manifest itself. This represents a characteristic difference between the two types of radiation.

**Pendellösung effects in transmitting crystals**

Returning to the characterization of the released Bragg intensity from the crystal slice shown in Fig. 2, dynamical diffraction theory for plane waves of extended width gives for the intensity distribution the expression \( I(γ) \) shown in Fig. 3. Here the parameter \( γ \) describes the energy-propagation direction within the crystal, \( N \) is the scattering-center density, \( F_H \) is the crystal structure factor and \( d_H \) the interplanar spacing value of the diffracting planes. We note the periodic term as being the source of interference Pendellösung effects. However, if an experiment is performed with a neutron beam in which different neutrons of too large a wavelength spread are Bragg diffracted (i.e. poor incident collimation) or if the crystal thickness \( t \) is undefined because of surface roughness, then the Pendellösung effects are averaged, and the simple limiting expression shown in Fig. 3 is expected, as was first shown by Kato (1960). This expression is characteristic of the non-absorbing crystal, not usually encountered in X-ray studies but which is quite valid for the case of silicon crystals (which we shall be considering) and neutron radiation in which true absorption is trivially small. Fig. 4 shows the intensity distribution from a silicon slice in (111) diffraction as obtained with neutrons of rather long wavelength (4.4 Å) by scanning over the Bragg reflection band with an exit slit. For comparison the distribution calculated from the Kato expression, with allowance for finite geometrical resolution, is also shown, and is in good agreement. There is no evidence in this distribution (sometimes referred to as the Kato profile) of Pendellösung effects, and this is because of the experimental limitations mentioned above, namely that the wavelength spread among different diffracting neutrons is too large and the surface structure is not well enough defined by Pendellösung interference standards. It is worth pointing out that this wavelength spread among different neutrons is a different quantity than the intrinsic wavelength band width of a single neutron as envisaged in Fig. 1.

If these experimental conditions are controlled, however, we can now recognize the Pendellösung effects as seen in Fig. 5. Here are shown two profile curves taken with rather shorter wavelength, 1.020 and 1.034 Å mean wavelength values, and one sees the characteristic edges of the Bragg beam, but now a very interesting structure has developed within the profile curve. Changing the wavelength by about 1% from the left to the right-hand case has changed the interference structure noticeably. In one case, we see an intensity minimum at the center position, and in the other it has grown into a maximum. This is a fringe structure and if we were to

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**Fig. 3. Dynamical diffraction by a Lau-ediffraction crystal.**

**Symmetric Laue Diffraction**

**Plane Wave**

\[
I(γ) = c(1-γ^2)^{-1/2} \sin^2 \left[ A \left(1 - γ^2 \right)^{-1/2} \tan θ \right]
\]

\[
A = 2πN(F_H R)^{1/2} d_H
\]

Without Pendellösung (Δλ too large)

\[
I(γ) = c(1-γ^2)^{-1/2}
\]

**Fig. 4. Kato profile of diffracted neutron beam from Laue crystal without absorption.**
take a photograph of this beam as it is released from the crystal under ideal conditions we would see the fringe structure the whole way across, the widest separation of the fringes being at the center, with a narrowing periodicity as we approach the edges. We are seeing only limited interference effects in Fig. 5, again because of finite resolution in the experiment. The slits are of finite size and that means that we are averaging over much of the structure.

The sensitivity to wavelength of the interference structure at the center suggests that we can perform illuminating experiments by studying the central radiation released from the crystal and then changing the wavelength in regular steps. The results of such an experiment are shown in Fig. 6 where the exit slit has been placed directly at the center, along the net planes of the crystal relative to the entrance-slit position, and we have now selectively changed the wavelength of the diffracted radiation in successive steps showing the modulation that develops in this fringe intensity. This is intensity as a function of the orientation angle of the crystal and, as we change the orientation of the crystal relative to the incident beam, then the Bragg relationship implies that we are selecting different wavelengths from the incident-beam radiation. For this case, the radiation that was delivered to the crystal was a full-spectrum neutron beam. Now neutrons are obtained from a nuclear reactor, as Professor Bacon has indicated, and this radiation is always full-spectrum radiation. In this direct reactor radiation there are additionally many fast neutrons (more energetic neutrons which are useless for these experiments), and many γ-rays.

