The cross-correlation $r * \check{\wp}_1^0 * \wp_2^0$ between the original electron densities is then obtained by further periodizing by $\mathbb{Z}^3$.

Note that these expressions are valid for any choice of 'atomic' density functions $\wp_{j_1}^{(1)}$ and $\wp_{j_2}^{(2)}$, which may be taken as molecular fragments if desired (see Section 1.3.4.4.8).

If G contains elements g such that $\mathbf{R}_g$ has an eigenspace E_1 with eigenvalue 1 and an invariant complementary subspace E_2, while $\mathbf{t}_g$ has a non-zero component $\mathbf{t}_g^{(1)}$ in E_1, then the Patterson function $r * \check{\wp}^0 * \wp^0$ will contain *Harker peaks* (Harker, 1936) of the form

$$S_g(\mathbf{x}) - \mathbf{x} = \mathbf{t}_g^{(1)} \oplus (S_g^{(2)}(\mathbf{x}) - \mathbf{x})$$

[where $S_g^{(s)}$ represent the action of g in E_2] in the translate of E_1 by $\mathbf{t}_g^{(1)}$.

1.3.4.3. *Crystallographic discrete Fourier transform algorithms*

1.3.4.3.1. *Historical introduction*

In 1929, W. L. Bragg demonstrated the practical usefulness of the Fourier transform relation between electron density and structure factors by determining the structure of diopside from three principal projections calculated numerically by 2D Fourier summation (Bragg, 1929). It was immediately realized that the systematic use of this powerful method, and of its extension to three dimensions, would entail considerable amounts of numerical computation which had to be organized efficiently. As no other branch of applied science had yet needed this type of computation, crystallographers had to invent their own techniques.

The first step was taken by Beevers & Lipson (1934) who pointed out that a 2D summation could be factored into successive 1D summations. This is essentially the tensor product property of the Fourier transform (Sections 1.3.2.4.2.4, 1.3.3.3.1), although its aspect is rendered somewhat complicated by the use of sines and cosines instead of complex exponentials. Computation is economized to the extent that the cost of an $N \times N$ transform grows with N as $2N^3$ rather than N^4. Generalization to 3D is immediate, reducing computation size from N^6 to $3N^4$ for an $N \times N \times N$ transform. The complication introduced by using expressions in terms of sines and cosines is turned to advantage when symmetry is present, as certain families of terms are systematically absent or are simply related to each other; multiplicity corrections must, however, be introduced. The necessary information was tabulated for each space group by Lonsdale (1936), and was later incorporated into Volume I of *International Tables*.

The second step was taken by Beevers & Lipson (1936) and Lipson & Beevers (1936) in the form of the invention of the 'Beevers–Lipson strips', a practical device which was to assist a whole generation of crystallographers in the numerical computation of crystallographic Fourier sums. The strips comprise a set of 'cosine strips' tabulating the functions

$$A \cos\left(\frac{2\pi hm}{60}\right) \quad (A = 1, 2, \ldots, 99;\ h = 1, 2, \ldots, 99)$$

and a set of 'sine strips' tabulating the functions

$$B \sin\left(\frac{2\pi hm}{60}\right) \quad (B = 1, 2, \ldots, 99;\ h = 1, 2, \ldots, 99)$$

for the 16 arguments $m = 0, 1, \ldots, 15$. Function values are rounded to the nearest integer, and those for other arguments m may be obtained by using the symmetry properties of the sine and cosine functions. A Fourier summation of the form

$$Y(m) = \sum_{j=1}^{n} \left[A_j \cos\left(\frac{2\pi h_j m}{60}\right) + B_j \sin\left(\frac{2\pi h_j m}{60}\right) \right]$$

is then performed by selecting the n cosine strips labelled (A_j, h_j) and the n sine strips labelled (B_j, h_j), placing them in register, and adding the tabulated values columnwise. The number 60 was chosen as the l.c.m. of 12 (itself the l.c.m. of the orders of all possible non-primitive translations) and of 10 (for decimal convenience). The limited accuracy imposed by the two-digit tabulation was later improved by Robertson's sorting board (Robertson, 1936*a*,*b*) or by the use of separate strips for each decimal digit of the amplitude (Booth, 1948*b*), which allowed three-digit tabulation while keeping the set of strips within manageable size. Cochran (1948*a*) found that, for most structures under study at the time, the numerical inaccuracies of the method were less than the level of error in the experimental data. The sampling rate was subsequently increased from 60 to 120 (Beevers, 1952) to cope with larger unit cells.

Further gains in speed and accuracy were sought through the construction of special-purpose mechanical, electro-mechanical, electronic or optical devices. Two striking examples are the mechanical computer RUFUS built by Robertson (1954, 1955, 1961) on the principle of previous strip methods (see also Robertson, 1932) and the electronic analogue computer X-RAC built by Pepinsky, capable of real-time calculation and display of 2D and 3D Fourier syntheses (Pepinsky, 1947; Pepinsky & Sayre, 1948; Pepinsky *et al.*, 1961; see also Suryan, 1957). The optical methods of Lipson & Taylor (1951, 1958) also deserve mention. Many other ingenious devices were invented, whose descriptions may be found in Booth (1948*b*), Niggli (1961), and Lipson & Cochran (1968).

Later, commercial punched-card machines were programmed to carry out Fourier summations or structure-factor calculations (Shaffer *et al.*, 1946*a*,*b*; Cox *et al.*, 1947, 1949; Cox & Jeffrey, 1949; Donohue & Schomaker, 1949; Grems & Kasper, 1949; Hodgson *et al.*, 1949; Greenhalgh & Jeffrey, 1950; Kitz & Marchington, 1953).

The modern era of digital electronic computation of Fourier series was initiated by the work of Bennett & Kendrew (1952), Mayer & Trueblood (1953), Ahmed & Cruickshank (1953*b*), Sparks *et al.* (1956) and Fowweather (1955). Their Fourier-synthesis programs used Beevers–Lipson factorization, the program by Sparks *et al.* being the first 3D Fourier program useable for all space groups (although these were treated as $P1$ or $P\bar{1}$ by data expansion). Ahmed & Barnes (1958) then proposed a 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 $P1$ 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 ρ 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 $\rho_{\mathbf{x}}$ indexed by $\mathbf{x}\in(\mathbf{N}^{-1}\mathbb{Z}^3)/\mathbb{Z}^3$; or equivalently the $\rho_{\mathbf{m}}$ 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:

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

or

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

and

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

or

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

In the presence of symmetry, the unique data are

– $\{\rho_{\mathbf{x}}\}_{\mathbf{x}\in D}$ or $\{\rho_{\mathbf{m}}\}_{\mathbf{m}\in D}$ 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:

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

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

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

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

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

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 $\rho(\mathbf{x}', \mathbf{x}'')$ 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 ρ 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 *P*622 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.

The ability of a given factoring method to accommodate crystallographic symmetry will thus be determined by the extent to which the crystallographic group action respects (or fails to respect) the partitioning of the index set into the substructures pertaining to that method. This remark justifies trying to gain an overall view of the algebraic structures involved, and of the possibilities of a crystallographic group acting 'naturally' on them.

The index sets $\{\mathbf{m}|\mathbf{m} \in \mathbb{Z}^3/\mathbf{N}\mathbb{Z}^3\}$ and $\{\mathbf{h}|\mathbf{h} \in \mathbb{Z}^3/\mathbf{N}^T\mathbb{Z}^3\}$ are finite Abelian groups under component-wise addition. If an iterated addition is viewed as an action of an integer scalar $n \in \mathbb{Z}$ *via*

$$\begin{aligned} n\mathbf{h} &= \mathbf{h} + \mathbf{h} + \ldots + \mathbf{h} \qquad (n \text{ times}) \qquad \text{for } n > 0, \\ &= \mathbf{0} \qquad\qquad\qquad\qquad\quad \text{for } n = 0, \\ &= -(\mathbf{h} + \mathbf{h} + \ldots + \mathbf{h}) \quad (|n| \text{ times}) \quad \text{for } n < 0, \end{aligned}$$

then an Abelian group becomes a *module* over the ring $\mathbb{Z}$ (or, for short, a $\mathbb{Z}$-module), a module being analogous to a vector space but with scalars drawn from a ring rather than a field. The left actions of a crystallographic group G by

$$g: \quad \mathbf{m} \longmapsto \mathbf{R}_g\mathbf{m} + \mathbf{N}\mathbf{t}_g \mod \mathbf{N}\mathbb{Z}^3$$

and by

$$g: \quad \mathbf{h} \longmapsto (\mathbf{R}_g^{-1})^T\mathbf{h} \mod \mathbf{N}^T\mathbb{Z}^3$$

can be combined with this $\mathbb{Z}$ action as follows:

$$\begin{aligned} \textstyle\sum_{g\in G} n_g g: &\quad \mathbf{m} \longmapsto \textstyle\sum_{g\in G} n_g(\mathbf{R}_g\mathbf{m} + \mathbf{N}\mathbf{t}_g) \mod \mathbf{N}\mathbb{Z}^3, \\ \textstyle\sum_{g\in G} n_g g: &\quad \mathbf{h} \longmapsto \textstyle\sum_{g\in G} n_g[(\mathbf{R}_g^{-1})^T\mathbf{h}] \mod \mathbf{N}^T\mathbb{Z}^3. \end{aligned}$$

This provides a left action, on the indexing sets, of the set

$$\mathbb{Z}G = \left\{ \sum_{g\in G} n_g g \middle| n_g \in \mathbb{Z} \text{ for each } g \in G \right\}$$

of symbolic linear combinations of elements of G with integral coefficients. If addition and multiplication are defined in $\mathbb{Z}G$ by

$$\left(\sum_{g_1\in G} a_{g_1}g_1\right) + \left(\sum_{g_2\in G} b_{g_2}g_2\right) = \sum_{g\in G}(a_g + b_g)g$$

and

$$\left(\sum_{g_1\in G} a_{g_1}g_1\right) \times \left(\sum_{g_2\in G} b_{g_2}g_2\right) = \sum_{g\in G} c_g g,$$

with

$$c_g = \sum_{g'\in G} a_{g'} b_{(g')^{-1}g},$$

then $\mathbb{Z}G$ is a *ring*, and the action defined above makes the indexing sets into $\mathbb{Z}G$-*modules*. The ring $\mathbb{Z}G$ is called the *integral group ring* of G (Curtis & Reiner, 1962, p. 44).

From the algebraic standpoint, therefore, the interaction between symmetry and factorization can be expected to be favourable whenever the indexing sets of partial transforms are $\mathbb{Z}G$-submodules of the main $\mathbb{Z}G$-modules.

1.3.4.3.4.1. *Multidimensional Cooley–Tukey factorization*

Suppose, as in Section 1.3.3.3.2.1, that the decimation matrix $\mathbf{N}$ may be factored as $\mathbf{N}_1\mathbf{N}_2$. Then any grid point index $\mathbf{m} \in \mathbb{Z}^3/\mathbf{N}\mathbb{Z}^3$ in real space may be written

$$\mathbf{m} = \mathbf{m}_1 + \mathbf{N}_1\mathbf{m}_2$$

with $\mathbf{m}_1 \in \mathbb{Z}^3/\mathbf{N}_1\mathbb{Z}^3$ and $\mathbf{m}_2 \in \mathbb{Z}^3/\mathbf{N}_2\mathbb{Z}^3$ determined by

$$\begin{aligned} \mathbf{m}_1 &= \mathbf{m} \mod \mathbf{N}_1\mathbb{Z}^3, \\ \mathbf{m}_2 &= \mathbf{N}_1^{-1}(\mathbf{m} - \mathbf{m}_1) \mod \mathbf{N}_2\mathbb{Z}^3. \end{aligned}$$

These relations establish a one-to-one correspondence $\mathbf{m} \leftrightarrow (\mathbf{m}_1, \mathbf{m}_2)$ between $I = \mathbb{Z}^3/\mathbf{N}\mathbb{Z}^3$ and the Cartesian product $I_1 \times I_2$ of $I_1 = \mathbb{Z}^3/\mathbf{N}_1\mathbb{Z}^3$ and $I_2 = \mathbb{Z}^3/\mathbf{N}_2\mathbb{Z}^3$, and hence $I \cong I_1 \times I_2$ as a set. However $I \ncong I_1 \times I_2$ as an Abelian group, since in general $\mathbf{m} + \mathbf{m}' \not\leftrightarrow (\mathbf{m}_1 + \mathbf{m}_1', \mathbf{m}_2 + \mathbf{m}_2')$ because there can be a 'carry' from the addition of the first components into the second components; therefore, $I \ncong I_1 \times I_2$ as a $\mathbb{Z}G$-module, which shows that the incorporation of symmetry into the Cooley–Tukey algorithm is not a trivial matter.

