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The Accurate Measurement of the Wavelength of Monochromatic Neutron Beams

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The wavelength of the monochromatic beam of two different neutron diffractometers has been measured with an extrapolation to eliminate systematic errors, similar to that used in X-ray lattice parameter measurements. An accuracy of 1 part in 5000 has been achieved for the instrument at a high-flux reactor. The wavelength measurement provides the first step towards the absolute determination of lattice parameters by neutron diffraction.

Introduction

Measurements of lattice parameters are seldom made by neutron diffraction, because of the low-beam intensities and poor resolution in comparison with X-ray experiments. If such measurements could be made, however, this would be of benefit for certain special experiments, such as some examinations of a minority phase in a two-phase sample, where measurements are difficult or impossible by X-ray methods. One obvious problem in neutron diffraction is lack of precision in the value of the wavelength of the neutron beam. For example, the wavelengths of the characteristic X-radiation, used in normal powder work, are invariant and known to about 1 part in 10^5 (*International Tables for X-ray Crystallography*, 1962). The wavelength of a neutron beam, on the other hand, depends on the instrument concerned, particularly its geometry and the monochromating crystal used. Although there seems to be no definite convention, it is unusual to find neutron wavelengths specified reliably to better than 1 part in 100, except that occasionally the figures given indicate the power of the adding machines used rather than the experimental accuracy achieved. The aims of the present experiment were not only to establish the value of wavelength for the diffractometers used, but also to find the error in the value. The error in lattice parameter in any subsequent experiment could then be evaluated from the differential form of the Bragg equation.

The determination of neutron wavelengths

The wavelength of the monochromatic beam of a neutron diffractometer is generally measured by experiment. The diffraction pattern of a standard sample is recorded and the neutron wavelength calculated from the angular positions of the most intense peaks and the (known) lattice parameter of the specimen. For example, a standard sample of nickel is frequently used and the calculations applied in turn to the intense 111 and 200 peaks. Although slightly different values of wavelength may result from these two calculations, an average value suffices to the limits of accuracy usually employed.

In accurate X-ray measurements of lattice parameters, the effect of various limitations in the experiments is to produce a systematic variation in the position of Bragg peaks from their expected positions. This results in an apparent variation of lattice parameter with Bragg angle, and the effects of this are corrected for by various extrapolation procedures (Nelson & Riley, 1945). The present measurements were intended to establish whether such extrapolation procedures were applicable to the accurate measurement of neutron wavelength and to establish the correct choice of sample and data analysis accordingly.

Experimental

Two accurate determinations of neutron wavelength have been made, one on the Curran powder diffractometer at the Dido Reactor, A.E.R.E., Harwell, the other on the D2 diffractometer at the high-flux reactor of the Institut Laue–Langevin, Grenoble.

In the A.E.R.E. experiment the ordered alloy Cu_9Al_4 (Bradley, Goldschmidt & Lipson, 1938) was used because of the even distribution of peaks in the diffraction pattern. Some difficulty was experienced, however, in resolving neighbouring peaks at high angles, and only eight peaks were resolved well enough to be used in the analysis. The instrumental resolution could not have been improved further to alleviate this, since the lower peak intensities which would have resulted could not have been tolerated, as the time taken to record the diffraction pattern was 48 h. Some advantage was gained because the diffractometer was a multicounter device and three distinct counter records were amalgamated to provide data.

For the I.L.L. experiment, an NaCl powder sample was used. The reflexions were more suitably spaced and 12 well resolved peaks were obtained. A smaller incremental step length was used, which helped the delineation of the peaks. In this case it took 20 h to record the diffraction pattern.

For both instruments, separate scans were made through the incident beam to evaluate the zero error of the 2θ scale and to examine the incident beam profile.

Extrapolation procedure

The results of the experiments were analysed as follows. The Bragg angle θ for each peak in the diffraction pattern was obtained from its position and the position of the incident beam. Both measurements were an average of mid-chords at several heights. A value of lattice parameter for each peak was calculated, using the approximate value of the wavelength (1·06 Å and 1·14 Å for Harwell and Grenoble respectively). The values of lattice parameter obtained had a systematic variation with θ, as shown in Figs. 1 and 2, where they are plotted against cot θ. In the equivalent X-ray experiment it is unusual, although not impossible, for such a graph to have a positive slope. Figs. 1 and 2 show that for the neutron case both positive and negative slopes may occur. Although the neutron and X-ray experiments are similar there are important differences, such as the fact that the neutron counter travels to just one side of the incident beam, namely the focusing side of the two-axis instrument (Wagner & Kulenkampff, 1922). Although the precise influence of the various limitations in the neutron experiment are not well known, Figs. 1 and 2 suggest that extrapolation against cot θ is a reasonable approximation.

