Acta Cryst. (1977). A33, 323–326

Elimination of the Finite-Range Effect on the Block-Size Distribution from the Fourier Transform

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(*Received* 16 *June* 1976; *accepted* 25 *October* 1976)

A diffraction profile can be recorded for only a finite range around the peak. This leads to spurious oscillations in the block-size distribution determined by means of the Fourier transform. The Bertaut [*Acta Cryst.* (1952), **5**, 117–121] correction gives an approximate distribution $g_1(m)$ related to the real one $g(m)$ by a convolution product with a function $d(m)$, of the type $(\sin x)/x$. A modified variant of the successive-convolution unfolding method is proposed by considering the convolution product only over a finite size range where the distribution could be non-zero. The method was tested for two hypothetical distributions. A procedure to set the limits of the size interval is suggested.

1. Introduction

It is well known that for a line broadened only by the size effect the distributions of diameters perpendicular to the plane of reflexion of the crystalline blocks scattering coherently, could, in principle, be determined. Let $I(X)$ be the line profile where $X=2d_{hkl}\cos(\theta-\theta_0)/\lambda$ with d_{hkl} the corresponding interplanar distance, λ the wavelength and $\theta-\theta_0$ the departure from the Bragg angle, θ_0, and $i(m)$ its Fourier transform. As shown by Bertaut (1950), the distribution in diameters (as defined above) is given by

$$g(M)\sim\left(\frac{\mathrm{d}^2 i}{\mathrm{d}m^2}\right)_{m=M} \tag{1}$$

or

$$g(M)\sim\int_{-\infty}^{+\infty}X^2 I(X)e^{-2\pi imX}\mathrm{d}X. \tag{2}$$

But, owing to both the presence of the other peaks and oscillations in the tails, where the experimental errors are the largest, the profile can be determined only over a finite range and the extrapolation [*e.g.* using a Cauchy function (de Bergevin & Germi, 1972)] is often a difficult matter. Therefore one has to introduce a 'cut-off', analytically equivalent to multiplying the profile, after subtracting the background, by a passing step function defined as

$$D(X)=\begin{cases}1 & X\in(-X_1,X_1)\\ 0 & \text{otherwise}\end{cases} \tag{3}$$

where $-X_1$ and $+X_1$ are the cut-off points, which in fact means using the function $[I(X)-I(X_1)]D(X)$ instead of $I(X)$. As Bertaut (1952) soon realized and Young, Gerdes & Wilson (1967) emphasized later, this cut-off causes a severe alteration of the distribution.

Bertaut has shown indeed that the second derivative of the 'observed'-profile transform is given by

$$i_1''(m)=-d(m)+\int g(M)d(m-M)\mathrm{d}M \tag{4}$$

where the subscript 1 stands for the entity affected by a measurement over a finite range only and $d(m)$ is the Fourier transform of D

$$d(m)=\frac{\sin(2\pi X_1 m)}{\pi m}. \tag{5}$$

The resulting 'observed' distribution,

$$g_1(m)=\int g(m)d(m-M)\mathrm{d}M, \tag{6}$$

is considered close to the real one, g, if the profile is measured over a large enough range, 'out to a distance equal to five or six times its width'. When this condition cannot be fulfilled, the size distribution determination is of doubtful value.

This paper reports on the possibility of improving the method at this stage by determining $g(m)$ from (6) by an appropriate method of deconvolution.

2. The method of deconvolution

The equation to be solved for $g(m)$ is the first-kind Fredholm integral equation

$$\int_R g(M)\left[\frac{\sin 2\pi(M+m)X_1}{\pi(M+m)}+\frac{\sin 2\pi(M-m)X_1}{\pi(M-m)}\right]\mathrm{d}M=g_1(m). \tag{7}$$

$g_1(m)$ is subjected to errors, and so the problem is [in Hadamard's terms (see, for example, Lavrentiev, 1967)] improperly posed, but since the weighting function, $d(m)$, is precisely known, we looked for a simpler method avoiding the difficulties of the regularization or statistical estimate methods (see, for example, Shaw, 1972). The successive-convolution (van Cittert) method (Ergun, 1968) has relatively little tendency to amplify the fluctuations, but it should apparently fail since it is readily shown that $d(m)*d(m)=d(m)$ ($*$ means convolution) and the process defined by

$$g_n=\left[\sum_{j=1}^{n}(-1)^{j-1}\binom{n}{j}d^{*(j-1)}\right]*g_1 \tag{8}$$

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where $*(j-1)$ in the exponent stands for $(j-1)$ times convolution, reduces to

$$g_n = ng_1 - (n-1)g_1 * d. \tag{9}$$

This can be noted too on Fig. 1 giving the results of the computer experiment (see below).

As Warren & Mozzi (1975) pointed out, the deconvolution by the Fourier transform or by the methods of moments is also impossible when one deals with the function $(\sin x)/x$.

