general programming technique to allow full use of symmetry elements (orthorhombic or lower) in the 3D Beevers–Lipson factorization process, including multiplicity corrections. Their method was later adopted by Shoemaker & Sly (1961), and by crystallographic program writers at large.

The discovery of the FFT algorithm by Cooley & Tukey in 1965, which instantly transformed electrical engineering and several other disciplines, paradoxically failed to have an immediate impact on crystallographic computing. A plausible explanation is that the calculation of large 3D Fourier maps was a relatively infrequent task which was not thought to constitute a bottleneck, as crystallographers had learned to settle most structural questions by means of cheaper 2D sections or projections. It is significant in this respect that the first use of the FFT in crystallography by Barrett & Zwick (1971) should have occurred as part of an iterative scheme for improving protein phases by density modification in real space, which required a much greater number of Fourier transformations than any previous method. Independently, Bondot (1971) had attracted attention to the merits of the FFT algorithm.

The FFT program used by Barrett & Zwick had been written for signal-processing applications. It was restricted to sampling rates of the form 2^n, and was not designed to take advantage of crystallographic symmetry at any stage of the calculation; Bantz & Zwick (1974) later improved this situation somewhat.

It was the work of Ten Eyck (1973) and Immirzi (1973, 1976) which led to the general adoption of the FFT in crystallographic computing. Immirzi treated all space groups as *P*1 by data expansion. Ten Eyck based his program on a versatile multi-radix FFT routine (Gentleman & Sande, 1966) coupled with a flexible indexing scheme for dealing efficiently with multidimensional transforms. He also addressed the problems of incorporating symmetry elements of order 2 into the factorization of 1D transforms, and of transposing intermediate results by other symmetry elements. He was thus able to show that in a large number of space groups (including the 74 space groups having orthorhombic or lower symmetry) it is possible to calculate only the unique results from the unique data within the logic of the FFT algorithm. Ten Eyck wrote and circulated a package of programs for computing Fourier maps and re-analysing them into structure factors in some simple space groups (*P*1, *P*1, *P*2, *P*2/*m*, $P2_1$, *P*222, $P2_12_12_1$, *Pmmm*). This package was later augmented by a handful of new space-group-specific programs contributed by other crystallographers ($P2_12_12$, *I*222, $P3_121$, $P4_12_12$). The writing of such programs is an undertaking of substantial complexity, which has deterred all but the bravest: the usual practice is now to expand data for a high-symmetry space group to the largest subgroup for which a specific FFT program exists in the package, rather than attempt to write a new program. Attempts have been made to introduce more modern approaches to the calculation of crystallographic Fourier transforms (Auslander, Feig & Winograd, 1982; Auslander & Shenefelt, 1987; Auslander *et al.*, 1988) but have not gone beyond the stage of preliminary studies.

The task of fully exploiting the FFT algorithm in crystallographic computations is therefore still unfinished, and it is the purpose of this section to provide a systematic treatment such as that (say) of Ahmed & Barnes (1958) for the Beevers–Lipson algorithm.

Ten Eyck's approach, based on the reducibility of certain space groups, is extended by the derivation of a universal transposition formula for intermediate results. It is then shown that space groups which are not completely reducible may nevertheless be treated by three-dimensional Cooley–Tukey factorization in such a way that their symmetry may be fully exploited, whatever the shape of their asymmetric unit. Finally, new factorization methods with built-in symmetries are presented. The unifying concept throughout this presentation is that of 'group action' on indexing sets, and of 'orbit exchange' when this action has a composite structure; it affords new ways of rationalizing the use of symmetry, or of improving computational speed, or both.

1.3.4.3.2. Defining relations and symmetry considerations

A finite set of reflections $\{F_{\mathbf{h}_l}\}_{l\in L}$ can be periodized without aliasing by the translations of a suitable sublattice $\mathbf{N}^T\Lambda^*$ of the reciprocal lattice Λ^*; the converse operation in real space is the sampling of ρ at points $\mathbf{X}$ of a grid of the form $\mathbf{N}^{-1}\Lambda$ (Section 1.3.2.7.3). In standard coordinates, $\{F_{\mathbf{h}_l}\}_{l\in L}$ is periodized by $\mathbf{N}^T\mathbb{Z}^3$, and $\mathscr{P}$ is sampled at points $\mathbf{x}\in\mathbf{N}^{-1}\mathbb{Z}^3$.

