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1.3. Fourier transforms in crystallography: theory, algorithms and applications

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1.3.1. General introduction

Since the publication of Volume II of *International Tables*, most aspects of the theory, computation and applications of Fourier transforms have undergone considerable development, often to the point of being hardly recognizable.

The mathematical analysis of the Fourier transformation has been extensively reformulated within the framework of distribution theory, following Schwartz's work in the early 1950s.

The computation of Fourier transforms has been revolutionized by the advent of digital computers and of the Cooley–Tukey algorithm, and progress has been made at an ever-accelerating pace in the design of new types of algorithms and in optimizing their interplay with machine architecture.

These advances have transformed both theory and practice in several fields which rely heavily on Fourier methods; much of electrical engineering, for instance, has become digital signal processing.

By contrast, crystallography has remained relatively unaffected by these developments. From the conceptual point of view, old-fashioned Fourier series are still adequate for the quantitative description of X-ray diffraction, as this rarely entails consideration of molecular transforms between reciprocal-lattice points. From the practical point of view, three-dimensional Fourier transforms have mostly been used as a tool for visualizing electron-density maps, so that only moderate urgency was given to trying to achieve ultimate efficiency in these relatively infrequent calculations.

Recent advances in phasing and refinement methods, however, have placed renewed emphasis on concepts and techniques long used in digital signal processing, *e.g.* flexible sampling, Shannon interpolation, linear filtering, and interchange between convolution and multiplication. These methods are iterative in nature, and thus generate a strong incentive to design new crystallographic Fourier transform algorithms making the fullest possible use of all available symmetry to save both storage and computation.

As a result, need has arisen for a modern and coherent account of Fourier transform methods in crystallography which would provide:

(i) a simple and foolproof means of switching between the three different guises in which the Fourier transformation is encountered (Fourier transforms, Fourier series and discrete Fourier transforms), both formally and computationally;

(ii) an up-to-date presentation of the most important algorithms for the efficient numerical calculation of discrete Fourier transforms;

(iii) a systematic study of the incorporation of symmetry into the calculation of crystallographic discrete Fourier transforms;

(iv) a survey of the main types of crystallographic computations based on the Fourier transformation.

The rapid pace of progress in these fields implies that such an account would be struck by quasi-immediate obsolescence if it were written solely for the purpose of compiling a catalogue of results and formulae 'customized' for crystallographic use. Instead, the emphasis has been placed on a mode of presentation in which most results and formulae are *derived* rather than listed. This does entail a substantial mathematical overhead, but has the advantage of preserving in its 'native' form the context within which these results are obtained. It is this context, rather than any particular set of results, which constitutes the most fertile source of new ideas and new applications, and as such can have any hope at all of remaining useful in the long run.

These conditions have led to the following choices:

(i) the mathematical theory of the Fourier transformation has been cast in the language of Schwartz's theory of distributions which has long been adopted in several applied fields, in particular electrical engineering, with considerable success; the extra work involved handsomely pays for itself by allowing the three different types of Fourier transformations to be treated together, and by making all properties of the Fourier transform consequences of a single property (the convolution theorem). This is particularly useful in all questions related to the sampling theorem;

(ii) the various numerical algorithms have been presented as the consequences of basic algebraic phenomena involving Abelian groups, rings and finite fields; this degree of formalization greatly helps the subsequent incorporation of symmetry;

(iii) the algebraic nature of space groups has been re-emphasized so as to build up a framework which can accommodate both the phenomena used to factor the discrete Fourier transform and those which underlie the existence (and lead to the classification) of space groups; this common ground is found in the notion of *module over a group ring* (*i.e.* integral representation theory), which is then applied to the formulation of a large number of algorithms, many of which are new;

(iv) the survey of the main types of crystallographic computations has tried to highlight the roles played by various properties of the Fourier transformation, and the ways in which a better exploitation of these properties has been the driving force behind the discovery of more powerful methods.

In keeping with this philosophy, the theory is presented first, followed by the crystallographic applications. There are 'forward references' from mathematical results to the applications which later invoke them (thus giving 'real-life' examples rather than artificial ones), and 'backward references' as usual. In this way, the internal logic of the mathematical developments – the surest guide to future innovations – can be preserved, whereas the alternative solution of relegating these to appendices tends on the contrary to obscure that logic by subordinating it to that of the applications.

