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The joint probability distribution function of structure factors with rational indices. IV. The $P1$ case

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Abstract

The method of the joint probability distribution functions of structure factors has been extended to reflections with rational indices. The most general case, space group $P1$, has been considered. The positional parameters are the primitive random variables of our probabilistic approach, while the reflection indices are kept fixed. Quite general joint probability distributions have been considered from which conditional distributions have been derived: these proved applicable to the accurate estimation of the real and imaginary parts of a structure factor, given prior information on other structure factors. The method is also discussed in relation to the Hilbert-transform techniques.

1. Symbols and notation

The following list defines some of the symbols used in this paper.

N: number of atoms in the unit cell.

f_j: scattering factor of the jth atom (thermal factor included).

$\mathbf{h}$: three-dimensional index with integral components (h, k, l).

$\mathbf{p}$, $\mathbf{q}$: three-dimensional indices with rational components (p_1, p_2, p_3), (q_1, q_2, q_3), respectively.

$p_s = p_1 + p_2 + p_3$.

$q_s = q_1 + q_2 + q_3$.

φ: phase of the structure factor.

$\sum_1(\mathbf{p})$, $\sum_1(\mathbf{q}) = \sum_{j=1}^N f_j$ calculated for the reflections with vectorial indices $\mathbf{p}$ and $\mathbf{q}$, respectively.

$\sum_2(\mathbf{p})$, $\sum_2(\mathbf{q}) = \sum_{j=1}^N f_j^2$ calculated for the reflections with vectorial indices $\mathbf{p}$ and $\mathbf{q}$, respectively.

$\sum_{11}(\mathbf{p}, \mathbf{q}) = \sum_{j=1}^N f_j(\mathbf{p}) f_j(\mathbf{q})$.

Papers by Giacovazzo & Siliqi (1998), Giacovazzo, Siliqi, Carrozzini *et al.* (1999) and Giacovazzo, Siliqi, Altomare *et al.* (1999) will be referred as papers I, II and III, respectively.

2. Introduction

In papers I and II of this series, the statistical properties of the structure factors with rational indices were investigated. In papers I and II, distributions $P(|F_{\mathbf{p}}|)$ were obtained for the $P1$ and $P\bar{1}$ cases, respectively, which show remarkable deviations from the distributions derived by Wilson (1942) for the standard index reflections. Distributions $P(\varphi_{\mathbf{p}})$ and $P(\varphi_{\mathbf{p}}||F_{\mathbf{p}}|)$ were also derived, which proved to be, in favourable cases, quite different from the uniform Wilson phase distribution.

Papers I and II can be considered as the first step for the development of direct-methods procedures involving rational index reflections. To this end, joint probability distributions of structure factors were derived for the $P\bar{1}$ case in paper III. An important result of that study is that phase and moduli estimates arise from joint probability distribution functions involving reflections whose indices do not necessarily give rise to structure invariants or seminvariants. This additional degree of freedom is allowed by the rationality of the indices and by the basic assumptions on the primitive random variables.

In this paper, we will derive in $P1$ the joint probability distribution functions of structure factors with rational indices. We will first focus our attention on the distribution $P(F_{\mathbf{p}}, F_{\mathbf{q}})$, where $\mathbf{p}$ and $\mathbf{q}$ can be any pair of vectors with rational components, the case of integral components included. We will show that the simplest formulae are obtained by including in the calculations the real and imaginary parts of the structure factors rather than their moduli and phases, *i.e.* it is easier to calculate $P(A_{\mathbf{p}}, B_{\mathbf{p}}, A_{\mathbf{q}}, B_{\mathbf{q}}, \ldots)$ than $P(|F_{\mathbf{p}}|, \varphi_{\mathbf{p}}, |F_{\mathbf{q}}|, \varphi_{\mathbf{q}}, \ldots)$. The formulae so obtained will be applied to the case in which $\mathbf{p}$ is a half-integral index reflection while the $\mathbf{q}$'s are standard indices, or *vice versa*. To follow all the mathematical calculations, the reader may wish to refer to the formulae collected in Appendix B of paper III.

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Special attention will also be devoted to the symmetry; indeed, a lack of symmetry occurs when rational indices are considered.

Recently, the discrete Hilbert transform has been evoked (Ramachandran, 1969; Mishnev, 1993, 1996; Zanotti *et al.*, 1996) to link structure amplitudes having half-integral Miller indices with structure amplitudes of standard reflections. The relation between such techniques and our probabilistic method is discussed.

