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The Resolution Function in Neutron Diffractometry. II. The Resolution Function of a Conventional Two-Crystal Neutron Diffractometer for Elastic Scattering*

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The relationship of the resolution function of a conventional two-crystal neutron diffractometer for elastic scattering to the resolution function of a three-crystal diffractometer is discussed. An analytic expression is derived for its form assuming Gaussian mosaic and collimation angles and the application of this to the measurement of elastic scattering is considered.

1. Introduction

The layout of a typical two-crystal diffractometer is illustrated in Fig. 1. $2\theta_M$ and $2\theta_S$ are the scattering angles of the monochromator and sample respectively and the settings of these angles define the point in **Q** space for which the probability of detection of a neutron is highest, according to the relations:

$$k_I = \pi/d_M \sin\theta_M \quad (1)$$

$$\mathbf{Q}_0 = \mathbf{k}_F - \mathbf{k}_I\,, \quad (2)$$

where d_M is the d spacing of the monochromator and the angle between $\mathbf{k}_F$ and $\mathbf{k}_I$ (the most probable wave vectors) is $2\theta_S$. The resolution function of the instrument is then the probability of detection of neutrons as a function of $\Delta\mathbf{Q}$ when the instrument has been set to measure a scattering process corresponding to the point $\mathbf{Q}_0$.

In an earlier paper (Cooper & Nathans, 1967), which we shall refer to as paper I*, we have considered the general features of the resolution function of a crystal neutron diffractometer and its experimental determination. An analytic expression was derived for its form for a three-crystal diffractometer assuming Gaussian mosaic and collimation functions. In the present paper we shall consider the case of a two-crystal diffractometer for elastic scattering and we shall derive the corresponding but more simple analytic expression.

Although we shall use the same notation as in paper I and show how the resolution function of a two-crystal diffractometer for elastic scattering is simply a special case of the resolution function derived for a three-crystal diffractometer, we shall derive the two-crystal resolution function independently so that a knowledge of the details of the three-crystal analysis is not necessary.

A detailed knowledge of the resolution function is essential in any study of diffuse scattering since the apparent value of the cross-section will in fact be the mean value obtained by weighting the cross-section throughout **Q** space by the value of the resolution function at each point. It is necessary, even in the measurement of integrated Bragg intensities, if an accurate correction for the contribution of diffuse scattering (*e.g.* thermal diffuse scattering) is required. In addition, the resolution function is directly applicable to the observation of Bragg peaks when details of the mosaic spread of the sample are being studied or when a direct determination of the resolution function is required. We shall therefore consider the application of the resolution function in §3.

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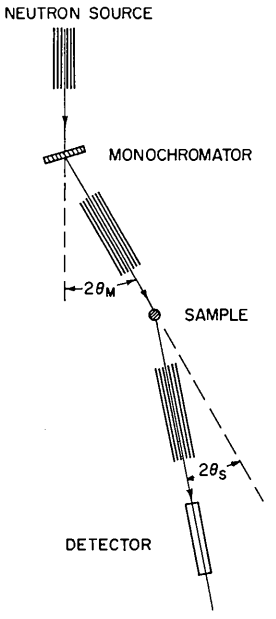

Fig. 1. Layout of a typical two-crystal diffractometer.

2. Derivation of the resolution function

The two-crystal diffractometer provides no energy resolution and in the present analysis we shall consider only elastic scattering processes, *i.e.* $k_F = k_I$.

* The authors are grateful to Dr J. Als-Nielsen for pointing out two minor errors in paper I: in equation (67*b*) there should be a closing bracket after M_{14} and the first $-c_1$ occurring in equation (67*d*) should be omitted, so that the second term becomes $-G_1(M_{14}-c_1M_{44})^2$.

Fig. 2 shows a vector diagram in reciprocal space. $\mathbf{k}_i$ and $\mathbf{k}_f$ are wave vectors corresponding to any elastic scattering process ($k_f = k_i$) in the sample and we shall define $\Delta\mathbf{k}_i = \mathbf{k}_i - \mathbf{k}_I$ and $\Delta\mathbf{k}_f = \mathbf{k}_f - \mathbf{k}_F$. We shall define divergence angles and mosaic and collimation parameters as before (paper I, derivation of the resolution function) but with region 2 as sample to detector.