It is hoped that this attempt at an overall presentation of the main features of Fourier transforms and of their ubiquitous role in crystallography will be found useful by scientists both within and outside the field.

1.3.2. The mathematical theory of the Fourier transformation

1.3.2.1. *Introduction*

The Fourier transformation and the practical applications to which it gives rise occur in three different forms which, although they display a similar range of phenomena, normally require distinct formulations and different proof techniques:

(i) *Fourier transforms*, in which both function and transform depend on continuous variables;

(ii) *Fourier series*, which relate a periodic function to a discrete set of coefficients indexed by n-tuples of integers;

(iii) *discrete Fourier transforms*, which relate finite-dimensional vectors by linear operations representable by matrices.

At the same time, the most useful property of the Fourier transformation – the exchange between multiplication and convolution – is mathematically the most elusive and the one which requires the greatest caution in order to avoid writing down meaningless expressions.

It is the unique merit of Schwartz's theory of distributions (Schwartz, 1966) that it affords complete control over all the troublesome phenomena which had previously forced mathematicians to settle for a piecemeal, fragmented theory of the Fourier transformation. By its ability to handle rigorously highly 'singular'

objects (especially δ-functions, their derivatives, their tensor products, their products with smooth functions, their translates and lattices of these translates), distribution theory can deal with all the major properties of the Fourier transformation as particular instances of a single basic result (the exchange between multiplication and convolution), *and* can at the same time accommodate the three previously distinct types of Fourier theories within a unique framework. This brings great simplification to matters of central importance in crystallography, such as the relations between

(*a*) periodization, and sampling or decimation;
(*b*) Shannon interpolation, and masking by an indicator function;
(*c*) section, and projection;
(*d*) differentiation, and multiplication by a monomial;
(*e*) translation, and phase shift.

All these properties become subsumed under the same theorem.

This striking synthesis comes at a slight price, which is the relative complexity of the notion of distribution. It is first necessary to establish the notion of topological vector space and to gain sufficient control (or, at least, understanding) over convergence behaviour in certain of these spaces. The key notion of *metrizability* cannot be circumvented, as it underlies most of the constructs and many of the proof techniques used in distribution theory. Most of Section 1.3.2.2 builds up to the fundamental result at the end of Section 1.3.2.2.6.2, which is basic to the definition of a distribution in Section 1.3.2.3.4 and to all subsequent developments.

The reader mostly interested in applications will probably want to reach this section by starting with his or her favourite topic in Section 1.3.4, and following the backward references to the relevant properties of the Fourier transformation, then to the proof of these properties, and finally to the definitions of the objects involved. Hopefully, he or she will then feel inclined to follow the forward references and thus explore the subject from the abstract to the practical. The books by Dieudonné (1969) and Lang (1965) are particularly recommended as general references for all aspects of analysis and algebra.

1.3.2.2. *Preliminary notions and notation*

Throughout this text, $\mathbb{R}$ will denote the set of real numbers, $\mathbb{Z}$ the set of rational (signed) integers and $\mathbb{N}$ the set of natural (unsigned) integers. The symbol $\mathbb{R}^n$ will denote the Cartesian product of n copies of $\mathbb{R}$:

$$\mathbb{R}^n = \mathbb{R} \times \ldots \times \mathbb{R} \quad (n \text{ times}, n \geq 1),$$

so that an element $\mathbf{x}$ of $\mathbb{R}^n$ is an n-tuple of real numbers:

$$\mathbf{x} = (x_1, \ldots, x_n).$$

Similar meanings will be attached to $\mathbb{Z}^n$ and $\mathbb{N}^n$.

The symbol $\mathbb{C}$ will denote the set of complex numbers. If $z \in \mathbb{C}$, its modulus will be denoted by $|z|$, its conjugate by $\bar{z}$ (not z^*), and its real and imaginary parts by $\mathscr{Re}\,(z)$ and $\mathscr{Im}\,(z)$:

$$\mathscr{Re}\,(z) = \tfrac{1}{2}(z + \bar{z}), \qquad \mathscr{Im}\,(z) = \frac{1}{2i}(z - \bar{z}).$$