A straight line was fitted to the points of the graphs by computer, by means of a least-squares method. The extrapolated value of the lattice parameter was then noted. The value assumed for the wavelength was then changed (effectively altering the values of ordinate of all the points) until the straight line extrapolated to the known value of lattice parameter of the specimen. Fig. 1 illustrates this for the case of the A.E.R.E. experiment, in which the value of the wavelength was required to be

$$\lambda = 1{\cdot}0612 \text{ Å} .$$

A separate X-ray experiment was carried out to find the lattice parameter of the Cu_9Al_4 specimen, which yielded $a_0 = 8{\cdot}7035 \pm 0{\cdot}0005$ Å, in good agreement with earlier work (Nelson & Riley, 1945).

Exactly the same procedure was followed with the data from the I.L.L. experiment, on the D2 instrument, except that the value of lattice parameter of the sample was taken to be $a = 5{\cdot}6406$ Å. This value was calculated for the measured ambient temperature with the values of lattice parameter and coefficient of thermal expansion provided in *International Tables for X-ray Crystallography* (1962). The wavelength value was found to be $\lambda = 1{\cdot}14981$ Å and in Fig. 2 the single set of points appropriate to this value is shown.

It was necessary to calculate the error in the values obtained in order to decide the number of significant figures, and this could be done with reference to the experimental *errors*, rather than to the systematic *variations* which the extrapolation procedure eliminates.

The points of Figs. 1 and 2 were accordingly fitted by a straight line by means of a least-squares method for three different schemes and this allowed the error in the value of wavelength to be found.

In the first case, all the points were given equal weight and fitted by a straight line as in the equivalent X-ray experiment. In the second case note was taken of the error in each point on the graph, by weighting each point appropriately prior to the fitting. This error was derived from $\Delta a_0 = -a_0 \cot\theta \Delta\theta$, where $\Delta\theta$ is the error in peak position caused by the statistical spread of the counts of that peak, evaluated directly from the counter record. These errors are shown on the points in Figs. 1 and 2, where the smaller error bars in Fig. 2 result from the better statistical reliability of the high-flux instrument and the smaller incremental step in 2θ. The way in which the sizes of these errors vary throughout the diffraction pattern depends on a number of factors, such as the resolution and the intensities of the peaks themselves. Fig. 2 illustrates clearly the distinction between these *errors* and the systematic *variations*, since for the low-angle peaks the systematic variation is largest (as expected) while the random error is small because the peaks are intense and well resolved. In the third case a weighting for the points proportional to the peak area was given, on the simple assumption that the more intense the peak the more accurately its position

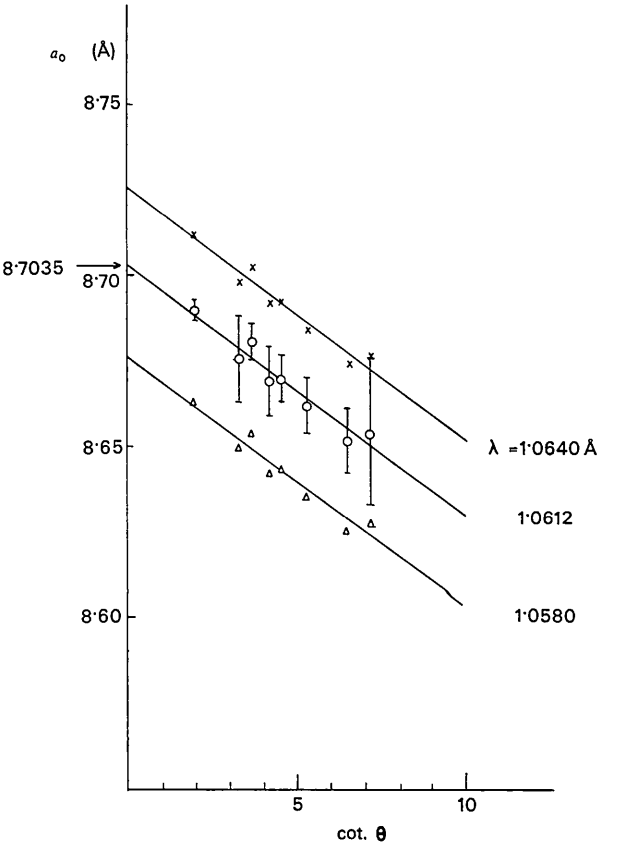

Fig. 1. Variation of measured a_0 with cot θ for Cu_9Al_4, Curran diffractometer. Points resulting from three different choices of wavelength are indicated.

could be found. Such a weighting would be useful in a more superficial analysis in which the real errors Δa_0 were not explicitly evaluated. For each of these three cases a straight line was fitted to the experimental points by means of a least-squares method [see Mulvey (1964), for example]. The resulting lines for the second experiment are shown in Fig. 2. The values of the intercept and the slope of the lines were recorded, and the error in the slope and the error in the intercept also calculated by the computer, with the equations derived from general statistical theory (Mulvey, 1964). The values obtained in the computation are given in Table 1, rounded off in accordance with the errors obtained. (We are grateful to Dr L. Gillott of this department for the use of his program for the above calculations).

