JOURNAL OF COMPUTATIONAL PHYSICS 3, 444–446 (1969)

NOTE

A Method of Alphabetic Plotting of Undistorted Fourier Maps in X-Ray Crystallography*[1]

One of the primary calculations in x-ray crystal structure analysis involves three-dimensional Fourier series where the summation represents the electron density distribution, $\rho(\bar{r})$, throughout the unit repeat cell of the crystal. This may be represented as:

$$\rho(\bar{r}) = \frac{1}{V} \sum_{H} F(\bar{H})\, e^{-2\pi i \bar{H}\cdot\bar{r}} \tag{1}$$

where V is the volume of the unit cell, H is the triplet of numbers (Miller indices) which define the orientation of the diffraction vector, $\bar{H}$, and $F(\bar{H})$ is the structure factor of the appropriate set of Bragg planes. A similar Fourier series is used to compute the "Patterson Function," which is often used in the initial stages of a structure investigation. Techniques for computing these functions efficiently have been well described [1], [2].

Alphabetic plotting has often been used as a convenient method of handling the output of Fourier programs. A letter of the alphabet is assigned to each of a number of predefined numerical ranges of the function, and this letter is printed for each calculated point.

Most Fourier programs have been written to calculate the values of the function at points which are evenly spaced as referred to the coordinate system which describes the unit repeat cell of the crystal. In the majority of cases the axial lengths of this system are not equal, and frequently the system is not orthogonal. Each section through the unit cell of the crystal must be printed by a printer whose coordinate system is orthogonal, but whose axial lengths are usually unequal. As a result the printed map is distorted and difficult to interpret.

Bennett and Kendrew [3], in one of the earliest papers on the use of computers to calculate Fourier series, mention the problem and suggest mechanical modification of the teleprinter in order to obtain undistorted maps. Donnay and Takeda [4] have presented a more practical solution. It involves rewriting the Fourier program

* This research is supported by the National Institute of Dental Research under Grant PHS 5T1 DE 117 05.

[1] The authors would like to thank Dr. Kenneth Brownstein for helpful discussions suggesting the method of interpolation.

so that it calculates the values of the function at the points where the printer can print them. The present authors have developed a simpler solution which makes it possible to use existing Fourier programs. It consists of an alphabetic plotting program which uses a special method of four point interpolation to expand or contract the horizontal scale. Provision is also made for horizontal offset of each line to allow for nonorthogonal sections.

The interpolation is accomplished by using a third-order spline function [5]. In this special case the function, $\phi(x)$, is fitted to four points and is defined as follows:

(i) ϕ is a cubic in each of the three intervals $(x_1, x_2) = (x_a, x_b), (x_b, x_c), (x_c, x_d)$.

(ii) $\phi(x_i) = \rho_i$, $i = a, b, c, d$.

(iii) ϕ' and ϕ'' are continuous at x_b and x_c.

(iv) $\phi'' = 0$ at x_a and x_d.

This definition provides the twelve conditions necessary to calculate the twelve coefficients of the three cubic equations. Calculation of these coefficients is simplified if the cubic equations are written:

$$\rho = \phi(x) = (A_j x + B_j)(x - x_{1j})(x - x_{2j}) + C_j x + D_j, \quad j = 1, 2, 3 \tag{2}$$

Because existing Fourier programs calculate the function on a fixed grid, it is possible to scale the coordinates so that the x's in the above equation are all integers. Further, it is possible to subtract a constant from all the x's so that the first point has an abscissa of zero. This simplifies the calculation of the coefficients as follows:

$$\begin{aligned}
C_1 &= \rho_b - \rho_a, & D_1 &= \rho_a, \\
C_2 &= \rho_c - \rho_b, & D_2 &= 2\rho_b - \rho_c, \\
C_3 &= \rho_d - \rho_c, & D_3 &= 3\rho_c - 2\rho_d, \\
A_2 &= (C_1 - 2C_2 + C_3)/3, & & \\
B_2 &= (5C_2 - 4C_1 - C_3)/5, & & \\
A_1 &= B_2/3, & B_1 &= B_2/3, \\
A_3 &= -(A_1 + A_2), & B_3 &= 4A_2 + B_1 + B_2.
\end{aligned} \tag{3}$$

The central interval is used wherever possible. The outside intervals are used at the extreme ends of the line. A better fit at the ends of the line can be obtained by letting the Fourier program calculate an extra point at each end of the line and using central intervals only.

This method of interpolation is suited to a rapidly changing function. A simple cubic fitted to four points would give values widely divergent from those of the input points.

In practice, sections are plotted which include the "a" and "b" axes of the crystal, and the scale is chosen so that the line spacing of the printer corresponds to the

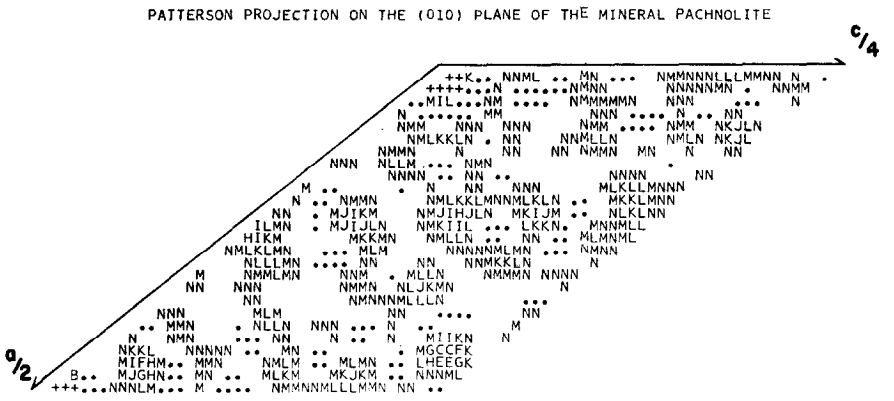

FIG. 1. Example of the output of the plotting program. Positive values of the function range from "N", the least positive, to "A", the most positive. Positive overflows are printed as + signs, zero values are suppressed, and negative values are printed as periods.

calculated interval along the "a" axis. The "y" coordinate of each print position along each line in the coordinate system of the crystal is then calculated, and the interpolation routine is used to find the value of ρ at each of these points. If the unit cell of the crystal is not orthogonal, each line of output will be offset from the one above it. Figure 1 is an example of the output of the program used in this laboratory.

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