3. The joint probability distribution $P(A_{\mathbf{p}}, B_{\mathbf{p}}, A_{\mathbf{q}}, B_{\mathbf{q}})$

Let us suppose that the reflection indices are fixed while the variables x_j, y_j and z_j ($j = 1, \ldots, N$) are independently and uniformly distributed in the interval (0, 1). The structure factors are then variables themselves. In $P1$,

$$\begin{aligned} F_{\mathbf{p}} &= |F_{\mathbf{p}}| \exp(i\varphi_{\mathbf{p}}) \\ &= \sum_{j=1}^{N} f_j \exp(2\pi i \mathbf{p} \cdot \mathbf{r}_j) \\ &= A_{\mathbf{p}} + iB_{\mathbf{p}}, \\ A_{\mathbf{p}} &= \sum_{j=1}^{N} f_j \cos(2\pi \mathbf{p} \cdot \mathbf{r}_j), \\ B_{\mathbf{p}} &= \sum_{j=1}^{N} f_j \sin(2\pi \mathbf{p} \cdot \mathbf{r}_j). \end{aligned}$$

The characteristic function $C(u_{\mathbf{p}}, v_{\mathbf{p}}, u_{\mathbf{q}}, v_{\mathbf{q}})$ of the distribution $P(A_{\mathbf{p}}, B_{\mathbf{p}}, A_{\mathbf{q}}, B_{\mathbf{q}})$ is given by

$$\begin{aligned} C(u_{\mathbf{p}}, v_{\mathbf{p}}, u_{\mathbf{q}}, v_{\mathbf{q}}) &= \langle \exp i(u_{\mathbf{p}}A_{\mathbf{p}} + v_{\mathbf{p}}B_{\mathbf{p}} + u_{\mathbf{q}}A_{\mathbf{q}} + v_{\mathbf{q}}B_{\mathbf{q}})\rangle \\ &\simeq \exp\big\{ i[u_{\mathbf{p}}K_{10}(\mathbf{p}) + v_{\mathbf{p}}K_{01}(\mathbf{p}) \\ &\quad + u_{\mathbf{q}}K_{10}(\mathbf{q}) + v_{\mathbf{q}}K_{01}(\mathbf{q})] \\ &\quad - \tfrac{1}{2}[u_{\mathbf{p}}^2K_{20}(\mathbf{p}) + v_{\mathbf{p}}^2K_{02}(\mathbf{p}) \\ &\quad + u_{\mathbf{q}}^2K_{20}(\mathbf{q}) + v_{\mathbf{q}}^2K_{02}(\mathbf{q}) \\ &\quad + 2u_{\mathbf{p}}v_{\mathbf{p}}K_{12}(\mathbf{p}) + 2u_{\mathbf{p}}u_{\mathbf{q}}K_{13}(\mathbf{p}, \mathbf{q}) \\ &\quad + 2u_{\mathbf{p}}v_{\mathbf{q}}K_{14}(\mathbf{p}, \mathbf{q}) + 2v_{\mathbf{p}}u_{\mathbf{q}}K_{23}(\mathbf{p}, \mathbf{q}) \\ &\quad + 2v_{\mathbf{p}}v_{\mathbf{q}}K_{24}(\mathbf{p}, \mathbf{q}) + 2u_{\mathbf{q}}v_{\mathbf{q}}K_{34}(\mathbf{q})]\big\}, \end{aligned}$$

where $u_{\mathbf{p}}$, $v_{\mathbf{p}}$, $u_{\mathbf{q}}$ and $v_{\mathbf{q}}$ are carrying variables associated with $A_{\mathbf{p}}$, $B_{\mathbf{p}}$, $A_{\mathbf{q}}$ and $B_{\mathbf{q}}$, respectively, and