Let $g \in G$ act on I through

$$g: \quad \mathbf{m} \longmapsto S_g(\mathbf{m}) = \mathbf{R}_g\mathbf{m} + \mathbf{N}\mathbf{t}_g \mod \mathbf{N}\mathbb{Z}^3$$

and suppose that $\mathbf{N}$ 'integerizes' all the non-primitive translations $\mathbf{t}_g$ so that we may write

$$\mathbf{N}\mathbf{t}_g = \mathbf{t}_g^{(1)} + \mathbf{N}_1\mathbf{t}_g^{(2)},$$

with $\mathbf{t}_g^{(1)} \in I_1$ and $\mathbf{t}_g^{(2)} \in I_2$ determined as above. Suppose further that $\mathbf{N}$, $\mathbf{N}_1$ and $\mathbf{N}_2$ commute with $\mathbf{R}_g$ for all $g \in G$, *i.e.* (by Schur's lemma, Section 1.3.4.2.2.4) that these matrices are integer multiples of the identity in each G-invariant subspace. The action of g on $\mathbf{m} = \mathbf{N}\mathbf{x} \mod \mathbf{N}\mathbb{Z}^3$ leads to

$$\begin{aligned} S_g(\mathbf{m}) &= \mathbf{N}[\mathbf{R}_g(\mathbf{N}^{-1}\mathbf{m}) + \mathbf{N}\mathbf{t}_g] &&\mod \mathbf{N}\mathbb{Z}^3 \\ &= \mathbf{N}\mathbf{R}_g\mathbf{N}^{-1}(\mathbf{m}_1 + \mathbf{N}_1\mathbf{m}_2) + \mathbf{t}_g^{(1)} + \mathbf{N}_1\mathbf{t}_g^{(2)} &&\mod \mathbf{N}\mathbb{Z}^3 \\ &= \mathbf{R}_g\mathbf{m}_1 + \mathbf{t}_g^{(1)} + \mathbf{N}_1(\mathbf{R}_g\mathbf{m}_2 + \mathbf{t}_g^{(2)}) &&\mod \mathbf{N}\mathbb{Z}^3, \end{aligned}$$

which we may decompose as

$$S_g(\mathbf{m}) = [S_g(\mathbf{m})]_1 + \mathbf{N}_1[S_g(\mathbf{m})]_2$$

with

$$[S_g(\mathbf{m})]_1 \equiv S_g(\mathbf{m}) \mod \mathbf{N}_1\mathbb{Z}^3$$

and

$$[S_g(\mathbf{m})]_2 \equiv \mathbf{N}_1^{-1}\{S_g(\mathbf{m}) - [S_g(\mathbf{m})]_1\} \mod \mathbf{N}_2\mathbb{Z}^3.$$

Introducing the notation

$$\begin{aligned} S_g^{(1)}(\mathbf{m}_1) &= \mathbf{R}_g\mathbf{m}_1 + \mathbf{t}_g^{(1)} \mod \mathbf{N}_1\mathbb{Z}^3, \\ S_g^{(2)}(\mathbf{m}_2) &= \mathbf{R}_g\mathbf{m}_2 + \mathbf{t}_g^{(2)} \mod \mathbf{N}_2\mathbb{Z}^3, \end{aligned}$$

the two components of $S_g(\mathbf{m})$ may be written

$$\begin{aligned} [S_g(\mathbf{m})]_1 &= S_g^{(1)}(\mathbf{m}_1), \\ [S_g(\mathbf{m})]_2 &= S_g^{(2)}(\mathbf{m}_2) + \boldsymbol{\mu}_2(g, \mathbf{m}_1) \mod \mathbf{N}_2\mathbb{Z}^3, \end{aligned}$$

with

$$\boldsymbol{\mu}_2(g, \mathbf{m}_1) = \mathbf{N}_1^{-1}\{(\mathbf{R}_g\mathbf{m}_1 + \mathbf{t}_g^{(1)}) - [S_g(\mathbf{m}_1)]_1\} \mod \mathbf{N}_2\mathbb{Z}^3.$$

The term $\boldsymbol{\mu}_2$ is the geometric equivalent of a *carry* or *borrow*: it arises because $\mathbf{R}_g\mathbf{m}_1 + \mathbf{t}_g^{(1)}$, calculated as a vector in $\mathbb{Z}^3/\mathbf{N}\mathbb{Z}^3$, may be outside the unit cell $\mathbf{N}_1[0, 1]^3$, and may need to be brought back into it by a 'large' translation with a non-zero component in the $\mathbf{m}_2$ space; equivalently, the action of g may need to be applied around different permissible origins for different values of $\mathbf{m}_1$, so as to map the unit cell into itself without any recourse to lattice translations. [Readers familiar with the cohomology of groups (see *e.g.* Hall, 1959; MacLane, 1963) will recognize $\boldsymbol{\mu}_2$ as the cocycle of the extension of $\mathbb{Z}G$-modules described by the exact sequence $0 \to I_2 \to I \to I_1 \to 0$.]

Thus G acts on I in a rather complicated fashion: although $g \longmapsto S_g^{(1)}$ does define a left action in I_1 alone, no action can be defined in I_2 alone because $\boldsymbol{\mu}_2$ depends on $\mathbf{m}_1$. However, because S_g, $S_g^{(1)}$ and $S_g^{(2)}$ *are* left actions, it follows that $\boldsymbol{\mu}_2$ satisfies the identity

$$\boldsymbol{\mu}_2(gg', \mathbf{m}_1) = S_g^{(2)}[\boldsymbol{\mu}_2(g', \mathbf{m}_1)] + \boldsymbol{\mu}_2[g, S_g^{(1)}(\mathbf{m}_1)] \qquad \text{mod } \mathbf{N}_2\mathbb{Z}^3$$

for all g, g' in G and all $\mathbf{m}_1$ in I_1. In particular, $\boldsymbol{\mu}_2(\mathrm{e}, \mathbf{m}_1) = \mathbf{0}$ for all $\mathbf{m}_1$, and

$$\boldsymbol{\mu}_2(g^{-1}, \mathbf{m}_1) = -S_{g^{-1}}^{(2)}\{\boldsymbol{\mu}_2[g, S_{g^{-1}}^{(1)}(\mathbf{m}_1)]\} \text{ mod } \mathbf{N}_2\mathbb{Z}^3.$$

This action will now be used to achieve optimal use of symmetry in the multidimensional Cooley–Tukey algorithm of Section 1.3.3.3.2.1. Let us form an array Y according to

$$Y(\mathbf{m}_1, \mathbf{m}_2) = \rho(\mathbf{m}_1 + \mathbf{N}_1\mathbf{m}_2)$$

for *all* $\mathbf{m}_2 \in I_2$ but only for the *unique* $\mathbf{m}_1$ under the action $S_g^{(1)}$ of G in I_1. Except in special cases which will be examined later, these vectors contain essentially an asymmetric unit of electron-density data, up to some redundancies on boundaries. We may then compute the partial transform on $\mathbf{m}_2$:

$$Y^*(\mathbf{m}_1, \mathbf{h}_2) = \frac{1}{|\det \mathbf{N}_2|} \sum_{\mathbf{m}_2 \in I_2} Y(\mathbf{m}_1, \mathbf{m}_2) e[\mathbf{h}_2 \cdot (\mathbf{N}_2^{-1}\mathbf{m}_2)].$$

Using the symmetry of ρ in the form $\rho = S_g^{\#}\rho$ yields by the procedure of Section 1.3.3.3.2 the transposition formula

$$Y^*(S_g^{(1)}(\mathbf{m}_1), \mathbf{h}_2) = e\{\mathbf{h}_2 \cdot [\mathbf{N}_2^{-1}(\mathbf{t}_g^{(2)} + \boldsymbol{\mu}_2(g, \mathbf{m}_1))]\} \times Y^*(\mathbf{m}_1, [\mathbf{R}_g^{(2)}]^T\mathbf{h}_2).$$

By means of this identity we can transpose intermediate results Y^* initially indexed by

$$(\text{unique } \mathbf{m}_1) \times (\text{all } \mathbf{h}_2),$$

so as to have them indexed by

$$(\text{all } \mathbf{m}_1) \times (\text{unique } \mathbf{h}_2).$$

We may then apply twiddle factors to get

$$Z(\mathbf{m}_1, \mathbf{h}_2) = e[\mathbf{h}_2 \cdot (\mathbf{N}^{-1}\mathbf{m}_1)] Y^*(\mathbf{m}_1, \mathbf{h}_2)$$

and carry out the second transform

$$Z^*(\mathbf{h}_1, \mathbf{h}_2) = \frac{1}{|\det \mathbf{N}_1|} \sum_{\mathbf{m}_1 \in I_1} Z(\mathbf{m}_1, \mathbf{h}_2) e[\mathbf{h}_1 \cdot (\mathbf{N}_1^{-1}\mathbf{m}_1)].$$

The final results are indexed by

$$(\text{all } \mathbf{h}_1) \times (\text{unique } \mathbf{h}_2),$$

which yield essentially an asymmetric unit of structure factors after unscrambling by:

$$F(\mathbf{h}_2 + \mathbf{N}_2^T\mathbf{h}_1) = Z^*(\mathbf{h}_1, \mathbf{h}_2).$$

The transposition formula above applies to intermediate results when going backwards from F to ρ, provided these results are considered *after* the twiddle-factor stage. A transposition formula applicable *before* that stage can be obtained by characterizing the action of G on $\mathbf{h}$ (including the effects of periodization by $\mathbf{N}^T\mathbb{Z}^3$) in a manner similar to that used for $\mathbf{m}$.

Let

$$\mathbf{h} = \mathbf{h}_2 + \mathbf{N}_2^T\mathbf{h}_1,$$

with

$$\mathbf{h}_2 = \mathbf{h} \qquad \text{mod } \mathbf{N}_2^T\mathbb{Z}^3,$$
$$\mathbf{h}_1 = (\mathbf{N}_2^{-1})^T(\mathbf{h} - \mathbf{h}_2) \quad \text{mod } \mathbf{N}_1^T\mathbb{Z}^3.$$

We may then write

$$\mathbf{R}_g^T\mathbf{h} = [\mathbf{R}_g^T\mathbf{h}]_2 + \mathbf{N}_2^T[\mathbf{R}_g^T\mathbf{h}]_1,$$

with

$$[\mathbf{R}_g^T\mathbf{h}]_2 = [\mathbf{R}_g^{(2)}]^T\mathbf{h}_2 \qquad \text{mod } \mathbf{N}_2^T\mathbb{Z}^3,$$
$$[\mathbf{R}_g^T\mathbf{h}]_1 = [\mathbf{R}_g^{(1)}]^T\mathbf{h}_1 + \boldsymbol{\eta}_1(g, \mathbf{h}_2) \quad \text{mod } \mathbf{N}_1^T\mathbb{Z}^3.$$

Here $[\mathbf{R}_g^{(2)}]^T$, $[\mathbf{R}_g^{(1)}]^T$ and $\boldsymbol{\eta}_1$ are defined by

$$[\mathbf{R}_g^{(2)}]^T\mathbf{h}_2 = \mathbf{R}_g^T\mathbf{h} \qquad \text{mod } \mathbf{N}_2^T\mathbb{Z}^3,$$
$$[\mathbf{R}_g^{(1)}]^T\mathbf{h}_1 = \mathbf{R}_g^T\mathbf{h} \qquad \text{mod } \mathbf{N}_1^T\mathbb{Z}^3$$

and

$$\boldsymbol{\eta}_1(g, \mathbf{h}_2) = (\mathbf{N}_2^{-1})^T(\mathbf{R}_g^T\mathbf{h}_2 - [\mathbf{R}_g^{(2)}]^T\mathbf{h}_2) \text{ mod } \mathbf{N}_1^T\mathbb{Z}^3.$$

Let us then form an array Z^* according to

$$Z^*(\mathbf{h}_1', \mathbf{h}_2') = F(\mathbf{h}_2' + \mathbf{N}_2^T\mathbf{h}_1')$$

for *all* $\mathbf{h}_1'$ but only for the *unique* $\mathbf{h}_2'$ under the action of G in $\mathbb{Z}^3/\mathbf{N}_2^T\mathbb{Z}^3$, and transform on $\mathbf{h}_1'$ to obtain

$$Z(\mathbf{m}_1, \mathbf{h}_2) = \sum_{\mathbf{h}_1' \in \mathbb{Z}^3/\mathbf{N}_1^T\mathbb{Z}^3} Z^*(\mathbf{h}_1', \mathbf{h}_2') e[-\mathbf{h}_1' \cdot (\mathbf{N}_1^{-1}\mathbf{m}_1)].$$

Putting $\mathbf{h}' = \mathbf{R}_g^T\mathbf{h}$ and using the symmetry of F in the form

$$F(\mathbf{h}') = F(\mathbf{h}) \exp(-2\pi i \mathbf{h} \cdot \mathbf{t}_g),$$

where

$$\mathbf{h} \cdot \mathbf{t}_g = (\mathbf{h}_2^T + \mathbf{h}_1^T\mathbf{N}_2)(\mathbf{N}_2^{-1}\mathbf{N}_1^{-1})(\mathbf{t}_g^{(1)} + \mathbf{N}_1\mathbf{t}_g^{(2)})$$
$$\equiv \mathbf{h}_2 \cdot \mathbf{t}_g + \mathbf{h}_2 \cdot (\mathbf{N}_1^{-1}\mathbf{t}_g^{(1)}) \text{ mod } 1$$

yields by a straightforward rearrangement

$$Z(\mathbf{m}_1, [\mathbf{R}_g^{(2)}]^T\mathbf{h}_2) = e[-\{\mathbf{h}_2 \cdot \mathbf{t}_g + \boldsymbol{\eta}_1(g, \mathbf{h}_2) \cdot (\mathbf{N}_1^{-1}\mathbf{m}_1)\}] \times Z\{S_g^{(1)}(\mathbf{m}_1), \mathbf{h}_2\}.$$

This formula allows the transposition of intermediate results Z from an indexing by

$$(\text{all } \mathbf{m}_1) \times (\text{unique } \mathbf{h}_2)$$

to an indexing by

$$(\text{unique } \mathbf{m}_1) \times (\text{all } \mathbf{h}_2).$$

We may then apply the twiddle factors to obtain

$$Y^*(\mathbf{m}_1, \mathbf{h}_2) = e[-\mathbf{h}_2 \cdot (\mathbf{N}^{-1}\mathbf{m}_1)] Z(\mathbf{m}_1, \mathbf{h}_2)$$

and carry out the second transform on $\mathbf{h}_2$

$$Y(\mathbf{m}_1, \mathbf{m}_2) = \sum_{\mathbf{h}_2 \in \mathbb{Z}^3/\mathbf{N}_2^T\mathbb{Z}^3} Y^*(\mathbf{m}_1, \mathbf{h}_2) e[-\mathbf{h}_2 \cdot (\mathbf{N}_2^{-1}\mathbf{m}_2)].$$

The results, indexed by

$$(\text{unique } \mathbf{m}_1) \times (\text{all } \mathbf{m}_2)$$

yield essentially an asymmetric unit of electron densities by the rearrangement

$$\rho(\mathbf{m}_1 + \mathbf{N}_1\mathbf{m}_2) = Y(\mathbf{m}_1, \mathbf{m}_2).$$

The equivalence of the two transposition formulae up to the intervening twiddle factors is readily established, using the relation

$$\mathbf{h}_2 \cdot [\mathbf{N}_2^{-1}\boldsymbol{\mu}_2(g,\mathbf{m}_1)] = \boldsymbol{\eta}_1(g,\mathbf{h}_2)\cdot(\mathbf{N}_1^{-1}\mathbf{m}_1) \bmod 1$$

which is itself a straightforward consequence of the identity

$$\mathbf{h}\cdot[\mathbf{N}^{-1}S_g(\mathbf{m})] = \mathbf{h}\cdot\mathbf{t}_g + (\mathbf{R}_g^T\mathbf{h})\cdot(\mathbf{N}^{-1}\mathbf{m}).$$

To complete the characterization of the effect of symmetry on the Cooley–Tukey factorization, and of the economy of computation it allows, it remains to consider the possibility that some values of $\mathbf{m}_1$ may be invariant under some transformations $g \in G$ under the action $\mathbf{m}_1 \longmapsto S_g^{(1)}(\mathbf{m}_1)$.

