Reciprocity and Shape Functions in Multiple Scattering Diagrams

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Dedicated to Prof. Dr. K. MOLIÈRE *on the occasion of his sixtieth birthday*

The scattering amplitude for fast electrons diffracted from a plate of crystal is analyzed in terms of multiple scattering diagrams. It is shown that to every individual scattering process in an arbitrary orientation there exists a process in the reciprocity configuration equal in amplitude and phase, that is, to every diagram there corresponds a reciprocal diagram.

It is shown that the geometric part of the diagram may be expressed in terms of the divided differences of the kinematic shape transform.

1. Introduction

The general solution for the scattering of fast electrons from a plate of crystal of thickness H [1], can be written, $U(h,k) = \sum_n U_n(h,k)$ where $U(h,k)$ is the diffraction amplitude of a beam scattered in a direction defined by the intersection of the Ewald sphere with that normal to the crystal surface which passes through the reciprocal lattice point $(h,k,0)$.

The quantity $U_n(h,k)$ can be considered, pictorially, as the diffraction amplitude resulting from the sum of amplitudes from all processes which involve n interactions. A multiple scattering diagram represents one of these processes [2]. The possibility of a simple, representation arises from the particular form of $U_n(h,k)$ which can be separated into the sum of products of structure dependent and geometric dependent parts. Specifically,

$$U_n(h,k) = \sum_l \sum_{h_1 k_1 l_1} \cdots \sum_{h_{n-1} k_{n-1} l_{n-1}} (i\sigma)^n V(h_1,k_1,l_1) \ldots V(h - \sum h_r, k - \sum k_r, l - \sum l_r) \times$$
$$\times \frac{\exp\{-2\pi i \zeta H\}}{(2\pi i)^n} \left[\exp\{i\pi\zeta H\} \left(\frac{\sin \pi \zeta H}{\zeta}\right) \{(\zeta-\zeta_1)\ldots(\zeta-\zeta_{n-1})\}^{-1} + \ldots \right.$$
$$\left. + \sum_{m=1}^{n-1} \exp\{i\pi\zeta_m H\} \frac{\sin \pi \zeta_m H}{\zeta_m} \{(\zeta_m-\zeta_1)\ldots(\zeta_m-\zeta_{m-1})(\zeta_m-\zeta_{m+1}) + \ldots (\zeta_m-\zeta)\}^{-1} \right], \quad (1)$$

where $\sigma = \frac{\pi}{W\lambda} \cdot \frac{2}{1+(1-\beta^2)^{1/2}}$,

W is the accelerating voltage, $\beta = v/c$, λ is the wavelength of the incident electrons with velocity v, and the $V(h_q,k_q,l_q)$ are the crystal structure amplitudes. The ζ_n are the n-beam excitation errors.

A single term in Eq. (1) can be represented by a linear graph in reciprocal space, terminating on the scattering vectors of the constituent processes, the sequence of the segments being that of the interactions. With the conventional construction for the Ewald sphere, the successive vertices of the graph have excitation errors appropriate to the term. Since a diagram has directed segments and labelled vertices, it represents a specific sequence of interactions at a particular angle of incidence.

These diagrams have been used in the classification of symmetries [2, 3]. In the present communication they are used in a discussion of reciprocity [4, 5] and of the shape function.

2. Reciprocity

POGANY and TURNER [6] have shown that reciprocity results from a symmetry inherent in the Green's function, and holds separately for each order of interaction in the Born series. In the present notation, they have shown that a reciprocal relationship holds not only for $U(h,k)$ but also for each $U_n(h,k)$, separately. It will be shown that this analysis can be carried further, and that to every scattering process, represented by a multiple scattering diagram, there exists a process in the reciprocity configuration equal in amplitude and phase.

To simplify the notation the scattering geometry is defined by specifying the centre of the Ewald sphere and the diffraction vector $\boldsymbol{g}$. This vector connects the origin to the intersection of the sphere with that normal to the crystal surface that passes

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through the reciprocal lattice point defined by the three dimensional vector $\boldsymbol{h}$. A simple construction for the reciprocity configuration is then obtained by reflecting the centre of the Ewald sphere in the vector $\boldsymbol{g}$ (Fig. 1).

