Application of Fast Fourier Transform Methods to Crystal Structure Analysis

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Abstract

A new type of Fourier transform in crystal structure analysis was introduced. It enables the calculation speed more radidly than that of ordinary Fourier transform. But it is not so effective for some cases. Asymmetric units suitable for the transformation are discussed.

Introduction

Fast Fourier transform (FFT) was introduced by Cooley and Tukey (1965), and was described by many authors. Their explanatory descriptions were focussed on the FFT written by Cooley and Tukey's algorithm. FFT of Cooley and Tukey's algorithm seems to be disadvantageous to apply for the crystal structure analysis due to the following reasons, (1) Fourier transform of structure factors can be limited within grid points in asymmetric units, on the other hand, the results by FFT method should be obtained after transforming for all grid points, and bit-reversed process is necessary in the final stage, (2) the number of the grid points in the unit cells are more than that of the reciprocal lattice points. To overcome these disadvantages, the present authors formulated a new kind of finite discrete Fourier transform adapting the principle of FFT.

Asymmetric units for Fourier transform of structure factors were discussed by Ten Eyck (1973). His method is ambiguous and the some results are not correct. The asymmetric units for Fourier transform should agree with the asymmetric units for space group symmetry in the unit cells which are listed in International Tables for Crystallography (1983). The only thing we should do is to design shapes of asymmetric units in order to

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make the subjects of grid points to be suitable for Fourier transform.

Fourier transform of electron density was discussed by Brünger (1989) for saving memory.

1. Finite discrete Fourier transform

The finite discrete Fourier transform $X(x_i)$ of a function of $\Phi(h)$ ($h=0, 1, ..., M-1$) is expressed by

$$X(x_i) = \sum_{h=0} \Phi(h) \exp(-2\pi ihx_i), \qquad (1.1)$$

where $x_i = i/N$ and $0 < i < N$. The number M and N are assumed to be powers of $M = 2^m$ and $N = 2^n$, respectively, for succeeding description. Equation (1.1) can be regarded as a linear transformation of the column vector Φ of which $(h+1)$st element is $\Phi(h)$ to the column vector Φ of which $(i+1)$st element is $X(x_i)$. The $(i+1)(h+1)$ element of the transformation matrix P is $\exp(-2\pi ihx_i)$. When $hi \equiv s \pmod{2^n}$, a matrix I of which $(i+1)(h+1)$ element is s, is introduced, the element s and t of the $(h+1)$st and $(2^{m-1}+h+1)$st columns of the $(i+1)$st row in I are

$$s = ih - 2^n u, \qquad (1.2)$$
$$t = i(2^{m-1}+h) - 2^n v, \qquad (1.3)$$

where u and v are integers. If $s = t$, then

$$2^{m-1}i = 2^n(u-v). \qquad (1.4)$$

Hence, if $i = 2^{n-m+1}$, equation (1.4) is always valid. The value of i satisfying equation (1.4) is denoted by p, $p = 2^{n-m+1}$.

When $k = i + qp$, the $(i+1)(j+1)$ element of I is a, where $i < p$ and q is an integer, and the $(p+1)(h+1)$ element is s, then the $(k+1)(h+1)$ element becomes $a+qs$. Similarly, when the $(i+1)(2^{m-1}+h+1)$ element of I is b, then the $(k+1)(2^{m-1}+h+1)$ element of I becomes $b+qs$. Hence, when $i = qp+i'$, $1 \leq i' < p$, the ij and $i(j+2^{m-1})$ elements in the ith row of P are those of the $i'j$ and $i'(j+2^{m-1})$ multiplied by $\exp(2\pi iqpj/2^n) = \exp(2\pi iqj/2^{m-1})$.

