1.3.4.5. *Related applications*

1.3.4.5.1. *Helical diffraction*

The theory of diffraction by helical structures (Cochran *et al.*, 1952; Klug *et al.*, 1958) has played an important part in the study of polypeptides, of nucleic acids and of tobacco mosaic virus.

1.3.4.5.1.1. *Circular harmonic expansions in polar coordinates*

Let $f = f(x, y)$ be a reasonably regular function in two-dimensional real space. Going over to polar coordinates

$$x = r\cos\varphi \quad y = r\sin\varphi$$

and writing, by slight misuse of notation, $f(r, \varphi)$ for $f(r\cos\varphi, r\sin\varphi)$ we may use the periodicity of f with respect to φ to expand it as a Fourier series (Byerly, 1893):

$$f(r,\varphi) = \sum_{n\in\mathbb{Z}} f_n(r)\exp(in\varphi)$$

with

$$f_n(r) = \frac{1}{2\pi}\int_0^{2\pi} f(r,\varphi)\exp(-in\varphi)\ \mathrm{d}\varphi.$$

Similarly, in reciprocal space, if $F = F(\xi, \eta)$ and if

$$\xi = R\cos\psi \quad \eta = R\sin\psi$$

then

$$F(R,\psi) = \sum_{n\in\mathbb{Z}} i^n F_n(R)\exp(in\psi)$$

with

$$F_n(R) = \frac{1}{2\pi i^n}\int_0^{2\pi} F(R,\psi)\exp(-in\psi)\ \mathrm{d}\psi,$$

where the phase factor i^n has been introduced for convenience in the forthcoming step.

1.3.4.5.1.2. *The Fourier transform in polar coordinates*

The Fourier transform relation between f and F may then be written in terms of f_n's and F_n's. Observing that $\xi x + \eta y = Rr\cos(\varphi - \psi)$, and that (Watson, 1944)

$$\int_0^{2\pi} \exp(iX\cos\theta + in\theta)\ \mathrm{d}\theta = 2\pi i^n J_n(X),$$

we obtain:

$$\begin{aligned} F(R,\psi) &= \int_0^\infty\int_0^{2\pi}\left[\sum_{n\in\mathbb{Z}} f_n(r)\exp(in\varphi)\right] \\ &\quad\times \exp[2\pi iRr\cos(\varphi-\psi)]r\ \mathrm{d}r\ \mathrm{d}\varphi \\ &= \sum_{n\in\mathbb{Z}} i^n\left[\int_0^\infty f_n(r)J_n(2\pi Rr)2\pi r\ \mathrm{d}r\right]\exp(in\psi); \end{aligned}$$

hence, by the uniqueness of the Fourier expansion of F:

$$F_n(R) = \int_0^\infty f_n(r)J_n(2\pi Rr)2\pi r\ \mathrm{d}r.$$

The inverse Fourier relationship leads to

$$f_n(r) = \int_0^\infty F_n(R)J_n(2\pi rR)2\pi R\ \mathrm{d}R.$$

The integral transform involved in the previous two equations is called the *Hankel transform* (see *e.g.* Titchmarsh, 1922; Sneddon, 1972) of order n.

1.3.4.5.1.3. *The transform of an axially periodic fibre*

Let ρ be the electron-density distribution in a fibre, which is assumed to have translational periodicity with period 1 along z, and to have compact support with respect to the (x, y) coordinates. Thus ρ may be written

$$\rho = \left[\delta_x \otimes \delta_y \otimes \left(\sum_{k\in\mathbb{Z}}\delta_{(k)}\right)_z\right] * \rho^0,$$

where $\rho^0 = \rho^0(x, y, z)$ is the motif.

By the tensor product property, the inverse Fourier transform $F = \bar{\mathscr{F}}_{xyz}[\rho]$ may be written

$$F = \left[1_\xi \otimes 1_\eta \otimes \left(\sum_{l\in\mathbb{Z}}\delta_{(l)}\right)_\zeta\right] \times \bar{\mathscr{F}}[\rho^0]$$

and hence consists of 'layers' labelled by l:

$$F = \sum_{l\in\mathbb{Z}} F(\xi,\eta,l)(\delta_{(l)})_\zeta$$

with

$$F(\xi,\eta,l) = \int_0^1 \bar{\mathscr{F}}_{xy}[\rho^0](\xi,\eta,z)\exp(2\pi ilz)\ \mathrm{d}z.$$