Suppose that $\mathbf{m}_1$ has a non-trivial isotropy subgroup $G_{\mathbf{m}_1}$, and let $g \in G_{\mathbf{m}_1}$. Then each subarray $Y_{\mathbf{m}_1}$ defined by

$$Y_{\mathbf{m}_1}(\mathbf{m}_2) = Y(\mathbf{m}_1,\mathbf{m}_2) = \rho(\mathbf{m}_1 + \mathbf{N}_1\mathbf{m}_2)$$

satisfies the identity

$$\begin{aligned} Y_{\mathbf{m}_1}(\mathbf{m}_2) &= Y_{S_g^{(1)}(\mathbf{m}_1)}[S_g^{(2)}(\mathbf{m}_2) + \boldsymbol{\mu}_2(g,\mathbf{m}_1)] \\ &= Y_{\mathbf{m}_1}[S_g^{(2)}(\mathbf{m}_2) + \boldsymbol{\mu}_2(g,\mathbf{m}_1)] \end{aligned}$$

so that the data for the transform on $\mathbf{m}_2$ have residual symmetry properties. In this case the identity satisfied by $\boldsymbol{\mu}_2$ simplifies to

$$\boldsymbol{\mu}_2(gg',\mathbf{m}_1) = S_g^{(2)}[\boldsymbol{\mu}_2(g',\mathbf{m}_1)] + \boldsymbol{\mu}_2(g,\mathbf{m}_1) \bmod \mathbf{N}_2\mathbb{Z}^3,$$

which shows that the mapping $g \longmapsto \boldsymbol{\mu}_2(g,\mathbf{m}_1)$ satisfies the Frobenius congruences (Section 1.3.4.2.2.3). Thus the internal symmetry of subarray $Y_{\mathbf{m}_1}$ with respect to the action of G on $\mathbf{m}_2$ is given by $G_{\mathbf{m}_1}$ acting on $\mathbb{Z}^3/\mathbf{N}_2\mathbb{Z}^3$ *via*

$$\mathbf{m}_2 \longmapsto S_g^{(2)}(\mathbf{m}_2) + \boldsymbol{\mu}_2(g,\mathbf{m}_1) \bmod \mathbf{N}_2\mathbb{Z}^3.$$

The transform on $\mathbf{m}_2$ needs only be performed for one out of $[G : G_{\mathbf{m}_1}]$ distinct arrays $Y_{\mathbf{m}_1}$ (results for the others being obtainable by the transposition formula), and this transforms is $G_{\mathbf{m}_1}$-symmetric. In other words, the following cases occur:

(i)	$G_{\mathbf{m}_1} = \{e\}$	maximum saving in computation (by $\lvert G\rvert$); $\mathbf{m}_2$-transform has no symmetry.
(ii)	$G_{\mathbf{m}_1} = G' < G$	saving in computation by a factor of $[G : G']$; $\mathbf{m}_2$-transform is G'-symmetric.
(iii)	$G_{\mathbf{m}_1} = G$	no saving in computation; $\mathbf{m}_2$-transform is G-symmetric.

The symmetry properties of the $\mathbf{m}_2$-transform may themselves be exploited in a similar way if $\mathbf{N}_2$ can be factored as a product of smaller decimation matrices; otherwise, an appropriate symmetrized DFT routine may be provided, using for instance the idea of 'multiplexing/demultiplexing' (Section 1.3.4.3.5). We thus have a recursive *descent procedure*, in which the deeper stages of the recursion deal with transforms on *fewer points*, or of *lower symmetry* (usually both).

The same analysis applies to the $\mathbf{h}_1$-transforms on the subarrays $Z^*_{\mathbf{h}_2}$, and leads to a similar descent procedure.

In conclusion, crystallographic symmetry can be fully exploited to reduce the amount of computation to the minimum required to obtain the unique results from the unique data. No such analysis was so far available in cases where the asymmetric units in real and reciprocal space are not parallelepipeds. An example of this procedure will be given in Section 1.3.4.3.6.5.

1.3.4.3.4.2. *Multidimensional Good factorization*

This procedure was described in Section 1.3.3.3.2.2. The main difference with the Cooley–Tukey factorization is that if $\mathbf{N} = \mathbf{N}_1\mathbf{N}_2\ldots\mathbf{N}_{d-1}\mathbf{N}_d$, where the different factors are pairwise coprime, then the Chinese remainder theorem reindexing makes $\mathbb{Z}^3/\mathbf{N}\mathbb{Z}^3$ isomorphic to a direct sum.

$$\mathbb{Z}^3/\mathbf{N}\mathbb{Z}^3 \cong (\mathbb{Z}^3/\mathbf{N}_1\mathbb{Z}^3)\oplus\ldots\oplus(\mathbb{Z}^3/\mathbf{N}_d\mathbb{Z}^3),$$

where each p-primary piece is endowed with an induced $\mathbb{Z}G$-module structure by letting G operate in the usual way but with the corresponding modular arithmetic. The situation is thus more favourable than with the Cooley–Tukey method, since there is no interference between the factors (no 'carry'). In the terminology of Section 1.3.4.2.2.2, G acts *diagonally* on this direct sum, and results of a partial transform may be transposed by orbit exchange as in Section 1.3.4.3.4.1 but without the extra terms $\boldsymbol{\mu}$ or $\boldsymbol{\eta}$. The analysis of the symmetry properties of partial transforms also carries over, again without the extra terms. Further simplification occurs for all p-primary pieces with p other than 2 or 3, since all non-primitive translations (including those associated to lattice centring) disappear modulo p.

Thus the cost of the CRT reindexing is compensated by the computational savings due to the absence of twiddle factors and of other phase shifts associated with non-primitive translations and with geometric 'carries'.

Within each p-primary piece, however, higher powers of p may need to be split up by a Cooley–Tukey factorization, or carried out directly by a suitably adapted Winograd algorithm.

1.3.4.3.4.3. *Crystallographic extension of the Rader/Winograd factorization*

As was the case in the absence of symmetry, the two previous classes of algorithms can only factor the global transform into partial transforms on prime numbers of points, but cannot break the latter down any further. Rader's idea of using the action of the group of units $U(p)$ to obtain further factorization of a p-primary transform has been used in 'scalar' form by Auslander & Shenefelt (1987), Shenefelt (1988), and Auslander *et al.* (1988). It will be shown here that it can be adapted to the crystallographic case so as to take advantage also of the possible existence of n-fold cyclic symmetry elements ($n = 3, 4, 6$) in a two-dimensional transform (Bricogne & Tolimieri, 1990). This adaptation entails the use of certain rings of *algebraic* integers rather than ordinary integers, whose connection with the handling of cyclic symmetry will now be examined.

Let G be the group associated with a threefold axis of symmetry: $G = \{e, g, g^2\}$ with $g^3 = e$. In a standard trigonal basis, G has matrix representation

$$\mathbf{R}_e = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} = \mathbf{I}, \quad \mathbf{R}_g = \begin{pmatrix} 0 & -1 \\ 1 & -1 \end{pmatrix}, \quad \mathbf{R}_{g^2} = \begin{pmatrix} -1 & 1 \\ -1 & 0 \end{pmatrix}$$

in real space,

$$\mathbf{R}^*_e = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} = \mathbf{I}, \quad \mathbf{R}^*_g = \begin{pmatrix} -1 & -1 \\ 1 & 0 \end{pmatrix}, \quad \mathbf{R}^*_{g^2} = \begin{pmatrix} 0 & 1 \\ -1 & -1 \end{pmatrix}$$

in reciprocal space. Note that

$$\mathbf{R}^*_{g^2} = [\mathbf{R}_{g^2}^{-1}]^T = \mathbf{R}_g^T,$$

and that

$$\mathbf{R}_g^T = \mathbf{J}^{-1}\mathbf{R}_g\mathbf{J}, \quad \text{where } \mathbf{J} = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

so that $\mathbf{R}_g$ and $\mathbf{R}_g^T$ are conjugate in the group of 2×2 unimodular

integer matrices. The group ring $\mathbb{Z}G$ is commutative, and has the structure of the polynomial ring $\mathbb{Z}[X]$ with the single relation $X^2 + X + 1 = 0$ corresponding to the minimal polynomial of $\mathbf{R}_g$. In the terminology of Section 1.3.3.2.4, the ring structure of $\mathbb{Z}G$ is obtained from that of $\mathbb{Z}[X]$ by carrying out polynomial addition and multiplication modulo $X^2 + X + 1$, then replacing X by any generator of G. This type of construction forms the very basis of algebraic number theory [see Artin (1944, Section IIc) for an illustration of this viewpoint], and $\mathbb{Z}G$ as just defined is isomorphic to the ring $\mathbb{Z}[\omega]$ of algebraic integers of the form $a + b\omega$ [$a, b \in \mathbb{Z}, \omega = \exp(2\pi i/3)$] under the identification $X \leftrightarrow \omega$. Addition in this ring is defined component-wise, while multiplication is defined by

$$(a_1 + b_1\omega) \times (a_2 + b_2\omega) = (a_1a_2 - b_1b_2) + [(a_1 - b_1)b_2 + b_1a_2]\omega.$$

In the case of a fourfold axis, $G = \{e, g, g^2, g^3\}$ with $g^4 = e$, and

$$\mathbf{R}_g = \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix} = \mathbf{R}_g^*, \quad \text{with again } \mathbf{R}_g^T = \mathbf{J}^{-1}\mathbf{R}_g\mathbf{J}.$$

$\mathbb{Z}G$ is obtained from $\mathbb{Z}[X]$ by carrying out polynomial arithmetic modulo $X^2 + 1$. This identifies $\mathbb{Z}G$ with the ring $\mathbb{Z}[i]$ of Gaussian integers of the form $a + bi$, in which addition takes place component-wise while multiplication is defined by

$$(a_1 + b_1 i) \times (a_2 + b_2 i) = (a_1a_2 - b_1b_2) + (a_1b_2 + b_1a_2)i.$$

In the case of a sixfold axis, $G = \{e, g, g^2, g^3, g^4, g^5\}$ with $g^6 = e$, and

$$\mathbf{R}_g = \begin{pmatrix} 1 & -1 \\ 1 & 0 \end{pmatrix}, \quad \mathbf{R}_g^* = \begin{pmatrix} 0 & -1 \\ 1 & 1 \end{pmatrix}, \quad \mathbf{R}_g^T = \mathbf{J}^{-1}\mathbf{R}_g\mathbf{J}.$$

$\mathbb{Z}G$ is isomorphic to $\mathbb{Z}[\omega]$ under the mapping $g \leftrightarrow 1 + \omega$ since $(1 + \omega)^6 = 1$.

Thus in all cases $\mathbb{Z}G \cong \mathbb{Z}[X]/P(X)$ where $P(X)$ is an irreducible quadratic polynomial with integer coefficients.

The actions of G on lattices in real and reciprocal space (Sections 1.3.4.2.2.4, 1.3.4.2.2.5) extend naturally to actions of $\mathbb{Z}G$ on $\mathbb{Z}^2$ in which an element $z = a + bg$ of $\mathbb{Z}G$ acts *via*

$$\mathbf{m} = \begin{pmatrix} m_1 \\ m_2 \end{pmatrix} \longmapsto z\mathbf{m} = (a\mathbf{I} + b\mathbf{R}_g)\begin{pmatrix} \mathbf{m}_1 \\ \mathbf{m}_2 \end{pmatrix}$$

in real space, and *via*

$$\mathbf{h} = \begin{pmatrix} h_1 \\ h_2 \end{pmatrix} \longmapsto z\mathbf{h} = (a\mathbf{I} + b\mathbf{R}_g^T)\begin{pmatrix} h_1 \\ h_2 \end{pmatrix}$$

in reciprocal space. These two actions are related by conjugation, since

$$(a\mathbf{I} + b\mathbf{R}_g^T) = \mathbf{J}^{-1}(a\mathbf{I} + b\mathbf{R}_g)\mathbf{J}$$

and the following identity (which is fundamental in the sequel) holds:

$$(z\mathbf{h}) \cdot \mathbf{m} = \mathbf{h} \cdot (z\mathbf{m}) \quad \text{for all } \mathbf{m}, \mathbf{h} \in \mathbb{Z}^2.$$

Let us now consider the calculation of a $p \times p$ two-dimensional DFT with n-fold cyclic symmetry ($n = 3, 4, 6$) for an odd prime $p \geq 5$. Denote $\mathbb{Z}/p\mathbb{Z}$ by $\mathbb{Z}_p$. Both the data and the results of the DFT are indexed by $\mathbb{Z}_p \times \mathbb{Z}_p$: hence the action of $\mathbb{Z}G$ on these indices is in fact an action of $\mathbb{Z}_pG$, the latter being obtained from $\mathbb{Z}G$ by carrying out all integer arithmetic in $\mathbb{Z}G$ modulo p. The algebraic structure of $\mathbb{Z}_pG$ combines the symmetry-carrying ring structure of $\mathbb{Z}G$ with the finite field structure of $\mathbb{Z}_p$ used in Section 1.3.3.2.3.1, and holds the key to a symmetry-adapted factorization of the DFT at hand.

The structure of $\mathbb{Z}_pG$ depends on whether $P(X)$ remains irreducible when considered as a polynomial over $\mathbb{Z}_p$. Thus two cases arise:

(1) $P(X)$ remains irreducible mod p, *i.e.* there is no nth root of unity in $\mathbb{Z}_p$;

(2) $P(X)$ factors as $(X - u)(X - v)$, *i.e.* there are nth roots of unity in $\mathbb{Z}_p$.

These two cases require different developments.

