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A Fast-Converging Refinement of One-Dimensional Convolution Square Roots

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A fast-converging mathematical method for calculation of convolution square roots of one-dimensional functions has been developed. Application to one-dimensional periodic structure projections as well as to non-periodic structures is possible. Computer calculations of some examples are given.

Introduction

The well known phase problem of diffraction theory to be solved usually in reciprocal space has its equivalent in computing the convolution square root (CSR) of the Patterson function in real space.

From a known structure the Patterson function or the distance statistic can be calculated through the convolution square (CS). This calculation works straightforwardly; the inversion CSR has been realized only in special cases (Hosemann & Bagchi, 1962), although it would be desirable for single-crystal structure analysis as well as for the analysis of non-periodic structures to have such a calculation method.

The calculation of the three-dimensional CSR is possible in principle, if sufficient computer time and storage are available, but has not been carried out yet.

If one tries to compute the one-dimensional CSR of Patterson projections a unique solution cannot be expected. The number of solutions depends on the number of axis reflections used. Independent of the sign or phase selection one obtains the same Patterson function. In reality the CSR can only be carried out for those projections of which a significant approximation is known. It can be shown (Hosemann & Bagchi, 1962) that a unique solution for the CSR of pseudo-periodic or non-periodic structures requires a fast diminution of the electron density to zero with increasing **x** values.

Mathematical description of the method

The CS of a structure $\rho(\mathbf{x})$ is defined by

$$\mathrm{CS}(\mathbf{x}) = \sum_{-\infty}^{+\infty} \rho(\mathbf{y})\,\rho(\mathbf{y}-\mathbf{x})\,\mathrm{d}V_y; \qquad (1)$$

x, **y** are vectors and $\mathrm{d}V_y$ the volume element in real space. If the structure is assumed to be given one-dimensionally, point by point, for example in the storage of a computer, (1) can be represented by the following expression:

$$\mathrm{CS}_j = \sum_{i=1}^{n} \rho_i \rho_{i+j-1} \quad j = 1, \dots m. \qquad (2)$$

This expression differs for periodic and non-periodic structures. In the periodic case the Patterson function has the same period as the structure itself. Therefore, only points of one unit cell have to be taken ($m = n$). For finite non-period structures we have $m = 2n$. Because of the CS centrosymmetry only half of the CS points are required. The known methods (Hosemann & Bagchi, 1962) start at one end of the distribution function CS in order to solve (2) for the ρ_i; the accuracy of those methods depends significantly on that of the starting points $\mathrm{CS}_1, \mathrm{CS}_2, \dots$ The method to be described here works totally differently.

Assuming a first approximation of the ρ_i is known, the equation system (2) may be written:

$$\mathrm{CS}_j = \sum_{i=1}^{n} \rho_i \rho^*_{i+j-1} \quad j = 1, \dots m\ (=n), \qquad (3)$$

where ρ_i denote the unknown structure and ρ^*_{i+j-1} the first approximation. The CS_j are the Patterson points of the experiment. Regarding (3) as a system of linear equations of order n for the ρ_i, the inversion of the matrix (ρ^*_i) leads to a unique solution in general, which can be called the second approximation. The input of the second approximation into (3) would not be very reasonable since one further step would result in the previous values ρ_i.

Taking the average

$$\rho^{**}_i = \frac{\rho_i + \rho^*_i}{2} \qquad (4)$$

as the second approximation, repetition of the procedure described above leads to a very-fast-converging refinement, which is almost independent of the first approximation if there is a unique solution of the problem. Otherwise, if different solutions are possible, the first approximation has to be close to the required solution.

Examples

The method described to calculate the CSR of a one-dimensional function has been tested against several examples of which the results of three calculations will be described here. In all three cases the x_1 projection of a 4-hydroxyindole derivative (Hecht & Luger, 1974;

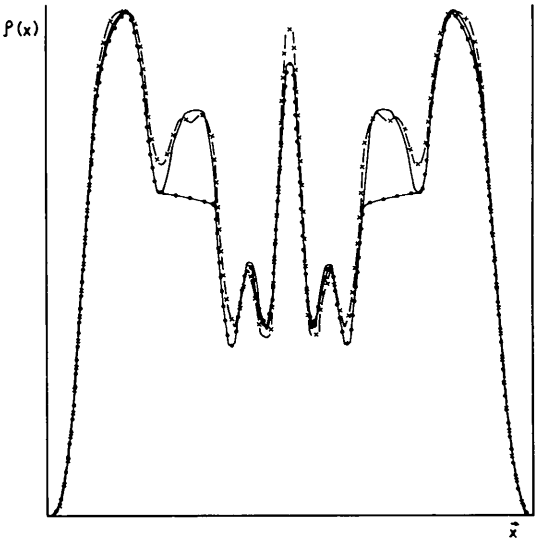

Fig. 1. Graphical representation of example (*a*). First approximation obtained by neglect of a medium-sized peak. —— Experimental density, ×—×—× refined approximation, •—•—• first approximation.