If X is a finite set, then $|X|$ will denote the number of its elements. If mapping f sends an element x of set X to the element $f(x)$ of set Y, the notation

$$f : x \longmapsto f(x)$$

will be used; the plain arrow $\rightarrow$ will be reserved for denoting limits, as in

$$\lim_{\rho \to \infty} \left(1 + \frac{x}{p}\right)^p = e^x.$$

If X is any set and S is a subset of X, the *indicator function* χ_S of S is the real-valued function on X defined by

$$\chi_S(x) = 1 \quad \text{if } x \in S$$
$$= 0 \quad \text{if } x \notin S.$$

1.3.2.2.1. *Metric and topological notions in* $\mathbb{R}^n$

The set $\mathbb{R}^n$ can be endowed with the structure of a vector space of dimension n over $\mathbb{R}$, and can be made into a Euclidean space by treating its standard basis as an orthonormal basis and defining the Euclidean norm:

$$\|\mathbf{x}\| = \left(\sum_{i=1}^{n} x_i^2\right)^{1/2}.$$

By misuse of notation, $\mathbf{x}$ will sometimes also designate the column vector of coordinates of $\mathbf{x} \in \mathbb{R}^n$; if these coordinates are referred to an orthonormal basis of $\mathbb{R}^n$, then

$$\|\mathbf{x}\| = (\mathbf{x}^T\mathbf{x})^{1/2},$$

where $\mathbf{x}^T$ denotes the transpose of $\mathbf{x}$.

The distance between two points $\mathbf{x}$ and $\mathbf{y}$ defined by $d(\mathbf{x}, \mathbf{y}) = \|\mathbf{x} - \mathbf{y}\|$ allows the topological structure of $\mathbb{R}$ to be transferred to $\mathbb{R}^n$, making it a *metric space*. The basic notions in a metric space are those of neighbourhoods, of open and closed sets, of limit, of continuity, and of convergence (see Section 1.3.2.2.6.1).

A subset S of $\mathbb{R}^n$ is *bounded* if $\sup \|\mathbf{x} - \mathbf{y}\| < \infty$ as $\mathbf{x}$ and $\mathbf{y}$ run through S; it is *closed* if it contains the limits of all convergent sequences with elements in S. A subset K of $\mathbb{R}^n$ which is both bounded and closed has the property of being *compact*, *i.e.* that whenever K has been covered by a family of open sets, a finite subfamily can be found which suffices to cover K. Compactness is a very useful topological property for the purpose of proof, since it allows one to reduce the task of examining infinitely many local situations to that of examining only finitely many of them.

1.3.2.2.2. *Functions over* $\mathbb{R}^n$

Let φ be a complex-valued function over $\mathbb{R}^n$. The *support* of φ, denoted Supp φ, is the smallest closed subset of $\mathbb{R}^n$ outside which φ vanishes identically. If Supp φ is compact, φ is said to have compact support.

If $\mathbf{t} \in \mathbb{R}^n$, the *translate* of φ by $\mathbf{t}$, denoted $\tau_{\mathbf{t}}\varphi$, is defined by

$$(\tau_{\mathbf{t}}\varphi)(\mathbf{x}) = \varphi(\mathbf{x} - \mathbf{t}).$$

Its support is the geometric translate of that of φ:

$$\text{Supp } \tau_{\mathbf{t}}\varphi = \{\mathbf{x} + \mathbf{t} | \mathbf{x} \in \text{Supp } \varphi\}.$$

If A is a non-singular linear transformation in $\mathbb{R}^n$, the *image* of φ by A, denoted $A^{\#}\varphi$, is defined by

$$(A^{\#}\varphi)(\mathbf{x}) = \varphi[A^{-1}(\mathbf{x})].$$

Its support is the geometric image of Supp φ under A:

$$\text{Supp } A^{\#}\varphi = \{A(\mathbf{x}) | \mathbf{x} \in \text{Supp } \varphi\}.$$

If S is a non-singular affine transformation in $\mathbb{R}^n$ of the form

$$S(\mathbf{x}) = A(\mathbf{x}) + \mathbf{b}$$

with A linear, the image of φ by S is $S^{\#}\varphi = \tau_{\mathbf{b}}(A^{\#}\varphi)$, *i.e.*

$$(S^{\#}\varphi)(\mathbf{x}) = \varphi[A^{-1}(\mathbf{x} - \mathbf{b})].$$

Its support is the geometric image of Supp φ under S:

$$\text{Supp } S^{\#}\varphi = \{S(\mathbf{x}) | \mathbf{x} \in \text{Supp } \varphi\}.$$

It may be helpful to visualize the process of forming the image of a function by a geometric operation as consisting of applying that operation to the *graph* of that function, which is equivalent to