Case 1. $\mathbb{Z}_pG$ is a finite field with p^2 elements. There is essentially (*i.e.* up to isomorphism) only one such field, denoted $GF(p^2)$, and its group of units is a cyclic group with $p^2 - 1$ elements. If γ is a generator of this group of units, the input data $\rho_{\mathbf{m}}$ with $\mathbf{m} \neq \mathbf{0}$ may be reordered as

$$\mathbf{m}_0, \gamma\mathbf{m}_0, \gamma^2\mathbf{m}_0, \gamma^3\mathbf{m}_0, \ldots, \gamma^{p^2-2}\mathbf{m}_0$$

by the *real-space action* of γ; while the results $F_{\mathbf{h}}$ with $\mathbf{h} \neq \mathbf{0}$ may be reordered as

$$\mathbf{h}_0, \gamma\mathbf{h}_0, \gamma^2\mathbf{h}_0, \gamma^3\mathbf{h}_0, \ldots, \gamma^{p^2-2}\mathbf{h}_0$$

by the *reciprocal-space action* of γ, where $\mathbf{m}_0$ and $\mathbf{h}_0$ are arbitrary non-zero indices.

The core $\mathbf{C}_{p\times p}$ of the DFT matrix, defined by

$$\mathbf{F}_{p\times p} = \begin{pmatrix} 1 & 1 & \ldots & 1 \\ 1 & & & \\ \vdots & & \mathbf{C}_{p\times p} & \\ 1 & & & \end{pmatrix},$$

will then have a skew-circulant structure (Section 1.3.3.2.3.1) since

$$(\mathbf{C}_{p\times p})_{jk} = e\left[\frac{(\gamma^j\mathbf{h}_0)\cdot(\gamma^k\mathbf{m}_0)}{p}\right] = e\left[\frac{\mathbf{h}_0\cdot(\gamma^{j+k}\mathbf{m}_0)}{p}\right]$$

depends only on $j + k$. Multiplication by $\mathbf{C}_{p\times p}$ may then be turned into a cyclic convolution of length $p^2 - 1$, which may be factored by two DFTs (Section 1.3.3.2.3.1) or by Winograd's techniques (Section 1.3.3.2.4). The latter factorization is always favourable, as it is easily shown that $p^2 - 1$ is divisible by 24 for any odd prime $p \geq 5$. This procedure is applicable even if no symmetry is present in the data.

Assume now that cyclic symmetry of order $n = 3$, 4 or 6 *is* present. Since n divides 24 hence divides $p^2 - 1$, the generator g of this symmetry is representable as $\gamma^{(p^2-1)/n}$ for a suitable generator γ of the group of units. The reordered data will then be $(p^2 - 1)/n$-periodic rather than simply $(p^2 - 1)$-periodic; hence the reindexed results will be *n-decimated* (Section 1.3.2.7.2), and the $(p^2 - 1)/n$ non-zero results can be calculated by applying the DFT to the $(p^2 - 1)/n$ unique input data. In this way, the n-fold symmetry can be used in full to calculate the core contributions from the unique data to the unique results by a DFT of length $(p^2 - 1)/n$.

It is a simple matter to incorporate non-primitive translations into this scheme. For example, when going from structure factors to electron densities, reordered data items separated by $(p^2 - 1)/n$ are not equal but differ by a phase shift proportional to their index mod p, whose effect is simply to shift the origin of the n-decimated transformed sequence. The same economy of computation can therefore be achieved as in the purely cyclic case.

Dihedral symmetry elements, which map g to g^{-1} (Section 1.3.4.2.2.3), induce extra one-dimensional symmetries of order 2 in the reordered data which can also be fully exploited to reduce computation.

Case 2. If $p \geq 5$, it can be shown that the two roots u and v are always distinct. Then, by the Chinese remainder theorem (CRT) for polynomials (Section 1.3.3.2.4) we have a ring isomorphism

$$\mathbb{Z}_p[X]/P(X) \cong \{\mathbb{Z}_p[X]/(X - u)\} \times \{\mathbb{Z}_p[X]/(X - v)\}$$

defined by sending a polynomial $Q(X)$ from the left-hand-side ring to its two residue classes modulo $X-u$ and $X-v$, respectively. Since the latter are simply the constants $Q(u)$ and $Q(v)$, the CRT reindexing has the particularly simple form

$$a+bX \longmapsto (a+bu, a+bv) = (\alpha, \beta)$$

or equivalently

$$\begin{pmatrix} a \\ b \end{pmatrix} \longmapsto \begin{pmatrix} \alpha \\ \beta \end{pmatrix} = \mathbf{M}\begin{pmatrix} a \\ b \end{pmatrix} \bmod p, \quad \text{with } \mathbf{M} = \begin{pmatrix} 1 & u \\ 1 & v \end{pmatrix}.$$

The CRT reconstruction formula similarly simplifies to

$$\begin{pmatrix} \alpha \\ \beta \end{pmatrix} \longmapsto \begin{pmatrix} a \\ b \end{pmatrix} = \mathbf{M}^{-1}\begin{pmatrix} \alpha \\ \beta \end{pmatrix} \bmod p,$$
$$\text{with } \mathbf{M}^{-1} = \frac{1}{v-u}\begin{pmatrix} v & -u \\ -1 & 1 \end{pmatrix}.$$

The use of the CRT therefore amounts to the *simultaneous diagonalization* (by $\mathbf{M}$) of all the matrices representing the elements of $\mathbb{Z}_pG$ in the basis $(1, X)$.

A first consequence of this diagonalization is that the internal structure of $\mathbb{Z}_pG$ becomes clearly visible. Indeed, $\mathbb{Z}_pG$ is mapped isomorphically to a direct product of two copies of $\mathbb{Z}_p$, in which arithmetic is carried out *component-wise* between eigenvalues α and β. Thus if

$$z = a + bX \overset{\text{CRT}}{\longleftrightarrow} (\alpha, \beta),$$
$$z' = a' + b'X \overset{\text{CRT}}{\longleftrightarrow} (\alpha', \beta'),$$

then

$$z + z' \overset{\text{CRT}}{\longleftrightarrow} (\alpha+\alpha', \beta+\beta'),$$
$$zz' \overset{\text{CRT}}{\longleftrightarrow} (\alpha\alpha', \beta\beta').$$

Taking in particular

$$z \overset{\text{CRT}}{\longleftrightarrow} (\alpha, 0) \neq (0,0),$$
$$z' \overset{\text{CRT}}{\longleftrightarrow} (0, \beta) \neq (0,0),$$

we have $zz' = 0$, so that $\mathbb{Z}_pG$ contains zero divisors; therefore $\mathbb{Z}_pG$ *is not a field.* On the other hand, if $z \overset{\text{CRT}}{\longleftrightarrow} (\alpha, \beta)$ with $\alpha \neq 0$ and $\beta \neq 0$, then α and β belong to the group of units $U(p)$ (Section 1.3.3.2.3.1) and hence have inverses α^{-1} and β^{-1}; it follows that z is a unit in $\mathbb{Z}_pG$, with inverse $z^{-1} \overset{\text{CRT}}{\longleftrightarrow} (\alpha^{-1}, \beta^{-1})$. Therefore, $\mathbb{Z}_pG$ consists of four distinct pieces:

$$0 \overset{\text{CRT}}{\longleftrightarrow} \{(0,0)\},$$
$$D_1 \overset{\text{CRT}}{\longleftrightarrow} \{(\alpha, 0) | \alpha \in U(p)\} \cong U(p),$$
$$D_2 \overset{\text{CRT}}{\longleftrightarrow} \{(0, \beta) | \beta \in U(p)\} \cong U(p),$$
$$U \overset{\text{CRT}}{\longleftrightarrow} \{(\alpha, \beta) | \alpha \in U(p), \beta \in U(p)\} \cong U(p) \times U(p).$$

A second consequence of this diagonalization is that the actions of $\mathbb{Z}_pG$ on indices $\mathbf{m}$ and $\mathbf{h}$ can themselves be brought to diagonal form by basis changes:

$$\mathbf{m} \longmapsto (a\mathbf{I} + b\mathbf{R}_g)\mathbf{m}$$
$$\text{becomes } \boldsymbol{\mu} \longmapsto \begin{pmatrix} \alpha & 0 \\ 0 & \beta \end{pmatrix}\boldsymbol{\mu} \quad \text{with } \boldsymbol{\mu} = \mathbf{M}\mathbf{m},$$
$$\mathbf{h} \longmapsto (a\mathbf{I} + b\mathbf{R}_g^T)\mathbf{h}$$
$$\text{becomes } \boldsymbol{\eta} \longmapsto \begin{pmatrix} \alpha & 0 \\ 0 & \beta \end{pmatrix}\boldsymbol{\eta} \quad \text{with } \boldsymbol{\eta} = \mathbf{M}\mathbf{J}\mathbf{h}.$$

Thus the sets of indices $\boldsymbol{\mu}$ and $\boldsymbol{\eta}$ can be split into four pieces as $\mathbb{Z}_pG$ itself, according as these indices have none, one or two of their coordinates in $U(p)$. These pieces will be labelled by the same symbols – 0, D_1, D_2 and U – as those of $\mathbb{Z}_pG$.

The scalar product $\mathbf{h} \cdot \mathbf{m}$ may be written in terms of $\boldsymbol{\eta}$ and $\boldsymbol{\mu}$ as

$$\mathbf{h} \cdot \mathbf{m} = [\boldsymbol{\eta} \cdot ((\mathbf{M}^{-1})^T \mathbf{J} \mathbf{M}^{-1})\boldsymbol{\mu}],$$

and an elementary calculation shows that the matrix $= (\mathbf{M}^{-1})^T\mathbf{J}\mathbf{M}^{-1}$ is *diagonal* by virtue of the relation

$$uv = \text{constant term in } P(X) = 1.$$

Therefore, $\mathbf{h} \cdot \mathbf{m} = 0$ if $\mathbf{h} \in D_1$ and $\boldsymbol{\mu} \in D_2$ or *vice versa.*

We are now in a position to rearrange the DFT matrix $\mathbf{F}_{p\times p}$. Clearly, the structure of $\mathbf{F}_{p\times p}$ is more complex than in case 1, as there are three types of 'core' matrices:

type 1: $D \times D$ (with $D = D_1$ or D_2);
type 2: $D \times U$ or $U \times D$;
type 3: $U \times U$.

(Submatrices of type $D_1 \times D_2$ and $D_2 \times D_1$ have all their elements equal to 1 by the previous remark.)

Let γ be a generator of $U(p)$. We may reorder the elements in D_1, D_2 and U – and hence the data and results indexed by these elements – according to powers of γ. This requires one exponent in each of D_1 and D_2, and two exponents in U. For instance, in the $\mathbf{h}$-index space:

$$D_1 = \left\{ \begin{pmatrix} \gamma & 0 \\ 0 & 0 \end{pmatrix}^j \begin{pmatrix} \eta_1 \\ 0 \end{pmatrix}_0 \Big| \, j = 1, \ldots, p-1 \right\}$$
$$D_2 = \left\{ \begin{pmatrix} 0 & 0 \\ 0 & \gamma \end{pmatrix}^j \begin{pmatrix} 0 \\ \eta_2 \end{pmatrix}_0 \Big| \, j = 1, \ldots, p-1 \right\}$$
$$U = \left\{ \begin{pmatrix} \gamma & 0 \\ 0 & 1 \end{pmatrix}^{j_1} \begin{pmatrix} 1 & 0 \\ 0 & \gamma \end{pmatrix}^{j_2} \begin{pmatrix} \eta_1 \\ \eta_2 \end{pmatrix}_0 \Big| \, j_1 = 1, \ldots, p-1; \; j_2 = 1, \ldots, p-1 \right\}$$

and similarly for the $\boldsymbol{\mu}$ index.

Since the diagonal matrix $\boldsymbol{\Delta}$ commutes with all the matrices representing the action of γ, this rearrangement will induce skew-circulant structures in all the core matrices. The corresponding cyclic convolutions may be carried out by Rader's method, *i.e.* by diagonalizing them by means of two $(p-1)$-point one-dimensional DFTs in the $D \times D$ pieces and of two $(p-1) \times (p-1)$-point two-dimensional DFTs in the $U \times U$ piece (the $U \times D$ and $D \times U$ pieces involve extra section and projection operations).

In the absence of symmetry, no computational saving is achieved, since the same reordering could have been applied to the initial $\mathbb{Z}_p \times \mathbb{Z}_p$ indexing, without the CRT reindexing.

In the presence of n-fold cyclic symmetry, however, the rearranged $\mathbf{F}_{p\times p}$ lends itself to an n-fold reduction in size. The basic fact is that whenever case 2 occurs, $p-1$ is divisible by n (*i.e.* $p-1$ is divisible by 6 when $n = 3$ or 6, and by 4 when $n = 4$), say

$p-1=nq$. If g is a generator of the cyclic symmetry, the generator γ of $U(p)$ may be chosen in such a way that $g=\gamma^q$. The action of g is then to increment the j index in D_1 and D_2 by q, and the (j_1,j_2) index in U by (q,q). Since the data items whose indices are related in this way have identical values, the DFTs used to diagonalize the Rader cyclic convolutions will operate on *periodized data*, hence yield *decimated results*; and the non-zero results will be obtained from the unique data by DFTs n times smaller than their counterparts in the absence of symmetry.

A more thorough analysis is needed to obtain a Winograd factorization into the normal from **CBA** in the presence of symmetry (see Bricogne & Tolimieri, 1990).

Non-primitive translations and dihedral symmetry may also be accommodated within this framework, as in case 1.

This reindexing by means of algebraic integers yields larger orbits, hence more efficient algorithms, than that of Auslander *et al.* (1988) which only uses ordinary integers acting by scalar dilation.

1.3.4.3.5. *Treatment of conjugate and parity-related symmetry properties*

Most crystallographic Fourier syntheses are real-valued and originate from Hermitian-symmetric collections of Fourier coefficients. Hermitian symmetry is closely related to the action of a centre of inversion in reciprocal space, and thus interacts strongly with all other genuinely crystallographic symmetry elements of order 2. All these symmetry properties are best treated by factoring by 2 and reducing the computation of the initial transform to that of a collection of smaller transforms with less symmetry or none at all.

1.3.4.3.5.1. *Hermitian-symmetric or real-valued transforms*

The computation of a DFT with Hermitian-symmetric or real-valued data can be carried out at a cost of half that of an ordinary transform, essentially by 'multiplexing' pairs of special partial transforms into general complex transforms, and then 'demultiplexing' the results on the basis of their symmetry properties. The treatment given below is for general dimension n; a subset of cases for $n=1$ was treated by Ten Eyck (1973).