On the other hand, as indicated by Jones & Misell (1970) or by Ruland (1971), the successive-convolution method has to converge when $0<D<2$ for all values of D. Since this condition stems from $\lim_{n\to\infty}(1-D)^n=0$, the convergence is fast if D is close to unity over the range where G_1 (transform of g_1) is not zero, *i.e.*

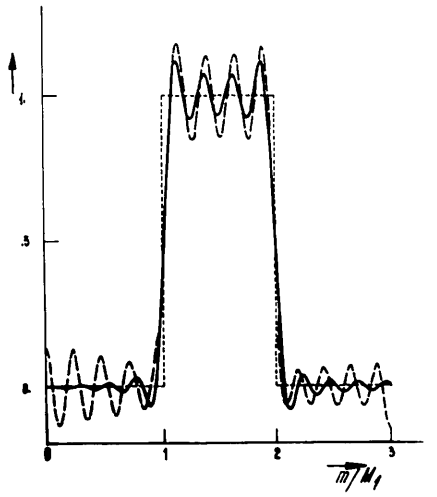


Fig. 1. 'Infinite-range' successive-folding method of deconvolution applied to the rectangular distribution for the case $X_1=4/M_1$. Hypothetical (dotted line), 'experimental' (solid line) and convolution-corrected (dashed line) distributions.

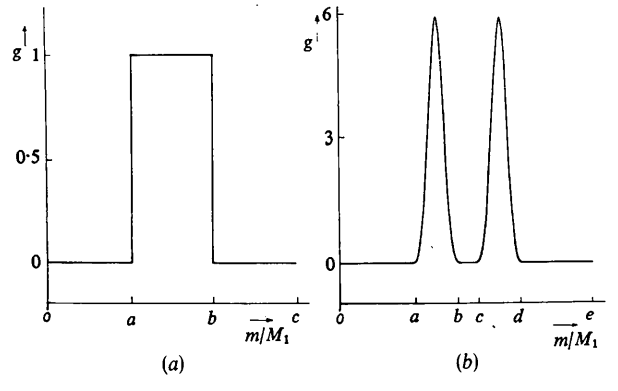


Fig. 2. Hypothetical distributions: (*a*) rectangular: $a=1$, $b=2a$, $c=3a$; (*b*) normal with two maxima: $a=0{\cdot}7$, $b=1{\cdot}1$, $c=1{\cdot}3$, $d=1{\cdot}7$, $e=2{\cdot}4$.

within $(-X_1, +X_1)$. Outside this range $D=0$ and the process is no longer convergent.

Considering (3), we see that the iterative procedure is unconditionally convergent and should not be stopped unless the condition imposed, *e.g.*

$$\int |g_1 - g_n * d| \mathrm{d}m \le \varepsilon \int |g_1| \mathrm{d}m \tag{10}$$

where ε is an arbitrary small constant, is fulfilled.

We note at first that for any selected (sieved) powder the range of the diameters has a lower and an upper limit, say α and β respectively.

Then, a contribution from outside this range (α, β), to the fold has no physical meaning. Let us introduce now a 'finite range' successive folding by defining the convolution product as

$$d * g_1 \equiv \int_\alpha^\beta d(m-M)g_1(M)\mathrm{d}M \tag{11}$$

and using it in (8) to compute the corrected function *within* the range (α, β) and then, by

$$g_n(m) = g_1(m) - \int_\alpha^\beta d(m-M)g_n(M)\mathrm{d}M \quad m \in R\backslash(\alpha,\beta) \tag{12}$$

the function outside that range [the sign \ denotes the subtraction of the (α, β) interval from the real manifold R]. Thus, after the iterative process has been completed for the values within (α, β), the function outside this range is computed by only one operation, (12).

3. Test of the method

A computer experiment has been performed to test the method by using two hypothetical distributions: a rectangular, $g_r(m)$ (Fig. 2*a*), and a normal with two maxima, $g_b(m)$ (Fig. 2*b*).

The rectangular distribution is just that used by Bertaut (1952) to illustrate the effect of the finite range

$$g_r(m) = \begin{cases} 1 & a \le m \le b \\ 0 & \text{elsewhere.} \end{cases}$$

The second distribution is defined as

$$g_b(m) = \begin{cases} A\{\exp -[\gamma(m-m_0)^2] - B\} & \\ \qquad m \in (a,b),\ m_0 = \dfrac{a+b}{2} & \\ \qquad \text{and } m \in (c,d),\ m_0 = \dfrac{c+d}{2} & \\ 0 \qquad \text{elsewhere.} & \end{cases}$$

The constants are chosen so that the function is continuous in a,b,c,d. (Actually, $\gamma = 4{\cdot}15883 \times 10^{-2}$, $B = 0{\cdot}015625$.)

