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Fast Fourier Transforms for Space Groups Containing Rotation Axes of Order Three and Higher

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Abstract

Methods are described for exploiting the symmetry of uniaxial space groups containing rotation axes of order three and higher to improve the efficiency of computation of Fourier transforms. Mapping a symmetrical two-dimensional section into four dimensions enables the selection of non-contiguous asymmetric units over which fast Fourier transforms can be performed that reduce the computation time by a factor of approximately the order of the rotation axis. The application of the procedure to plane group $p3$ and its extension to $p4$ and $p6$ are described.

Introduction

Diffraction intensities and atomic distributions in crystals are related to one another by Fourier transforms, which therefore play a major role in structural crystallography. Because of this a large fraction of the work on computational methods, throughout the history of the application of diffraction techniques to crystallography, has been directed toward improving the efficiency of computation of Fourier transforms. A major advance came with the development by Cooley & Tukey (1965; also Gentleman & Sande, 1966) of a procedure that has become known as the fast Fourier transform, or FFT. Whereas previously used methods had required numbers of operations proportional to the square of the number of Fourier coefficients, N, the number of operations required by the FFT procedure is proportional approximately to $N \log N$, which, for moderately large values of N, increases only slightly more rapidly than linearly with increasing N.

Further savings in both time and computer capacity can be achieved if a FFT routine can make use of space-group symmetry to avoid storing redundant data and

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performing redundant computations. The application of space-group symmetry to FFT routines was discussed by Ten Eyck (1973), who classified space groups as 'solved' or 'almost solved', depending on whether all or almost all of the redundancy could be eliminated. All triclinic, monoclinic and orthorhombic space groups can be solved (even $P1$, because of Friedel's law, requires only half of the possible structure factors) and many of the uniaxial space groups that contain screw axes can also be solved or almost solved. As long as asymmetric units can be defined that extend the entire length of the unit cell in all three dimensions, which essentially means that no two-dimensional section contains rotation symmetry of order three or higher, computation of a density map from a unique set of structure factors, or computation of structure factors by inversion from an asymmetric unit of a density map, is fairly straightforward. For example, in space group $P4_1$ the output of two-dimensional transforms of sections in the region $0 \leq x < 1, 0 \leq y < 1, 0 \leq z < 1/4$ contains all of the information necessary for input to one-dimensional transforms along rows parallel to z in the region $0 \leq x \leq 1/2, 0 \leq y \leq 1/2, 0 \leq z < 1$, but in space group $P4$ there is no way to choose an asymmetric unit that extends the entire length of the unit cell in the a or b direction.

Wilson (1988, 1990) has studied the distribution of space groups in which organic molecules crystallize and has shown that crystals with high-order rotation axes, presumably because of packing problems, are extremely rare. However, proteins are known that crystallize in such space groups as $P6_3$ (McRee, Tainer, Meyer, Van Beeumen, Cusanovich & Getzoff, 1989) and $P6_322$ (Sussman, 1992) and symmetrical molecular complexes, such as viruses, can crystallize in space groups with higher-order rotation axes, which suggests that there is a practical need to address the problem of FFT routines that exploit the symmetry properties of space groups containing higher-order rotation axes. In this paper we show that, under certain easily satisfied conditions, it is in fact possible, by defining an asymmetric unit that is not contiguous, to perform a FFT whose time and memory requirements are reduced from those of the entire unit cell by a factor that is close to the order of the group for space groups that contain threefold rotation axes. We shall emphasize a procedure for the two-dimensional space group $p3$, which then can be incorporated in procedures for three-dimensional space groups containing threefold rotation axes and 6_3 screw axes. We conclude with a brief discussion of the extension of the procedures to fourfold and sixfold rotation axes.

Definitions

1. For a positive integer N the *ring of integers modulo* N, denoted by $\mathcal{Z}/N$, is the set of integers $\{0, 1, \ldots, N-1\}$, with all arithmetic operations defined modulo N.

2. For two sets of numbers, X and Y, the *Cartesian product* of X and Y, denoted by $X \times Y$, is the set of ordered pairs (x, y) such that x includes all elements of X, and y includes all elements of Y.

$$X \times Y = \{(x, y) | x \in X, y \in Y\}.$$

3. For positive integers N_1 and N_2, $\mathcal{Z}/N_1 \times \mathcal{Z}/N_2$ is the Cartesian product of $\mathcal{Z}/N_1$ and $\mathcal{Z}/N_2$, with componentwise arithmetic defined modulo N_1 and N_2, respectively.