Table 1. *Lattice parameter and wavelength values*

	Curran Diffractometer – Cu_9Al_4		D2 Diffractometer – NaCl	
	a_0 (Å)	δa_0 (Å)	a_0 (Å)	δa_0 (Å)
Unweighted	8·703	±0·004	5·641	±0·001
Weighted (1)	8·706	±0·002	5·640	±0·002
Weighted (2)	8·703	±0·003	5·642	±0·001
Mean	8·704	±0·003	5·641	±0·001
Corresponding Wavelength	1·0612	±0·0004	1·1498	±0·0002

The errors in the value of wavelength selected can be evaluated from Table 1, since for a small change in the slope of the fitted line (and hence intercept) there is a corresponding small change in wavelength required to ensure that the line extrapolates to the known value of lattice parameter. From the fractional variation in extrapolated value of lattice parameter we find the error in wavelength.

$$\lambda_{\text{CURRAN}} = 1{\cdot}0612 \pm 0{\cdot}0004 \text{ Å}$$
$$\lambda_{\text{D2}} \quad = 1{\cdot}1498 \pm 0{\cdot}0002 \text{ Å}.$$

Thus by the use of an extrapolation techinque the wavelength of the neutron beams of the two instruments has been measured to about 1 part in 2500 for the A.E.R.E. instrument and to 1 part in 5000 for the I.L.L. instrument. Although the error is worse than the equivalent X-ray value, the experiment does indicate that the neutron diffraction determinations can be made to a greater accuracy than suggested by the approximate results which are normally published. The two experiments described above were wavelength calibrations performed within the course of other investigations. The samples and experimental times were comparable with those of the main experiments. The aim was to find the wavelength value to an accuracy comparable with that of the rest of the investigation rather than to consider the absolute limits of the technique. For example, similar experiments using a high-resolution instrument and an appropriate sample (giving of the order of 100 peaks in the diffraction pattern) might show that the limits of accuracy ultimately rest with the constancy of the reactor itself, rather than with the neutron instrument. One aim in presenting the results is to show that due cognisance must be taken of the possible systematic variations and of methods for their removal. In this context it may be remarked that whilst the growing use of powerful computer methods for the analysis of powder diffraction patterns (Rietveld, 1967) enables structures to be refined to greater accuracy than hitherto, such techniques demand in turn a greater precision in the specification of such input data as the value of the neutron wavelength.

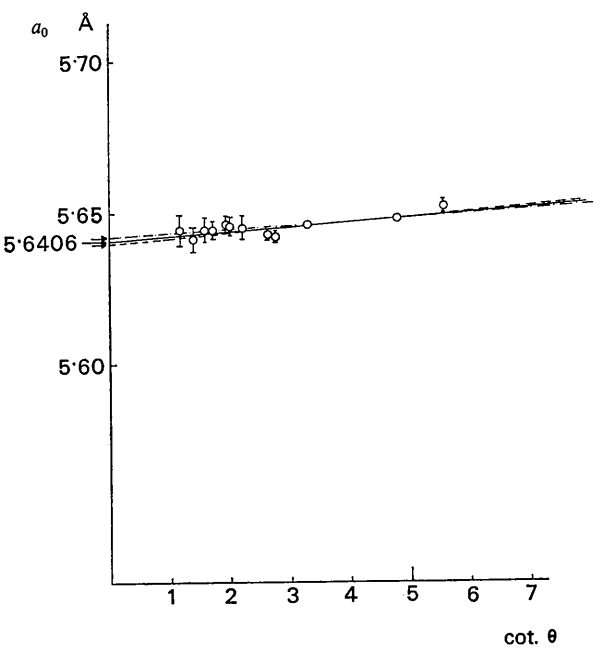

Fig. 2. Variation of measured a_0 with cot θ for NaCl, D2 diffractometer, for value of wavelength $\lambda = 1{\cdot}1498$ Å. The variation in intercept caused by different line fitting procedures is illustrated. ———— all points of equal weight; – – – – – points weighted according to Δa_0; –·–·–·–·– points weighted in proportion to peak intensity.

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References

BRADLEY, A. J., GOLDSCHMIDT, H. J. & LIPSON, H. (1938). *J. Inst. Met.* **63**, 149–161.

International Tables for X-ray Crystallography (1962). Vol. 3. Birmingham: The Kynoch Press.

MULVEY, J. (1964). *High Energy and Nuclear Physics Data Handbook*, chap. 14. The National Institute for Research in Nuclear Science.

NELSON, J. B. & RILEY, D. P., (1945). *Proc. Phys. Soc.* **57**, 160–177.

RIETVELD, H. M. (1969). *J. Appl. Cryst.* **2**, 65–71.

WAGNER, E. & KULENKAMPFF, H. (1922). *Ann. Phys.* **68**, 369–413.