First of all, the values of $g_1(m)$ were computed from (7) in a series of equidistant points covering the whole range of interest by considering three values of X_1: $1/M_1$, $2/M_1$ and $4/M_1$, with $M_1 = \beta - \alpha$.

Then, the 'finite range' successive-convolution

method as defined by (11) and (12) was applied to obtain the 'corrected' distributions. The procedure was carried out until the fraction integral residue, as defined in (10), became lower than $\varepsilon = 0{\cdot}05\%$. The hypothetical, 'experimental' and convolution-corrected distributions are plotted in Figs. 3 and 4 for the rectangular and normal distributions, respectively.

In order to see how the assessment of α and β in (11) and (12) determines the reliability of the proposed method, the procedure has been applied to the rectangular distribution by considering three different (α, β) ranges: narrowed, enlarged and displaced. The narrowing and enlargement were by 25% and the displacement by 12·5% towards the low values, the width being unaltered in the last case. The results are plotted in Fig. 5.

4. Discussion

The failure of the 'infinite' successive-convolution method of unfolding is shown in Fig. 1. For the same case the 'finite' method results are plotted in Fig. 3(*c*).

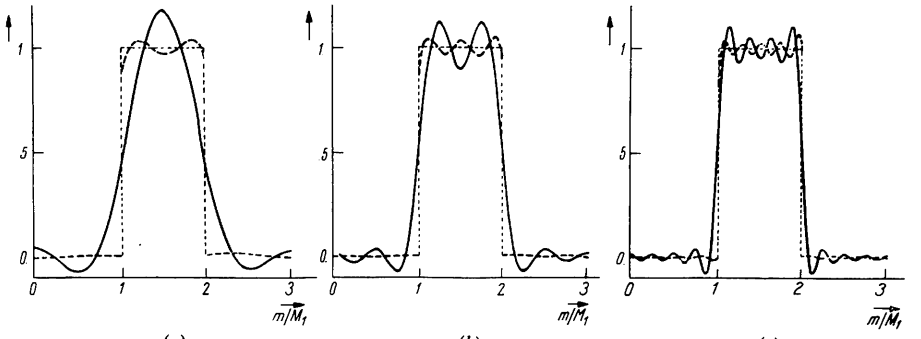
As a rule, for the rectangular case the distribution has been considered over a total range of $3M_1$. After the first iteration that produces a decrease of the fractional integral residue (10) from 1 to 0·8%, the next 27 iterations decreased it only to 0·5% and the spurious oscillations are higher than that in the 'experimental' curve.

For the 'exactly set' α and β, the process has a fast convergence, decreasing the residue to 0·048% [from 2·59; this figure is higher than that corresponding to Fig. 1 since the fractional integral residue is now defined as

$$\int_{\alpha}^{\beta} |g_n(m) * d - g_1(m)| \mathrm{d}m \Big/ \int_{\alpha}^{\beta} |g_1(m)| \mathrm{d}m]$$

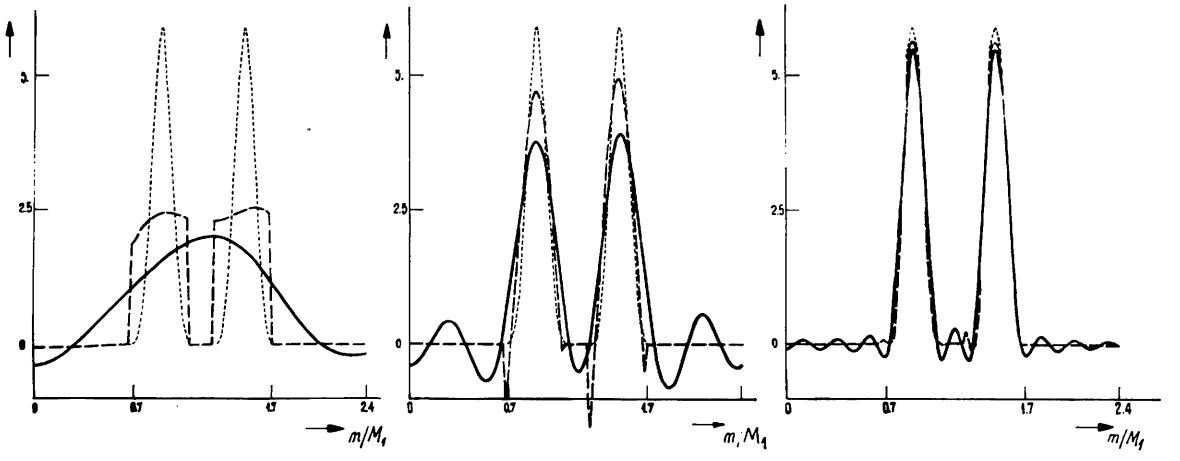
in 15 iterations and leading thus to a distribution extremely close to the hypothetical one (Fig. 3*c*).

