International Tables for Crystallography (2006). Vol. B, Section 1.3.2.6.7, pp. 42–43.

$$\text{(i)} \qquad w_{\boldsymbol{\mu}} = \langle T^0_{\mathbf{x}}, \exp(-2\pi i \boldsymbol{\mu} \cdot \mathbf{x})\rangle$$

$$\text{(ii)} \qquad T_{\mathbf{x}} = \sum_{\boldsymbol{\mu} \in \mathbb{Z}^n} w_{\boldsymbol{\mu}} \exp(+2\pi i \boldsymbol{\mu} \cdot \mathbf{x})$$

are referred to as the *Fourier analysis* and the *Fourier synthesis* of T, respectively (there is a discrepancy between this terminology and the crystallographic one, see Section 1.3.4.2.1.1). In other words, any periodic distribution $T \in \mathscr{S}'$ may be represented by a Fourier series (ii), whose coefficients are calculated by (i). The convergence of (ii) towards T in $\mathscr{S}'$ will be investigated later (Section 1.3.2.6.10).

1.3.2.6.5. *The case of non-standard period lattices*

Let Λ denote the non-standard lattice consisting of all vectors of the form $\sum_{j=1} m_j \mathbf{a}_j$, where the m_j are rational integers and $\mathbf{a}_1, \ldots, \mathbf{a}_n$ are n linearly independent vectors in $\mathbb{R}^n$. Let R be the corresponding lattice distribution: $R = \sum_{x \in \Lambda} \delta_{(\mathbf{x})}$.

Let $\mathbf{A}$ be the non-singular $n \times n$ matrix whose successive columns are the coordinates of vectors $\mathbf{a}_1, \ldots, \mathbf{a}_n$ in the standard basis of $\mathbb{R}^n$; $\mathbf{A}$ will be called the *period matrix* of Λ, and the mapping $\mathbf{x} \longmapsto \mathbf{Ax}$ will be denoted by A. According to Section 1.3.2.3.9.5 we have

$$\langle R, \varphi\rangle = \sum_{\mathbf{m} \in \mathbb{Z}^n} \varphi(\mathbf{Am}) = \langle r, (A^{-1})^{\#}\varphi\rangle = |\det \mathbf{A}|^{-1} \langle A^{\#} r, \varphi\rangle$$

for any $\varphi \in \mathscr{S}$, and hence $R = |\det \mathbf{A}|^{-1} A^{\#} r$. By Fourier transformation, according to Section 1.3.2.5.5,

$$\mathscr{F}[R] = |\det \mathbf{A}|^{-1} \mathscr{F}[A^{\#} r] = [(\mathbf{A}^{-1})^T]^{\#} \mathscr{F}[r] = [(\mathbf{A}^{-1})^T]^{\#} r,$$

which we write:

$$\mathscr{F}[R] = |\det \mathbf{A}|^{-1} R^*$$

with

$$R^* = |\det \mathbf{A}| [(\mathbf{A}^{-1})^T]^{\#} r.$$

R^* is a lattice distribution:

$$R^* = \sum_{\boldsymbol{\mu} \in \mathbb{Z}^n} \delta_{[(\mathbf{A}^{-1})^T \boldsymbol{\mu}]} = \sum_{\boldsymbol{\xi} \in \Lambda^*} \delta_{(\boldsymbol{\xi})}$$

associated with the *reciprocal lattice* Λ^* whose basis vectors $\mathbf{a}_1^*, \ldots, \mathbf{a}_n^*$ are the columns of $(\mathbf{A}^{-1})^T$. Since the latter matrix is equal to the adjoint matrix (*i.e.* the matrix of co-factors) of $\mathbf{A}$ divided by $\det \mathbf{A}$, the components of the reciprocal basis vectors can be written down explicitly (see Section 1.3.4.2.1.1 for the crystallographic case $n = 3$).