The probability of a neutron being detected is then as given in paper I, equation (6), equating the analyzer term $P(\Delta k_f, \gamma_2)$ [paper I, equation (5)] to unity ($P_A = 1$, $\tan\theta_A = 0$, $\eta_A = \infty$), and omitting the analyzer to detector terms, *i.e.*

$$P(\Delta k_i, \gamma_1, \gamma_2, \delta_1, \delta_2) = P_M P_0 \exp\left\{-\frac{1}{2}\left[\left(\frac{(\Delta k_i/k_I)\tan\theta_M + \gamma_1}{\eta_M}\right)^2 + \left(\frac{2(\Delta k_i/k_I)\tan\theta_M + \gamma_1}{\alpha_0}\right)^2 + \frac{\gamma_1^2}{\alpha_1^2} + \frac{\gamma_2^2}{\alpha_2^2} + \left(\frac{\delta_1^2}{4\tan^2\theta_M \eta'^2_M + \beta_0^2}\right) + \frac{\delta_1^2}{\beta_1^2} + \frac{\delta_2^2}{\beta_2^2}\right]\right\}. \quad (3)$$

As before, the expression for the probability can be separated into two independent terms, a horizontal term $P_H(\Delta k_i, \gamma_1, \gamma_2)$ and a vertical term $P_V(\delta_1, \delta_2)$ such that the total probability $P = P_H \times P_V$, and the value of the resolution function at a given point in reciprocal space is obtained by integrating the probability over all possible paths $(\mathbf{k}_i, \mathbf{k}_f)$ to that point:

$$R(\mathbf{Q}) = \int P(\mathbf{Q}) d\mathbf{k}_i \,. \quad (4)$$

The resolution function can be written in the form:

$$R(\mathbf{Q}_0 + \Delta\mathbf{Q}) = R_0 \exp\left\{-\tfrac{1}{2}\sum_{k=1}^{3}\sum_{l=1}^{3} M_{kl} X_k X_l\right\}, \quad (5)$$

where $X_1 = \Delta Q_x, X_2 = \Delta Q_y, X = \Delta Q_z$ and for convenience X_1 is taken parallel to $\mathbf{Q}_0$ and X_3 is taken to be vertical. R_0 is the optimum value of the resolution function $R(\mathbf{Q}_0)$. R_0 and M_{kl} are functions of $k_I, \mathbf{Q}_0, \eta_m, d_m, \alpha_0, \alpha_1, \alpha_2, \beta_0, \beta_1, \beta_2$.

The resolution function can thus be calculated as a special case of that derived in paper I, Appendix II, by putting $\omega = 0, P_A = 1, \tan\theta_A = 0, \alpha_3 = \beta_3 = \eta_A = \infty$. On the other hand, the derivation is considerably simplified by these conditions and is therefore given in the Appendix to the present paper to avoid the necessity of considering this as a special case of a three-crystal diffractometer.