Experimentalists usually do not work with such beams because there is so much contaminant radiation that complicates the experimentation one might want to do. In our case we are utilizing full radiation from the reactor source and this flood of radiation falls on the crystal through the entrance slit. The crystal is extremely selective of what it Bragg diffracts and it selects from this incident beam only about one part in 10⁷ of the incident radiation intensity. In the experiment, the incident beam was collimated by a converging channel of width 3 minutes arc and entered the crystal through a gadolinium-edged slit opening of width 0.13 mm. The wavelength range that is selected by the crystal for Bragg diffraction at a given orientation is dictated then by this collimation angular width and this was purposely arranged to be small.

In Fig. 6 the orders of interference between the two propagating waves passing through the crystal are identified at the fringe maxima. Thus the designation $n = 28$ implies that the two propagating waves have gotten out of phase by 28 de Broglie wavelengths by the time they have reached the exit face of the crystal. Such patterns can be studied over a wide range of wavelengths across the full spectrum that is available from the reactor source. In practice this means that we can measure fringes from the lowest-wavelength position, about 0.6 or 0.7 Å, up to perhaps 2 Å, where the intensity from the source has fallen so much that the fringe contrast becomes troublesomely small. These fringes can be measured over a wide range of interference; our experiments have covered the range from 7 to about 100, as I have indicated. Moreover they can be measured on both sides of the incident-beam direction in this one-axis spectrometer and one then compares the full fringe spectrum that is observed on the right-hand side of the incident beam with that obtained on the left-hand side of the incident beam, correlating the fringe orders on the two sides, thereby obviating the need to know the exact angular position of the incident beam. Thus, many maxima and minima positions are determined as a result of this full-spectrum fringe scanning and considerable precision in the fringe periodicity becomes available.

**Neutron wave characteristics**

We want to interpret this fringe periodicity in terms of physical quantities and here we face some embarrassment. The expression given in Fig. 3 was characteristic of plane-wave radiation but there has also been an equivalent dynamical diffraction theory wherein one envisages the radiation as being of spherical wave character. This again is due to Kato (1961).

![Fig. 5. Pendellösung interference effects within diffracted beam from Laue crystal.](image)

![Fig. 6. Intensity modulation of central rays in diffracted beam as wavelength is varied.](image)
At the beginning of this experimentation we were not prepared to say whether the neutrons in these experiments were of one character or of the other character and it thus became necessary in making the proper interpretation of these fringe locations to establish the character of the neutron radiation as it propagates through the crystal. Fig. 7 illustrates the differences between these two approaches, plane wave theory and spherical wave theory.

In the top section of Fig. 7 I have reproduced the expression that we have seen, and if you study this expression you find that the quantity \((n + \frac{1}{2})/t \tan \theta_n\), with \(\theta_n\) the Bragg angle corresponding to fringe order interference \(n\) at maxima positions, should be a constant across the full fringe spectrum and constant also for different thickness slices. On the other hand with spherical wave theory the intensity distribution becomes a little different, varying as the square of a Bessel function of zero order. It seems to be the rule in physics that in progressing from plane wave to spherical or cylindrical wave cases you change the analytical description from nice well behaved trigonometric functions to Bessel functions, and this is no exception. This Bessel function can be used for all of the present experimental results in limiting form which returns it to the trigonometric form, but now with a somewhat altered argument. The argument of this \(\sin^2\) function has an extra factor \(\pi/4\) which did not appear in the plane wave expression and additionally there is a subtle difference in the appearance of the \((1 - \gamma^2)\) term. The difference between the two expressions is not large but must nevertheless be considered in interpreting the results of the full-spectrum fringe display. I should mention one other experimental detail at this stage about the \((1 - \gamma^2)\) term appearing in the above expressions. The fringe spectrum shown in Fig. 6 is taken with the two slit openings placed accurately along a common net plane in the crystal implying unit value for the term. However, the slit openings are necessarily of finite dimension and thus an averaged value for this quantity deviating slightly from unity is characteristic of the experimental conditions. This average value moreover changes across the spectrum from fringe to fringe and this serves to shift the fringe positions slightly from the ideal geometry case. The magnitude of this correction can be evaluated by computer calculation and allowed for. It is to be noticed that this correction figures differently for the two wave-character cases.

