Short Communications

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An alternative method for the calculation of a phase-grating function for use in dynamic electron-diffraction calculations.

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A method is described which permits multislice computation of n-beam diffraction intensities directly from the structure amplitudes. This is achieved by successive convolution, a numerical technique compatible with the multislice procedure.

The multislice method for the calculation of the scattered wave function, $\psi_{hk}(H)$, from a crystal of thickness H, is

$$\psi_{hk}(H) = \ldots[Q(\Delta z) * [Q(\Delta z) * [Q(\Delta z)]P(\Delta z)]P(\Delta z)] \quad \ldots \text{ to } n \text{ convolutions,} \quad (1)$$

where $H = n \,.\, \Delta z$, $P(\Delta z)$ is the propagation function, $\exp(2\pi i \zeta_{hk} \Delta z)$, ζ_{hk} being the excitation error of the hk reflexion, and $Q(\Delta z) = \mathscr{F}_{hk}\{\exp[i\varphi_p(x,y)\sigma\Delta z]\}$ is the phase-grating function for a slice Δz thick. Here the symbol $*$ has been used to denote convolution, and $\mathscr{F}_{hk}$ used to denote Fourier transformation. $\varphi_p(x,y)$ is the projected potential in the slice of thickness Δz and σ is the interaction parameter, $\sigma = \pi/\lambda W \,.\, 2/[1+(1-\beta^2)^{1/2}]$, λ the relativistic wavelength, W the accelerating potential, and $\beta = v/c$, v being the electron velocity and c the velocity of light (Cowley & Moodie, 1957; Goodman & Moodie, 1974; Lynch, 1971).

In the numerical calculation of equation (1) the usual practice has been to evaluate $Q(\Delta z)$ by summation of the structure amplitudes, V_{hk0}, to obtain $\varphi_p(x,y)$, calculation of $\cos[\varphi_p(x,y)\sigma\Delta z] + i \sin[\varphi_p(x,y)\sigma\Delta z]$ at each sampling point, and then back-Fourier transformation. It is the purpose of this communication to demonstrate a method which evaluates $Q(\Delta z)$ directly from the V_{hk0}.

The expression

$$\exp[i\varphi_p(x,y)\sigma\Delta z] = \lim_{s\to\infty}\left[1 + i\frac{\sigma\Delta z}{s}\varphi_p(x,y)\right]^s \quad (2)$$

may be calculated for finite s when the error to second order becomes $1/2!\ 1/s\,[\sigma\Delta z\varphi_p(x,y)]^2$. Typically $\varphi_p(x,y) \sim 10^2$ V, $\Delta z \sim 1$ Å, $\sigma \sim 10^{-3}$ so that for $s \sim 10^3$ the error is of the order of 10^{-5}, which is acceptable.

It is convenient to work with the Fourier transform of equation (2), *viz.*

$$Q_{hk}(\Delta z) = \left[\delta(h,k) + i\sigma V_{hk}\frac{\Delta z}{s}\right] * \left[\delta(h,k) + i\sigma V_{hk}\frac{\Delta z}{s}\right] * \quad \ldots \text{ to } s \text{ convolutions.} \quad (3)$$

The calculation of this expression is reduced considerably by a device equivalent to that used by Sturkey (1962) for the matrix method whereby if $s = 2^r$, then

$$Q_{hk}\left[\frac{\Delta z}{(2^r)}\right] = \delta(h,k) + i\sigma V_{hk}\frac{\Delta z}{s} = Q_1$$

and

$$Q_{hk}\left[\frac{2\,.\,\Delta z}{(2)^r}\right] = Q_1 * Q_1 = Q_2$$

and

$$Q_{hk}\left[\frac{2(2\,.\,\Delta z)}{(2)^r}\right] = Q_2 * Q_2 = Q_3 \ldots \quad (4)$$

and only r convolutions are required.

The equations (4) have been used as a numerical procedure to evaluate the phase-grating function. Table 1 shows the scattered amplitudes of the first seven reflexions calculated by the two procedures. The calculation was made for $W_4Nb_{26}O_{77}$ (Anstis, Lynch, Moodie & O'Keefe, 1973), systematics case, 00l reflexions, $\Delta z = 5$ Å. The only significant differences in the calculated values are in the 5th figure, and further, even out to the 30th order, the differences remain of the same order.

Table 1. *Scattered amplitudes calculated by both methods for a* 5 Å *slice of* $W_4Nb_{26}O_{77}$

	Usual method		Convolution method	
l	Real	Imaginary	Real	Imaginary
0	0·997786	0·063879	0·997787	0·063870
1	−0·000068	0·000794	−0·000068	0·000804
2	0·000266	−0·003017	0·000266	−0·003022
3	0·000238	−0·002835	0·000237	−0·002825
4	−0·000221	0·002726	−0·000220	0·002710
5	−0·000343	0·004239	−0·000344	0·004255
6	0·000012	0·000074	0·000013	0·000061
7	0·000491	−0·006289	0·000491	−0·006288

The phase grating calculated by this convolution method has been used in a 29-beam multislice procedure to obtain scattered amplitudes as a function of thickness. Again, results using the new procedure compared with the results using the conventionally calculated phase grating are identical to one part in 10^5 up to crystal thicknesses of 1000 Å. Thus the accumulation of error is still within acceptable limits.