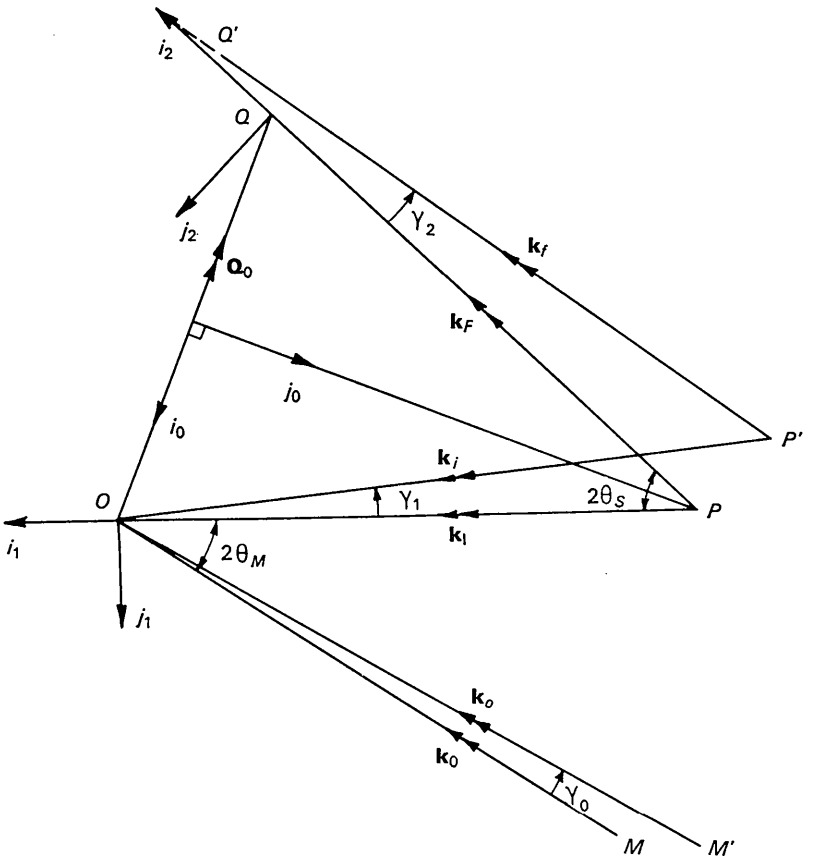

Fig. 2. Vector diagram in reciprocal space for an elastic scattering process.

3. Discussion

As in the case of the three-crystal diffractometer the form of the resolution function derived for elastic scattering from a two-crystal diffractometer, assuming Gaussian mosaic and collimation functions, is such that the loci of constant probability are ellipsoids in **Q** space and the resolution function has a Gaussian dependence on ΔQ for any straight line through $\mathbf{Q}_0$.

Using the derivation given in the Appendix a computer program can readily be written to evaluate the resolution function at any point in **Q** space. The integral of the product of the resolution function and the scattering cross-section will then give the value of the intensity for any setting of the diffractometer:

$$I(\mathbf{Q}_0)=\int R(\mathbf{Q}_0+\Delta\mathbf{Q})\sigma_m(\mathbf{Q}_0+\Delta\mathbf{Q})\Delta\mathbf{Q}\,, \qquad (6)$$

where σ_m is the cross-section integrated over the mosaic spread of the sample, $M(\mathbf{q}_m)$, and $\mathbf{q}_m$ is the reciprocal lattice vector defining translation from the point of optimum Bragg reflection:

$$\sigma_m(\mathbf{Q}_0+\Delta\mathbf{Q})=\int\sigma(\mathbf{Q}_0+\Delta\mathbf{Q}+\mathbf{q}_m)M(\mathbf{q}_m)d\mathbf{q}_m\,. \qquad (7)$$

Equation (6) can thus be used to calculate the theoretical intensity for an experimental scan with any type of elastic scattering cross-section. In the case of a Bragg reflection we may note that if the mosaic function of the sample is also Gaussian in shape then any linear scan in **Q** space through the Bragg reflection will be Gaussian in shape.

We should also point out that P_M in equation (3) is a function of the wavelength since we have

$$P_M\propto\frac{\lambda_I^3F^2}{\sin 2\theta_M}\,, \qquad (8)$$

where $\lambda_I(=2\pi/k_I)$ is the incident wavelength and F is the structure factor for the appropriate reflection.

The dependence of the peak intensity on the incident wavelength will also depend on the geometry of the monochromator and on the wavelength distribution in the primary beam. However, it should be possible in a particular case to determine the optimum wavelength so that the magnitude of the resolution function can be reduced without undue loss in intensity. This factor may be particularly important in the case of a three-crystal diffractometer for the type of phonon measurements discussed in paper I.

APPENDIX
Derivation of the resolution matrix

A vector diagram in reciprocal space is shown in Fig. 2. We shall define sets of orthogonal axes i_1,j_1,l_1; i_2,j_2,l_2 and i_0,j_0,l_0 such that i_1, i_2 and i_0 are parallel to $\mathbf{k}_I$, $\mathbf{k}_F$ and $-\mathbf{Q}_0$ respectively and l_1, l_2 and l_0 are vertical.

