$$\chi_M \times \wp^0 = \wp^0.$$

It then follows that $\wp = r * (\chi_M \times \wp)$. Defining the 'interference function' G as the normalized indicator function of M according to

$$G(\boldsymbol{\eta}) = \frac{1}{\mathrm{vol}(M)} \bar{\mathscr{F}}[\chi_M](\boldsymbol{\eta})$$

we may invoke Shannon's theorem to calculate the value $\bar{\mathscr{F}}[\wp^0](\boldsymbol{\xi})$ at an arbitrary point $\boldsymbol{\xi}$ of reciprocal space from its sample values $F(\mathbf{h}) = \bar{\mathscr{F}}[\wp^0](\mathbf{h})$ at points of the reciprocal lattice as

$$\bar{\mathscr{F}}[\wp^0](\boldsymbol{\xi}) = \sum_{\mathbf{h}\in\mathbb{Z}^3} \mathbf{G}(\boldsymbol{\xi} - \mathbf{h}) F(\mathbf{h}).$$

This aspect of Shannon's theorem constitutes the mathematical basis of phasing methods based on geometric redundancies created by solvent regions and/or noncrystallographic symmetries (Bricogne, 1974). The connection between Shannon's theorem and the phase problem was first noticed by Sayre (1952*b*). He pointed out that the Patterson function of $\wp$, written as $\kappa[\wp, \wp] = r * (\check{\wp}^0 * \wp^0)$, may be viewed as consisting of a motif $\kappa^0 = \check{\wp}^0 * \wp^0$ (containing all the internal interatomic vectors) which is periodized by convolution with r. As the translates of κ^0 by vectors of $\mathbb{Z}^3$ *do* overlap, the sample values of the intensities $|F(\mathbf{h})|^2$ at nodes of the reciprocal lattice do not provide enough data to interpolate intensities $|F(\boldsymbol{\xi})|^2$ at arbitrary points of reciprocal space. Thus the loss of phase is intimately related to the impossibility of intensity interpolation, implying in return that any indication of intensity values attached to non-integral points of the reciprocal lattice is a potential source of phase information.

1.3.4.2.1.8. *Sections and projections*

It was shown at the end of Section 1.3.2.5.8 that the convolution theorem establishes, under appropriate assumptions, a duality between sectioning a smooth function (viewed as a multiplication by a δ-function in the sectioning coordinate) and projecting its transform (viewed as a convolution with the function **1** everywhere equal to 1 as a function of the projection coordinate). This duality follows from the fact that $\mathscr{F}$ and $\bar{\mathscr{F}}$ map $\mathbf{1}_{x_i}$ to δ_{x_i} and δ_{x_i} to $\mathbf{1}_{x_i}$ (Section 1.3.2.5.6), and from the tensor product property (Section 1.3.2.5.5).

In the case of periodic distributions, projection and section must be performed with respect to directions or subspaces which are integral with respect to the period lattice if the result is to be periodic; furthermore, projections must be performed only on the contents of *one* repeating unit along the direction of projection, or else the result would diverge. The same relations then hold between principal central sections and projections of the electron density and the dual principal central projections and sections of the weighted reciprocal lattice, *e.g.*

$$\wp(x_1, 0, 0) \leftrightarrow \sum_{h_1,\, h_2} F(h_1, h_2, h_3),$$

$$\wp(x_1, x_2, 0) \leftrightarrow \sum_{h_3} F(h_1, h_2, h_3),$$

$$\wp_{1,\,2}(x_3) = \int_{\mathbb{R}^2/\mathbb{Z}^2} \wp(x_1, x_2, x_3)\, \mathrm{d}x_1\, \mathrm{d}x_2 \leftrightarrow F(0, 0, h_3),$$

$$\wp_1(x_2, x_3) = \int_{\mathbb{R}/\mathbb{Z}} \wp(x_1, x_2, x_3)\, \mathrm{d}x_1 \qquad \leftrightarrow F(0, h_2, h_3)$$

etc.

When the sections are principal but not central, it suffices to use the shift property of Section 1.3.2.5.5. When the sections or projections are not principal, they can be made principal by changing to new primitive bases B and B^* for Λ and Λ^*, respectively, the transition matrices $\mathbf{P}$ and $\mathbf{P}^*$ to these new bases being related by $\mathbf{P}^* = (\mathbf{P}^{-1})^T$ in order to preserve duality. This change of basis must be such that one of these matrices (say, $\mathbf{P}$) should have a given integer vector $\mathbf{u}$ as its first column, $\mathbf{u}$ being related to the line or plane defining the section or projection of interest.