4. The concept of Cartesian product extends to any finite number of factors. In particular, $\mathcal{Z}/N_1 \times \mathcal{Z}/N_2 \times \mathcal{Z}/N_3$ is the ordered set $\{(N_1, N_2, N_3) | N_j \in \mathcal{Z}/N_j, j = 1, 2, 3\}$.

5. We shall use the elements of $\mathcal{Z}/N_1 \times \mathcal{Z}/N_2$ and $\mathcal{Z}/N_1 \times \mathcal{Z}/N_2 \times \mathcal{Z}/N_3$ to label the nodes of a sublattice within a unit cell in two and three dimensions, respectively. A function defined at the nodes of this sublattice will be referred to as an *associated function.*

6. Two positive integers are *relatively prime* if they have no common factor greater than 1.

Consider a crystal whose structure conforms to the symmetry of a space group, G. A set of sublattice nodes that are equivalent under the operations of G is an *orbit*, and an associated function will have the same value at each node of an orbit. A subset of the nodes that contains one node from each orbit in the unit cell is an *asymmetric unit*. A space group is said to *act diagonally* if all matrices in a two- or three-dimensional representation of the group are diagonal, so that, within an orbit, x coordinates depend only on x coordinates, y coordinates depend only on y coordinates, and z coordinates depend only on z coordinates. Ten Eyck (1973) showed that efficient FFT procedures can be described if the space-group symmetries of all two-dimensional sections act diagonally.

The Chinese remainder theorem

Several consequences of a result known as the *Chinese remainder theorem* play a major role in the development of algorithms for FFTs with higher-order rotation axes. Consider a composite number $N = N_1 N_2$ and define a mapping from $\mathcal{Z}/N$ to $\mathcal{Z}/N_1 \times \mathcal{Z}/N_2$ by

$$M(x) = (x \bmod N_1, x \bmod N_2).$$

$M(x)$ associates an element x in $\mathcal{Z}/N$ with an ordered pair (x_1, x_2) in $\mathcal{Z}/N_1 \times \mathcal{Z}/N_2$. For a pair of elements, both in $\mathcal{Z}/N$, it is readily verified that

$$\begin{aligned} M(x+y) &= ([x+y] \bmod N_1, [x+y] \bmod N_2) \\ &= M(x) + M(y). \\ M(xy) &= (xy \bmod N_1, xy \bmod N_2) \\ &= M(x)M(y). \end{aligned}$$

Thus the mapping preserves the arithmetic structure of the rings and is an example of *ring homomorphism.* If a homomorphic mapping is invertible, that is if there is also a unique correspondence between (x_1, x_2) and x, the

Table 1. *Chinese remainder theorem mapping between* $\mathcal{Z}/2 \times \mathcal{Z}/5$ *and* $\mathcal{Z}/10$

	x_2				
x_1	0	1	2	3	4
0	0	6	2	8	4
1	5	1	7	3	9

rings are said to be *isomorphic*. The rings $\mathcal{Z}/N_1N_2$ and $\mathcal{Z}/N_1 \times \mathcal{Z}/N_2$ are not in general isomorphic.

The Chinese remainder theorem states that, if N_1 and N_2 are relatively prime, there exists a unique pair of elements of $\mathcal{Z}/N_1N_2$, e_1 and e_2, known as the *system of idempotents* corresponding to the factorization N_1N_2, such that $e_1 \bmod N_1 = 1$; $e_1 \bmod N_2 = 0$; $e_2 \bmod N_1 = 0$; $e_2 \bmod N_2 = 1$. It is readily verified that the set $\{e_1, e_2\}$ has the following additional properties.

1. $(e_1 + e_2) \bmod N_1N_2 = 1$;
2. $e_1e_2 \bmod N_1N_2 = 0$;
3. $e_1^2 \bmod N_1N_2 = e_1$; $e_2^2 \bmod N_1N_2 = e_2$.

From these properties it follows that

$$M^{-1}(x_1, x_2) = (x_1e_1 + x_2e_2) \bmod N$$

is a unique mapping from $\mathcal{Z}/N_1 \times \mathcal{Z}/N_2$ into $\mathcal{Z}/N_1N_2$ inverse to the mapping $M(x)$, and these rings are therefore isomorphic. Table 1 shows an example of this mapping for which $N_1 = 2$; $N_2 = 5$. For this case $e_1 = 5$ and $e_2 = 6$. It is apparent from inspection of the pattern that the fact that 2 and 5 are relatively prime is a necessary (and sufficient) condition for the mapping to be unique.