Several arguments can be presented leading to the conclusion that spherical wave theory is applicable in our case. First, in the Kato profile curves of Fig. 5 one should expect interference effects only over the central region defined as the overlap region characteristic of the finite size of the two defining slits if plane wave theory were effective. In practice, however, interference effects are observed over a region several times more extended than this geometrically defined central region, thus suggesting the applicability of the spherical-wave treatment, which has no restriction on the location of the interference effects. Secondly, the two expressions for the intensity distribution predict inverted shifts of the fringe pattern when the exit slit of Fig. 2 is purposely displaced from the exact center position within the central region. Experiments testing this fringe shift have been performed with the result shown in Fig. 8, where the central region extends out to a value for the displacement of about 0.13 mm. It is seen that the direction of the fringe shift is consistent with that characteristic of spherical wave theory and that fringes are observed beyond the central region. Additionally, one expects the requirements of spherical wave theory, namely full coherent excitation of the crystal diffraction process over the Darwin range by a single neutron wave, to be met, since the single-slit diffraction broadening of the incident wave by the entrance slit is about three times larger than the Darwin range for the conditions of the experiment. All of these arguments lead one to accept the applicability of spherical-wave theory and that the coherent wave front of the travelling neutron waves must be curved.

Symmetric Laue Diffraction

Plane wave theory

\[
I(\gamma, \lambda) = c(1 - \gamma^2)^{-1/2} \sin^2 [A(1 - \gamma^2)^{-1/2} \tan \theta]
\]

\[
A = 2 + \tan \theta_n \sqrt{\frac{d_H}{f_H}}
\]

and \((n + 1/2)/t \tan \theta_n\) = constant

Spherical wave theory

\[
I(\gamma, \lambda) = c'(1 - \gamma^2)^{-1/2} J_\nu^2 [A(1 - \gamma^2)^{-1/2} \tan \theta]
\]

\[
c'(1 - \gamma^2)^{-1/2} \sin^2 [\pi/4 + A(1 - \gamma^2)^{-1/2} \tan \theta]
\]

and \((n + 1/4)/t \tan \theta_n\) = constant

Fig. 7. Differences between dynamical diffraction theory with plane-wave and spherical-wave radiation.

Fig. 8. Comparison between experiment and theory for phase shift of fringe pattern for plane and spherical wave character.
Interpretation of Pendell"osung fringe patterns for silicon and germanium

With this assessment of the wave characteristics we can return to the analysis of the fringe pattern which is expected to display a constant value for the variable \((n+\frac{1}{4})/t \tan \theta_a\) across the spectrum and for crystals of different thickness \(t\). We have obtained full-spectrum fringe patterns for five cases, with silicon crystals of three thickness values, and Fig. 9 shows a display of the fringe-order constant \(versus\) spectral wavelength for four of these cases (the fifth case superimposes upon the others and is omitted for clarity). It is seen on this expanded scale drawing that this quantity is \textit{not constant} across the spectrum and moreover there are differences between the different cases. Well, this was very annoying when it was first uncovered. It implied for one thing that the crystal structure factor \(F_{\text{th}}\) or equivalently the atomic scattering amplitude of a silicon atom was dependent upon \(\lambda\), a conclusion which we were not prepared to accept. Considerable effort was expended in trying to understand this unexpected variation from the viewpoint both of experimental artifact and of some misinterpretation of neutron dynamical diffraction. This took different forms ranging from that as crude as the effect of an incorrect angle scale on the spectrometer to that as exotic as the presence of a neutron-spin Borrmann action in which neutrons of one spin state experienced full Borrmann absorption while the other spin state passed properly through the crystal while the other spin state experienced full Borrmann absorption. None of these attempts was successful until the effects produced by residual curvature in the crystals were investigated – we would have arrived at this explanation at an earlier stage if our lines of communication with our friends, the topographers, had been closer.