The problem of constructing a matrix $\mathbf{P}$ given $\mathbf{u}$ received an erroneous solution in Volume II of *International Tables* (Patterson, 1959), which was subsequently corrected in 1962. Unfortunately, the solution proposed there is complicated and does not suggest a general approach to the problem. It therefore seems worthwhile to record here an effective procedure which solves this problem in any dimension n (Watson, 1970).

Let

$$\mathbf{u} = \begin{pmatrix} u_1 \\ \vdots \\ u_n \end{pmatrix}$$

be a primitive integral vector, *i.e.* g.c.d. $(u_1, \ldots, u_n) = 1$. Then an $n \times n$ integral matrix $\mathbf{P}$ with $\det \mathbf{P} = 1$ having $\mathbf{u}$ as its first column can be constructed by induction as follows. For $n = 1$ the result is trivial. For $n = 2$ it can be solved by means of the Euclidean algorithm, which yields z_1, z_2 such that $u_1 z_2 - u_2 z_1 = 1$, so that we may take $\mathbf{P} = \begin{pmatrix} u_1 & z_1 \\ u_2 & z_2 \end{pmatrix}$. Note that, if $\mathbf{z} = \begin{pmatrix} z_1 \\ z_2 \end{pmatrix}$ is a solution, then $\mathbf{z} + m\mathbf{u}$ is another solution for any $m \in \mathbb{Z}$. For $n \geq 3$, write $\mathbf{u} = \begin{pmatrix} u_1 \\ d\mathbf{z} \end{pmatrix}$ with $d =$ g.c.d. $(u_2, \ldots, u_n)$ so that both $\mathbf{z} = \begin{pmatrix} z_2 \\ \vdots \\ z_n \end{pmatrix}$ and $\begin{pmatrix} u_1 \\ d \end{pmatrix}$ are primitive. By the inductive hypothesis there is an integral 2×2 matrix $\mathbf{V}$ with $\begin{pmatrix} u_1 \\ d \end{pmatrix}$ as its first column, and an integral $(n-1) \times (n-1)$ matrix $\mathbf{Z}$ with $\mathbf{z}$ as its first column, with $\det \mathbf{V} = 1$ and $\det \mathbf{Z} = 1$.

Now put

$$\mathbf{P} = \begin{pmatrix} 1 & \\ & \mathbf{Z} \end{pmatrix} \begin{pmatrix} \mathbf{V} & \\ & \mathbf{I}_{n-2} \end{pmatrix},$$

i.e.

$$\mathbf{P} = \begin{pmatrix} 1 & 0 & 0 & . & 0 \\ 0 & z_2 & * & . & * \\ 0 & z_3 & * & . & * \\ . & . & . & . & . \\ 0 & z_n & * & . & * \end{pmatrix} \begin{pmatrix} u_1 & * & 0 & . & 0 \\ d & * & 0 & . & 0 \\ 0 & 0 & 1 & . & 0 \\ . & . & . & . & . \\ 0 & 0 & 0 & . & 1 \end{pmatrix}.$$

The first column of $\mathbf{P}$ is

$$\begin{pmatrix} u_1 \\ dz_2 \\ . \\ . \\ dz_n \end{pmatrix} = \mathbf{u},$$

and its determinant is 1, QED.

The incremental step from dimension $n-1$ to dimension n is the construction of 2×2 matrix $\mathbf{V}$, for which there exist infinitely many solutions labelled by an integer m_{n-1}. Therefore, the collection of matrices $\mathbf{P}$ which solve the problem is labelled by $n-1$ arbitrary integers $(m_1, m_2, \ldots, m_{n-1})$. This freedom can be used to adjust the shape of the basis B.

Once $\mathbf{P}$ has been chosen, the calculation of general sections and projections is transformed into that of *principal* sections and projections by the changes of coordinates:

$$\mathbf{x} = \mathbf{P}\mathbf{x}', \qquad \mathbf{h} = \mathbf{P}^*\mathbf{h}',$$

and an appeal to the tensor product property.