When the matrices $P^1, P^2, ..., P^{M/2}$, $M = 2^m$, are introduced, where P^j is $px2$ rectangular matrix and the ij element is $\exp(2\pi i(i-1)(j-1)/2^n)$, and the $i(j+M/2)$ element is $\exp(2\pi i(i-1)(j+M/2-1)/2^n)$, the results of the product of P^j and Φ are denoted by F^0_j. When the unit matrix with dimension $2^{i-1}p$ ($i \geq 1$) is denoted by E_i, matrices $W(i)_j$ are introduced and expressed by

$$W(i)_j = \begin{vmatrix} E_i & E_i \\ w_j E_i & -w_j E_i \end{vmatrix}, \qquad (1.5)$$

where $w_j = \exp(2\pi i(j-1)/2^{m-i})$ ($j=1, ..., 2^{m-i-1}$). When vectors A_j consisting of two subvectors, where the first subvector is $F^{i-1}j$ and the second subvector is F^{i-1}_{j+s} ($s = 2^{m-i-1}$), and F^i_j is defined by

$$F^i_j = W(i)_j A_j, \qquad (1.6)$$

then, the subvector of X of which elements are $X(x_0), X(x_1), ..., X(x_{p-1})$ ($p = 2^{n-m+i}$) are

obtained by the sum of the vectors $F^{i-1}{}_j, j=1, 2, .., 2^{m-i}$, and the subvector of X of which elements are $X(x_p), X(x_{p+1}), .., X(x_{2p-1})$ is obtained by the sum of $w_j(F^{i-1}{}_j - F^{i-1}{}_{j+s}), j=1, 2,$ $.., s=2^{m-i-1}$. When $i=m-1$, the dimension of $F^{m-2}{}_j$ is $2^{m-2} \cdot 2^{n-m+1} = 2^{n-1}, 2^{m-i-1} = 1$ and $w_1 = 1$, hence, $F^{m-1}{}_1$ becomes equal to X, where X is as follows,

$$X = F^{m-1}{}_1 = \begin{vmatrix} E_i & E_i \\ E_i & -E_i \end{vmatrix} \begin{vmatrix} F^{m-2}{}_1 \\ F^{m-2}{}_2 \end{vmatrix}, \quad (1.7)$$

For this calculation, a two-dimensional array with length $N=2^n$, for example, $F(2, N)$ is prepared and the jth element of $F^0{}_i$ is stored at $F(1, j+p(i-1))$. Then, the elements of $F^s{}_t$ are calculated by the following ways:

    1. $ia = (t-1)p$

        $ia = ia - N[ia/N] + 1$

    2. $ja = j + (i-1)p$

        $jj = j + 2(i-1)p$

        $F(isa, jj) = F(isb, ja) + F(isb, ja+NH)$

        $F(isa, jj+p) = (F(isb, ja) - F(isb, ja+NH)) w(ia)$

where $NH = N/2; p = 2^{n-m+s}$, and $w(ia) = \exp(ia 2\pi i/N)$. The number isa and isb are set to 2 and 1 at first, and change alternately along the change of the value of s. The final output is vector X.

When the number of the grid points, NG, is smaller than N, the value $IS = [NG/p]$ is calculated for each s where $p = 2^{n-m+i}$. If the IS becomes smaller than 2, $X(x_i)$ are calculated. The flow chart of this Fourier transform is given in Fig.1.

Although the dimensions of the vectors are not limited within the power of 2, it is desirable for M and N to be the multiples of powers of two.

When $M = 5 \cdot 2^m$ and $N = 5 \cdot 2^n$, the value of p is also 2^{n-m+1}. The vectors $F^i{}_j$ are obtained by quite the same ways as the case of $M = 2^m$ and $N = 2^n$ until $i = m-1$. The vectors $F^{m-1}{}_j$ are of dimension $2^{m-1}p$, where $j = 1, 2, .., 5$. Then, $W(m)_j$ $(j=1)$ matrix consists of submatrices with dimension $2^{m-1}p$ and the ij element is $w^{(j-1)(i-1)}E$, where $w^{(j-1)(i-1)} = \exp(2\pi i (i-1)(j-1)/5)$ and E is unit matrix of the dimension $2^{m-1}p$. The result of the product of $W(m)_j$ and the vector of which the ith element is $F^{m-1}{}_i$ is X.