A distribution T will be called Λ-*periodic* if $\tau_{\boldsymbol{\xi}} T = T$ for all $\boldsymbol{\xi} \in \Lambda$; as previously, T may be written $R * T^0$ for some motif distribution T^0 with compact support. By Fourier transformation,

$$\begin{aligned} \mathscr{F}[T] &= |\det \mathbf{A}|^{-1} R^* \cdot \mathscr{F}[T^0] \\ &= |\det \mathbf{A}|^{-1} \sum_{\boldsymbol{\xi} \in \Lambda^*} \mathscr{F}[T^0](\boldsymbol{\xi}) \delta_{(\boldsymbol{\xi})} \\ &= |\det \mathbf{A}|^{-1} \sum_{\boldsymbol{\mu} \in \mathbb{Z}^n} \mathscr{F}[T^0][(\mathbf{A}^{-1})^T \boldsymbol{\mu}] \delta_{[(\mathbf{A}^{-1})^T \boldsymbol{\mu}]} \end{aligned}$$

so that $\mathscr{F}[T]$ is a weighted reciprocal-lattice distribution, the weight attached to node $\boldsymbol{\xi} \in \Lambda^*$ being $|\det \mathbf{A}|^{-1}$ times the value $\mathscr{F}[T^0](\boldsymbol{\xi})$ of the Fourier transform of the motif T^0.

This result may be further simplified if T and its motif T^0 are referred to the standard period lattice $\mathbb{Z}^n$ by defining t and t^0 so that $T = A^{\#} t$, $T^0 = A^{\#} t^0$, $t = r * t^0$. Then

$$\mathscr{F}[T^0](\boldsymbol{\xi}) = |\det \mathbf{A}| \mathscr{F}[t^0](\mathbf{A}^T \boldsymbol{\xi}),$$

hence

$$\mathscr{F}[T^0][(\mathbf{A}^{-1})^T \boldsymbol{\mu}] = |\det \mathbf{A}| \mathscr{F}[t^0](\boldsymbol{\mu}),$$

so that

$$\mathscr{F}[T] = \sum_{\boldsymbol{\mu} \in \mathbb{Z}^n} \mathscr{F}[t^0](\boldsymbol{\mu}) \delta_{[(\mathbf{A}^{-1})^T \boldsymbol{\mu}]}$$

in non-standard coordinates, while

$$\mathscr{F}[t] = \sum_{\boldsymbol{\mu} \in \mathbb{Z}^n} \mathscr{F}[t^0](\boldsymbol{\mu}) \delta_{(\boldsymbol{\mu})}$$

in standard coordinates.

The reciprocity theorem may then be written:

$$\text{(iii)} \qquad W_{\boldsymbol{\xi}} = |\det \mathbf{A}|^{-1} \langle T^0_{\mathbf{x}}, \exp(-2\pi i \boldsymbol{\xi} \cdot \mathbf{x})\rangle, \quad \boldsymbol{\xi} \in \boldsymbol{\Lambda}^*$$

$$\text{(iv)} \qquad T_{\mathbf{x}} = \sum_{\boldsymbol{\xi} \in \Lambda^*} W_{\boldsymbol{\xi}} \exp(+2\pi i \boldsymbol{\xi} \cdot \mathbf{x})$$

in non-standard coordinates, or equivalently:

$$\text{(v)} \qquad w_{\boldsymbol{\mu}} = \langle t^0_{\mathbf{x}}, \exp(-2\pi i \boldsymbol{\mu} \cdot \mathbf{x})\rangle, \quad \boldsymbol{\mu} \in \mathbb{Z}^n$$

$$\text{(vi)} \qquad t_{\mathbf{x}} = \sum_{\boldsymbol{\mu} \in \mathbb{Z}^n} w_{\boldsymbol{\mu}} \exp(+2\pi i \boldsymbol{\mu} \cdot \mathbf{x})$$

in standard coordinates. It gives an n-dimensional Fourier series representation for any periodic distribution over $\mathbb{R}^n$. The convergence of such series in $\mathscr{S}'(\mathbb{R}^n)$ will be examined in Section 1.3.2.6.10.