Booth (1945*a*) made use of the convolution theorem to form the Fourier coefficients of 'bounded projections', which provided a compromise between 2D and 3D Fourier syntheses. If it is desired to compute the projection on the (x, y) plane of the electron density lying between the planes $z = z_1$ and $z = z_2$, which may be written as

$$[\rho \times (\mathbf{1}_x \otimes \mathbf{1}_y \otimes \chi_{[z_1,\, z_2]})] * (\delta_x \otimes \delta_y \otimes \mathbf{1}_z).$$

The transform is then

$$[F * (\delta_h \otimes \delta_k \otimes \bar{\mathscr{F}}[\chi_{[z_1,\, z_2]}])] \times (\mathbf{1}_h \otimes \mathbf{1}_k \otimes \delta_l),$$

giving for coefficient (h, k):

$$\sum_{l\in\mathbb{Z}} F(h,k,l)\exp\{2\pi i l[(z_1+z_2)/2]\} \times \frac{\sin \pi l(z_1 - z_2)}{\pi l}.$$

1.3.4.2.1.9. *Differential syntheses*

Another particular instance of the convolution theorem is the duality between differentiation and multiplication by a monomial (Sections 1.3.2.4.2.8, 1.3.2.5.8).

In the present context, this result may be written

$$\bar{\mathscr{F}}\left[\frac{\partial^{m_1+m_2+m_3}\rho}{\partial X_1^{m_1}\partial X_2^{m_2}\partial X_3^{m_3}}\right](\mathbf{H}) = (-2\pi i)^{m_1+m_2+m_3} H_1^{m_1} H_2^{m_2} H_3^{m_3} F(\mathbf{A}^T\mathbf{H})$$

in Cartesian coordinates, and

$$\bar{\mathscr{F}}\left[\frac{\partial^{m_1+m_2+m_3}\rho}{\partial x_1^{m_1}\partial x_2^{m_2}\partial x_3^{m_3}}\right](\mathbf{h}) = (-2\pi i)^{m_1+m_2+m_3} h_1^{m_1} h_2^{m_2} h_3^{m_3} F(\mathbf{h})$$

in crystallographic coordinates.

A particular case of the first formula is

$$-4\pi^2 \sum_{\mathbf{H}\in\Lambda^*} \|\mathbf{H}\|^2 F(\mathbf{A}^T\mathbf{H})\exp(-2\pi i\mathbf{H}\cdot\mathbf{X}) = \Delta\rho(\mathbf{X}),$$

where

$$\Delta\rho = \sum_{j=1}^{3} \frac{\partial^2\rho}{\partial X_j^2}$$

is the Laplacian of ρ.

The second formula has been used with $|\mathbf{m}| = 1$ or 2 to compute 'differential syntheses' and refine the location of maxima (or other stationary points) in electron-density maps. Indeed, the values at $\mathbf{x}$ of the gradient vector $\nabla\rho$ and Hessian matrix $(\nabla\nabla^T)\rho$ are readily obtained as

$$(\nabla\rho)(\mathbf{x}) = \sum_{\mathbf{h}\in\mathbb{Z}^3} (-2\pi i\mathbf{h}) F(\mathbf{h})\exp(-2\pi i\mathbf{h}\cdot\mathbf{x}),$$

$$[(\nabla\nabla^T)\rho](\mathbf{x}) = \sum_{\mathbf{h}\in\mathbb{Z}^3} (-4\pi^2\mathbf{h}\mathbf{h}^T) F(\mathbf{h})\exp(-2\pi i\mathbf{h}\cdot\mathbf{x}),$$

and a step of Newton iteration towards the nearest stationary point of ρ will proceed by

$$\mathbf{x} \longmapsto \mathbf{x} - \{[(\nabla\nabla^T)\rho](\mathbf{x})\}^{-1}(\nabla\rho)(\mathbf{x}).$$

The modern use of Fourier transforms to speed up the computation of derivatives for model refinement will be described in Section 1.3.4.4.7.