In the absence of symmetry, the unique data are

– the $F_{\mathbf{h}}$ indexed by $\mathbf{h}\in\mathbb{Z}^3/\mathbf{N}^T\mathbb{Z}^3$ in reciprocal space;

– the $\mathscr{P}_{\mathbf{x}}$ indexed by $\mathbf{x}\in(\mathbf{N}^{-1}\mathbb{Z}^3)/\mathbb{Z}^3$; or equivalently the $\mathscr{P}_{\mathbf{m}}$ indexed by $\mathbf{m}\in\mathbb{Z}^3/\mathbf{N}\mathbb{Z}^3$, where $\mathbf{x}=\mathbf{N}^{-1}\mathbf{m}$.

They are connected by the ordinary DFT relations:

$$F_{\mathbf{h}}=\frac{1}{|\det\mathbf{N}|}\sum_{\mathbf{x}\in(\mathbf{N}^{-1}\mathbb{Z}^3)/\mathbb{Z}^3}\mathscr{P}_{\mathbf{x}}\exp(2\pi i\mathbf{h}\cdot\mathbf{x})$$

or

$$F_{\mathbf{h}}=\frac{1}{|\det\mathbf{N}|}\sum_{\mathbf{m}\in\mathbb{Z}^3/\mathbf{N}\mathbb{Z}^3}\mathscr{P}_{\mathbf{m}}\exp[2\pi i\mathbf{h}\cdot(\mathbf{N}^{-1}\mathbf{m})]$$

and

$$\mathscr{P}_{\mathbf{x}}=\sum_{\mathbf{h}=\mathbb{Z}^3/\mathbf{N}^T\mathbb{Z}^3}F_{\mathbf{h}}\exp(-2\pi i\mathbf{h}\cdot\mathbf{x})$$

or

$$\mathscr{P}_{\mathbf{m}}=\sum_{\mathbf{h}\in\mathbb{Z}^3/\mathbf{N}^T\mathbb{Z}^3}F_{\mathbf{h}}\exp[-2\pi i\mathbf{h}\cdot(\mathbf{N}^{-1}\mathbf{m})].$$

In the presence of symmetry, the unique data are

– $\{\mathscr{P}_{\mathbf{x}}\}_{\mathbf{x}\in D}$ or $\{\mathscr{P}_{\mathbf{m}}\}_{\mathbf{m}\in D}$ in real space (by abuse of notation, D will denote an asymmetric unit for $\mathbf{x}$ or for $\mathbf{m}$ indifferently);

– $\{F_{\mathbf{h}}\}_{\mathbf{h}\in D^*}$ in reciprocal space.

The previous summations may then be subjected to orbital decomposition, to yield the following 'crystallographic DFT' (CDFT) defining relations:

$$\begin{aligned}F_{\mathbf{h}}&=\frac{1}{|\det\mathbf{N}|}\sum_{\mathbf{x}\in D}\mathscr{P}_{\mathbf{x}}\left[\sum_{\gamma\in G/G_{\mathbf{x}}}\exp\{2\pi i\mathbf{h}\cdot[S_\gamma(\mathbf{x})]\}\right]\\&=\frac{1}{|\det\mathbf{N}|}\sum_{\mathbf{x}\in D}\mathscr{P}_{\mathbf{x}}\left[\frac{1}{|G_{\mathbf{x}}|}\sum_{g\in G}\exp\{2\pi i\mathbf{h}\cdot[S_g(\mathbf{x})]\}\right],\\\mathscr{P}_{\mathbf{x}}&=\sum_{\mathbf{h}\in D^*}F_{\mathbf{h}}\left[\sum_{\gamma\in G/G_{\mathbf{h}}}\exp\{-2\pi i\mathbf{h}\cdot[S_\gamma(\mathbf{x})]\}\right]\\&=\sum_{\mathbf{h}\in D^*}F_{\mathbf{h}}\left[\frac{1}{|G_{\mathbf{h}}|}\sum_{g\in G}\exp\{-2\pi i\mathbf{h}\cdot[S_g(\mathbf{x})]\}\right],\end{aligned}$$

