A Note on the Integral Equation Formulation of Dynamical Theory *

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The coupled integral equations for dynamical scattering are developed from the general integral equation. The results are given in the forward scattering approximation. Extension to back scattering is briefly mentioned. Expressions for distorted crystals are derived both in the column approximation and beyond. The formulation is suggested to be very useful as a basis for perturbation methods.

Introduction

The development of the dynamical theory of electron diffraction into a sophisticated tool for the calculation of diffracted beam intensities and electron microscope contrast has largely been based upon differential equations. (For reviews, see HIRSCH, HOWIE, NICHOLSON, PASHLEY and WHELAN [1], or KAMBE and MOLIÈRE [2].)

Although integral equation methods have been derived by several authors (FUJIWARA [3], TOURNARIE [4], KAMBE [5]), they do not appear to have become of the same practical importance; especially in contrast calculations, where the HOWIE and WHELAN [6] formulation may be the most widely used. It is the aim of this note to present a formulation in terms of coupled integral equations in a way which may exhibit the advantages of these, particularly as a basis for perturbation methods.

The Coupled Integral Equations

As is shown in standard textbooks in quantum theory, the Schrödinger equation for scattering can be formally solved by introducing the free particle Greens function:

$$\psi(\boldsymbol{r}) = \exp\{i\,\boldsymbol{k}_0\,\boldsymbol{r}\} + \int G(\boldsymbol{r},\boldsymbol{r}')\;U(\boldsymbol{r}')\;\psi(\boldsymbol{r}')\;\mathrm{d}\boldsymbol{r}' \tag{1}$$

where $U(\boldsymbol{r})$ is the scattering potential, $\exp\{i\,\boldsymbol{k}_0\,\boldsymbol{r}\}$ the incident wave and

$$G(\boldsymbol{r},\boldsymbol{r}') = \frac{1}{4\pi}\,\frac{\exp\{i\,k\,|\,\boldsymbol{r}-\boldsymbol{r}'\,|\}}{|\,\boldsymbol{r}\,|\,\boldsymbol{r}'\,|} = \frac{i}{8\pi^2\,k}\int \frac{\exp\{i\,k\,(\boldsymbol{r}-\boldsymbol{r}')\}}{k^2-\varkappa^2}\,\mathrm{d}\boldsymbol{\varkappa}\,. \tag{1 a}$$

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The radial integration through $\varkappa$ is here understood to be taken along a specified contour in the complex $\varkappa$-plane around the pole of the integrand, i. e. at the Ewald sphere. Thus G corresponds to a spherical wave which describes the propagation of a disturbance ψ from a point $\boldsymbol{r}'$ to another point $\boldsymbol{r}$.

Now, spherical waves are unconvenient in the theory of transmission electron diffraction. Here we wish to describe the propagation of the disturbance from a level z' to another level z. This suggests that the z-component, $\varkappa_z$, in wave vector-space should be treated differently from the others. In order to accomplish this, let us write the vectors involved in (1) in terms of a z-component and components normal to the z-axis, viz

$$\boldsymbol{k}_0 = (k_z, \boldsymbol{k}_{0\varrho}),\quad \boldsymbol{r} = (z,\varrho),\quad \boldsymbol{\varkappa} = \boldsymbol{k}_0 + \boldsymbol{\sigma},\quad \boldsymbol{\sigma} = (\zeta, \boldsymbol{s})$$

and, at the outset, perform the integration over the component k_z in the Greens function (1 a) in such a way as to take care of the poles at the Ewald sphere, i. e.