The converse property is also useful: it relates the derivatives of the continuous transform $\bar{\mathscr{F}}[\rho^0]$ to the moments of ρ^0:

$$\frac{\partial^{m_1+m_2+m_3}\bar{\mathscr{F}}[\rho^0]}{\partial X_1^{m_1}\partial X_2^{m_2}\partial X_3^{m_3}}(\mathbf{H}) = \bar{\mathscr{F}}[(2\pi i)^{m_1+m_2+m_3} X_1^{m_1} X_2^{m_2} X_3^{m_3}\rho^0_{\mathbf{x}}](\mathbf{H}).$$

For $|\mathbf{m}| = 2$ and $\mathbf{H} = \mathbf{0}$, this identity gives the well known relation between the Hessian matrix of the transform $\bar{\mathscr{F}}[\rho^0]$ at the origin of reciprocal space and the inertia tensor of the motif ρ^0. This is a particular case of the moment-generating properties of $\bar{\mathscr{F}}$, which will be further developed in Section 1.3.4.5.2.

1.3.4.2.1.10. *Toeplitz forms, determinantal inequalities and Szegö's theorem*

The classical results presented in Section 1.3.2.6.9 can be readily generalized to the case of triple Fourier series; no new concept is needed, only an obvious extension of the notation.

Let ρ be real-valued, so that Friedel's law holds and $F(-\mathbf{h}) = \overline{F(\mathbf{h})}$. Let H be a finite set of indices comprising the origin: $\mathsf{H} = \{\mathbf{h}_0 = \mathbf{0}, \mathbf{h}_1, \ldots, \mathbf{h}_n\}$. Then the Hermitian form in $n+1$ complex variables

$$T_{\mathsf{H}}[\rho](\mathbf{u}) = \sum_{j,\ k=0}^{n} F(\mathbf{h}_j - \mathbf{h}_k)\overline{u}_j u_k$$

is called the Toeplitz form of order H associated to ρ. By the convolution theorem and Parseval's identity,

$$T_{\mathsf{H}}[\rho](\mathbf{u}) = \int_{\mathbb{R}^3/\mathbb{Z}^3} \rho(\mathbf{x}) \left|\sum_{j=0}^{n} u_j \exp(2\pi i\mathbf{h}_j\cdot\mathbf{x})\right|^2 \mathrm{d}^3\mathbf{x}.$$

If ρ is almost everywhere non-negative, then for all H the forms $T_{\mathsf{H}}[\rho]$ are positive semi-definite and therefore all Toeplitz determinants $D_{\mathsf{H}}[\rho]$ are non-negative, where

$$D_{\mathsf{H}}[\rho] = \det\{[F(\mathbf{h}_j - \mathbf{h}_k)]\}.$$

The Toeplitz–Carathéodory–Herglotz theorem given in Section 1.3.2.6.9.2 states that the converse is true: if $D_{\mathsf{H}}[\rho] \geq 0$ for all H, then ρ is almost everywhere non-negative. This result is known in the crystallographic literature through the papers of Karle & Hauptman (1950), MacGillavry (1950), and Goedkoop (1950), following previous work by Harker & Kasper (1948) and Gillis (1948*a*,*b*).

Szegö's study of the asymptotic distribution of the eigenvalues of Toeplitz forms as their order tends to infinity remains valid. Some precautions are needed, however, to define the notion of a sequence (H_k) of finite subsets of indices tending to infinity: it suffices that the H_k should consist essentially of the reciprocal-lattice points $\mathbf{h}$ contained within a domain of the form $k\Omega$ (k-fold dilation of Ω) where Ω is a convex domain in $\mathbb{R}^3$ containing the origin (Widom, 1960). Under these circumstances, the eigenvalues $\lambda_\nu^{(n)}$ of the Toeplitz forms $T_{\mathsf{H}_k}[\rho]$ become equidistributed with the sample values $\rho_{\nu'}^{(n)}$ of ρ on a grid satisfying the Shannon sampling criterion for the data in H_k (*cf.* Section 1.3.2.6.9.3).

A particular consequence of this equidistribution is that the geometric means of the $\lambda_\nu^{(n)}$ and of the $\rho_{\nu'}^{(n)}$ are equal, and hence as in Section 1.3.2.6.9.4

$$\lim_{k\to\infty}\{D_{\mathsf{H}_k}[\rho]\}^{1/|\mathsf{H}_k|} = \exp\left\{\int_{\mathbb{R}^3/\mathbb{Z}^3} \log\rho(\mathbf{x})\ \mathrm{d}^3\mathbf{x}\right\},$$

where $|\mathsf{H}_k|$ denotes the number of reflections in H_k. Complementary terms giving a better comparison of the two sides were obtained by Widom (1960, 1975) and Linnik (1975).

references