(*a*) *Underlying group action*

Hermitian symmetry is not a geometric symmetry, but it is defined in terms of the action in reciprocal space of point group $G=\bar{1}$, *i.e.* $G=\{e,-e\}$, where e acts as $\mathbf{I}$ (the $n\times n$ identity matrix) and $-e$ acts as $-\mathbf{I}$.

This group action on $\mathbb{Z}^n/\mathbf{N}\mathbb{Z}^n$ with $\mathbf{N}=\mathbf{N}_1\mathbf{N}_2$ will now be characterized by the calculation of the cocycle $\boldsymbol{\eta}_1$ (Section 1.3.4.3.4.1) under the assumption that $\mathbf{N}_1$ and $\mathbf{N}_2$ are both *diagonal.* For this purpose it is convenient to associate to any integer vector $\mathbf{v}=\begin{pmatrix}v_1\\ \vdots\\ v_n\end{pmatrix}$ in $\mathbb{Z}^n$ the vector $\boldsymbol{\zeta}(\mathbf{v})$ whose jth component is

$$\begin{cases}0 \text{ if} & v_j=0\\ 1 \text{ if} & v_j\neq 0.\end{cases}$$

Let $\mathbf{m}=\mathbf{m}_1+\mathbf{N}_1\mathbf{m}_2$, and hence $\mathbf{h}=\mathbf{h}_2+\mathbf{N}_2\mathbf{h}_1$. Then

$$-\mathbf{h}_2 \bmod \mathbf{N}\mathbb{Z}^n=\mathbf{N}\boldsymbol{\zeta}(\mathbf{h}_2)-\mathbf{h}_2,$$
$$-\mathbf{h}_2 \bmod \mathbf{N}_2\mathbb{Z}^n=\mathbf{N}_2\boldsymbol{\zeta}(\mathbf{h}_2)-\mathbf{h}_2,$$

hence

$$\begin{aligned}\boldsymbol{\eta}_1(-e,\mathbf{h}_2)&=\mathbf{N}_2^{-1}\{[\mathbf{N}\boldsymbol{\zeta}(\mathbf{h}_2)-\mathbf{h}_2]-[\mathbf{N}_2\boldsymbol{\zeta}(\mathbf{h}_2)-\mathbf{h}_2]\} \bmod \mathbf{N}_1\mathbb{Z}^n\\ &=-\boldsymbol{\zeta}(\mathbf{h}_2) \bmod \mathbf{N}_1\mathbb{Z}^n.\end{aligned}$$

Therefore $-e$ acts by

$$(\mathbf{h}_2,\mathbf{h}_1)\longmapsto[\mathbf{N}_2\boldsymbol{\zeta}(\mathbf{h}_2)-\mathbf{h}_2,\mathbf{N}_1\boldsymbol{\zeta}(\mathbf{h}_1)-\mathbf{h}_1-\boldsymbol{\zeta}(\mathbf{h}_2)].$$

Hermitian symmetry is traditionally dealt with by factoring by 2, *i.e.* by assuming $\mathbf{N}=2\mathbf{M}$. If $\mathbf{N}_2=2\mathbf{I}$, then each $\mathbf{h}_2$ is invariant under G, so that each partial vector $\mathbf{Z}^*_{\mathbf{h}_2}$ (Section 1.3.4.3.4.1) inherits the symmetry internally, with a 'modulation' by $\boldsymbol{\eta}_1(g,\mathbf{h}_2)$. The 'multiplexing–demultiplexing' technique provides an efficient treatment of this singular case.

(*b*) *Calculation of structure factors*

The computation may be summarized as follows:

$$\rho \overset{\mathbf{dec}(\mathbf{N}_1)}{\longmapsto} \mathbf{Y} \overset{\bar{F}(\mathbf{N}_2)}{\longmapsto} \mathbf{Y}^* \overset{\text{TW}}{\longmapsto} \mathbf{Z} \overset{\bar{F}(\mathbf{N}_1)}{\longmapsto} \mathbf{Z}^* \overset{\mathbf{rev}(\mathbf{N}_2)}{\longmapsto} \mathbf{F}$$

where $\mathbf{dec}(\mathbf{N}_1)$ is the initial decimation given by $Y_{\mathbf{m}_1}(\mathbf{m}_2)=\rho(\mathbf{m}_1+\mathbf{N}_1\mathbf{m}_2)$, TW is the transposition and twiddle-factor stage, and $\mathbf{rev}(\mathbf{N}_2)$ is the final unscrambling by coset reversal given by $F(\mathbf{h}_2+\mathbf{N}_2\mathbf{h}_1)=\mathbf{Z}^*_{\mathbf{h}_2}(\mathbf{h}_1)$.

(i) *Decimation in time* $(\mathbf{N}_1=2\mathbf{I},\mathbf{N}_2=\mathbf{M})$

The decimated vectors $\mathbf{Y}_{\mathbf{m}_1}$ are real and hence have Hermitian transforms $\mathbf{Y}^*_{\mathbf{m}_1}$. The 2^n values of $\mathbf{m}_1$ may be grouped into 2^{n-1} pairs $(\mathbf{m}_1',\mathbf{m}_1'')$ and the vectors corresponding to each pair may be multiplexed into a general complex vector

$$\mathbf{Y}=\mathbf{Y}_{\mathbf{m}_1'}+i\mathbf{Y}_{\mathbf{m}_1''}.$$

The transform $\mathbf{Y}^*=\bar{F}(\mathbf{M})[\mathbf{Y}]$ can then be resolved into the separate transforms $\mathbf{Y}^*_{\mathbf{m}_1'}$ and $\mathbf{Y}^*_{\mathbf{m}_1''}$ by using the Hermitian symmetry of the latter, which yields the demultiplexing formulae

$$Y^*_{\mathbf{m}_1'}(\mathbf{h}_2)+iY^*_{\mathbf{m}_1''}(\mathbf{h}_2)=Y^*(\mathbf{h}_2)$$
$$\overline{Y^*_{\mathbf{m}_1'}(\mathbf{h}_2)}+i\overline{Y^*_{\mathbf{m}_1''}(\mathbf{h}_2)}=Y^*[\mathbf{M}\boldsymbol{\zeta}(\mathbf{h}_2)-\mathbf{h}_2].$$

The number of partial transforms $\bar{F}(\mathbf{M})$ is thus reduced from 2^n to 2^{n-1}. Once this separation has been achieved, the remaining steps need only be carried out for a unique half of the values of $\mathbf{h}_2$.

(ii) *Decimation in frequency* $(\mathbf{N}_1=\mathbf{M},\mathbf{N}_2=2\mathbf{I})$

Since $\mathbf{h}_2\in\mathbb{Z}^n/2\mathbb{Z}^n$ we have $-\mathbf{h}_2=\mathbf{h}_2$ and $\boldsymbol{\zeta}(\mathbf{h}_2)=\mathbf{h}_2 \bmod 2\mathbb{Z}^n$. The vectors of decimated and scrambled results $\mathbf{Z}^*_{\mathbf{h}_2}$ then obey the symmetry relations

$$Z^*_{\mathbf{h}_2}(\mathbf{h}_1-\mathbf{h}_2)=\overline{Z^*_{\mathbf{h}_2}[\mathbf{M}\boldsymbol{\zeta}(\mathbf{h}_1)-\mathbf{h}_1]}$$

which can be used to halve the number of $\bar{F}(\mathbf{M})$ necessary to compute them, as follows.

Having formed the vectors $\mathbf{Z}_{\mathbf{h}_2}$ given by

$$Z_{\mathbf{h}_2}(\mathbf{m}_1)=\left[\sum_{\mathbf{m}_2\in\mathbb{Z}^n/2\mathbb{Z}^n}\frac{(-1)^{\mathbf{h}_2\cdot\mathbf{m}_2}}{2^n}\rho(\mathbf{m}_1+\mathbf{M}\mathbf{m}_2)\right]e[\mathbf{h}_2\cdot(\mathbf{N}^{-1}\mathbf{m}_1)],$$

we may group the 2^n values of $\mathbf{h}_2$ into 2^{n-1} pairs $(\mathbf{h}_2',\mathbf{h}_2'')$ and for each pair form the multiplexed vector:

$$\mathbf{Z}=\mathbf{Z}_{\mathbf{h}_2'}+i\mathbf{Z}_{\mathbf{h}_2''}.$$

After calculating the 2^{n-1} transforms $\mathbf{Z}^*=\bar{F}(\mathbf{M})[\mathbf{Z}]$, the 2^n individual transforms $\mathbf{Z}^*_{\mathbf{h}_2'}$ and $\mathbf{Z}^*_{\mathbf{h}_2''}$ can be separated by using for each pair the demultiplexing formulae

$$Z^*_{\mathbf{h}_2'}(\mathbf{h}_1)+iZ^*_{\mathbf{h}_2''}(\mathbf{h}_1)=Z^*(\mathbf{h}_1)$$
$$Z^*_{\mathbf{h}_2'}(\mathbf{h}_1-\mathbf{h}_2')+iZ^*_{\mathbf{h}_2''}(\mathbf{h}_1-\mathbf{h}_2'')=\overline{Z^*[\mathbf{M}\boldsymbol{\zeta}(\mathbf{h}_1)-\mathbf{h}_1]}$$

which can be solved recursively. If all pairs are chosen so that they differ only in the jth coordinate $(\mathbf{h}_2)_j$, the recursion is along $(\mathbf{h}_1)_j$ and can be initiated by introducing the (real) values of $Z^*_{\mathbf{h}_2'}$ and $Z^*_{\mathbf{h}_2''}$ at $(\mathbf{h}_1)_j=0$ and $(\mathbf{h}_1)_j=M_j$, accumulated *e.g.* while forming $\mathbf{Z}$ for that pair. Only points with $(\mathbf{h}_1)_j$ going from 0 to $\frac{1}{2}M_j$ need be resolved,

and they contain the unique half of the Hermitian-symmetric transform $\mathbf{F}$.

(*c*) *Calculation of electron densities*

The computation may be summarized as follows:

$$\mathbf{F} \overset{\mathbf{scr}(\mathbf{N}_2)}{\longmapsto} \mathbf{Z}^* \overset{F(\mathbf{N}_1)}{\longmapsto} \mathbf{Z} \overset{\mathrm{TW}}{\longmapsto} \mathbf{Y}^* \overset{F(\mathbf{N}_2)}{\longmapsto} \mathbf{Y} \overset{\mathbf{nat}(\mathbf{N}_1)}{\longmapsto} \rho$$

where $\mathbf{scr}(\mathbf{N}_2)$ is the decimation with coset reversal given by $\mathbf{Z}^*_{\mathbf{h}_2}(\mathbf{h}_1) = F(\mathbf{h}_2 + \mathbf{N}_2\mathbf{h}_1)$, TW is the transposition and twiddle-factor stage, and $\mathbf{nat}(\mathbf{N}_1)$ is the recovery in natural order given by $\rho(\mathbf{m}_1 + \mathbf{N}_1\mathbf{m}_2) = Y_{\mathbf{m}_1}(\mathbf{m}_2)$.

(i) *Decimation in time* $(\mathbf{N}_1 = \mathbf{M}, \mathbf{N}_2 = 2\mathbf{I})$

The last transformation $F(2\mathbf{I})$ has a real-valued matrix, and the final result ρ is real-valued. It follows that the vectors $\mathbf{Y}^*_{\mathbf{m}_1}$ of intermediate results after the twiddle-factor stage are real-valued, hence lend themselves to multiplexing along the real and imaginary components of half as many general complex vectors.

Let the 2^n initial vectors $\mathbf{Z}^*_{\mathbf{h}_2}$ be multiplexed into 2^{n-1} vectors

$$\mathbf{Z}^* = \mathbf{Z}^*_{\mathbf{h}'_2} + i\mathbf{Z}^*_{\mathbf{h}''_2}$$

[one for each pair $(\mathbf{h}'_2, \mathbf{h}''_2)$], each of which yields by $F(\mathbf{M})$ a vector

$$\mathbf{Z} = \mathbf{Z}_{\mathbf{h}'_2} + i\mathbf{Z}_{\mathbf{h}''_2}.$$

The real-valuedness of the $\mathbf{Y}^*_{\mathbf{m}_1}$ may be used to recover the separate result vectors for $\mathbf{h}'_2$ and $\mathbf{h}''_2$. For this purpose, introduce the abbreviated notation

$$\begin{aligned} e[-\mathbf{h}'_2 \cdot (\mathbf{N}^{-1}\mathbf{m}_1)] &= (c' + is')(\mathbf{m}_1) \\ e[-\mathbf{h}''_2 \cdot (\mathbf{N}^{-1}\mathbf{m}_1)] &= (c'' + is'')(\mathbf{m}_1) \\ R_{\mathbf{h}_2}(\mathbf{m}_1) &= Y^*_{\mathbf{m}_1}(\mathbf{h}_2) \\ \mathbf{R}' = \mathbf{R}_{\mathbf{h}'_2}, &\quad \mathbf{R}'' = \mathbf{R}_{\mathbf{h}''_2}. \end{aligned}$$

Then we may write

$$\begin{aligned} \mathbf{Z} &= (c' + is')\mathbf{R}' + i(c'' + is'')\mathbf{R}'' \\ &= (c'\mathbf{R}' + s''\mathbf{R}'') + i(-s'\mathbf{R}' + c''\mathbf{R}'') \end{aligned}$$

or, equivalently, for each $\mathbf{m}_1$,

$$\begin{pmatrix} \mathscr{R}e\, Z \\ \mathscr{I}m\, Z \end{pmatrix} = \begin{pmatrix} c' & s'' \\ -s' & c'' \end{pmatrix} \begin{pmatrix} R' \\ R'' \end{pmatrix}.$$

Therefore $\mathbf{R}'$ and $\mathbf{R}''$ may be retrieved from $\mathbf{Z}$ by the 'demultiplexing' formula:

$$\begin{pmatrix} R' \\ R'' \end{pmatrix} = \frac{1}{c'c'' + s's''} \begin{pmatrix} c'' & -s'' \\ s' & c' \end{pmatrix} \begin{pmatrix} \mathscr{R}e\, Z \\ \mathscr{I}m\, Z \end{pmatrix}$$

which is valid at all points $\mathbf{m}_1$ where $c'c'' + s's'' \neq 0$, *i.e.* where

$$\cos[2\pi(\mathbf{h}'_2 - \mathbf{h}''_2) \cdot (\mathbf{N}^{-1}\mathbf{m}_1)] \neq 0.$$

Demultiplexing fails when

$$(\mathbf{h}'_2 - \mathbf{h}''_2) \cdot (\mathbf{N}^{-1}\mathbf{m}_1) = \tfrac{1}{2} \bmod 1.$$

If the pairs $(\mathbf{h}'_2, \mathbf{h}''_2)$ are chosen so that their members differ only in one coordinate (the jth, say), then the exceptional points are at $(\mathbf{m}_1)_j = \frac{1}{2}M_j$ and the missing transform values are easily obtained *e.g.* by accumulation while forming $\mathbf{Z}^*$.