Diagonalization of space group $p3$

Although it is possible to construct a one-dimensional faithful representation of point group 3 using complex numbers, the smallest faithful representation (Prince, 1982) using real numbers (electron density must be invariant under real transformations) is two-dimensional. The representation referred to crystal axes, with a γ angle of 120°, is

$$\begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \begin{pmatrix} 0 & -1 \\ 1 & -1 \end{pmatrix}, \begin{pmatrix} -1 & 1 \\ -1 & 0 \end{pmatrix},$$

which is not diagonal. If the number of grid points along a cell edge can be factored into two relatively prime factors, however, the Chinese remainder theorem may be used to map the two-dimensional unit cell in space group $p3$ into a four-dimensional unit cell in which asymmetric units may be chosen that satisfy the conditions for performing a FFT that contains all of the information required to compute the transformation over the full cell. The procedure goes as follows:

1. Map the grid points (x, y) in $\mathcal{Z}/N_1N_2 \times \mathcal{Z}/N_1N_2$ into $(\mathcal{Z}/N_1 \times \mathcal{Z}/N_2) \times (\mathcal{Z}/N_1 \times \mathcal{Z}/N_2)$, giving $((x_1, x_2), (y_1, y_2))$. (Note that if N_1 and N_2 are relatively prime, N_1^2 and N_2^2 are also.)

2. Interchange x_2 and y_1, giving $((x_1, y_1), (x_2, y_2))$ in $(\mathcal{Z}/N_1 \times \mathcal{Z}/N_1) \times (\mathcal{Z}/N_2 \times \mathcal{Z}/N_2)$.

3. Choose an asymmetric unit that extends the entire range of (x_1, y_1) and Fourier transform it.

4. The output of this FFT contains all of the information required to fill in the transform of the entire cell. From this choose an asymmetric unit that contains the entire range of (x_2, y_2) [this procedure is known as *orbit exchange* and it is equivalent to the procedures described by Ten Eyck (1973) for space groups that are 'solved' or 'almost solved'] and transform it.

5. Perform the inverses of the transformations in steps 2 and 1 to recover the transform of the original (x, y).

To clarify how this procedure works in practice, consider again the case of $N_1 = 2$, $N_2 = 5$, so that the unit cell in $p3$ is 10×10. The 100 points divide into 34 orbits, with the one at (0, 0) being degenerate because of the special position. [Note that if the grid size had contained 3 as a factor, there would have been three degenerate orbits because of the special positions at (1/3,2/3) and (2/3,1/3).] Fig. 1 shows a 10×10 unit cell, with the grid points labeled by the numbers from 0 to 33 according to $p3$. Table 2 shows the remapping into a $2 \times 2 \times 5 \times 5$ four-dimensional space. Each entry in the table gives the orbit number and the coordinates of the points in the original cell that mapped to that point. The (x_2, y_2) rows of the table have been ordered according to a rule described by An, Tian & Tolimieri (1992), which emphasizes the underlying threefold symmetry. Examination of the table reveals that the rows divide the cell into 25 interpenetrating 2×2 lattices and the columns divide it into four interpenetrating 5×5 lattices. The second, third and fourth columns each contain one member of each of 25 orbits and they are transformed from one to another by the threefold rotation. It is also apparent that the first nine rows and the first two columns contain sufficient asymmetric units to allow computation of the full transform. The first row and the first column, which contain the point (0, 0), are subsets of the full cell with all of its symmetry. An algorithm

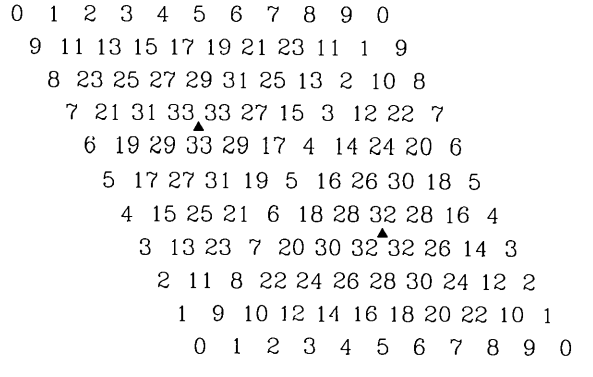

Fig. 1. A 10×10 unit cell with the grid points labeled by orbit numbers from 0 to 33 according to the two-dimensional space group $p3$. The positions of threefold axes other than the corners of the cell are shown.