$$G(\boldsymbol{r},\boldsymbol{r}') = \int \exp\{i(\boldsymbol{s}+\boldsymbol{k}_{0\varrho})\,(\boldsymbol{\varrho}-\boldsymbol{\varrho}')\} \cdot \int \frac{\exp\{i\,(k_z+\zeta)\,(z-z')\}}{(k_z+\zeta)^2+(\boldsymbol{s}+\boldsymbol{k}_{0\varrho})^2-k^2}\,\mathrm{d}\zeta\,\mathrm{d}\boldsymbol{s} \tag{2}$$

where the ζ-integration is understood to follow the contour given by FUJIWARA [3] (1959) in his derivation of the Born series for diffraction from a parallel plate crystal (Figure 1). When $z>z'$, that is for forward scattering, the integration contour can be closed in the upper half plane. There results:

$$G_{\mathrm{forw}} = 2\pi i \int \exp\{i\,\boldsymbol{k}_0(\boldsymbol{r}-\boldsymbol{r}')\} \cdot \exp\{i\,\boldsymbol{s}\,(\boldsymbol{\varrho}-\boldsymbol{\varrho}')\}\,\frac{\exp\{i\,\zeta_{\mathrm{s}}\,(z-z')\}}{\zeta_{\mathrm{s}}-\zeta^{\mathrm{s}}}\,\mathrm{d}\boldsymbol{s}$$

* To Professor K. MOLIÈRE on his 60-th birthday.

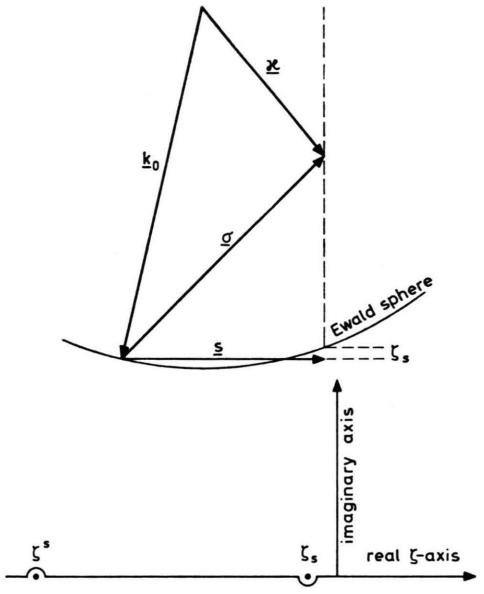

Fig. 1. a) Vectors in reciprocal space, the z-axis points downwards. b) Integration contour for the ζ-integration, Equation (2).

where $\zeta_s = -k_z + \sqrt{k^2 - (\boldsymbol{s} + \boldsymbol{k}_{0\varrho})^2}$ is the height of the Ewald sphere above the point $\boldsymbol{k}_0 + \boldsymbol{s}$ in reciprocal space, that is, an excitation error. The other pole: $\zeta^s = -k_z - \sqrt{k^2 - (\boldsymbol{s} + \boldsymbol{k}_{0\varrho})^2}$. The denominator $\zeta_s - \zeta^s$ can, as a rule, be substituted by $2\,k_z$ (or $2\,k$ when the z-axis is close to the direction of incidence).

The back scattering Greens function is obtained for $z < z'$; the integration contour should then be closed in the lower half plane:

$$G_{\text{back}} = -2\pi i \int \exp\{i\,\boldsymbol{k}_0(\boldsymbol{r}-\boldsymbol{r}')\} \cdot \exp\{i\,\boldsymbol{s}(\boldsymbol{\rho}-\boldsymbol{\rho}')\}\,\frac{\exp\{i\,\zeta^s(z-z')\}}{\zeta_s - \zeta^s}\,\mathrm{d}\boldsymbol{s}\,.$$

The choice of integration path evidently serves to define the directions of forward and backward scattering.

Let us now derive coupled integral equations for forward scattering in a perfect crystal. On taking

$$U(\boldsymbol{r}) = \sum_g U_g \exp\{i\,\boldsymbol{g}\,\boldsymbol{r}\}$$

and

$$\psi(\boldsymbol{r}) = \sum_g \psi_g(z)\,\exp\{i(\boldsymbol{k}_0 + \boldsymbol{g})\,\boldsymbol{r}\}$$

and neglecting back scattering, we obtain from (1) and (2)