The final stage of the calculation is then

$$\rho(\mathbf{m}_1 + \mathbf{M}\mathbf{m}_2) = \sum_{\mathbf{h}_2 \in \mathbf{Z}^n/2\mathbf{Z}^n} (-1)^{\mathbf{h}_2 \cdot \mathbf{m}_2} R_{\mathbf{h}_2}(\mathbf{m}_1).$$

(ii) *Decimation in frequency* $(\mathbf{N}_1 = 2\mathbf{I}, \mathbf{N}_2 = \mathbf{M})$

The last transformation $F(\mathbf{M})$ gives the real-valued results ρ, therefore the vectors $\mathbf{Y}^*_{\mathbf{m}_1}$ after the twiddle-factor stage each have Hermitian symmetry.

A first consequence is that the intermediate vectors $\mathbf{Z}_{\mathbf{h}_2}$ need only be computed for the unique half of the values of $\mathbf{h}_2$, the other half being related by the Hermitian symmetry of $\mathbf{Y}^*_{\mathbf{m}_1}$.

A second consequence is that the 2^n vectors $\mathbf{Y}^*_{\mathbf{m}_1}$ may be condensed into 2^{n-1} general complex vectors

$$\mathbf{Y}^* = \mathbf{Y}^*_{\mathbf{m}'_1} + i\mathbf{Y}^*_{\mathbf{m}''_1}$$

[one for each pair $(\mathbf{m}'_1, \mathbf{m}''_1)$] to which a general complex $F(\mathbf{M})$ may be applied to yield

$$\mathbf{Y} = \mathbf{Y}_{\mathbf{m}'_1} + i\mathbf{Y}_{\mathbf{m}''_1}$$

with $\mathbf{Y}_{\mathbf{m}'_1}$ and $\mathbf{Y}_{\mathbf{m}''_1}$ *real-valued.* The final results can therefore be retrieved by the particularly simple demultiplexing formulae:

$$\begin{aligned} \rho(\mathbf{m}'_1 + 2\mathbf{m}_2) &= \mathscr{R}e\, Y(\mathbf{m}_2), \\ \rho(\mathbf{m}''_1 + 2\mathbf{m}_2) &= \mathscr{I}m\, Y(\mathbf{m}_2). \end{aligned}$$

1.3.4.3.5.2. *Hermitian-antisymmetric or pure imaginary transforms*

A vector $\mathbf{X} = \{X(\mathbf{k}) | \mathbf{k} \in \mathbb{Z}^n/\mathbf{N}\mathbb{Z}^n\}$ is said to be Hermitian-antisymmetric if

$$X(\mathbf{k}) = -\overline{X(-\mathbf{k})} \text{ for all } \mathbf{k}.$$

Its transform $\mathbf{X}^*$ then satisfies

$$X^*(\mathbf{k}^*) = -\overline{X^*(\mathbf{k}^*)} \text{ for all } \mathbf{k}^*,$$

i.e. is purely imaginary.

If $\mathbf{X}$ is Hermitian-antisymmetric, then $\mathbf{F} = \pm i\mathbf{X}$ is Hermitian-symmetric, with $\rho = \pm i\mathbf{X}^*$ real-valued. The treatment of Section 1.3.4.3.5.1 may therefore be adapted, with trivial factors of i or -1, or used as such in conjunction with changes of variable by multiplication by $\pm i$.

1.3.4.3.5.3. *Complex symmetric and antisymmetric transforms*

The matrix $-\mathbf{I}$ is its own contragredient, and hence (Section 1.3.2.4.2.2) the transform of a symmetric (respectively antisymmetric) function is symmetric (respectively antisymmetric). In this case the group $G = \{e, -e\}$ acts in both real and reciprocal space as $\{\mathbf{I}, -\mathbf{I}\}$. If $\mathbf{N} = \mathbf{N}_1\mathbf{N}_2$ with both factors diagonal, then $-e$ acts by

$$\begin{aligned} (\mathbf{m}_1, \mathbf{m}_2) &\longmapsto [\mathbf{N}_1\boldsymbol{\zeta}(\mathbf{m}_1) - \mathbf{m}_1, \mathbf{N}_2\boldsymbol{\zeta}(\mathbf{m}_2) - \mathbf{m}_2 - \boldsymbol{\zeta}(\mathbf{m}_1)], \\ (\mathbf{h}_2, \mathbf{h}_1) &\longmapsto [\mathbf{N}_2\boldsymbol{\zeta}(\mathbf{h}_2) - \mathbf{h}_2, \mathbf{N}_1\boldsymbol{\zeta}(\mathbf{h}_1) - \mathbf{h}_1 - \boldsymbol{\zeta}(\mathbf{h}_2)], \end{aligned}$$

i.e.

$$\begin{aligned} \boldsymbol{\mu}_2(-e, \mathbf{m}_1) &= -\boldsymbol{\zeta}(\mathbf{m}_1) \bmod \mathbf{N}_2\mathbb{Z}^n, \\ \boldsymbol{\eta}_1(-e, \mathbf{h}_2) &= -\boldsymbol{\zeta}(\mathbf{h}_2) \bmod \mathbf{N}_1\mathbb{Z}^n. \end{aligned}$$

The symmetry or antisymmetry properties of $\mathbf{X}$ may be written

$$X(-\mathbf{m}) = -\varepsilon X(\mathbf{m}) \text{ for all } \mathbf{m},$$

with $\varepsilon = +1$ for symmetry and $\varepsilon = -1$ for antisymmetry.

The computation will be summarized as

$$\mathbf{X} \overset{\mathbf{dec}(\mathbf{N}_1)}{\longmapsto} \mathbf{Y} \overset{\bar{F}(\mathbf{N}_2)}{\longmapsto} \mathbf{Y}^* \overset{\mathrm{TW}}{\longmapsto} \mathbf{Z} \overset{\bar{F}(\mathbf{N}_1)}{\longmapsto} \mathbf{Z}^* \overset{\mathbf{rev}(\mathbf{N}_2)}{\longmapsto} \mathbf{X}^*$$

with the same indexing as that used for structure-factor calculation. In both cases it will be shown that a transform $F(\mathbf{N})$ with $\mathbf{N} = \mathbf{2M}$ and $\mathbf{M}$ diagonal can be computed using only 2^{n-1} partial transforms $F(\mathbf{M})$ instead of 2^n.

(i) *Decimation in time* $(\mathbf{N}_1 = 2\mathbf{I}, \mathbf{N}_2 = \mathbf{M})$

Since $\mathbf{m}_1 \in \mathbb{Z}^n/2\mathbb{Z}^n$ we have $-\mathbf{m}_1 = \mathbf{m}_1$ and $\boldsymbol{\zeta}(\mathbf{m}_1) = \mathbf{m}_1 \bmod 2\mathbb{Z}^n$, so that the symmetry relations for each parity class of data $\mathbf{Y}_{\mathbf{m}_1}$ read

$$Y_{\mathbf{m}_1}[\mathbf{M}\boldsymbol{\zeta}(\mathbf{m}_2) - \mathbf{m}_2 - \mathbf{m}_1] = \varepsilon Y_{\mathbf{m}_1}(\mathbf{m}_2)$$

or equivalently

$$\tau_{\mathbf{m}_1}\mathbf{Y}_{\mathbf{m}_1} = \varepsilon\breve{\mathbf{Y}}_{\mathbf{m}_1}.$$

Transforming by $F(\mathbf{M})$, this relation becomes

$$e[-\mathbf{h}_2 \cdot (\mathbf{M}^{-1}\mathbf{m}_1)]\mathbf{Y}^*_{\mathbf{m}_1} = \varepsilon\mathbf{Y}^*_{\mathbf{m}_1}.$$

Each parity class thus obeys a different symmetry relation, so that we may multiplex them in pairs by forming for each pair $(\mathbf{m}'_1, \mathbf{m}''_1)$ the vector

$$\mathbf{Y} = \mathbf{Y}_{\mathbf{m}'_1} + \mathbf{Y}_{\mathbf{m}''_1}.$$

Putting

$$e[-\mathbf{h}_2 \cdot (\mathbf{M}^{-1}\mathbf{m}'_1)] = (c' + is')(\mathbf{h}_2)$$
$$e[-\mathbf{h}_2 \cdot (\mathbf{M}^{-1}\mathbf{m}''_1)] = (c'' + is'')(\mathbf{h}_2)$$

we then have the demultiplexing relations for each $\mathbf{h}_2$:

$$Y^*_{\mathbf{m}'_1}(\mathbf{h}_2) + Y^*_{\mathbf{m}''_1}(\mathbf{h}_2) = Y^*(\mathbf{h}_2)$$
$$(c' + is')(\mathbf{h}_2)Y^*_{\mathbf{m}'_1}(\mathbf{h}_2) + (c'' + is'')(\mathbf{h}_2)Y^*_{\mathbf{m}''_1}(\mathbf{h}_2) = \varepsilon Y^*[\mathbf{M}\boldsymbol{\zeta}(\mathbf{h}_2) - \mathbf{h}_2]$$

which can be solved recursively. Transform values at the exceptional points $\mathbf{h}_2$ where demultiplexing fails (*i.e.* where $c' + is' = c'' + is''$) can be accumulated while forming $\mathbf{Y}$.

Only the unique half of the values of $\mathbf{h}_2$ need to be considered at the demultiplexing stage and at the subsequent TW and $F(2\mathbf{I})$ stages.

(ii) *Decimation in frequency* $(\mathbf{N}_1 = \mathbf{M}, \mathbf{N}_2 = 2\mathbf{I})$

The vectors of final results $\mathbf{Z}^*_{\mathbf{h}_2}$ for each parity class $\mathbf{h}_2$ obey the symmetry relations

$$\tau_{\mathbf{h}_2}\mathbf{Z}^*_{\mathbf{h}_2} = \varepsilon\breve{\mathbf{Z}}^*_{\mathbf{h}_2},$$

which are different for each $\mathbf{h}_2$. The vectors $\mathbf{Z}_{\mathbf{h}_2}$ of intermediate results after the twiddle-factor stage may then be multiplexed in pairs as

$$\mathbf{Z} = \mathbf{Z}_{\mathbf{h}'_2} + \mathbf{Z}_{\mathbf{h}''_2}.$$

After transforming by $F(\mathbf{M})$, the results $\mathbf{Z}^*$ may be demultiplexed by using the relations

$$Z^*_{\mathbf{h}'_2}(\mathbf{h}_1) + Z^*_{\mathbf{h}''_2}(\mathbf{h}_1) = Z^*(\mathbf{h}_1)$$
$$Z^*_{\mathbf{h}'_2}(\mathbf{h}_1 - \mathbf{h}'_2) + Z^*_{\mathbf{h}''_2}(\mathbf{h}_1 - \mathbf{h}''_2) = \varepsilon Z^*[\mathbf{M}\boldsymbol{\zeta}(\mathbf{h}_1) - \mathbf{h}_1]$$

which can be solved recursively as in Section 1.3.4.3.5.1(*b*)(ii).

1.3.4.3.5.4. *Real symmetric transforms*

Conjugate symmetric (Section 1.3.2.4.2.3) implies that if the data $\mathbf{X}$ are real and symmetric [*i.e.* $X(\mathbf{k}) = \overline{X(\mathbf{k})}$ and $X(-\mathbf{k}) = X(\mathbf{k})$], then so are the results $\mathbf{X}^*$. Thus if $\mathcal{P}$ contains a centre of symmetry, $\mathbf{F}$ is real symmetric. There is no distinction (other than notation) between structure-factor and electron-density calculation; the algorithms will be described in terms of the former. It will be shown that if $\mathbf{N} = 2\mathbf{M}$, a real symmetric transform can be computed with only 2^{n-2} partial transforms $F(\mathbf{M})$ instead of 2^n.

(i) *Decimation in time* $(\mathbf{N}_1 = 2\mathbf{I}, \mathbf{N}_2 = \mathbf{M})$

Since $\mathbf{m}_1 \in \mathbb{Z}^n/2\mathbb{Z}^n$ we have $-\mathbf{m}_1 = \mathbf{m}_1$ and $\boldsymbol{\zeta}(\mathbf{m}_1) = \mathbf{m}_1 \bmod 2\mathbb{Z}^n$. The decimated vectors $\mathbf{Y}_{\mathbf{m}_1}$ are not only real, but have an internal symmetry expressed by

$$\mathbf{Y}_{\mathbf{m}_1}[\mathbf{M}\boldsymbol{\zeta}(\mathbf{m}_2) - \mathbf{m}_2 - \mathbf{m}_1] = \varepsilon\mathbf{Y}_{\mathbf{m}_1}(\mathbf{m}_2).$$

This symmetry, however, is different for each $\mathbf{m}_1$ so that we may multiplex two such vectors $\mathbf{Y}_{\mathbf{m}'_1}$ and $\mathbf{Y}_{\mathbf{m}''_1}$ into a general *real* vector

$$\mathbf{Y} = \mathbf{Y}_{\mathbf{m}'_1} + \mathbf{Y}_{\mathbf{m}''_1},$$

for each of the 2^{n-1} pairs $(\mathbf{m}'_1, \mathbf{m}''_1)$. The 2^{n-1} Hermitian-symmetric transform vectors

$$\mathbf{Y}^* = \mathbf{Y}^*_{\mathbf{m}'_1} + \mathbf{Y}^*_{\mathbf{m}''_1}$$

can then be evaluated by the methods of Section 1.3.4.3.5.1(*b*) at the cost of only 2^{n-2} general complex $F(\mathbf{M})$.