Changing to polar coordinates in the (x, y) and (ξ, η) planes decomposes the calculation of F from ρ into the following steps:

$$\begin{aligned} g_{nl}(r) &= \frac{1}{2\pi}\int_0^{2\pi}\int_0^1 \rho(r,\varphi,z)\exp[i(-n\varphi + 2\pi lz)]\ \mathrm{d}\varphi\ \mathrm{d}z \\ G_{nl}(R) &= \int_0^\infty g_{nl}(r)J_n(2\pi Rr)2\pi r\ \mathrm{d}r \\ F(R,\psi,l) &= \sum_{n\in\mathbb{Z}} i^n G_{nl}(R)\exp(in\psi) \end{aligned}$$

and the calculation of ρ from F into:

$$\begin{aligned} G_{nl}(R) &= \frac{1}{2\pi i^n}\int_0^{2\pi} F(R,\psi,l)\exp(-in\psi)\ \mathrm{d}\psi \\ g_{nl}(r) &= \int_0^\infty G_{nl}(R)J_n(2\pi rR)2\pi R\ \mathrm{d}R \\ \rho(r,\varphi,z) &= \sum_{n\in\mathbb{Z}}\sum_{l\in\mathbb{Z}} g_{nl}(r)\exp[i(n\varphi - 2\pi lz)]. \end{aligned}$$

These formulae are seen to involve a 2D Fourier series with respect to the two periodic coordinates φ and z, and Hankel transforms along the radial coordinates. The two periodicities in φ and z are independent, so that all combinations of indices (n, l) occur in the Fourier summations.

1.3.4.5.1.4. *Helical symmetry and associated selection rules*

Helical symmetry involves a 'clutching' between the two (hitherto independent) periodicities in φ (period 2π) and z (period 1) which causes a subdivision of the period lattice and hence a decimation (governed by 'selection rules') of the Fourier coefficients.

Let $\mathbf{i}$ and $\mathbf{j}$ be the basis vectors along $\varphi/2\pi$ and z. The integer lattice with basis $(\mathbf{i}, \mathbf{j})$ is a period lattice for the (φ, z) dependence of the electron density ρ of an axially periodic fibre considered in Section 1.3.4.5.1.3:

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$$\rho(r, \varphi + 2\pi k_1, z + k_2) = \rho(r, \varphi, z).$$

Suppose the fibre now has *helical* symmetry, with u copies of the same molecule in t turns, where g.c.d. $(u, t) = 1$. Using the Euclidean algorithm, write $u = \lambda t + \mu$ with λ and μ positive integers and $\mu < t$. The period lattice for the (φ, z) dependence of ρ may be defined in terms of the new basis vectors:

I, joining subunit 0 to subunit l in the same turn;

J, joining subunit 0 to subunit λ after wrapping around.

In terms of the original basis

$$\mathbf{I} = \frac{t}{u}\mathbf{i} + \frac{1}{u}\mathbf{j}, \quad \mathbf{J} = \frac{-\mu}{u}\mathbf{i} + \frac{\lambda}{u}\mathbf{j}.$$

If α and β are coordinates along **I** and **J**, respectively,

$$\begin{pmatrix} \varphi/2\pi \\ z \end{pmatrix} = \frac{1}{u}\begin{pmatrix} t & -\mu \\ 1 & \lambda \end{pmatrix}\begin{pmatrix} \alpha \\ \beta \end{pmatrix}$$

or equivalently

$$\begin{pmatrix} \alpha \\ \beta \end{pmatrix} = \begin{pmatrix} \lambda & \mu \\ -1 & t \end{pmatrix}\begin{pmatrix} \varphi/2\pi \\ z \end{pmatrix}.$$

By Fourier transformation,

$$\left(\frac{\varphi}{2\pi}, z\right) \Leftrightarrow (-n, l)$$
$$(\alpha, \beta) \Leftrightarrow (m, p)$$

with the transformations between indices given by the contragredients of those between coordinates, *i.e.*

$$\begin{pmatrix} n \\ l \end{pmatrix} = \begin{pmatrix} -\lambda & 1 \\ -\mu & t \end{pmatrix}\begin{pmatrix} m \\ p \end{pmatrix}$$

and

$$\begin{pmatrix} m \\ p \end{pmatrix} = \frac{1}{u}\begin{pmatrix} -t & 1 \\ \mu & \lambda \end{pmatrix}\begin{pmatrix} n \\ l \end{pmatrix}.$$

It follows that

$$l = tn + um,$$

or alternatively that

$$\mu n = up - \lambda l,$$

which are two equivalent expressions of the *selection rules* describing the decimation of the transform. These rules imply that only certain orders n contribute to a given layer l.