If $\Delta\mathbf{k}_i$ has components x_1,y_1,z_1 along axes i_1,j_1,l_1 and $\Delta\mathbf{k}_f$ has components x_2,y_2,z_2 along axes i_2,j_2,l_2 then we can readily determine their components along i_0,j_0,l_0. We have

$$\Delta\mathbf{k}_i=(x_1b+y_1a)\mathbf{i}_0+(-x_1a+y_1b)\mathbf{j}_0+z_1\mathbf{l}_0 \qquad (9a)$$

$$\Delta\mathbf{k}_f=(-x_2b+y_2a)\mathbf{i}_0+(-x_2a-y_2b)\mathbf{j}_0+z_2\mathbf{l}_0\,, \qquad (9b)$$

where

$$a=\cos\theta_S \qquad (10a)$$

$$b=\sin\theta_S\,. \qquad (10b)$$

Then for elastic scattering $x_2=x_1$ and the vector $\Delta\mathbf{Q}=\Delta\mathbf{k}_f-\Delta\mathbf{k}_i$ will have the following components:

$$\Delta Q_x=-x_1 2b+y_2a-y_1a \qquad (11a)$$

$$\Delta Q_y=-y_2b-y_1b \qquad (11b)$$

$$\Delta Q_z=z_2-z_1\,. \qquad (11c)$$

Eliminating y_2 and y_1 from equations (11a) and (11b) respectively we obtain

$$y_1=-\frac{1}{2a}\Delta Q_x-\frac{1}{2b}\Delta Q_y-\frac{b}{a}x_1 \qquad (12a)$$

$$y_2=\frac{1}{2a}\Delta Q_x-\frac{1}{2b}\Delta Q_y+\frac{b}{a}x_1\,. \qquad (12b)$$

If we write

$$y_1=Cx_1+D \qquad (13a)$$

$$y_2=Ex_1+F \qquad (13b)$$

then we have

$$C=-b/a=-\tan\theta_S \qquad (14a)$$

$$E=b/a=\tan\theta_S \qquad (14b)$$

and

$$D=d_1\Delta Q_x+d_2\Delta Q_y \qquad (15a)$$

$$F=f_1\Delta Q_x+f_2\Delta Q_y \qquad (15b)$$

where

$$d_1=-1/2a \qquad d_2=-1/2b \qquad (16a)$$

$$f_1=1/2a \qquad f_2=-1/2b\,. \qquad (16b)$$

The horizontal term in the resolution function is given by

$$R_H\propto\int_{-\infty}^{\infty}\exp\{-\tfrac{1}{2}[(a_1x_1+a_2y_1)^2+a_3^2y_1^2+a_4^2y_2^2+(a_7x_1+a_8y_1)^2]\}dx_1dy_1dy_2\,, \qquad (17)$$

where a_j is given in terms of the collimation parameters:

$$a_1=\tan\theta_M/\eta_Mk_I \qquad a_4=1/\alpha_2k_I$$

$$a_2=1/\eta_Mk_I \qquad a_7=2\tan\theta_M/\alpha_0k_I$$

$$a_3=1/\alpha_1k_I \qquad a_8=1/\alpha_0k_I \qquad (18)$$

[see equation (3)]. We are assuming that Δk_i and Δk_f are small compared with k_I and that the usual small angle approximations are valid, so that $y_1=k_I\gamma_1$, *etc.*

Writing equation (17) as

$$R_H \propto \int_{-\infty}^{\infty} \exp\{-\tfrac{1}{2}[A'x_1^2+B'x_1+C']\}dx_1 \quad (19)$$

and putting

$$\begin{aligned} b_0&=a_1a_2+a_7a_8\\ b_1&=a_2^2+a_3^2+a_8^2\\ b_2&=a_4^2\\ b_5&=a_1^2+a_7^2 \end{aligned} \quad (20)$$

we have

$$A'=2b_0C+b_1C^2+b_2E^2+b_5 \quad (21a)$$

$$B'=2[(b_0+b_1C)D+b_2F] \quad (21b)$$

$$C'=b_1D^2+b_2F^2 \quad (21c)$$

and on integration equation (19) becomes

$$R_H=R_o^h \exp\{-\tfrac{1}{2}[C'-B'^2/4A']\}. \quad (22)$$

From equations (21) it can be seen that

$$C'-B'^2/4A'=g_0D^2+g_1F^2+g_4DF \quad (23)$$

where

$$\begin{aligned} g_0&=b_1-(b_0+b_1C)^2/A'\\ g_1&=b_2-(b_2E)^2/A'\\ g_4&=-2(b_0+b_1C)b_2E/A'. \end{aligned} \quad (24)$$