The demultiplexing relations by which the separate vectors $\mathbf{Y}^*_{\mathbf{m}'_1}$ and $\mathbf{Y}^*_{\mathbf{m}''_1}$ may be recovered are most simply obtained by observing that the vectors $\mathbf{Z}$ after the twiddle-factor stage are real-valued since $F(2\mathbf{I})$ has a real matrix. Thus, as in Section 1.3.4.3.5.1(*c*)(i),

$$\mathbf{Y}^*_{\mathbf{m}'_1} = (c' - is')\mathbf{R}'$$
$$\mathbf{Y}^*_{\mathbf{m}''_1} = (c'' - is'')\mathbf{R}'',$$

where $\mathbf{R}'$ and $\mathbf{R}''$ are real vectors and where the multipliers $(c' - is')$ and $(c'' - is'')$ are the inverse twiddle factors. Therefore,

$$\mathbf{Y}^* = (c' - is')\mathbf{R}' + (c'' - is'')\mathbf{R}''$$
$$= (c'\mathbf{R}' + c''\mathbf{R}'') - i(s'\mathbf{R}' + s''\mathbf{R}'')$$

and hence the demultiplexing relation for each $\mathbf{h}_2$:

$$\begin{pmatrix} R' \\ R'' \end{pmatrix} = \frac{1}{c's'' - s'c''}\begin{pmatrix} s'' & -c'' \\ -s' & c' \end{pmatrix}\begin{pmatrix} \mathscr{R}e\ Y^* \\ -\mathscr{I}m\ Y^* \end{pmatrix}.$$

The values of $R'_{\mathbf{h}_2}$ and $R''_{\mathbf{h}_2}$ at those points $\mathbf{h}_2$ where $c's'' - s'c'' = 0$ can be evaluated directly while forming $\mathbf{Y}$. This demultiplexing and the final stage of the calculation, namely

$$F(\mathbf{h}_2 + \mathbf{M}\mathbf{h}_1) = \frac{1}{2^n}\sum_{\mathbf{m}_1 \in \mathbf{Z}^n/2\mathbf{Z}^n}(-1)^{\mathbf{h}_1 \cdot \mathbf{m}_1}R_{\mathbf{m}_1}(\mathbf{h}_2)$$

need only be carried out for the unique half of the range of $\mathbf{h}_2$.

(ii) *Decimation in frequency* $(\mathbf{N}_1 = \mathbf{M}, \mathbf{N}_2 = 2\mathbf{I})$

Similarly, the vectors $\mathbf{Z}^*_{\mathbf{h}_2}$ of decimated and scrambled results are real and obey internal symmetries

$$\tau_{\mathbf{h}_2}\mathbf{Z}^*_{\mathbf{h}_2} = \varepsilon\breve{\mathbf{Z}}^*_{\mathbf{h}_2}$$

which are different for each $\mathbf{h}_2$. For each of the 2^{n-1} pairs $(\mathbf{h}'_2, \mathbf{h}''_2)$ the multiplexed vector

$$\mathbf{Z} = \mathbf{Z}_{\mathbf{h}'_2} + \mathbf{Z}_{\mathbf{h}''_2}$$

is a Hermitian-symmetric vector without internal symmetry, and the 2^{n-1} real vectors

$$\mathbf{Z}^* = \mathbf{Z}^*_{\mathbf{h}'_2} + \mathbf{Z}^*_{\mathbf{h}''_2}$$

may be evaluated at the cost of only 2^{n-2} general complex $F(\mathbf{M})$ by the methods of Section 1.3.4.3.5.1(*c*). The individual transforms $\mathbf{Z}_{\mathbf{h}'_2}$ and $\mathbf{Z}_{\mathbf{h}''_2}$ may then be retrieved *via* the demultiplexing relations

$$Z^*_{\mathbf{h}'_2}(\mathbf{h}_1) + Z^*_{\mathbf{h}''_2}(\mathbf{h}_1) = Z^*(\mathbf{h}_1)$$
$$Z^*_{\mathbf{h}'_2}(\mathbf{h}_1 - \mathbf{h}'_2) + Z^*_{\mathbf{h}''_2}(\mathbf{h}_1 - \mathbf{h}''_2) = Z^*[\mathbf{M}\boldsymbol{\zeta}(\mathbf{h}_1) - \mathbf{h}_1]$$

which can be solved recursively as described in Section 1.3.4.3.5.1(*b*)(ii). This yields the unique half of the real symmetric results $\mathbf{F}$.

1.3.4.4.3.5.5. *Real antisymmetric transforms*

If $\mathbf{X}$ is real antisymmetric, then its transform $\mathbf{X}^*$ is purely imaginary and antisymmetric. The double-multiplexing techniques used for real symmetric transforms may therefore be adapted with only minor changes involving signs and factors of i.

1.3.4.4.3.5.6. *Generalized multiplexing*

So far the multiplexing technique has been applied to pairs of vectors with similar types of parity-related and/or conjugate symmetry properties, in particular the same value of ε.

It can be generalized so as to accommodate mixtures of vectors with different symmetry characteristics. For example if $\mathbf{X}_1$ is Hermitian-symmetric and $\mathbf{X}_2$ is Hermitian-antisymmetric, so that $\mathbf{X}_1^*$ is real-valued while $\mathbf{X}_2^*$ has purely imaginary values, the multiplexing process should obviously form $\mathbf{X} = \mathbf{X}_1 + \mathbf{X}_2$ (instead of $\mathbf{X} = \mathbf{X}_1 + i\mathbf{X}_2$ if both had the same type of symmetry), and demultiplexing consists in separating

$$\mathbf{X}_1^* = \mathscr{Re}\,\mathbf{X}^*$$
$$\mathbf{X}_2^* = i\mathscr{Im}\,\mathbf{X}^*.$$

The general multiplexing formula for pairs of vectors may therefore be written

$$\mathbf{X} = \mathbf{X}_1 + \omega\mathbf{X}_2,$$

where ω is a phase factor (*e.g.* 1 or i) chosen in such a way that all non-exceptional components of $\mathbf{X}_1$ and $\mathbf{X}_2$ (or $\mathbf{X}_1^*$ and $\mathbf{X}_2^*$) be embedded in the complex plane $\mathbb{C}$ along linearly independent directions, thus making multiplexing possible.

It is possible to develop a more general form of multiplexing/demultiplexing for more than two vectors, which can be used to deal with symmetry elements of order 3, 4 or 6. It is based on the theory of group characters (Ledermann, 1987).

1.3.4.4.3.6. *Global crystallographic algorithms*

All the necessary ingredients are now available for calculating the CDFT for any given space group.

1.3.4.4.3.6.1. *Triclinic groups*

Space group $P1$ is dealt with by the methods of Section 1.3.4.4.3.5.1 and $P\bar{1}$ by those of Section 1.3.4.4.3.5.4.

1.3.4.4.3.6.2. *Monoclinic groups*

A general monoclinic transformation is of the form

$$S_g : \mathbf{x} \longmapsto \mathbf{R}_g\mathbf{x} + \mathbf{t}_g$$

with $\mathbf{R}_g$ a diagonal matrix whose entries are $+1$ or -1, and $\mathbf{t}_g$ a vector whose entries are 0 or $\frac{1}{2}$. We may thus decompose both real and reciprocal space into a direct sum of a subspace $\mathbb{Z}^{n_+}$ where $\mathbf{R}_g$ acts as the identity, and a subspace $\mathbb{Z}^{n_-}$ where $\mathbf{R}_g$ acts as minus the identity, with $n_+ + n_- = n = 3$. All usual entities may be correspondingly written as direct sums, for instance:

$$\mathbf{R}_g = \mathbf{R}_g^+ \oplus \mathbf{R}_g^-, \quad \mathbf{N} = \mathbf{N}^+ \oplus \mathbf{N}^-, \quad \mathbf{M} = \mathbf{M}^+ \oplus \mathbf{M}^-,$$
$$\mathbf{t}_g = \mathbf{t}_g^+ \oplus \mathbf{t}_g^-, \quad \mathbf{t}_g^{(1)} = \mathbf{t}_g^{(1)+} \oplus \mathbf{t}_g^{(1)-}, \quad \mathbf{t}_g^{(2)} = \mathbf{t}_g^{(2)+} \oplus \mathbf{t}_g^{(2)-},$$
$$\mathbf{m} = \mathbf{m}^+ \oplus \mathbf{m}^-, \quad \mathbf{m}_1 = \mathbf{m}_1^+ \oplus \mathbf{m}_1^-, \quad \mathbf{m}_2 = \mathbf{m}_2^+ \oplus \mathbf{m}_2^-,$$
$$\mathbf{h} = \mathbf{h}^+ \oplus \mathbf{h}^-, \quad \mathbf{h}_1 = \mathbf{h}_1^+ \oplus \mathbf{h}_1^-, \quad \mathbf{h}_2 = \mathbf{h}_2^+ \oplus \mathbf{h}_2^-.$$

We will use factoring by 2, with decimation in frequency when computing structure factors, and decimation in time when computing electron densities; this corresponds to $\mathbf{N} = \mathbf{N}_1\mathbf{N}_2$ with $\mathbf{N}_1 = \mathbf{M}$, $\mathbf{N}_2 = 2\mathbf{I}$. The non-primitive translation vector $\mathbf{N}\mathbf{t}_g$ then belongs to $\mathbf{M}\mathbb{Z}^n$, and thus

$$\mathbf{t}_g^{(1)} = \mathbf{0} \bmod \mathbf{M}\mathbb{Z}^n, \quad \mathbf{t}_g^{(2)} \in \mathbb{Z}^n/2\mathbb{Z}^n.$$

The symmetry relations obeyed by $\wp$ and F are as follows: for electron densities

$$\wp(\mathbf{m}^+, \mathbf{m}^-) = \wp(\mathbf{m}^+ + \mathbf{N}^+\mathbf{t}_g^+, -\mathbf{m}^- - \mathbf{N}^-\mathbf{t}_g^-)$$

or, after factoring by 2,

$$\wp(\mathbf{m}_1^+, \mathbf{m}_2^+, \mathbf{m}_1^-, \mathbf{m}_2^-) = \wp(\mathbf{m}_1^+, \mathbf{m}_2^+ + \mathbf{t}_g^{(2)+}, \mathbf{M}^-\boldsymbol{\zeta}(\mathbf{m}_1^-) - \mathbf{m}_1^- - \mathbf{m}_2^-, \mathbf{m}_2^- + \mathbf{t}_g^{(2)-});$$

while for structure factors

$$F(\mathbf{h}^+, \mathbf{h}^-) = \exp[2\pi i(\mathbf{h}^+ \cdot \mathbf{t}_g^+ + \mathbf{h}^- \cdot \mathbf{t}_g^-)]F(\mathbf{h}^+, -\mathbf{h}^-)$$

with its Friedel counterpart

$$F(\mathbf{h}^+, \mathbf{h}^-) = \exp[2\pi i(\mathbf{h}^+ \cdot \mathbf{t}_g^+ + \mathbf{h}^- \cdot \mathbf{t}_g^-)]\overline{F(-\mathbf{h}^+, \mathbf{h}^-)}$$

or, after factoring by 2,

$$F(\mathbf{h}_1^+, \mathbf{h}_2^+, \mathbf{h}_1^-, \mathbf{h}_2^-) = (-1)^{\mathbf{h}_2^+ \cdot \mathbf{t}_g^{(2)+} + \mathbf{h}_2^- \cdot \mathbf{t}_g^{(2)-}} \times F(\mathbf{h}_1^+, \mathbf{h}_2^+, \mathbf{M}^-\boldsymbol{\zeta}(\mathbf{h}_1^-) - \mathbf{h}_1^- - \mathbf{h}_2^-, \mathbf{h}_2^-)$$

with Friedel counterpart

$$F(\mathbf{h}_1^+, \mathbf{h}_2^+, \mathbf{h}_1^-, \mathbf{h}_2^-) = (-1)^{\mathbf{h}_2^+ \cdot \mathbf{t}_g^{(2)+} + \mathbf{h}_2^- \cdot \mathbf{t}_g^{(2)-}} \overline{F[\mathbf{M}^+\boldsymbol{\zeta}(\mathbf{h}_1^+) - \mathbf{h}_1^+ - \mathbf{h}_2^+, \mathbf{h}_2^+, \mathbf{h}_1^-, \mathbf{h}_2^-]}.$$

When calculating electron densities, two methods may be used.

(i) Transform on $\mathbf{h}^-$ first.

The partial vectors defined by $X_{\mathbf{h}^+,\,\mathbf{h}_2^-} = F(\mathbf{h}^+, \mathbf{h}_1^-, \mathbf{h}_2^-)$ obey symmetry relations of the form

$$X(\mathbf{h}_1^- - \mathbf{h}_2^-) = \varepsilon X[\mathbf{M}^-\boldsymbol{\zeta}(\mathbf{h}_1^-) - \mathbf{h}_1^-]$$

with $\varepsilon = \pm 1$ independent of $\mathbf{h}_1^-$. This is the same relation as for the same parity class of data for a (complex or real) symmetric ($\varepsilon = +1$) or antisymmetric ($\varepsilon = -1$) transform. The same techniques can be used to decrease the number of $F(\mathbf{M}^-)$ by multiplexing pairs of such vectors and demultiplexing their transforms. Partial vectors with different values of ε may be mixed in this way (Section 1.3.4.4.3.5.6).

Once $F(\mathbf{N}^-)$ is completed, its results have Hermitian symmetry with respect to $\mathbf{h}^+$, and the methods of Section 1.3.4.4.3.5.1 may be used to obtain the unique electron densities.

(ii) Transform on $\mathbf{h}^+$ first.

The partial vectors defined by $X_{\mathbf{h}^-,\,\mathbf{h}_2^+} = F(\mathbf{h}_1^+, \mathbf{h}_2^+, \mathbf{h}^-)$ obey symmetry relations of the form

$$X(\mathbf{h}_1^+ - \mathbf{h}_2^+) = \overline{\varepsilon X[\mathbf{M}^+\boldsymbol{\zeta}(\mathbf{h}_1^+) - \mathbf{h}_1^+]}$$

with $\varepsilon = \pm 1$ independent of $\mathbf{h}_1^+$. This is the same relation as for the same parity class of data for a Hermitian symmetric ($\varepsilon = +1$) or antisymmetric ($\varepsilon = -1$) transform. The same techniques may be used to decrease the number of $F(\mathbf{M}^+)$. This generalizes the procedure described by Ten Eyck (1973) for treating dyad axes, *i.e.* for the case $n_+ = 1, \mathbf{t}_g^{(2)-} = \mathbf{0}$, and $\mathbf{t}_g^{(2)+} = \mathbf{0}$ (simple dyad) or $\mathbf{t}_g^{(2)+} \neq \mathbf{0}$ (screw dyad).