Table 2. *Chinese remainder theorem mapping from a* 10×10 *unit cell with p3 symmetry into a* $2 \times 2 \times 5 \times 5$ *unit cell in four-dimensional space*

	(x_1, y_1)			
(x_2, y_2)	(0,0)	(1,0)	(0,1)	(1,1)
(0,0)	0(0,0)	5(5,0)	5(0,5)	(5,5)
(1,0)	6(6,0)	1(1,0)	16(6,5)	17(1,5)
(3,1)	24(8,6)	33(3,6)	22(8,1)	12(3,1)
(3,0)	8(8,0)	3(3,0)	30(8,5)	31(3,5)
(4,3)	29(4,8)	10(9,8)	20(4,3)	14(9,3)
(4,0)	4(4,0)	9(9,0)	19(4,5)	18(9,5)
(2,4)	25(2,4)	32(7,4)	13(2,9)	23(7,9)
(2,0)	2(2,0)	7(7,0)	27(2,5)	26(7,5)
(1,2)	28(6,2)	11(1,2)	15(6,7)	21(1,7)
(4,4)	6(4,4)	16(9,4)	17(4,9)	1(9,9)
(3,2)	24(8,2)	22(3,2)	12(8,7)	33(3,7)
(2,2)	8(2,2)	30(7,2)	31(2,7)	3(7,7)
(4,1)	29(4,6)	20(9,6)	14(4,1)	10(9,1)
(1,1)	4(6,6)	19(1,6)	18(6,1)	9(1,1)
(2,3)	25(2,8)	13(7,8)	23(2,3)	32(7,3)
(3,3)	2(8,8)	27(3,8)	26(8,3)	7(3,3)
(1,4)	28(6,4)	15(1,4)	21(6,9)	11(1,9)
(0,1)	6(0,6)	17(5,6)	1(0,1)	16(5,1)
(4,2)	24(4,2)	12(9,2)	33(4,7)	22(9,7)
(0,3)	8(0,8)	31(5,8)	3(0,3)	30(5,3)
(2,1)	29(2,6)	14(7,6)	10(2,1)	20(7,1)
(0,4)	4(0,4)	18(5,4)	9(0,9)	19(5,9)
(1,3)	25(6,8)	23(1,8)	32(6,3)	13(1,3)
(0,2)	2(0,2)	26(5,2)	7(0,7)	27(5,7)
(3,4)	28(8,4)	21(3,4)	11(8,9)	15(3,9)

described by An, Tian & Tolimieri (1992) allows further reduction in the transform of the first column, taking advantage of its symmetry. Note, however, that if the cell were 20×20 rather than 10×10, the asymmetric unit would require six columns out of sixteen, and still higher powers of 2 would come even closer to the ideal of requiring only one third of the computational effort.

Higher-order rotation axes

Although Wilson's (1990) studies suggest that rotation axes of order higher than three rarely, if ever, occur in crystal structures of organic compounds, it is of some interest to consider under what conditions the techniques applied to $p3$ may be extended to $p4$ and $p6$. From Table 2, it is apparent that one of the reasons the technique works for $p3$ with $N_1 = 2$ and $N_2 = 5$ is that the symmetry operators cycle the coordinates of equivalent points among the parity combinations odd-odd, odd-even and even-odd, thereby ensuring that the points lie in disjoint sets. A similar two-dimensional real faithful representation of point group 4 is

$$\begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix}, \begin{pmatrix} -1 & 0 \\ 0 & -1 \end{pmatrix}, \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}.$$

If a unit cell in $p4$ is subdivided by a 10×10 grid, two undesirable things happen. First, the sample includes the fourfold special position at (1/2,1/2) and the twofold special positions at (1/2,0) and (0,1/2), so that there are three degenerate orbits and, even more importantly, the group operations do not change the parities of the coordinates, so that, although even-odd and odd-even interchange, odd-odd, as well as even-even, forms a fully symmetric set of its own. If, however, the unit cell is subdivided by a 15×15 grid, so that $N_1 = 3$ and $N_2 = 5$, the 225 points, in 57 orbits, divide into nine interpenetrating 5×5 lattices or 25 interpenetrating 3×3 lattices, and three out of nine columns or seven out of 25 rows of a table similar to Table 2 would form sufficient asymmetric units. This suggests that for efficient computation of FFTs the factors N_1 and N_2 of the subdividing grid should be relatively prime not only to each other but to the order of the rotation axis. The smallest number for $p4$ is therefore 15, and the smallest number for $p6$ would then be 35.

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