with the obvious alternatives in terms of $\mathscr{P}_{\mathbf{m}}$, $\mathbf{m}=\mathbf{N}\mathbf{x}$. Our problem is to evaluate the CDFT for a given space group as efficiently as possible, in spite of the fact that the group action has spoilt the simple tensor-product structure of the ordinary three-dimensional DFT (Section 1.3.3.3.1).

Two procedures are available to carry out the 3D summations involved as a succession of smaller summations:

Copyright © 2010 International Union of Crystallography

(1) *decomposition* into successive transforms of *fewer dimensions* but on the *same number of points* along these dimensions. This possibility depends on the reducibility of the space group, as defined in Section 1.3.4.2.2.4, and simply invokes the tensor product property of the DFT;

(2) *factorization* of the transform into transforms of the *same number of dimensions* as the original one, but on *fewer points* along each dimension. This possibility depends on the arithmetic factorability of the decimation matrix **N**, as described in Section 1.3.3.3.2.

Clearly, a symmetry expansion to the largest fully reducible subgroup of the space group will give maximal decomposability, but will require computing more than the unique results from more than the unique data. Economy will follow from factoring the transforms in the subspaces within which the space group acts irreducibly.

For irreducible subspaces of dimension 1, the group action is readily incorporated into the factorization of the transform, as first shown by Ten Eyck (1973).

For irreducible subspaces of dimension 2 or 3, the ease of incorporation of symmetry into the factorization depends on the type of factorization method used. The multidimensional Cooley–Tukey method (Section 1.3.3.3.1) is rather complicated; the multidimensional Good method (Section 1.3.3.3.2.2) is somewhat simpler; and the Rader/Winograd factorization admits a generalization, based on the arithmetic of certain rings of *algebraic* integers, which accommodates 2D crystallographic symmetries in a most powerful and pleasing fashion.

At each stage of the calculation, it is necessary to keep track of the definition of the asymmetric unit and of the symmetry properties of the numbers being manipulated. This requirement applies not only to the initial data and to the final results, where these are familiar; but also to all the intermediate quantities produced by partial transforms (on subsets of factors, or subsets of dimensions, or both), where they are less familiar. Here, the general formalism of transposition (or 'orbit exchange') described in Section 1.3.4.2.2.2 plays a central role.

1.3.4.3.3. Interaction between symmetry and decomposition

Suppose that the space-group action is reducible, *i.e.* that for each $g \in G$

$$\mathbf{R}_g = \begin{pmatrix} \mathbf{R}'_g & \mathbf{0} \\ \mathbf{0} & \mathbf{R}''_g \end{pmatrix}, \qquad \mathbf{t}_g = \begin{pmatrix} \mathbf{t}'_g \\ \mathbf{t}''_g \end{pmatrix};$$

by Schur's lemma, the decimation matrix must then be of the form

$$\mathbf{N} = \begin{pmatrix} \mathbf{N}' & \mathbf{0} \\ \mathbf{0} & \mathbf{N}'' \end{pmatrix}$$

if it is to commute with all the $\mathbf{R}_g$.