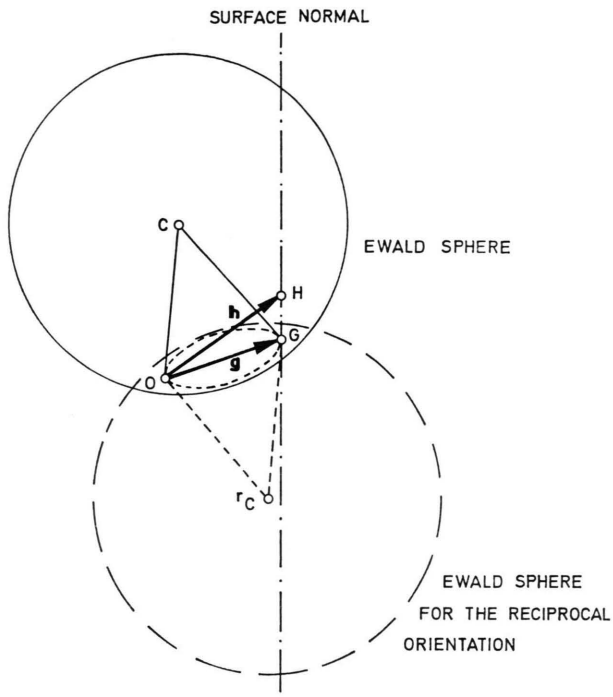

Fig. 1. The reciprocity configuration. The centre of the Ewald sphere C is reflected across the vector g to give the centre of the Ewald sphere ^{r}C for the reciprocal orientation, so that the points O, C, G, ^{r}C all lie in the same plane. It is not necessary for the point H to be in this plane.

As can be seen from Eq. (1), the component of the scattering amplitude due to one n-th order process having the sequence of interactions

$$V(\boldsymbol{h}_1),\ V(\boldsymbol{h}_2),\ldots V(\boldsymbol{h}-\Sigma\,\boldsymbol{h}_p)$$

is of the form

$$\mathbf{V}\,Z \tag{2}$$

where Z is a symetric function of the excitation errors $\zeta_1, \zeta_2, \ldots, \zeta$ at the lattice points

$$\boldsymbol{h}_1,\ \boldsymbol{h}_1+\boldsymbol{h}_2,\ldots,\boldsymbol{h},$$

and $$\mathbf{V}=V(\boldsymbol{h}_1)\,V(\boldsymbol{h}_2)\ldots V(\boldsymbol{h}-\Sigma\,\boldsymbol{h}_p)\,.$$

The proces with the reversed sequence of interactions

$$V(\boldsymbol{h}-\Sigma\,\boldsymbol{h}_p),\quad V(\boldsymbol{h}_{n-1})\ldots V(\boldsymbol{h}_1)$$

has, in the reciprocity configuration, excitation errors

$$^{r}\zeta_1, {}^{r}\zeta_2, \ldots, {}^{r}\zeta$$

at the lattice points

$$\boldsymbol{h}-\sum_1^{n-1}\boldsymbol{h}_p,\ \boldsymbol{h}-\sum_1^{n-2}\boldsymbol{h}_p,\ldots,\boldsymbol{h}\,.$$

The relation between excitation errors at a given lattice point in diagrams related by reciprocity is,

$$\begin{aligned} {}^{r}\zeta(\boldsymbol{p}) &= \zeta(\boldsymbol{h}-\boldsymbol{p})-\zeta \\ \zeta &= -\zeta \end{aligned} \tag{3}$$

where $^{r}\zeta(\boldsymbol{p})$ is the excitation error in the reciprocity configuration at the lattice point $\boldsymbol{p}$. Hence, for every excitation error ζ_m in a diagram D there exists an excitation error $\zeta_m-\zeta$ in the reciprocity configuration of the reversed sequence diagram ^{r}D. The function Z must therefore be evaluated under the transformation,

$$\begin{aligned} \zeta_m &\to \zeta_m-\zeta\,, \\ \zeta &\to -\zeta\,. \end{aligned} \tag{4}$$

For this purpose the appropriate term in Eq. (1) may be written,

$$Z=\frac{\exp\{i\,2\,\pi\,H\,\zeta/2\}-\exp\{-i\,2\,\pi\,H\,\zeta/2\}}{\zeta(\zeta-\zeta_1)\ldots(\zeta-\zeta_{n-1})}+\sum_{m=1}^{n-1}\frac{\exp\{i\,2\,\pi\,H(\zeta_m-\zeta/2)\}-\exp\{-i\,2\,\pi\,H\,\zeta/2\}}{\zeta_m(\zeta_m-\zeta_1)\ldots(\zeta_m-\zeta_{m-1})(\zeta_m-\zeta_{m+1})\ldots(\zeta_m-\zeta)}\,.$$