1.3.2.6.6. *Duality between periodization and sampling*

Let T^0 be a distribution with compact support (the 'motif'). Its Fourier transform $\bar{\mathscr{F}}[T^0]$ is analytic (Section 1.3.2.5.4) and may thus be used as a multiplier.

We may rephrase the preceding results as follows:

(i) if T^0 is 'periodized by R' to give $R * T^0$, then $\bar{\mathscr{F}}[T^0]$ is 'sampled by R^*' to give $|\det \mathbf{A}|^{-1} R^* \cdot \bar{\mathscr{F}}[T^0]$;

(ii) if $\bar{\mathscr{F}}[T^0]$ is 'sampled by R^*' to give $R^* \cdot \bar{\mathscr{F}}[T^0]$, then T^0 is 'periodized by R' to give $|\det \mathbf{A}| R * T^0$.

Thus the Fourier transformation establishes a duality between the periodization of a distribution by a period lattice Λ and the sampling of its transform at the nodes of lattice Λ^* reciprocal to Λ. This is a particular instance of the convolution theorem of Section 1.3.2.5.8.

At this point it is traditional to break the symmetry between $\mathscr{F}$ and $\bar{\mathscr{F}}$ which distribution theory has enabled us to preserve even in the presence of periodicity, and to perform two distinct identifications:

(i) a Λ-periodic distribution T will be handled as a distribution $\tilde{T}$ on $\mathbb{R}^n/\Lambda$, was done in Section 1.3.2.6.3;

(ii) a weighted lattice distribution $W = \sum_{\boldsymbol{\mu} \in \mathbb{Z}^n} W_{\boldsymbol{\mu}} \delta_{[(\mathbf{A}^{-1})^T \boldsymbol{\mu}]}$ will be identified with the collection $\{W_{\boldsymbol{\mu}} | \boldsymbol{\mu} \in \mathbb{Z}^n\}$ of its n-tuply indexed coefficients.

1.3.2.6.7. *The Poisson summation formula*

Let $\varphi \in \mathscr{S}$, so that $\mathscr{F}[\varphi] \in \mathscr{S}$. Let R be the lattice distribution associated to lattice Λ, with period matrix $\mathbf{A}$, and let R^* be associated to the reciprocal lattice Λ^*. Then we may write:

$$\begin{aligned} \langle R, \varphi\rangle &= \langle R, \bar{\mathscr{F}}[\mathscr{F}[\varphi]]\rangle \\ &= \langle \bar{\mathscr{F}}[R], \mathscr{F}[\varphi]\rangle \\ &= |\det \mathbf{A}|^{-1} \langle R^*, \mathscr{F}[\varphi]\rangle \end{aligned}$$

i.e.

$$\sum_{\mathbf{x}\in\Lambda}\varphi(\mathbf{x}) = |\det \mathbf{A}|^{-1}\sum_{\boldsymbol{\xi}\in\Lambda^*}\mathscr{F}[\varphi](\boldsymbol{\xi}).$$

This identity, which also holds for $\bar{\mathscr{F}}$, is called the *Poisson summation formula*. Its usefulness follows from the fact that the speed of decrease at infinity of φ and $\mathscr{F}[\varphi]$ are inversely related (Section 1.3.2.4.4.3), so that if one of the series (say, the left-hand side) is slowly convergent, the other (say, the right-hand side) will be rapidly convergent. This procedure has been used by Ewald (1921) [see also Bertaut (1952), Born & Huang (1954)] to evaluate lattice sums (Madelung constants) involved in the calculation of the internal electrostatic energy of crystals (see Chapter 3.4 in this volume on convergence acceleration techniques for crystallographic lattice sums).