In the case of a low-range width [Figs. 3(*a*) and 4(*a*)] which the method is intended to deal with, the unfolded curves are realistic representations of the dis-



(*a*) (*b*) (*c*)

Fig. 3. 'Finite-range' successive-folding method of deconvolution applied to the rectangular distribution for the cases: (*a*) $X_1 = 1/M_1$, (*b*) $X_1 = 2/M_1$ and (*c*) $X_1 = 4/M_1$. The curve notation is as in Fig. 1.



(*a*) (*b*) (*c*)

Fig. 4. 'Finite-range' deconvolution method applied to a normal distribution with two maxima. The cases are as in Fig. 3 and the curve notation as in Fig. 1.

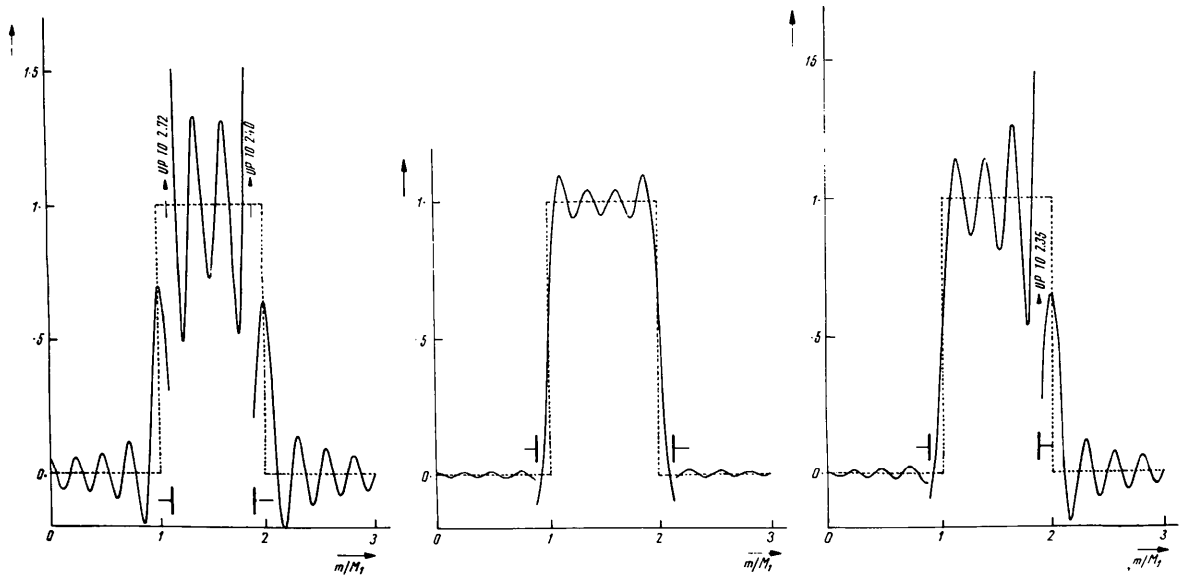


Fig. 5. 'Finite-range' successive-folding method of deconvolution applied to the rectangular distribution for $X_1 = 4/M_1$: (*a*) range narrowed by 25%, (*b*) range enlarged by 25% and (*c*) range displaced by 12·5% towards the low values. Hypothetical (dotted line) and convolution-corrected (solid line) distributions. (Range shown by ⊣ ⊢).

tributions, giving a correct area and an approximate width, and qualitatively, the shape. By comparison of the 'realistic' curves in Fig. 4, one can see, however, that a recording over a range as large as two or three line widths is required in order to determine accurately the block-size distribution.

The initial residue is, as expected, decreasing with the range of intensity recording but the rate of convergence is decreasing. This is a feature of the method, the iterative process leading asymptotically to the real function (disregarding error accumulation).

It is to be noted by considering Fig. 5 that if one of the ends assumed to be limiting the range of non-zero values of the distribution is inside the correct range, high spurious oscillations have been introduced. If the assessed range is too broad, except for the behaviour at the ends (Fig. 5*b*) the shape is almost correct.

These would suggest an iterative method for the correct assessment of α and β when applying the 'finite range' unfolding. A low multiple of d_{hkl} and the size of a (polycrystalline) grain could be taken respectively as initial values of α and β. Noting that the first few iterations are the most 'efficient', the 'finite-range' unfolding has to be carried out with, say, three or five steps by narrowing each time the (α, β) range until the measure of oscillation magnitude outside the distribution range becomes too high (this would show the range is too narrow and the last assessed α and β are to be considered correct). For instance, one could try to minimize

$$\int_{R_+ \setminus (\alpha, \beta)} |g(m)| \mathrm{d}m \quad [R_+ = (0, \infty)]$$

and stop the rangenarrowing when this function begins to increase.

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