The 2D Fourier analysis may now be performed by analysing a *single* subunit referred to coordinates α and β to obtain

$$h_{m,p}(r) = \int_0^1\int_0^1 \rho(r, \alpha, \beta)\exp[2\pi i(m\alpha + p\beta)]\ \mathrm{d}\alpha\ \mathrm{d}\beta$$

and then reindexing to get only the allowed g_{nl}'s by

$$g_{nl}(r) = uh_{-\lambda m+p,\ \mu m+tp}(r).$$

This is u times faster than analysing u subunits with respect to the (φ, z) coordinates.

1.3.4.5.2. *Application to probability theory and direct methods*

The Fourier transformation plays a central role in the branch of probability theory concerned with the limiting behaviour of sums of large numbers of independent and identically distributed random variables or random vectors. This privileged role is a consequence of the convolution theorem and of the 'moment-generating' properties which follow from the exchange between differentiation and multiplication by monomials. When the limit theorems are applied to the calculation of joint probability distributions of structure factors, which are themselves closely related to the Fourier transformation, a remarkable phenomenon occurs, which leads to the saddlepoint approximation and to the maximum-entropy method.

1.3.4.5.2.1. *Analytical methods of probability theory*

The material in this section is not intended as an introduction to probability theory [for which the reader is referred to Cramér (1946), Petrov (1975) or Bhattacharya & Rao (1976)], but only as an illustration of the role played by the Fourier transformation in certain specific areas which are used in formulating and implementing direct methods of phase determination.

(*a*) *Convolution of probability densities*

The addition of independent random variables or vectors leads to the convolution of their probability distributions: if $\mathbf{X}_1$ and $\mathbf{X}_2$ are two n-dimensional random vectors independently distributed with probability densities P_1 and P_2, respectively, then their sum $\mathbf{X} = \mathbf{X}_1 + \mathbf{X}_2$ has probability density $\mathscr{P}$ given by

$$\begin{aligned}\mathscr{P}(\mathbf{X}) &= \int_{\mathbf{R}^n} P_1(\mathbf{X}_1)P_2(\mathbf{X} - \mathbf{X}_1)\ \mathrm{d}^n\mathbf{X}_1 \\ &= \int_{\mathbf{R}^n} P_1(\mathbf{X} - \mathbf{X}_2)P_2(\mathbf{X}_2)\ \mathrm{d}^n\mathbf{X}_2\end{aligned}$$

i.e.

$$\mathscr{P} = P_1 * P_2.$$

This result can be extended to the case where P_1 and P_2 are singular measures (distributions of order zero, Section 1.3.2.3.4) and do not have a density with respect to the Lebesgue measure in $\mathbb{R}^n$.

(*b*) *Characteristic functions*

This convolution can be turned into a simple multiplication by considering the Fourier transforms (called the *characteristic functions*) of P_1, P_2 and $\mathscr{P}$, defined with a slightly different normalization in that there is no factor of 2π in the exponent (see Section 1.3.2.4.5), *e.g.*

$$C(\mathbf{t}) = \int_{\mathbf{R}^n} P(\mathbf{X})\exp(i\mathbf{t}\cdot\mathbf{X})\ \mathrm{d}^n\mathbf{X}.$$

Then by the convolution theorem

$$\mathscr{C}(\mathbf{t}) = C_1(\mathbf{t}) \times C_2(\mathbf{t}),$$

so that $\mathscr{P}(\mathbf{X})$ may be evaluated by Fourier inversion of its characteristic function as

$$\mathscr{P}(\mathbf{X}) = \frac{1}{(2\pi)^n}\int_{\mathbb{R}^n} C_1(\mathbf{t})C_2(\mathbf{t})\exp(-i\mathbf{t}\cdot\mathbf{X})\ \mathrm{d}^n\mathbf{t}$$

(see Section 1.3.2.4.5 for the normalization factors).

It follows from the differentiation theorem that the partial derivatives of the characteristic function $C(\mathbf{t})$ at $\mathbf{t} = \mathbf{0}$ are related to the moments of a distribution $\boldsymbol{P}$ by the identities

$$\begin{aligned}\mu_{r_1 r_2 \ldots r_n} &\equiv \int_D P(\mathbf{X})X_1^{r_1}X_2^{r_2}\ldots X_n^{r_n}\ \mathrm{d}^n\mathbf{X} \\ &= i^{-(r_1+\ldots+r_n)}\frac{\partial^{r_1+\ldots+r_n}C}{\partial t_1^{r_1}\ldots\partial t_n^{r_n}}\bigg|_{\mathbf{t}=\mathbf{0}}\end{aligned}$$

for any n-tuple of non-negative integers $(r_1, r_2, \ldots, r_n)$.