In the case of Fourier transform of electron densities, the roles of the grid points and the reciprocal lattice points are reversed. The matrix I of which $(h+1)(i+1)$ is s $(hi \equiv s (\bmod N))$ is the transposed one of that of Fourier transform of structure factors. The value of p becomes two in this case. This can be seen easily as follows. The elements s and t of the $(i+1)$ st and $(2^{n-1}+i+1)$ st columns of the $(h+1)$ st row in I are

    $s = hi - 2^n u,$

    $t = h(2^{n-1}+i) - 2^n v.$

If $s = t$, then

    $2^{n-1}h = 2^n(u-v),$

hence, p becomes 2. The first and second elements of $F^0{}_{j+1}$ become

    $X(x_j) + X(x_{j+NH})$ and $|X(x_j) - X(x_{j+NH})| w_j$
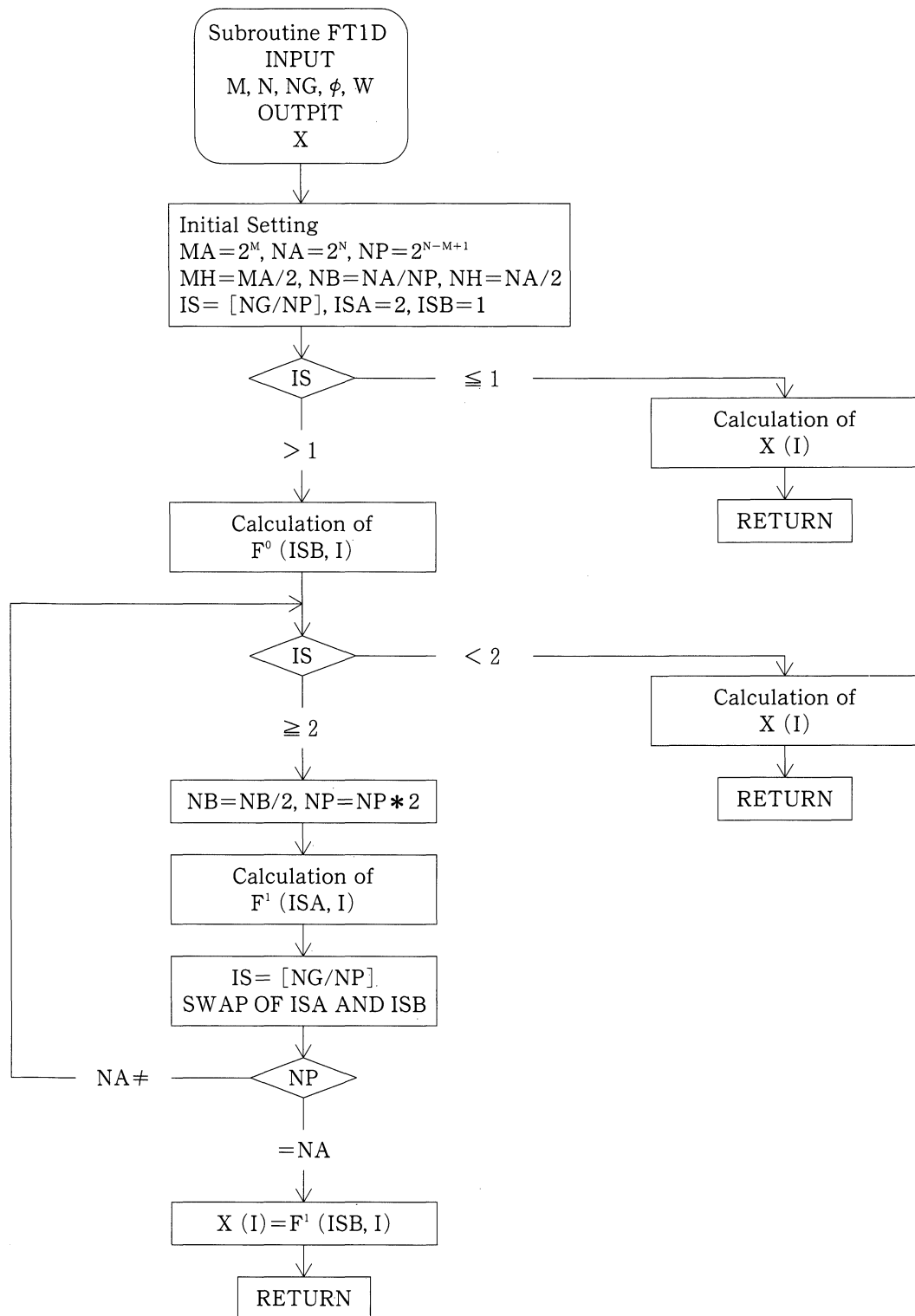
Fig.1   Flow chart of subroutine FT1D

where $NH = 2^{n-1}$ and $w_j = \exp(2\pi ij/N)$. The succeeding calculation is quite the same as the Fourier transform of structure factors.