The choice of s and hence of r, is controlled by calculation time limitations and by the number of significant bits available for floating-point calculation. The latter consideration arises from the relative magnitudes of the variables. For example, a weak V_{hk0} is of the order of 10^{-1} V, σ is of the order of 10^{-3} and $s = 10^3$ gives terms of the order of 10^{-6}. Thus the convolution procedure results in terms of the order of 1 (the central beam) added to terms of the order of 10^{-12}. Hence, this sets a limit of 10^3 for s for a computer with a numerical precision of 12 decimal places.

There are several criteria that may be used to evaluate the relative merits of the two forms of calculation. First there is the question of calculation time. A fast-Fourier transform will only require $n \cdot \log_2 n$ operations for n reflexions. However, there is still the necessity to evaluate $\sin[\sigma\varphi(x,y)\Delta z]$ and $\cos[\sigma\varphi(x,y)\Delta z]$ at n sampling points. Thus, although there are required $r \cdot n^2$ operations for the convolution method, the calculation times remain remarkably close together for a range of values of n (29 to 435). Thus there is no particular advantage in either method on this point.

Second, there are the questions of convenience and amount of memory storage required. In this case, the convolution method is far better as it requires far less storage and the procedure is identical with that used in the kernel of the multislice method, and thus a considerable economy in computer code is achieved. In terms of non-specialist investigators requiring their own routines for calculation, the convolution method is felt to be simpler to set up in the computer.

Finally, there are questions of physical insight gained from intermediate steps in the calculation. In this respect, it is felt that the potential distribution that is calculated in the course of the usual method is a great help, particularly in symmetry considerations and hence the convolution method is at a disadvantage in this case.

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A comment on Tiwari, Prasad & Srivastava's paper *The arcing of X-ray diffracion spots in X-ray photographs of cadmium iodide crystals.*

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It is shown that the arcing in X-ray photographs of CdI_2 crystals cannot be attributed to paracrystalline distortions in crystals.

In the paper quoted (Tiwari, Prasad & Srivastava, 1973) the authors attempted to explain the arcing on *a*-axis oscillation photographs of CdI_2 crystals in terms of paracrystalline distortions. Earlier Prasad & Srivastava (1971) had explained the arcing observed on X-ray *c*-axis oscillation photographs as crystal shape and size effects; this has already been shown to be wrong (Agrawal, 1973). The latter explained it in terms of tilt boundaries consisting of unit and partial edge dislocations created during crystal growth, as Agrawal & Trigunayat (1969*a*,*b*; 1970) and Agrawal, Chadha & Trigunayat (1970) had already suggested in the cases of arcing in X-ray *a*-axis oscillation and Laue photographs of CdI_2, $CdBr_2$ and PbI_2 crystals. Agrawal (1970, 1971) had also established the correlation between the phenomena of arcing and polytypism on the basis of experimental results. However, it is not necessary that the compounds displaying polytypism should also exhibit arcing, whereas the opposite may be true.

The diffraction patterns observed in CdI_2 crystals are quite different from those observed in chain molecules or fibrous crystals. In general, the patterns for natural fibres do not yield the reciprocal lattice of the crystal because, instead of a single crystal, one has a 'two-dimensional powder' resulting from the grouping of crystallites of random orientation along the axis of the fibre. The pattern therefore gives the figure of revolution obtained by rotating the reciprocal lattice for a single crystal around the fibre axis (Guinier, 1963). The crystals of cadmium iodide exhibit arcs of various shapes, *e.g.* fork, cross, square, semi-elliptical, *etc.*, each consisting of two or more spots, corresponding to each reciprocal lattice point (Agrawal & Trigunayat, 1969*a*), whereas in chain molecules or fibrous crystals each reciprocal lattice point gives rise to its own elongated and broadened spot. The intensity and broadening of the spots are a maximum near or at the centre and decrease towards their ends. The CdI_2 crystals also exhibit closed rings on Laue photographs each corresponding to a reciprocal lattice point, whereas in the latter case the spots are bridged by intensity ridges forming a ring because various reciprocal-lattice points or nodes at equal distances from the centre of the net are connected to form a ring due to paracrystalline distortions (Vainshtein, 1966). Besides, in the latter case the geometry of the diffraction pattern would not change if the crystal is irradiated either wholly or partially by the X-ray beam because the lattice cells which are different from one another are randomly distributed, whereas in the crystal exhibiting arcing or rings on the photographs, the diffraction pattern does change (Agrawal & Trigunayat, 1969*a*, Figs. 4 and 5; 1969*b*, Figs. 12 and 13).

In the paper quoted the paracrystallinity in CdI_2 crystals had been introduced during growth in two possible ways. Firstly, the fluctuations in axial parameters due to water molecules adsorbed on the surface of layers forming the crystal during growth, which itself is unrealistic, would give rise to (i) streaking joining the reflexions on the layer lines due to the *c*-axis fluctuations and (ii) extra spots occurring

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