Once $F(\mathbf{N}^+)$ is completed, its results have Hermitian symmetry properties with respect to $\mathbf{h}^-$ which can be used to obtain the unique electron densities.

Structure factors may be computed by applying the reverse procedures in the reverse order.

1.3.4.4.3.6.3. *Orthorhombic groups*

Almost all orthorhombic space groups are generated by two monoclinic transformations g_1 and g_2 of the type described in Section 1.3.4.4.3.6.2, with the addition of a centre of inversion $-e$ for centrosymmetric groups. The only exceptions are *Fdd*2 and *Fddd*

which contain diamond glides, in which some non-primitive translations are 'square roots' not of primitive lattice translations, but of centring translations. The generic case will be examined first.

To calculate electron densities, the unique octant of data may first be transformed on $\mathbf{h}^+$ (respectively $\mathbf{h}^-$) as in Section 1.3.4.3.6.2 using the symmetry pertaining to generator g_1. These intermediate results may then be expanded by generator g_2 by the formula of Section 1.3.4.3.3 prior to the final transform on $\mathbf{h}^-$ (respectively $\mathbf{h}^+$). To calculate structure factors, the reverse operations are applied in the reverse order.

The two exceptional groups *Fdd*2 and *Fddd* only require a small modification. The *F*-centring causes the systematic absence of parity classes with mixed parities, leaving only (000) and (111). For the former, the phase factors $\exp[2\pi i(\mathbf{h}^+ \cdot \mathbf{t}_g^+ + \mathbf{h}^- \cdot \mathbf{t}_g^-)]$ in the symmetry relations of Section 1.3.4.3.6.2 become powers of (-1) so that one is back to the generic case. For the latter, these phase factors are odd powers of i which it is a simple matter to incorporate into a modified multiplexing/demultiplexing procedure.

1.3.4.3.6.4. *Trigonal, tetragonal and hexagonal groups*

All the symmetries in this class of groups can be handled by the generalized Rader/Winograd algorithms of Section 1.3.4.3.4.3, but no implementation of these is yet available.

In groups containing axes of the form n_m with g.c.d. $(m, n) = 1$ (*i.e.* $3_1, 3_2, 4_1, 4_3, 6_1, 6_5$) along the c direction, the following procedure may be used (Ten Eyck, 1973):

(i) to calculate electron densities, the unique structure factors indexed by

$$[\text{unique } (h,k)] \times (\text{all } l)$$

are transformed on l; the results are rearranged by the transposition formula of Section 1.3.4.3.3 so as to be indexed by

$$[\text{all } (h,k)] \times \left[\text{unique } \left(\frac{1}{n}\right)\text{th of } z\right]$$

and are finally transformed on (h, k) to produce an asymmetric unit. For a dihedral group, the extra twofold axis may be used in the transposition to produce a unique $(1/2n)$th of z.

(ii) to calculate structure factors, the unique densities in $(1/n)$th of z [or $(1/2n)$th for a dihedral group] are first transformed on x and y, then transposed by the formula of Section 1.3.4.3.3 to reindex the intermediate results by

$$[\text{unique } (h,k)] \times (\text{all } z);$$

the last transform on z is then carried out.

1.3.4.3.6.5. *Cubic groups*

These are usually treated as their orthorhombic or tetragonal subgroups, as the body-diagonal threefold axis cannot be handled by ordinary methods of decomposition.

The three-dimensional factorization technique of Section 1.3.4.3.4.1 allows a complete treatment of cubic symmetry. Factoring by 2 along all three dimensions gives four types (*i.e.* orbits) of parity classes:

(000)	with residual threefold symmetry,
(100), (010), (001)	related by threefold axis,
(110), (101), (011)	related by threefold axis,
(111)	with residual threefold symmetry.

Orbit exchange using the threefold axis thus allows one to reduce the number of partial transforms from 8 to 4 (one per orbit). Factoring by 3 leads to a reduction from 27 to 11 (in this case, further reduction to 9 can be gained by multiplexing the three diagonal classes with residual threefold symmetry into a single class; see Section 1.3.4.3.5.6). More generally, factoring by q leads to a reduction from q^3 to $\frac{1}{3}(q^3 - q) - q$. Each of the remaining transforms then has a symmetry induced from the orthorhombic or tetragonal subgroup, which can be treated as above.

No implementation of this procedure is yet available.

1.3.4.3.6.6. *Treatment of centred lattices*

Lattice centring is an instance of the duality between periodization and decimation: the extra translational periodicity of ρ induces a decimation of $\mathbf{F} = \{F_\mathbf{h}\}$ described by the 'reflection conditions' on $\mathbf{h}$. As was pointed out in Section 1.3.4.2.2.3, non-primitive lattices are introduced in order to retain the same matrix representation for a given geometric symmetry operation in all the arithmetic classes in which it occurs. From the computational point of view, therefore, the main advantage in using centred lattices is that it *maximizes decomposability* (Section 1.3.4.2.2.4); reindexing to a primitive lattice would for instance often destroy the diagonal character of the matrix representing a dyad.

In the usual procedure involving three successive one-dimensional transforms, the loss of efficiency caused by the duplication of densities or the systematic vanishing of certain classes of structure factors may be avoided by using a multiplexing/demultiplexing technique (Ten Eyck, 1973):

(i) for base-centred or body-centred lattices, two successive planes of structure factors may be overlaid into a single plane; after transformation, the results belonging to each plane may be separated by parity considerations;

(ii) for face-centred lattices the same method applies, using four successive planes (the third and fourth with an origin translation);

(iii) for rhombohedral lattices in hexagonal coordinates, three successive planes may be overlaid, and the results may be separated by linear combinations involving cube roots of unity.

The three-dimensional factorization technique of Section 1.3.4.3.4.1 is particularly well suited to the treatment of centred lattices: if the decimation matrix of $\mathbf{N}$ contains as a factor $\mathbf{N}_1$ a matrix which 'integerizes' all the non-primitive lattice vectors, then centring is reflected by the systematic vanishing of certain classes of vectors of decimated data or results, which can simply be omitted from the calculation. An alternative possibly is to reindex on a primitive lattice and use different representative matrices for the symmetry operations: the loss of decomposability is of little consequence in this three-dimensional scheme, although it substantially complicates the definition of the cocycles $\boldsymbol{\mu}_2$ and $\boldsymbol{\eta}_1$.

1.3.4.3.6.7. *Programming considerations*

The preceding sections have been devoted to showing how the raw computational efficiency of a crystallographic Fourier transform *algorithm* can be maximized. This section will briefly discuss another characteristic (besides speed) which a crystallographic Fourier transform *program* may be required to possess if it is to be useful in various applications: a convenient and versatile mode of presentation of input data or output results.

The standard crystallographic FFT programs (Ten Eyck, 1973, 1985) are rather rigid in this respect, and use rather rudimentary data structures (lists of structure-factor values, and two-dimensional arrays containing successive sections of electron-density maps). It is frequently the case that considerable reformatting of these data or results must be carried out before they can be used in other computations; for instance, maps have to be converted from 2D sections to 3D 'bricks' before they can be inspected on a computer graphics display.

The explicitly three-dimensional approach to the factorization of the DFT and the use of symmetry offers the possibility of richer and more versatile data structures. For instance, the use of 'decimation in frequency' in real space and of 'decimation in time' in reciprocal

space leads to data structures in which real-space coordinates are handled by blocks (thus preserving, at least locally, the three-dimensional topological connectivity of the maps) while reciprocal-space indices are handled by parity classes or their generalizations for factors other than 2 (thus making the treatment of centred lattices extremely easy). This global three-dimensional indexing also makes it possible to carry symmetry and multiplicity characteristics for each subvector of intermediate results for the purpose of automating the use of the orbit exchange mechanism.

Brünger (1989) has described the use of a similar three-dimensional factoring technique in the context of structure-factor calculations for the refinement of macromolecular structures.

1.3.4.4. *Basic crystallographic computations*

1.3.4.4.1. *Introduction*

Fourier transform (FT) calculations play an indispensable role in crystallography, because the Fourier transformation is inherent in the diffraction phenomenon itself.

Besides this obligatory use, the FT has numerous other applications, motivated more often by its mathematical properties than by direct physical reasoning (although the latter can be supplied after the fact). Typically, many crystallographic computations turn out to be convolutions in disguise, which can be speeded up by orders of magnitude through a judicious use of the FT. Several recent advances in crystallographic computation have been based on this kind of observation.

1.3.4.4.2. *Fourier synthesis of electron-density maps*

Bragg (1929) was the first to use this type of calculation to assist structure determination. Progress in computing techniques since that time was reviewed in Section 1.3.4.3.1.

The usefulness of the maps thus obtained can be adversely affected by three main factors:

(i) limited resolution;

(ii) errors in the data;

(iii) computational errors.

Limited resolution causes 'series-termination errors' first investigated by Bragg & West (1930), who used an optical analogy with the numerical aperture of a microscope. James (1948*b*) gave a quantitative description of this phenomenon as a convolution with the 'spherical Dirichlet kernel' (Section 1.3.4.2.1.3), which reflects the truncation of the Fourier spectrum by multiplication with the indicator function of the limiting resolution sphere. Bragg & West (1930) suggested that the resulting ripples might be diminished by applying an artificial temperature factor to the data, which performs a further convolution with a Gaussian point-spread function. When the electron-density map is to be used for model refinement, van Reijen (1942) suggested using Fourier coefficients calculated from the model when no observation is available, as a means of combating series-termination effects.

Errors in the data introduce errors in the electron-density maps, with the same mean-square value by virtue of Parseval's theorem. Special positions accrue larger errors (Cruickshank & Rollett, 1953; Cruickshank, 1965*a*). To minimize the mean-square electron-density error due to large phase uncertainties, Blow & Crick (1959) introduced the 'best Fourier' which uses centroid Fourier coefficients; the associated error level in the electron-density map was evaluated by Blow & Crick (1959) and Dickerson *et al.* (1961*a*,*b*).

Computational errors used to be a serious concern when Beevers–Lipson strips were used, and Cochran (1948*a*) carried out a critical evaluation of the accuracy limitations imposed by strip methods. Nowadays, the FFT algorithm implemented on digital computers with a word size of at least 32 bits gives results accurate to six decimal places or better in most applications (see Gentleman & Sande, 1966).

1.3.4.4.3. *Fourier analysis of modified electron-density maps*

Various approaches to the phase problem are based on certain modifications of the electron-density map, followed by Fourier analysis of the modified map and extraction of phase information from the resulting Fourier coefficients.

1.3.4.4.3.1. *Squaring*

Sayre (1952*a*) derived his 'squaring method equation' for structures consisting of equal, resolved and spherically symmetric atoms by observing that squaring such an electron density is equivalent merely to sharpening each atom into its square. Thus

$$F_{\mathbf{h}} = \theta_{\mathbf{h}} \sum_{\mathbf{k}} F_{\mathbf{k}} F_{\mathbf{h}-\mathbf{k}},$$

where $\theta_{\mathbf{h}} = f(\mathbf{h})/f^{\mathrm{sq}}(\mathbf{h})$ is the ratio between the form factor $f(\mathbf{h})$ common to all the atoms and the form factor $f^{\mathrm{sq}}(\mathbf{h})$ for the squared version of that atom.

Most of the central results of direct methods, such as the tangent formula, are an immediate consequence of Sayre's equation. Phase refinement for a macromolecule by enforcement of the squaring method equation was demonstrated by Sayre (1972, 1974).

1.3.4.4.3.2. *Other non-linear operations*

A category of phase improvement procedures known as 'density modification' is based on the pointwise application of various quadratic or cubic 'filters' to electron-density maps after removal of negative regions (Hoppe & Gassmann, 1968; Hoppe *et al.*, 1970; Barrett & Zwick, 1971; Gassmann & Zechmeister, 1972; Collins, 1975; Collins *et al.*, 1976; Gassmann, 1976). These operations are claimed to be equivalent to reciprocal-space phase-refinement techniques such as those based on the tangent formula. Indeed the replacement of

$$\rho(\mathbf{x}) = \sum_{\mathbf{h}} F_{\mathbf{h}} \exp(-2\pi i \mathbf{h} \cdot \mathbf{x})$$

by $P[\rho(\mathbf{x})]$, where P is a polynomial

$$P(\rho) = a_0 + a_1\rho + a_2\rho^2 + a_3\rho^3 + \ldots$$

yields

$$P[\rho(\mathbf{x})] = a_0 + \sum_{\mathbf{h}} \left[a_1 F_{\mathbf{h}} + a_2 \sum_{\mathbf{k}} F_{\mathbf{k}} F_{\mathbf{h}-\mathbf{k}} + a_3 \sum_{\mathbf{k}} \sum_{\mathbf{l}} F_{\mathbf{k}} F_{\mathbf{l}} F_{\mathbf{h}-\mathbf{k}-\mathbf{l}} + \ldots \right] \exp(-2\pi i \mathbf{h} \cdot \mathbf{x})$$

and hence gives rise to the convolution-like families of terms encountered in direct methods. This equivalence, however, has been shown to be rather superficial (Bricogne, 1982) because the 'uncertainty principle' embodied in Heisenberg's inequality (Section 1.3.2.4.4.3) imposes severe limitations on the effectiveness of any procedure which operates *pointwise* in *both* real and reciprocal space.

In applying such methods, sampling considerations must be given close attention. If the spectrum of ρ extends to resolution Δ and if the pointwise non-linear filter involves a polynomial P of degree n, then $P(\rho)$ should be sampled at intervals of at most $\Delta/2n$ to accommodate the full bandwidth of its spectrum.

1.3.4.4.3.3. *Solvent flattening*

Crystals of proteins and nucleic acids contain large amounts of mother liquor, often in excess of 50% of the unit-cell volume,