2. Fourier transforms in crystal structure analysis

The electron density obtained by the Fourier transform of structure factors should satisfy the space group symmetry. When space group symmetry operation (R, t) transforms invaliantly the crystal structure, the electron density $\rho(Rr+t)$ is equal to $\rho(r)$. Hence, the points where the electron density to be calculated by Fourier transform can be confined within the asymmtric unit from which the all points in the unit cell can be obtained by space group symmetry. Hence, the Fourier transform is sufficient for the points in an asymmetric unit. The ratio of the volume of the primitive unit cell to that of the asymmetric unit is the order of the point group of the crystal. The volumes of asymmetric units are not related with the vectors t of space group symmetries. For example, asymmetric units of both space groups $P222$ and $P2_1 2_1 2_1$ can be

$$\{0 \leq x \leq 1/2,\ 0 \leq y \leq 1/2,\ 0 \leq z \leq 1\}.$$

The positions of asymmetric unit of the different space groups belonging to the same point group happen to be different, for example, the asymmetric unit of $P4_2 32$ is shifted by $z = -1/4$ from that of $P432$. Asymmetric units of space groups are listed in *International Tables for Crystallography, Vol.A.*

Asymmetric units should be chosen in order that all the points in the unit cells can be equivalent with the points in the asymmetric units. To keep the efficiency of the Fourier transform, it is desirable that the edges of the asymmetric unit are parallel to the edges of the unit cell.

Although symmetric units of higher symmetry have very complicated shapes, so-called asymmetric units for Fourier transform can be chosen so as to the domains obtained from union (in meanings of set theory) of the domains and its equivalent domains agree with the unit cell. The domain in unit cells of the space groups $P3$ and $P3_1$ can be chosen as,

$$\{0 \leq x \leq 1/3,\ 0 \leq y \leq 2/3,\ 0 \leq z \leq 1\}\ \cup\ \{0 \leq x \leq 1/3,\ 0 \leq y \leq 1/3,\ 0 \leq z \leq 1\}.$$

This domain is larger than the proper asymmetric units in these space groups, but the electron densities in the unit cells can be derived from the values at the points in the region.

3. Discussion

The merit of the formalism like equation (1.6) is that the successive order of the elements of X is not changed by the transformation and that the summation in equation (1.1) is done step by step by making partial sums. Hence, Fourier transform described in this article is suitable for subsets of grid points which represent the points in asymmetric units. Speeds of calculations of the present authors' method and FFT of Cooley and Tukey's algorithm may be nearly equal.

The result of the examination of the present authors' Fourier transform was as follows.

The total CPU time of full Fourier transform (not approximation given in *International Tables*) of the structure factors with space group I222 by ordinary Fourier transform was 5089.32 sec. for the range $-(M-1) \leq h, k, l \leq (M-1)$ and $0 \leq i/N, j/N, k/N \leq N-1$ where $M = N = 16$, the total CPU time by the present authors' Fourier transform was only 7.22 sec. When the ordinary transform is done utilizing the subcell, the total CPU time may be expected to become one eighth of the CPU time for the full transformation, hence the CPU time may become about 640 seconds. The total CPU time was 6.00 sec. for the present authors' Fourier transform. The effect utilizing subcell is small for FFT in general.

Ten Eyck (1973) discussed FFT and forms of asymmetric units. His discussions on asymmetric units is somewhat ambiguous and not exact as shown by the example of *P*222 and *P*2_1 2_1 2_1. The present authors could not determine the form of the asymmetric unit of P3 by his description. The ambiguity is brought by discussing the symmetry in reciprocal lattice for determination of asymmetric units. All the reciprocal lattice points are necessary to carry out Fourier transform of structure factors in principle. The volume of asymmetrical units is determined only from point group symmetry of the crystals.

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