It turns out that the theory of diffraction of curved or distorted crystals has been studied by a number of people again including Kato (1964) and Fig. 10 illustrates some of the implications for the case of a uniformly curved crystal. We start with the fundamental relationship characteristic of spherical wave theory, but now there is an extra function \(f(p)\) introduced into the argument of the periodic term with \(p\) depending upon various things but significantly upon the radius of curvature \(R\) of the crystal. This is based upon the treatment for a simple model of uniform curvature within a crystal and the expression shows that the quantity that we are calling the fringe-order variable \((n+\frac{1}{4})/t \tan \theta_a\) has a wavelength dependence in first approximation of the form \(a + \beta(\lambda \cos \theta)^{-2}\). This suggests that an examination of this variable as a function of the inverse square of \(\lambda \cos \theta\) for the four cases shown in Fig. 9. The fringe-order variable shows the predicted linear dependence and all of the lines extrapolate to a common intersection point with the different slopes implying different radii of curvature for the various cases. This is rather satisfying and it implies that we can account for the fringe distribution on the basis of a simple model of uniform curvature within the crystal slices as they were examined. I point out that the ordinate scale on this graph is a very expanded one, and even though some of the lines appear to have strong slope values, the actual slope values are not so large and are well determined. The lines are least-squares fitted with higher-order terms being included in the final fitting. One can obtain very precise information from this common extrapolated intersection value on the crystal structure factor and the results for Si are tabulated in Table 1 for the five cases that have been studied.

Fig. 9. Variation of fringe-order variable with neutron wavelength within Si(111) reflection for different crystal slices. The ordinate quantity should be a constant if the crystal possesses no curvature distortion.

Fig. 10. Effect of uniform crystal curvature on diffracted-intensity distribution.

\[
I(\lambda) = c' \sin^2 \left[ \pi/4 + A f(p) \tan \theta \right]
\]

\[
f(p) = \sqrt{2/\left(1 + p^2\right)^{1/2} + p^{-1} \arcsinh p}
\]

\[
= 1 + p^{2/6} - p^{2/3} + p^{5/2} + \cdots
\]

\[
p = \pi^2/RA \lambda \cos \theta
\]

and \((n+1/4)/t \tan \theta = a + \beta(\lambda \cos \theta)^{-2}\)
Three crystal slices $A$, $B$, $C$ of different thickness were examined. Precision thickness values are shown and the fringe-order ranges that were studied are indicated in the Table. These range from 7 to 77 for these cases, and the last column is the quantity, essentially the crystal structure factor, that is determined from the extrapolated intersection of the different lines of Fig. 11. There is some variation here and one is entitled to take a weighted mean of these as shown in the lower part of the Table. Thus the results indicate that the crystal structure factor is determined to a precision of one part in five thousand. Also shown in the Table is the radius of curvature that is calculated from the slopes of the lines that have been obtained. These radii are very large, ranging from 8 to 22 km, indicating a very trivial curvature but yet showing up very sensitively in these full-spectrum fringe patterns. Finally, if one wants to interpret this crystal structure-factor term as a scattering amplitude, an atomic scattering amplitude for neutron radiation, we need to extract the Debye-Waller temperature factor. For Si in 111 reflection it turns out that this factor is quite well known and one can apply the correction given in the Table. The atomic scattering amplitude is given with a precision of about 1 part in 4000. When you have determined the neutron scattering amplitude to this precision you have to worry about its interpretation in terms of contributing components.

There is nuclear scattering; there is also some electronic and nuclear charge scattering that arises because of an interaction between the charge distribution within the atom and the neutron magnetic moment and the neutron intrinsic charge structure. This is labelled as a charge scattering amplitude in the Table. In the Si case, the magnitude of the charge-scattering amplitude in units of the significant figures is about 43, some 4 or 5 times larger than the uncertainty in the value. So this small residual charge-scattering effect is showing up well outside the precision of the determination.