When φ is a multivariate Gaussian

$$\varphi(\mathbf{x}) = G_{\mathbf{B}}(\mathbf{x}) = \exp(-\tfrac{1}{2}\mathbf{x}^T\mathbf{B}\mathbf{x}),$$

then

$$\mathscr{F}[\varphi](\boldsymbol{\xi}) = |\det(2\pi\mathbf{B}^{-1})|^{1/2}G_{\mathbf{B}^{-1}}(\boldsymbol{\xi}),$$

and Poisson's summation formula for a lattice with period matrix $\mathbf{A}$ reads:

$$\sum_{\mathbf{m}\in\mathbb{Z}^n}G_{\mathbf{B}}(\mathbf{A}\mathbf{m}) = |\det \mathbf{A}|^{-1}|\det(2\pi\mathbf{B}^{-1})|^{1/2} \times \sum_{\boldsymbol{\mu}\in\mathbb{Z}^n}G_{4\pi^2\mathbf{B}^{-1}}[(\mathbf{A}^{-1})^T\boldsymbol{\mu}]$$

or equivalently

$$\sum_{\mathbf{m}\in\mathbb{Z}^n}G_C(\mathbf{m}) = |\det(2\pi\mathbf{C}^{-1})|^{1/2}\sum_{\boldsymbol{\mu}\in\mathbb{Z}^n}G_{4\pi^2\mathbf{C}^{-1}}(\boldsymbol{\mu})$$

with $\mathbf{C} = \mathbf{A}^T\mathbf{B}\mathbf{A}$.

1.3.2.6.8. *Convolution of Fourier series*

Let $S = R * S^0$ and $T = R * T^0$ be two Λ-periodic distributions, the motifs S^0 and T^0 having compact support. The convolution $S * T$ does not exist, because S and T do not satisfy the support condition (Section 1.3.2.3.9.7). However, the three distributions R, S^0 and T^0 do satisfy the generalized support condition, so that their convolution is defined; then, by associativity and commutativity:

$$R * S^0 * T^0 = S * T^0 = S^0 * T.$$

By Fourier transformation and by the convolution theorem:

$$\begin{aligned} R^* \times \mathscr{F}[S^0 * T^0] &= (R^* \times \mathscr{F}[S^0]) \times \mathscr{F}[T^0] \\ &= \mathscr{F}[T^0] \times (R^* \times \mathscr{F}[S^0]). \end{aligned}$$

Let $\{U_{\boldsymbol{\xi}}\}_{\boldsymbol{\xi}\in\Lambda^*}$, $\{V_{\boldsymbol{\xi}}\}_{\boldsymbol{\xi}\in\Lambda^*}$ and $\{W_{\boldsymbol{\xi}}\}_{\boldsymbol{\xi}\in\Lambda^*}$ be the sets of Fourier coefficients associated to S, T and $S * T^0 (= S^0 * T)$, respectively. Identifying the coefficients of $\delta_{\boldsymbol{\xi}}$ for $\boldsymbol{\xi} \in \Lambda^*$ yields the forward version of the convolution theorem for Fourier series:

$$W_{\boldsymbol{\xi}} = |\det \mathbf{A}|U_{\boldsymbol{\xi}}V_{\boldsymbol{\xi}}.$$

The backward version of the theorem requires that T be infinitely differentiable. The distribution $S \times T$ is then well defined and its Fourier coefficients $\{Q_{\boldsymbol{\xi}}\}_{\boldsymbol{\xi}\in\Lambda^*}$ are given by

$$Q_{\boldsymbol{\xi}} = \sum_{\boldsymbol{\eta}\in\Lambda^*}U_{\boldsymbol{\eta}}V_{\boldsymbol{\xi}-\boldsymbol{\eta}}.$$

1.3.2.6.9. *Toeplitz forms, Szegö's theorem*

Toeplitz forms were first investigated by Toeplitz (1907, 1910, 1911*a*). They occur in connection with the 'trigonometric moment problem' (Shohat & Tamarkin, 1943; Akhiezer, 1965) and probability theory (Grenander, 1952) and play an important role in several direct approaches to the crystallographic phase problem [see Sections 1.3.4.2.1.10, 1.3.4.5.2.2(*e*)]. Many aspects of their theory and applications are presented in the book by Grenander & Szegö (1958).