Hence,

$$^{r}Z=\frac{\exp\{-i\,2\,\pi\,H\,\zeta/2\}-\exp\{i\,2\,\pi\,H\,\zeta/2\}}{(-\zeta)(-\zeta_1)\ldots(-\zeta_{n-1})}+\sum_{m=1}^{n-1}\frac{\exp\{i\,2\,\pi\,H(\zeta_m-\zeta/2)\}-\exp\{i\,2\,\pi\,H\,\zeta/2\}}{\zeta_m(\zeta_m-\zeta_1)\ldots(\zeta_m-\zeta_{m-1})(\zeta_m-\zeta_{m+1})\ldots(\zeta_m-\zeta)}$$

$$=Z-2\,i\,\sin 2\,\pi\,H\,\zeta/2\left[\{\zeta(\zeta-\zeta_1)\ldots(\zeta-\zeta_{n-1})\}^{-1}+\right.$$

$$\left.+\sum_{m=1}^{n-1}\{\zeta_m(\zeta_m-\zeta_1)\ldots(\zeta_m-\zeta_{m-1})(\zeta_m-\zeta_{m+1})\ldots(\zeta_m-\zeta_1)\}^{-1}+\{(-\zeta)(-\zeta_1)\ldots(-\zeta_{n-1})\}^{-1}\right].$$

But, for any q

$$\sum_{s=1}^{p+1}\{q_s(q_s-q_1)\ldots(q_s-q_{s-1})(q_s-q_{s+1})\ldots(q_s-q_{p+1})\}^{-1}=(-1)^p\{q_1,q_2,\ldots,q_{p+1}\}^{-1},$$

since the term on the left hand side is the S-th divided difference of q^{-1}. Hence,

$$^{r}Z = Z\,.$$

Thus the Z function for an arbitrary process is equal to the Z function for the reversed sequence process in the reciprocity configuration. Since the $\mathbf{V}$ function is the same for both processes, the amplitudes and the phases of the processes represented by the two diagram are equal.

The symmetries of the crystal lattice may be associated with reciprocity by combining the appropriate diagrams.

Scattering diagrams for the central beam form over-all closed loops and, accordingly, exhibit special symmetries. For instance, if the crystal has a plane of reflection parallel to the bounding face but no centre of symmetry; for every arbitrary closed loop there exists another having reversed directions of interactions and equal Z function. The sum for these diagrams is therefore $(\mathbf{V}+\mathbf{V}^*)\,Z$, so that the antisymmetric components of the structure amplitudes cancel in pairs. Thus, under these conditions, the rocking curve for the central beam is centrosymmetric, not only in intensity but also in amplitude and phase. This result can also be obtained by summing the pair with reversed sequence, that is, the reciprocally related pair, and applying the operation for the plane of reflection. Summation of all four diagrams is sufficient to guarantee the result when phenomenological absorption[7] is included. GOODMAN and LEHMPFUHL[3] have shown how this result may be exploited in their development of the MÖLLENSTEDT[8] convergent beam technique.

3. Dynamical Shape Function

If the divided differences of the function $f(x)$ at the points $x_1, x_2, \ldots, x_n$ are defined recursively as,

$$\delta^1 f[x_1\,x_2] = \frac{f(x_1)-f(x_2)}{(x_1-x_2)}\,,$$

$$\delta^2 f[x_1\,x_2\,x_3] = \frac{\delta' f[x_1\,x_2]-\delta' f[x_2\,x_3]}{(x_1-x_3)}$$

$$\vdots \qquad\qquad \vdots$$

$$\delta^n f[x_1\,x_2,\ldots,x_n] = \frac{\delta^{n-1} f[x_1\,x_2\ldots x_{n-1}]-\delta^{n-1} f[x_2\,x_3\ldots x_n]}{(x_1-x_n)}$$

then the contribution to the scattering wave amplitude from one diagram can be written

$$_{\mathrm{d}}[U] = \mathbf{V}\,Z = [i\,\sigma\,H\,V(\boldsymbol{h}_1)]\,[i\,\sigma\,H\,V(\boldsymbol{h}_2)]\ldots\left[i\,\sigma\,H\,V\left(\boldsymbol{h}-\sum_{r=1}^{n-1}\boldsymbol{h}_r\right)\right]\times$$

$$\times\,\delta^{n-1}\frac{\exp\{i\,\pi\,H\,\zeta\}}{(2\,i)^{n-1}}\left(\frac{\sin\pi\,H\,\zeta}{\pi\,H\,\zeta}\right)[\pi\,H\,\zeta,\,\pi\,H\,\zeta_1,\ldots,\pi\,H\,\zeta_{n-1}]\,. \qquad (5)$$