Putting

$$\mathbf{x} = \begin{pmatrix} \mathbf{x}' \\ \mathbf{x}'' \end{pmatrix} \quad \text{and} \quad \mathbf{h} = \begin{pmatrix} \mathbf{h}' \\ \mathbf{h}'' \end{pmatrix},$$

we may define

$$S'_g(\mathbf{x}') = \mathbf{R}'_g\mathbf{x}' + \mathbf{t}'_g,$$
$$S''_g(\mathbf{x}'') = \mathbf{R}''_g\mathbf{x}'' + \mathbf{t}''_g,$$

and write $S_g = S'_g \oplus S''_g$ (direct sum) as a shorthand for

$$S_g(\mathbf{x}) = \begin{pmatrix} S'_g(\mathbf{x}') \\ S''_g(\mathbf{x}'') \end{pmatrix}.$$

We may also define the representation operators $S'^{\#}_g$ and $S''^{\#}_g$ acting on functions of $\mathbf{x}'$ and $\mathbf{x}''$, respectively (as in Section 1.3.4.2.2.4), and the operators S'^{*}_g and S''^{*}_g acting on functions of $\mathbf{h}'$ and $\mathbf{h}''$, respectively (as in Section 1.3.4.2.2.5). Then we may write

$$S^{\#}_g = (S'_g)^{\#} \oplus (S''_g)^{\#}$$

and

$$S^{*}_g = (S'_g)^{*} \oplus (S''_g)^{*}$$

in the sense that g acts on $f(\mathbf{x}) \equiv f(\mathbf{x}', \mathbf{x}'')$ by

$$(S^{\#}_g f)(\mathbf{x}', \mathbf{x}'') = f[(S'_g)^{-1}(\mathbf{x}'), (S''_g)^{-1}(\mathbf{x}'')]$$

and on $\Phi(\mathbf{h}) \equiv \Phi(\mathbf{h}', \mathbf{h}'')$ by

$$(S^{*}_g \Phi)(\mathbf{h}', \mathbf{h}'') = \exp(2\pi i \mathbf{h}' \cdot \mathbf{t}'_g) \exp(2\pi i \mathbf{h}'' \cdot \mathbf{t}''_g) \times \Phi[\mathbf{R}'^{T}_g \mathbf{h}', \mathbf{R}''^{T}_g \mathbf{h}''].$$

Thus equipped we may now derive concisely a general identity describing the symmetry properties of intermediate quantities of the form

$$T(\mathbf{x}', \mathbf{h}'') = \sum_{\mathbf{h}'} F(\mathbf{h}', \mathbf{h}'') \exp(-2\pi i \mathbf{h}' \cdot \mathbf{x}') = \frac{1}{|\det \mathbf{N}'|} \sum_{\mathbf{x}''} \rho(\mathbf{x}', \mathbf{x}'') \exp(+2\pi i \mathbf{h}'' \cdot \mathbf{x}''),$$

which arise through partial transformation of F on $\mathbf{h}'$ or of ρ on $\mathbf{x}''$. The action of $g \in G$ on these quantities will be

(i) through $(S'_g)^{\#}$ on the function $\mathbf{x}' \longmapsto T(\mathbf{x}', \mathbf{h}'')$,

(ii) through $(S''_g)^{*}$ on the function $\mathbf{h}'' \longmapsto T(\mathbf{x}', \mathbf{h}'')$,

and hence the symmetry properties of T are expressed by the identity

$$T = [(S'_g)^{\#} \oplus (S''_g)^{*}]T.$$

Applying this relation not to T but to $[(S'_{g^{-1}})^{\#} \oplus (S''_e)^{*}]T$ gives

$$[(S'_{g^{-1}})^{\#} \oplus (S''_e)^{*}]T = [(S'_e)^{\#} \oplus (S''_g)^{*}]T,$$

i.e.

$$\boxed{T(S'_g(\mathbf{x}'), \mathbf{h}'') = \exp(2\pi i \mathbf{h}'' \cdot \mathbf{t}''_g) T(\mathbf{x}', \mathbf{R}''^{T}_g \mathbf{h}'').}$$

If the unique $F(\mathbf{h}) \equiv F(\mathbf{h}', \mathbf{h}'')$ were initially indexed by

$$(\text{all } \mathbf{h}') \times (\text{unique } \mathbf{h}'')$$

(see Section 1.3.4.2.2.2), this formula allows the reindexing of the intermediate results $T(\mathbf{x}', \mathbf{h}'')$ from the initial form

$$(\text{all } \mathbf{x}') \times (\text{unique } \mathbf{h}'')$$