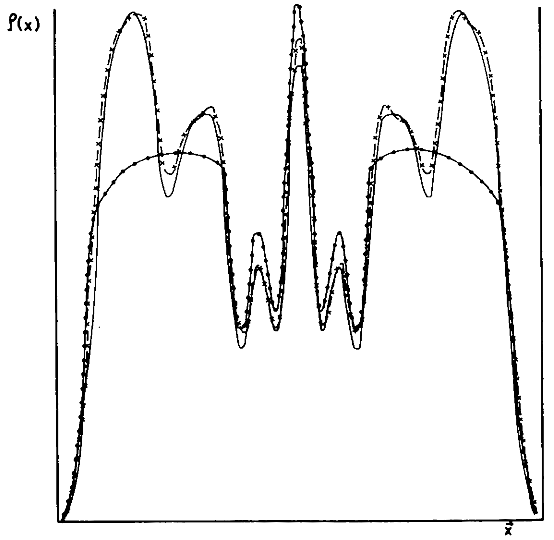

Fig. 2. Graphical representation of example (*b*). First approximation obtained by cutting off one major and one medium-sized peak. Curves as in Fig. 1.

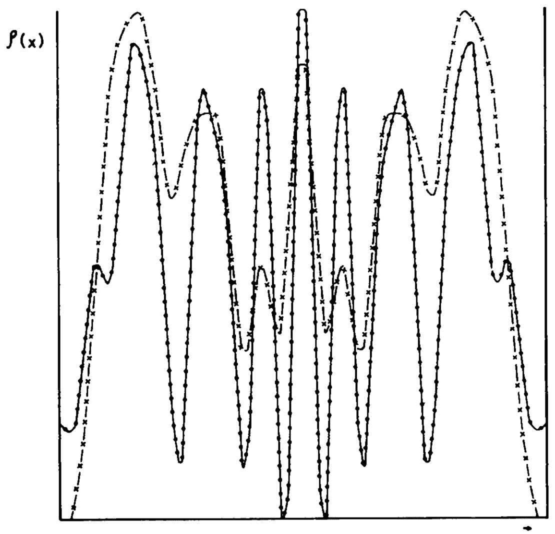

Fig. 3. Graphical representation of example (*c*). First approximation $\rho_n(x)$ obtained from 'unit' reflections. Refined and experimental distribution are identical within the graphical error. ×—×—× Refined approximation and experimental density, •—•—• first approximation.

monoclinic, space group $P2_1/c$) and its Fourier transform, the $F(h_100)$ reflections, have been used as a test object. The different examples were obtained by a more or less relevant modification of the known density projection $\rho(x_1)$ to be used as a first approximation $\rho^*(x_1)$.

(*a*) $\rho^*(x_1)$ was obtained by neglecting one of the medium-sized peaks of $\rho(x_1)$ (Fig. 1). By a four-cycle refinement a good fit to the original $\rho(x_1)$ distribution was obtained. However, it should be noted that the minor discrepancies between refinement and experimental density could not be improved by additional refinement cycles.

(*b*) $\rho^*(x_1)$ was obtained by cutting off one of the major maxima and the medium maximum of case (*a*) (Fig. 2). The result of a four-cycle refinement is in principle equal to that of (*a*).

(*c*) From the real $F(h_100)$ reflection series, 'unit' reflections $F_n(h_100)$ were obtained by taking the average of the $F(h_100)$ series for the magnitude of all F_n's:

$$|F_n(h_100)| = c = \frac{1}{n} \sum_{h_1} |F(h_100)|,$$

where n denotes the number of reflections in the (h_100) series. The sign of $F_n(h_100)$ was set equal to the sign of the corresponding $F(h_100)$ reflection, so that $F_n(h_100)$ is finally defined by

$$F_n(h_100) = c \text{ sign } [F(h_100)].$$

The density projection $\rho_n(x_1)$, calculated as the Fourier transform from the $F_n(h_100)$ series, was then used as the first approximation of $\rho(x_1)$ (Fig. 3). As expected, $\rho_n(x_1)$ differs totally from $\rho(x_1)$. A four-cycle CSR refinement results in a distribution which is, within the graphical error, indistinguishable from the experimental density $\rho(x)$.

In all the test examples, the CS of the refined densities fitted exactly the experimental Patterson function.

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