Similar studies have been carried out for Ge crystal slices and Fig. 12 shows a summary of these results. The fringe-order variable again exhibits a linear dependence upon the characteristic wavelength variable with a common intercept for the three cases studied. Again we see this curvature action, and again we interpret the common intercept value at infinite wavelength at the left-hand side of the graph in terms of the crystal structure factor. Here, for these three cases, the fringe-order range extended from 14 out to 94, and Table 2 summarizes the Ge results leading to a mean scattering amplitude. Again we have to apply the Debye-Waller factor and this is not known so well in the Ge case to my knowledge as it is in the Si case. Additionally, we have to recognize that this quantity, the atomic scattering amplitude, is a composite of nuclear scattering and charge scattering. This charge scattering is larger in the Ge case than in the Si case. So we can determine structure factors in practice to high precision by studying these fringe distributions.

### Table 1. Summary of results from neutron Pendellösung fringe study in Si(111) reflection

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Thickness (cm)</th>
<th>Observed fringe-order range</th>
<th>Radius of curvature $(m)$</th>
<th>$b e^{-w}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>0.55524 (8)</td>
<td>17 $\rightarrow$ 44</td>
<td>22000</td>
<td>0.41030 (10)</td>
</tr>
<tr>
<td>$B$</td>
<td>0.23179 (3)</td>
<td>7 $\rightarrow$ 18</td>
<td>8470</td>
<td>0.41041 (25)</td>
</tr>
<tr>
<td>$C$</td>
<td>0.97511 (8)</td>
<td>33 $\rightarrow$ 76</td>
<td>23100</td>
<td>0.41064 (7)</td>
</tr>
<tr>
<td>$C$</td>
<td>0.97511 (8)</td>
<td>42 $\rightarrow$ 73</td>
<td>8700</td>
<td>0.41034 (15)</td>
</tr>
<tr>
<td>$C$</td>
<td>0.97511 (8)</td>
<td>32 $\rightarrow$ 77</td>
<td>15090</td>
<td>0.41069 (10)</td>
</tr>
</tbody>
</table>

Weighted mean $b e^{-w} = 0.41053 (8)$ with $e^{-w} = 0.98842 (12)$, Si(111)

$b_{atom} = 0.41534 (10)$. $10^{-12}$ cm $= b_{nuc} + b_{chg}$
Table 2. Summary of results from neutron Pendellösung fringe study in Ge(111) reflection

<table>
<thead>
<tr>
<th>Thickness (cm)</th>
<th>Fringe-Radius of curvature $b_{e-W}$</th>
<th>(m)</th>
<th>$10^{-12}$ cm/atom</th>
</tr>
</thead>
<tbody>
<tr>
<td>A 0.64745 (10)</td>
<td>36 $\rightarrow$ 94</td>
<td>18250</td>
<td>0.80813 (14)</td>
</tr>
<tr>
<td>B 0.34610 (3)</td>
<td>20 $\rightarrow$ 51</td>
<td>40350</td>
<td>0.80856 (20)</td>
</tr>
<tr>
<td>C 0.24557 (10)</td>
<td>14 $\rightarrow$ 36</td>
<td>2200</td>
<td>0.80854 (42)</td>
</tr>
</tbody>
</table>

Weighted mean $b_{e-W}$ = 0.80829 (15)
with $e-W$ = 0.9868 (3), Ge(111)