$$\psi(\boldsymbol{r}) = \exp\{i\,\boldsymbol{k}_0\,\boldsymbol{r}\} + \frac{i}{8\pi^2 k}\sum_g\sum_g \int\int\int \exp\{i\,\boldsymbol{k}_0(\boldsymbol{r}-\boldsymbol{r}')\} \times \exp\{i\,\boldsymbol{s}(\boldsymbol{\rho}-\boldsymbol{\rho}')\}\,\exp\{i(\boldsymbol{k}_0 + \boldsymbol{g} + \boldsymbol{g}')\,\boldsymbol{r}'\} \times \exp\{i\,\zeta_s(z-z')\}\,U_{g'}\,\psi_g(z')\,\mathrm{d}z'\,\mathrm{d}\boldsymbol{s}\,\mathrm{d}\boldsymbol{\rho}\,. \tag{3}$$

The integration over $\boldsymbol{\rho}'$ yields

$$(2\pi)^2\,\delta\,(\boldsymbol{s} - (\boldsymbol{g}_0 + \boldsymbol{g}')_o)\,,$$

so that the integration over $\boldsymbol{s}$ becomes trivial. On identifying coefficients of $\exp\{i(\boldsymbol{k}_0 + \boldsymbol{h})\,\boldsymbol{r}\}$ on both sides of the equation, we obtain:

$$\psi_h(z) = \delta_{0h} + (i/2\,k)\sum_g \int_0^z \exp\{i\,\zeta_h(z-z')\} \times U_{h-g}\,\psi_g(z')\,\mathrm{d}z' \tag{4 a}$$

where ζ_h is the excitation error for the reflection h, see Figure 1. From these coupled integral equations, for the scattering, the Born series expansions derived by FUJIWARA [3] and by COWLEY and MOODIE [7] can be obtained readily. On multiplication by $\exp\{-i\,\zeta_h z\}$ and derivation on both sides, we obtain the linear differential equations introduced by HOWIE and WHELAN [6].

Sometimes it is convenient to introduce amplitudes

$$\varphi_h(z) = \exp\{-i\,\zeta_h z\}\,\psi_h(z)$$

referred to wave vectors of equal length, rather than of equal z-components. From (4 a) there results

$$\varphi_h(z) = \delta_{0h} + (i/2\,k)\sum_g \int_0^z \exp\{i(\zeta_g - \zeta_h)\,z'\} \times U_{h-g}\,\varphi_g(z')\,\mathrm{d}z'.$$

This latter form is particularly convenient when sums and differences of amplitudes are considered in patterns of high symmetry.

Distorted Crystals

Let us now turn to diffraction by distorted crystals. Let the distortion be described by a spatially varying displacement $\boldsymbol{R}(\boldsymbol{r})$; the potential is then given by

$$U(\boldsymbol{r}) = \sum_h U_h(\boldsymbol{\rho}, z)\,\exp\{i\,\boldsymbol{h}\,\boldsymbol{r}\}\,,$$

where

$$U_h(\boldsymbol{\rho}, z) = U_h^0\,\exp\{i\,\boldsymbol{h}\,\boldsymbol{R}(\boldsymbol{r})\}\,,$$

U_h^0 referring to the undistorted crystal. The variation of the Fourier potentials, U_h, with z does not

raise any problems; the above derivation of Eq. (4) holds equally for this case. The difficulties are connected with the $\boldsymbol{\rho}$-variation in U_h, and hence in ψ_h. Returning to the integration over $\boldsymbol{s}$ and $\boldsymbol{\rho}'$ in Eq. (3), where U_h and ψ_h now depend on $\boldsymbol{\rho}'$, we may introduce the so-called "column approximation" (HOWIE and WHELAN[6] 1962, see also HIRSCH et al.[1] 1965) in two ways: Either we may take the ϱ'-integration first, assuming U_h and ψ_h to vary only slowly with $\boldsymbol{\rho}'$. Such an approach is discussed, in relation to the differential equation, by HOWIE and BASINSKI[8]. Alternatively, we can start with the $\boldsymbol{s}$-integration and, at first, neglect the dependence of ζ_s with s around $\boldsymbol{s}=\boldsymbol{h}_0$. The integration then yields $\delta(\boldsymbol{\rho}-\boldsymbol{\rho}')$ – which is the column approximation – as the first term. Higher order terms can be included by expanding the integral in orders of $(\boldsymbol{s}-\boldsymbol{h}_0)$.

