1. GENERAL RELATIONSHIPS AND TECHNIQUES

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 $P1$ 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 ($P1$, $P\bar{1}$, $P2$, $P2/m$, $P2_1$, $P222$, $P2_12_12_1$, $Pmmm$). This package was later augmented by a handful of new space-group-specific programs contributed by other crystallographers ($P2_12_12$, $I222$, $P3_121$, $P4_12_12$). 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 $\wp$ 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 $\wp_{\mathbf{x}}$ indexed by $\mathbf{x}\in(\mathbf{N}^{-1}\mathbb{Z}^3)/\mathbb{Z}^3$; or equivalently the $\wp_{\mathbf{m}}$ 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:

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

or

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

and

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

or

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

In the presence of symmetry, the unique data are
– $\{\wp_{\mathbf{x}}\}_{\mathbf{x}\in D}$ or $\{\wp_{\mathbf{m}}\}_{\mathbf{m}\in D}$ 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:

$$F_{\mathbf{h}} = \frac{1}{|\det\mathbf{N}|}\sum_{\mathbf{x}\in D}\wp_{\mathbf{x}}\left[\sum_{\gamma\in G/G_{\mathbf{x}}}\exp\{2\pi i\mathbf{h}\cdot[S_\gamma(\mathbf{x})]\}\right]$$

$$= \frac{1}{|\det\mathbf{N}|}\sum_{\mathbf{x}\in D}\wp_{\mathbf{x}}\left[\frac{1}{|G_{\mathbf{x}}|}\sum_{g\in G}\exp\{2\pi i\mathbf{h}\cdot[S_g(\mathbf{x})]\}\right],$$

$$\wp_{\mathbf{x}} = \sum_{\mathbf{h}\in D^*}F_{\mathbf{h}}\left[\sum_{\gamma\in G/G_{\mathbf{h}}}\exp\{-2\pi i\mathbf{h}\cdot[S_\gamma(\mathbf{x})]\}\right]$$

$$= \sum_{\mathbf{h}\in D^*}F_{\mathbf{h}}\left[\frac{1}{|G_{\mathbf{h}}|}\sum_{g\in G}\exp\{-2\pi i\mathbf{h}\cdot[S_g(\mathbf{x})]\}\right],$$

with the obvious alternatives in terms of $\wp_{\mathbf{m}}$, $\mathbf{m}=\mathbf{N}\mathbf{x}$. 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:

(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 $\mathbf{N}$, 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 $\mathbf{x} = \begin{pmatrix} \mathbf{x}' \\ \mathbf{x}'' \end{pmatrix}$ and $\mathbf{h} = \begin{pmatrix} \mathbf{h}' \\ \mathbf{h}'' \end{pmatrix}$, 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

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

which arise through partial transformation of F on $\mathbf{h}'$ or of $\wp$ 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

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

to the final form

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

on which the second transform (on $\mathbf{h}''$) may now be performed, giving the final results $\wp(\mathbf{x}', \mathbf{x}'')$ indexed by

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

which is an asymmetric unit. An analogous interpretation holds if one is going from $\wp$ to F.

The above formula solves the general problem of transposing from one invariant subspace to another, and is the main device for decomposing the CDFT. Particular instances of this formula were derived and used by Ten Eyck (1973); it is useful for orthorhombic groups, and for dihedral groups containing screw axes n_m with g.c.d. $(m, n) = 1$. For comparison with later uses of orbit exchange, it should be noted that the type of intermediate results just dealt with is obtained after transforming on *all* factors in *one* summand.

A central piece of information for driving such a decomposition is the definition of the full asymmetric unit in terms of the asymmetric units in the invariant subspaces. As indicated at the end of Section 1.3.4.2.2.2, this is straightforward when G acts without fixed points, but becomes more involved if fixed points do exist. To this day, no systematic 'calculus of asymmetric units' exists which can automatically generate a complete description of the asymmetric unit of an arbitrary space group in a form suitable for directing the orbit exchange process, although Shenefelt (1988) has outlined a procedure for dealing with space group $P622$ and its subgroups. The asymmetric unit definitions given in Volume A of *International Tables* are incomplete in this respect, in that they do not specify the possible residual symmetries which may exist on the boundaries of the domains.

1.3.4.3.4. *Interaction between symmetry and factorization*

Methods for factoring the DFT in the absence of symmetry were examined in Sections 1.3.3.2 and 1.3.3.3. They are based on the observation that the finite sets which index both data and results are endowed with certain algebraic structures (*e.g.* are Abelian groups, or rings), and that subsets of indices may be found which are not merely subsets but *substructures* (*e.g.* subgroups or subrings). Summation over these substructures leads to partial transforms, and the way in which substructures fit into the global structure indicates how to reassemble the partial results into the final results. As a rule, the richer the algebraic structure which is identified in the indexing set, the more powerful the factoring method.

references