$\theta$ = 55°, $\lambda$ = 1.17 Å

Neutron wave-packet considerations

There is other interesting information available from these fringe-pattern studies, and that reflects upon my lecture title, having to do with neutron imperfections. Fig. 13 illustrates what we would expect from uncertainty-principle arguments about the neutron wave-packet dimension that is experienced in this sort of experiment. We have the crystal slice with two slits, one in front and one in back, and there is a geometrical angular range of propagation direction through which neutrons can travel from the entrance side to the exit side as dictated by the size of the slits and by the thickness of the crystal. In our usual situation this angular range is 1-8°, illustrated here as the quantity $\Delta \ell$. This uncertainty in propagation direction within the crystal implies that any one incident neutron, as it approached the crystal from the outside, had an uncertainty in its approach direction, and that can be calculated using the expression given in the Figure. There is a very large de-enhancement factor, 1-8° uncertainty in the propagation direction within the crystal, but only 0.063° of arc as the uncertainty in the approach direction and this characteristic factor, 5 orders of magnitude, permeates all of the dynamical diffraction effects. This uncertainty in approach direction for a single neutron is recognized as being far smaller than the incident-beam collimation through which separate neutrons are introduced to the crystal. This uncertainty in the approach direction can then be correlated with uncertainty in the wavelength of the approaching neutron by the Bragg relation, and finally the uncertainty of the approaching neutron wavelength can be interpreted in terms of the packet dimensions of the approaching neutron. Upon insertion of the numerical quantities, it turns out that the approaching neutron should have been defined by a de Broglie wave packet of longitudinal extent about $2 \times 10^5$ de Broglie wavelengths as that expected from uncertainty-principle arguments.

For comparison with this, the experiments themselves provide some information on this quantity and Fig. 14 shows how the experiment can be interpreted. If one has determined the fringe contrast, namely the intensity ratio at the top and at the bottom of the fringe pattern, you can say something about this coherency dimension. For a typical case which was studied rather carefully, at the 55th fringe order corresponding to a neutron wavelength of a little over 1 Å, the intensity contrast ratio was measured to be at least 50 with this lower limit being set by the unknown contributions to the low intensity at a fringe minimum. Since we have to think of two partial neutrons as having been propagated through the crystal with one lagging behind the other by a distance 55 de Broglie wavelengths as they

![Figure 15](image-url)
reach the exit face for this particular case, we can now interpret the intensity contrast in terms of the packet length. This packet length turns out to be a lower limit; the number of de Broglie waves in the packet according to the experiment should be larger than 2750 corresponding to a packet dimension of about 3000 Å. That is a lower limit and if a more careful assessment of the intensity ratio had been made, we could have defined it better. This is not in disagreement with the prediction of 200 000 waves per packet according to the uncertainty-principle arguments since the experiment asks only for a value larger than 2750. The test fails in a quantitative sense but we have been able to say something about the coherence length of these neutrons. It may well be that a true quantitative comparison can be effected in the future.

**Fig. 16.** Comparison between calculated and experimental back-face diffracted intensity.

**Fig. 17.** Comparison between calculated and experimental end-face diffracted intensity.

### Bragg-reflecting crystals

I have been concentrating on neutron observations made by Laue geometrical conditions, so let me say a few things about neutron release from Bragg-reflecting crystals. In the inset diagram of Fig. 15 is shown the entrance of a rather long wavelength beam on to the face of a Bragg-reflecting crystal with Bragg scattering at an angle of 90°. The intensity profile of the Bragg-reflecting radiation can be studied with a scanning slit and this distribution is shown in the Figure. Fiducial positions A–D are indicated in the figure identifying where the incident beam has struck the entrance face, $A$, or its undeviated projection to the back face, $B$, along with far-corner positions $C$ and $D$. A strong Bragg component is seen to be released at $A$ as well as a weaker component at $B$ followed by an extended tail out to $D$ with localized perturbations at $C$ and $D$. Here we are seeing front-face reflection, back-face reflection and end-face reflection from this crystal slice. Perhaps the most interesting feature in this intensity distribution is the complete absence of any intensity (above a background base line) in the region between $A$ and $B$. This is just what dynamical diffraction theory would predict for the intensity release. If the crystal is perfect there exists no mechanism for intensity release in the Bragg direction from volume points within the crystal, i.e. intensity is released only at surface points to which energy flow from the entrance position has occurred. Imperfections within the crystal volume (a mosaic crystal would be a limiting case) would, of course, modify this distribution.

