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The Calculation of Electron Diffraction Intensities by the Multislice Method

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The calculation of wave functions of scattered electrons by the multislice method of Cowley and Moodie with a finite number of beams is shown to lead to the solution of a finite, closed set of differential equations in the limit that the slice thickness approaches zero. The solution is normalized but differs from the exact wave function unless sufficient beams are included in the calculation. Hence, normalization is not sufficient to ensure that the computed wave function equals the exact wave function. The implications of this result for numerical work are discussed.

Introduction

The multislice method of Cowley and Moodie has been shown by Goodman & Moodie (1974) to give the solution of the form of Schrödinger's equation in which backscattering is neglected. If one imposes the condition that the electrons are scattered by a periodic potential then this equation is equivalent to a countably infinite set of coupled differential equations describing the amplitudes and phases of diffracted beams as a function of position in the scatterer.

Since numerical methods of solving Schrödinger's equation can account for the effects of only a finite number of beams it is worth examining the properties of approximate solutions obtained by methods involving a finite number of beams, and to see how the accuracy of these solutions might be estimated. While it is often not possible to determine analytic solutions to problems in which the effects of three or more beams are important, certain properties of the solutions can be obtained.

One method of determining approximate solutions to Schrödinger's equation is to consider a closed, finite subset of the differential equations which are equivalent to Schrödinger's equation. It can then be shown that the solution of this finite set of equations is normalized, a term which is defined later and which implies that the number of electrons incident on the scatterer equals the number leaving it. This is also a property of the exact solution of Schrödinger's equation. To determine whether the solution is an adequate approximation to an unknown exact solution it must be compared with the solution to a different set of differential equations.

Goodman & Moodie (1974) have suggested that the multislice method has an advantage over the method of truncating the set of differential equations in that it is possible to determine the accuracy of a wave function obtained by the multislice method with a finite number of beams without having to compare the results of calculations with different numbers of beams. Since for any non-zero slice thickness it is only in the limit that the number of beams becomes infinitely large that a multislice calculation results in

a normalized wave function, it is argued that the accuracy of a wave function can be determined by considering how close it is to being normalized.

In this note it will be shown that the multislice method leads to a normalized wave function also in the limit that the slice thickness approaches zero. This result holds irrespective of the number of beams that are considered. The normalized function is just the solution to the set of differential equations which result when all but those beams used in the multislice calculation are equated to zero. The implication of this result is that the condition of normalization is not sufficient to determine that a wave function obtained by the multislice method is accurate, so calculations involving different numbers of beams must be compared.

We first show that using the multislice method with a finite number of beams results in the solution to a finite set of differential equations in the limit that the slice thickness approaches zero. This can be proved by the method used by Goodman & Moodie (1974) to show that the multislice method yields the solution to Schrödinger's equation. However, in that work no restriction was placed on the number of beams. The property of normalization is then defined and it is shown that the solution of the differential equations is normalized. The consequences of this property for numerical work are discussed and some examples are given.

Mathematical formulation

In the following, for notational convenience, we consider a one-dimensional problem in which the scattering potential is given by

$$\varphi(x)=\sum_{h=-\infty}^{\infty} V(h)\exp(-2\pi i h x/a). \qquad (1)$$

The complex amplitude of a diffracted beam at a distance z from some reference line is denoted by $u(h, z)$ and the excitation error of a particular reflexion is written as $\zeta(h)$. The hth Fourier component of $\exp[i\sigma\varphi(x)\Delta z]$ is denoted by $q(h,\Delta z)$. The interaction constant σ is given by

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

where W is the accelerating voltage, λ the wavelength and $\beta=v/c$, v being the velocity of the electron. The multislice method enables $u(h, z+\Delta z)$ to be calculated given $u(h, z)$ by the following expression

$$u(h, z+\Delta z)=\exp[2\pi i\zeta(h)\Delta z]\times\sum_{h'=-\infty}^{\infty} q(h-h', \Delta z)u(h', z). \qquad (3)$$

To describe the way in which the multislice method is used in numerical calculations when only a finite number of beams is considered we introduce an operator $P(h)$ which has the value unity for those values of h which are included in the calculation and which is zero otherwise.

Numerically then one proceeds as follows: assume that at some thickness z all but a finite number of beams have zero amplitudes. The amplitudes of the remaining beams are denoted by $w(h, z)$. For instance, at the entrance face of the crystal this condition will apply if the incident electron beam is represented by a plane wave. Similarly, if the amplitudes of the diffracted beams are calculated by the method described below, there will be only a finite number of non-zero beams. We note here that $P(h)w(h, z)=w(h, z)$.