We can then write the resolution function in the form

$$R(\mathbf{Q}_0+\Delta\mathbf{Q})=R_0 \exp\{-\tfrac{1}{2}\sum_{k=1}^{3}\sum_{l=1}^{3} M_{kl}X_kX_l\}, \quad (25)$$

where $X_1=\Delta Q_x, X_2=\Delta Q_y, X_3=\Delta q_z$ and we have

$$M_{kl}=\tfrac{1}{2}[2g_0d_kd_l+2g_1f_kf_l+g_4(d_kf_l+d_lf_k)] \quad (26)$$

for k and $l=1$ or 2.

Since the vertical term in the resolution function is independent of the horizontal term we have

$$M_{k3}=M_{3l}=0 \quad (27)$$

for $k,l\neq 3$

and the matrix element M_{33} is as derived in paper I, equation (57):

$$M_{33}=\frac{a_{11}^2a_{12}^2}{a_{11}^2+a_{12}^2} \quad (28)$$

where

$$\begin{aligned} a_{11}^2&=\frac{1}{(4\tan^2\theta_M\eta'^2_M+\beta_0^2)k_I^2}+\frac{1}{\beta_1^2k_1^2}\\ a_{12}^2&=\frac{1}{\beta_2^2k_I^2}. \end{aligned} \quad (29)$$

Reference

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Neutron Diffraction Studies of Anharmonic Temperature Factors in BaF_2

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Accurate integrated intensities for the Bragg reflexion of neutrons from BaF_2 have been measured at various temperatures in the range 20–600°C and corrections for the effects of both thermal diffuse scattering and extinction have been applied. The experimental data show systematic deviations from the predictions of a model which assumes harmonic temperature factors for both types of atom, with observed intensities differing by up to ±60% from the calculated 'harmonic' values. Vibrational anharmonicity can be allowed for by using an effective one-particle potential of the form: $V_j(\mathbf{r})=V_{0j}+\frac{1}{2}\alpha_j(x^2+y^2+z^2)+\beta_jxyz$, where x, y and z are the coordinates defining the instantaneous displacement $\mathbf{r}$ of the nucleus of the jth atom and α_j and β_j are the coefficients of the quadratic and cubic terms respectively in the potential expansion. The site symmetry of the atoms allows the anharmonicity parameter β_j to be non-zero only for the fluorine atoms, and introduction of this single parameter brings the observed and calculated structure factors into very good agreement ($R\sim 1\%$) at all temperatures: the R value obtained for a harmonic model increases from 1·8% at room temperature to 8·9% at 600°C. The value obtained for β_F is $-3{\cdot}06\times10^{-12}$ erg.Å^{-3} at room temperature, falling to $-2{\cdot}52\times10^{-12}$ erg.Å^{-3} at 600°C. The present measurements provide no evidence for the existence of any appreciable anisotropic quartic, or higher-order, contributions to the temperature factors. The ratio of nuclear scattering lengths, b_{Ba}/b_F, was refined to 0·932 (±0·004), which gives a value for the nuclear scattering length of barium of $b_{Ba}=0{\cdot}522\ (\pm0{\cdot}011)\times10^{-12}$ cm, assuming $b_F=0{\cdot}560\ (\pm0{\cdot}010)\times10^{-12}$ cm.

1. Introduction

Anisotropy in the temperature factors of cubic crystals with atoms at sites of non-centric cubic symmetry has previously been observed by Willis and co-workers in their studies of UO_2 (Willis, 1963; Rouse, Willis & Pryor, 1968) and CaF_2 (Willis, 1965). In this paper we present the results of a neutron diffraction study of