Such intensity distribution can be interpreted in a very quantitative way with respect to the predictions of dynamical diffraction theory, and Fig. 16 illustrates the comparison between observation and calculation for the $B$ peak, the one that appears to originate at back-face reflection points. The abscissa variable is a parameter describing the scanning-slit position and the calculated curve has included the effects of geometrical resolution determined from the $A$ peak. Moreover, the intensity scale of the calculated curve has been normalized to the intensity of the $A$ peak and good agreement is seen to occur with respect to both the distribution and the absolute intensity level. Finally, Fig. 17 shows the intensity distribution as released from the end face of a crystal slice (there being no back-face reflection for the conditions shown) as compared to that which is calculated. Thus we see that dynamical diffraction theory can provide us with a very good quantitative description of the Bragg reflection effects.

### Concluding comments

This completes what I had to present about crystals and neutrons. Let me say that it has been intriguing to me that we can say something about fundamental neutron characteristics since this is novel information that has not been available from other forms of experimenta-
tion. We can say something about our crystals, not as much as we should like nor as much perhaps as our X-ray friends can, but then we are looking at bulk crystals with, of course, averaging over rather large volumes. Finally, we have seen that we can determine crystal structure factors, or atomic scattering amplitudes, with high precision and these quantities can be useful as standards in the neutron-technology field. It appears to me that there remains an untapped wealth of experimental findings in neutron and crystal physics open for study in such dynamical diffraction investigations.

References


Automatic Peak Determination in X-ray Powder Patterns

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A new technique is presented for the automatic determination of peak positions, intensities and widths from complex numerically recorded X-ray powder diagrams. The method uses standard powders for the determination of the actual repartition of incoming radiations and the determination of the apparatus function (aberrations, calibration etc.). The first trials give results the accuracy of which is much better than one could except from manual determinations.

Automatic recording of X-ray diffractograms has become now very usual, either by means of specific hardware, or by means of a small computer connected in real time to the experimental devices. Notwithstanding the very conventional case where recording consists in a simple strip-chart, most of the automated X-ray diffractometers produce a set of numerical data (count, angles, etc.) which can be transmitted to a computer for further analysis.

On the other hand, many programs have been written by X-ray crystallographers to analyse the data from an X-ray powder diagram. Some of these handle peak profiles to determine some properties of the specimen [Wagner (1967) for example] such as particle size, strain, etc.; obviously this kind of data-reduction program can use raw data from the diffractogram. However, more numerous are the crystallographic programs which do not use the raw data from the diffractogram but elaborated data expressed in terms of peak positions and intensities [such as the well known Johnson & Vand (1969) program for search and match in the ASTM Powder Data File, or the various powder-diagram direct-indexation programs by Ito (1950), Visser (1969), Taupin (1968) etc.].

Unfortunately, only few attempts seem to have been made up to now to automate the intermediate step, that is, determining the accurate peak positions and intensities from a complete diffractogram consisting eventually in a number of peaks which is unknown at first and may be large (ten to hundreds) with possible overlaps. Indeed the most simple cases can be treated by just searching for the counting maxima, stating that a peak is present each time a measurement is higher than the two or four neighbouring points; this is done for instance by Jenkins, Haas & Paolini (1971), but this method has the drawback of not being able to resolve peak clusters, and considering that the peak position is the position of the maximum of the count (which neglects the variation of shape due to the $K\alpha_1$ and $K\alpha_2$ doublet, as well as its influence on the height of the peak). However, the present technique, related to the general deconvolution problem, has been used already in some other domains of the physics involving spectra (Robaux, 1971; Deniz, Janink & Taupin, 1971).

The method we describe here is then an attempt to implement a general and satisfactory method bridging the gap between the automatic-diffractogram recording and the various established programs which require accurate peak positions.

I. Description of the technique

The present method can be divided into three major steps:
(1) determination of the wavelength distribution;
(2) determination of the geometric and instrumental aberrations;
(3) determination of the peak positions and intensities of the specimen.