To calculate an approximation to the amplitude of the hth diffracted beam at $z+\Delta z$, given that its amplitude at z is $w(h, z)$, one computes the expression

$$\exp[2\pi i\zeta(h)\Delta z]\sum_{h'=-\infty}^{\infty} q(h-h', \Delta z)w(h', z) \qquad (4)$$

for those values of h to be included in the calculation. The amplitudes of beams corresponding to other values of h are set equal to zero. The resultant function of h is denoted by $w(h, z+\Delta z)$ and equals $P(h)w(h, z+\Delta z)$. The above operations may be described by the equation

$$w(h, z+\Delta z)=P(h)\exp[2\pi i\zeta(h)\Delta z]\times\sum_{h'=-\infty}^{\infty} q(h-h', \Delta z)w(h', z). \qquad (5)$$

For sufficiently small values of Δz it is a good approximation to write

$$w(h, z+\Delta z)=w(h, z)+2\pi i\zeta(h)\Delta z w(h, z)+i\sigma\Delta z P(h)\sum_{h'=-\infty}^{\infty} V(h-h')w(h', z) \qquad (6)$$

so that in the limit that $\Delta z\to 0$ one obtains the differential equation

$$\frac{\mathrm{d}}{\mathrm{d}z}w(h, z)=i\sigma\sum_{h'=-\infty}^{\infty} P(h)V(h-h')P(h')w(h', z)+2\pi i\zeta(h)w(h, z). \qquad (7)$$

In deriving (7) the relation $P(h)w(h, z)=w(h, z)$ has been used.

We have shown that the diffracted beams, $w(h, z)$, calculated by the multislice method in the limit that the slice thickness approaches zero satisfy a closed finite set of differential equations.

It should be noted that no restriction has been placed on the index $h-h'$ in (7). For numerical work it may be convenient to restrict $h-h'$ to those values of the indices of beams the intensities of which are not set equal to zero. This is equivalent to considering a potential function with Fourier coefficients $V(h)$ which satisfy $P(h)V(h)=V(h)$.

We now show that the sum of the intensities of the beams described by (7) is independent of z. If a wave

function satisfies this property it will be called normalized. The differential equations (7) may be written in the form

$$\frac{\mathrm{d}\mathbf{w}}{\mathrm{d}z} = i\mathsf{M}\mathbf{w} \tag{8}$$

where $\mathbf{w}$ is a vector, the components of which are the amplitudes of the diffracted beams and M is a matrix which is Hermitian as a consequence of the Fourier coefficients of a real potential function φ satisfying $V(-h)=V^*(h)$. The sum of the intensities of the beams is given by $\mathbf{w}^*\mathbf{w}$. This quantity can be seen to be independent of z by considering $\mathrm{d}(\mathbf{w}^*\mathbf{w})/\mathrm{d}z$ and noting that $\mathrm{d}\mathbf{w}^*/\mathrm{d}z = -i\mathbf{w}^*\mathsf{M}$.

This result shows that in the limit as the slice thickness approaches zero one obtains a normalized wave function by the multislice method irrespective of the number of beams used in the calculation. The solution to (7), $w(h, z)$, is not equal to $P(h)u(h, z)$ where $u(h, z)$ is the exact value of the amplitude of the hth diffracted beam unless $P(h)=1$ for all values of h. Hence, it is not a sufficient condition to calculate a normalized wave function to ensure that the exact solution has been obtained.

Consider now a numerical computation using the multislice method with some non-zero value of the slice thickness and some finite number of beams. If the sum of intensities of the beams at the exit face of the crystal is close to the intensity of the incident beam then the wave function may be considered as being nearly normalized. If a computed wave function is not nearly normalized it does not follow that more beams are required to obtain an adequate approximation to the exact wave function since it may be necessary only to reduce the slice thickness.

On the other hand, if a nearly normalized function is computed it will still be necessary to compare that function with the results of a calculation involving a different number of beams to determine that an adequate number of beams has been considered.

Examples

These points are illustrated by some examples. First, if only one beam $w(0)$ is included in a calculation, the solution $w(0, z)=\exp[i\sigma V(0)z]w(0, 0)$ is obtained by the multislice method in the limit that the slice thickness approaches zero. This solution is normalized but, for a potential function which is not a constant, it is not equal to the exact wave function.

Consider next the case $\varphi(x)=V\cos(2\pi x)$, $\zeta(h)=0$ and $\lambda=0$, when the exact values of the amplitudes of the diffracted beams are given by

$$u(h, z)=(i)^h J_h(2\sigma Vz) \tag{9}$$

where J_h is a Bessel function of order h. In the case that the multislice method is used with three beams we obtain the functions

$$\begin{aligned} w(0,z)&=\cos(\sqrt{2}\sigma Vz)\\ w(\pm 1,2)&=(i/\sqrt{2})\sin(\sqrt{2}\sigma Vz) \end{aligned} \tag{10}$$

which are the solution to the three differential equations that result when only three beams are taken as non-zero. This solution is normalized but not equal to the solution (9).

These two examples show that normalization of a wave function obtained by the multislice method does not imply that it is the exact wave function. As an example that it may not be necessary to consider more beams if a computed wave function is not close to being normalized, consider the case $\varphi(x)=V\cos(2\pi x)$, $\zeta(1)=0$ and $\lambda=0{\cdot}07$ Å, the wavelength corresponding to an energy of 30 keV. A two-beam multislice computation when $V=10$ V, $z=100$Å and $\Delta z=10$ Å results in the sum of the intensities of the two beams being only 0·8 of the intensity of the incident beam. However, it can be demonstrated by numerical calculations that no more beams are required to obtain a good approximation to the exact values of the amplitudes of those beams.

Finally, it should be mentioned that while the multislice method may be considered as simply a method for integrating a system of differential equations it is, as pointed out by Goodman & Moodie (1974), a very efficient means for calculating wave functions.

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