1.3.2.6.9.1. *Toeplitz forms*

Let $f \in L^1(\mathbb{R}/\mathbb{Z})$ be real-valued, so that its Fourier coefficients satisfy the relations $c_{-m}(f) = \overline{c_m(f)}$. The Hermitian form in $n+1$ complex variables

$$T_n[f](\mathbf{u}) = \sum_{\mu=0}^{n}\sum_{\nu=0}^{n}\overline{u_\mu}c_{\mu-\nu}u_\nu$$

is called the nth *Toeplitz form* associated to f. It is a straightforward consequence of the convolution theorem and of Parseval's identity that $T_n[f]$ may be written:

$$T_n[f](\mathbf{u}) = \int_0^1\left|\sum_{\nu=0}^{n}u_\nu\exp(2\pi i\nu x)\right|^2 f(x)\,\mathrm{d}x.$$

1.3.2.6.9.2. *The Toeplitz–Carathéodory–Herglotz theorem*

It was shown independently by Toeplitz (1911*b*), Carathéodory (1911) and Herglotz (1911) that a function $f \in L^1$ is almost everywhere non-negative *if and only if* the Toeplitz forms $T_n[f]$ associated to f are positive semidefinite for all values of n.

This is equivalent to the infinite system of determinantal inequalities

$$D_n = \det\begin{pmatrix} c_0 & c_{-1} & \cdot & \cdot & c_{-n} \\ c_1 & c_0 & c_{-1} & \cdot & \cdot \\ \cdot & c_1 & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & c_{-1} \\ c_n & \cdot & \cdot & c_1 & c_0 \end{pmatrix} \geq 0 \quad \text{for all } n.$$

The D_n are called *Toeplitz determinants*. Their application to the crystallographic phase problem is described in Section 1.3.4.2.1.10.

1.3.2.6.9.3. *Asymptotic distribution of eigenvalues of Toeplitz forms*

The eigenvalues of the Hermitian form $T_n[f]$ are defined as the $n+1$ real roots of the characteristic equation $\det\{T_n[f-\lambda]\} = 0$. They will be denoted by

$$\lambda_1^{(n)}, \lambda_2^{(n)}, \ldots, \lambda_{n+1}^{(n)}.$$

It is easily shown that if $m \leq f(x) \leq M$ for all x, then $m \leq \lambda_\nu^{(n)} \leq M$ for all n and all $\nu = 1, \ldots, n+1$. As $n \to \infty$ these bounds, and the distribution of the $\lambda^{(n)}$ within these bounds, can be made more precise by introducing two new notions.

(i) *Essential bounds*: define ess inf f as the largest m such that $f(x) \geq m$ except for values of x forming a set of measure 0; and define ess sup f similarly.

(ii) *Equal distribution*. For each n, consider two sets of $n+1$ real numbers:

$$a_1^{(n)}, a_2^{(n)}, \ldots, a_{n+1}^{(n)}, \quad \text{and} \quad b_1^{(n)}, b_2^{(n)}, \ldots, b_{n+1}^{(n)}.$$

Assume that for each ν and each n, $|a_\nu^{(n)}| < K$ and $|b_\nu^{(n)}| < K$ with K independent of ν and n. The sets $\{a_\nu^{(n)}\}$ and $\{b_\nu^{(n)}\}$ are said to be equally distributed in $[-K, +K]$ if, for any function F over $[-K, +K]$,

$$\lim_{n\to\infty}\frac{1}{n+1}\sum_{\nu=1}^{n+1}[F(a_\nu^{(n)}) - F(b_\nu^{(n)})] = 0.$$

references