Let us follow this latter approach and consider the contribution to $\psi_h(\varrho, z)$ from $\psi_g(\boldsymbol{\rho}', z')$; i. e. the integral

$$\frac{\pi i}{\varkappa} \int \exp\{i(\boldsymbol{s}-\boldsymbol{h})(\boldsymbol{\rho}-\boldsymbol{\rho}')\} \times \exp\{i(\zeta_s-\zeta_h)(z-z')\}\, \mathrm{d}\boldsymbol{s} \exp\{i\zeta_h(z-z')\} \times U_{h-g}(\boldsymbol{\rho}', z')\, \psi_g(\boldsymbol{\rho}'\cdot z')\, \mathrm{d}z',$$

where

$$\zeta_s-\zeta_h = -\frac{1}{2k_z}(\boldsymbol{s}^2-2\boldsymbol{k}_0\cdot\boldsymbol{s}) + \tfrac{1}{2}k_z(\boldsymbol{h}^2-2\boldsymbol{k}_0\cdot\boldsymbol{h}) = \frac{+1}{2k_z}\left(2(\boldsymbol{k}_0+\boldsymbol{h})(\boldsymbol{h}-\boldsymbol{s}) + (\boldsymbol{h}-\boldsymbol{s})^2\right),$$

$\boldsymbol{h}$ is here assumed to have no z-component.

We can now classify our approximations according to the order of $(\boldsymbol{s}-\boldsymbol{h})$ retained in the argument of the exponential $\exp\{i(\zeta_s-\zeta_h)(z-z')\}$. Carrying out the two-dimensional $\boldsymbol{s}$-integration, we thus obtain

$$\psi_h(\varrho, z) = \delta_{0h} + (i/2k)\sum_g \int_0^z \exp\{i\zeta_h(z-z')\} \times \int U_{h-g}(\boldsymbol{\rho}')\, F_h(\boldsymbol{\rho}-\boldsymbol{\rho})\, \mathrm{d}\varrho'\, \mathrm{d}z'$$

where $F_h(\boldsymbol{\rho}-\boldsymbol{\rho}')$ in the different approximations is given by

i) *zeroth or column approximation,*

$$F_h(\boldsymbol{\rho}-\boldsymbol{\rho}') = \delta(\boldsymbol{\rho}-\boldsymbol{\rho}')$$

ii) *first order,*

$$F_h(\boldsymbol{\rho}-\boldsymbol{\rho}') = \delta\left((\boldsymbol{\rho}-\boldsymbol{\rho}') - \frac{\boldsymbol{k}_0\varrho+\boldsymbol{h}}{k_z}(z-z')\right)$$

This can be described as columns along the different rays; in the two beam case, the "Takagi triangle" (TAKAGI[9])

iii) *second order*

$$F_h(\boldsymbol{\rho}-\boldsymbol{\rho}') = \frac{k\,i}{2\pi(z-z')}\exp\left[i\frac{\left(\boldsymbol{\rho}-\boldsymbol{\rho}'-\frac{\boldsymbol{k}_0\varrho+\boldsymbol{h}}{k_z}(z-z')\right)^2}{(z-z')/k_z}\right]$$

This expression includes Fresnel diffraction effects and may be used for a more accurate refinement of column approximation results.

Concluding Remarks

The integral equations studied in this note can, in many ways, be regarded as the integral form of the linear differential equations extensively used in contrast calculations, see HIRSCH et al.[1]. The integral form may most likely offer advantages when perturbation methods are to be applied. These are particularly important in two fields of application: One is the treatment of diffuse scattering, where a general formulation in terms of the integral equation has been given previously (GJØNNES[10]) and also of weak beams, such as superlattice reflections.

Another field, which is receiving increasing attention is the analysis of fine details in the electron microscope contrast, e. g. from small defects. It may appear that the present formulation offers a convenient basis for such perturbation calculations.

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