The Z function is thus the divided difference of the phase shifted kinematic shape transform at the appropriate vertices. In confluent cases the divided difference operator includes the appropriate partial differential operators to the orders of the confluences. Specifically, if n_1 excitation errors are equal to ζ_1, n_2 are equal to ζ_2, n_p are equal to ζ_p,

$$_{\mathrm{d}}[U] = \mathbf{V}\,\frac{1}{(n_1-1)\,!}\cdot\frac{1}{(n_2-1)\,!}\cdots\frac{1}{(n_p-1)\,!}\cdot\frac{\partial^{n_1+n_2+\ldots+n_p-p}}{\partial x_1^{n_1-1}\ldots\partial x_p^{n_p-1}}\cdot\delta^{n-1}\,S[x_1,\ldots,x_p] \qquad (6)$$

where $x_q = \pi\,\zeta_q\,H$ and S is the phase shifted kinematic shape function. Relations and equalities among the ζ_q may therefore be incorporated in Z and combined with symmetries in $\mathbf{V}$. In the extreme case when $\zeta_1=\zeta_2=\ldots=\zeta=0$, the Z function, by Eq. (6) becomes a factorial so that,

$$U(h,k) = \mathcal{F}_{hk}\exp\{i\,\sigma\int_0^H \varphi(x,y,z)\,\mathrm{d}z\}\,,$$

where $\varphi(x,y,z)$ is the potential in the crystal and

$\mathcal{F}_{hk}$ is the Fourier transform operator. This has the form of a high voltage limit [9, 10]

$$U_{HVL}(h,k) = \lim_{\lambda\to 0} U(h,k)$$

since $U(h,k)$ can be written [11]

$$U(h,k) = \sum_{n=1}^{\infty}\sum_{r=0}^{\infty} \mathbf{V}_n \frac{(i\,2\pi H)^{n+r}}{(n+r)!}\, h_r(\zeta,\zeta_1,\ldots,\zeta_{n-1})$$

where $h_r(\zeta,\zeta_1,\ldots,\zeta_{n-1})$ is the complete homogeneous symmetric polynomial of order r in n variables. Thus the wavelength dependent part of $U_n(h,k)$ is $\sigma^n \lambda^r$, and,

$$\lim_{\lambda\to 0} \sigma^n \lambda^r = \lim_{\lambda\to 0} \left(\frac{2\pi m \lambda}{h^2}\right)^n \lambda^r,$$

$$= \left.\begin{array}{ll} 0 & r>0 \\ \left(\dfrac{2\pi m_0 \lambda_c}{h^2}\right)^n = \sigma_c{}^n & r=0 \end{array}\right\},$$

where $\lambda_c = (h/m_0 c)$ is the Compton wavelength. Thus,

$$U_{HVL}(h,k) = \mathcal{F}_{hk} \exp\{i\,\sigma_c \int_0^H \varphi(x,y,z)\,\mathrm{d}z\}.$$

Similarly the two beam approximation can be obtained as a Taylor's expansion in $V(h)\,V(\bar{h})$, the coefficients being differentials of the kinematic shape transform. By comparison with this form, reductions of the type investigated by NIEHRS [12], may be made. Details will be given elsewhere.

In these reductions it is sometimes easier to express the divided differences as Vandermonde determinants, so that

$$_{\mathrm{d}}[U] = \mathbf{V} \begin{vmatrix} S(x_1) & S(x_2) \ldots S(x_{n-1}) \\ x_1^{n-2} & x_2^{n-2} \quad x_{n-1}^{n-2} \\ \cdot & \cdot \quad \cdot \\ \cdot & \cdot \quad \cdot \\ \cdot & \cdot \quad \cdot \\ x_1 & x_2 \quad x_{n-1} \\ 1 & 1 \quad 1 \end{vmatrix} \div \begin{vmatrix} x_1^{n-1} \ldots x_{n-1}^{n-1} \\ x_1^{n-2} \quad x_{n-1}^{n-2} \\ \cdot \quad \cdot \\ \cdot \quad \cdot \\ \cdot \quad \cdot \\ x_1 \quad x_{n-1} \\ 1 \quad 1 \end{vmatrix},$$

Expressions for the confluent forms are available.

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