$$\begin{aligned} K_{10}(\mathbf{p}) &= \langle A_{\mathbf{p}}\rangle = \textstyle\sum_1(\mathbf{p})c_{\mathbf{p}}, \\ K_{01}(\mathbf{p}) &= \langle B_{\mathbf{p}}\rangle = \textstyle\sum_1(\mathbf{p})s_{\mathbf{p}}, \\ K_{10}(\mathbf{q}) &= \langle A_{\mathbf{q}}\rangle = \textstyle\sum_1(\mathbf{q})c_{\mathbf{q}}, \\ K_{01}(\mathbf{q}) &= \langle B_{\mathbf{q}}\rangle = \textstyle\sum_1(\mathbf{q})s_{\mathbf{q}}, \\ K_{20}(\mathbf{p}) &= \langle A_{\mathbf{p}}^2\rangle - \langle A_{\mathbf{p}}\rangle^2 = \textstyle\sum_2(\mathbf{p})[1 + c_{2\mathbf{p}} - 2c_{\mathbf{p}}^2]/2, \\ K_{02}(\mathbf{p}) &= \langle B_{\mathbf{p}}^2\rangle - \langle B_{\mathbf{p}}\rangle^2 = \textstyle\sum_2(\mathbf{p})[1 - c_{2\mathbf{p}} - 2s_{\mathbf{p}}^2]/2, \\ K_{20}(\mathbf{q}) &= \langle A_{\mathbf{q}}^2\rangle - \langle A_{\mathbf{q}}\rangle^2 = \textstyle\sum_2(\mathbf{q})[1 + c_{2\mathbf{q}} - 2c_{\mathbf{q}}^2]/2, \\ K_{02}(\mathbf{q}) &= \langle B_{\mathbf{q}}^2\rangle - \langle B_{\mathbf{q}}\rangle^2 = \textstyle\sum_2(\mathbf{q})[1 - c_{2\mathbf{q}} - 2s_{\mathbf{q}}^2]/2, \\ K_{12}(\mathbf{p}) &= \langle A_{\mathbf{p}}B_{\mathbf{p}}\rangle - \langle A_{\mathbf{p}}\rangle\langle B_{\mathbf{p}}\rangle \\ &= \textstyle\sum_2(\mathbf{p})[s_{2\mathbf{p}} - 2c_{\mathbf{p}}s_{\mathbf{p}}]/2, \\ K_{13}(\mathbf{p}, \mathbf{q}) &= \langle A_{\mathbf{p}}A_{\mathbf{q}}\rangle - \langle A_{\mathbf{p}}\rangle\langle A_{\mathbf{q}}\rangle \\ &= \textstyle\sum_{11}(\mathbf{p}, \mathbf{q})[c_{\mathbf{p}+\mathbf{q}} + c_{\mathbf{p}-\mathbf{q}} - 2c_{\mathbf{p}}c_{\mathbf{q}}]/2, \\ K_{14}(\mathbf{p}, \mathbf{q}) &= \langle A_{\mathbf{p}}B_{\mathbf{q}}\rangle - \langle A_{\mathbf{p}}\rangle\langle B_{\mathbf{q}}\rangle \\ &= \textstyle\sum_{11}(\mathbf{p}, \mathbf{q})[s_{\mathbf{p}+\mathbf{q}} + s_{\mathbf{q}-\mathbf{p}} - 2c_{\mathbf{p}}s_{\mathbf{q}}]/2, \\ K_{23}(\mathbf{p}, \mathbf{q}) &= \langle B_{\mathbf{p}}A_{\mathbf{q}}\rangle - \langle B_{\mathbf{p}}\rangle\langle A_{\mathbf{q}}\rangle \\ &= \textstyle\sum_{11}(\mathbf{p}, \mathbf{q})[s_{\mathbf{p}+\mathbf{q}} + s_{\mathbf{p}-\mathbf{q}} - 2s_{\mathbf{p}}c_{\mathbf{q}}]/2, \\ K_{24}(\mathbf{p}, \mathbf{q}) &= \langle B_{\mathbf{p}}B_{\mathbf{q}}\rangle - \langle B_{\mathbf{p}}\rangle\langle B_{\mathbf{q}}\rangle \\ &= \textstyle\sum_{11}(\mathbf{p}, \mathbf{q})[c_{\mathbf{p}-\mathbf{q}} - c_{\mathbf{p}+\mathbf{q}} - 2s_{\mathbf{p}}s_{\mathbf{q}}]/2, \\ K_{34}(\mathbf{q}) &= \langle A_{\mathbf{q}}B_{\mathbf{q}}\rangle - \langle A_{\mathbf{q}}\rangle\langle B_{\mathbf{q}}\rangle \\ &= \textstyle\sum_2(\mathbf{q})[s_{2\mathbf{q}} - 2c_{\mathbf{q}}s_{\mathbf{q}}]/2. \end{aligned}$$

In agreement with paper I,

$$\begin{aligned} c_p &= \cos(\pi p_s)c_{p_1/2}c_{p_2/2}c_{p_3/2} \\ s_p &= \sin(\pi p_s)c_{p_1/2}c_{p_2/2}c_{p_3/2} \\ c_{p_i} &= \sin(2\pi p_i)/(2\pi p_i) \\ s_{p_i} &= [1 - \cos(2\pi p_i)]/(2\pi p_i), \quad i = 1, 2, 3. \end{aligned}$$

All the cumulants were derived in paper I, except for $K_{13}(\mathbf{p}, \mathbf{q})$, $K_{14}(\mathbf{p}, \mathbf{q})$, $K_{23}(\mathbf{p}, \mathbf{q})$ and $K_{24}(\mathbf{p}, \mathbf{q})$ (cumulants involving both $\mathbf{p}$ and $\mathbf{q}$ reflections). In Appendix A, $K_{14}(\mathbf{p}, \mathbf{q})$ is derived, as a representative of such cumulants. The required joint probability distribution is

$$\begin{aligned} &P(A_{\mathbf{p}}, B_{\mathbf{p}}, A_{\mathbf{q}}, B_{\mathbf{q}}) \\ &\simeq (2\pi)^{-4} \int_{-\infty}^{\infty} \ldots \int_{-\infty}^{\infty} \exp\big(-i\{u_{\mathbf{p}}[A_{\mathbf{p}} - K_{10}(\mathbf{p})] \\ &\quad + v_{\mathbf{p}}[B_{\mathbf{p}} - K_{01}(\mathbf{p})] + u_{\mathbf{q}}[A_{\mathbf{q}} - K_{10}(\mathbf{q})] \\ &\quad + v_{\mathbf{q}}[B_{\mathbf{q}} - K_{01}(\mathbf{q})]\}\big) \\ &\quad \times \exp\big\{-\tfrac{1}{2}[u_{\mathbf{p}}^2K_{20}(\mathbf{p}) + v_{\mathbf{p}}^2K_{02}(\mathbf{p}) \\ &\quad + u_{\mathbf{q}}^2K_{20}(\mathbf{q}) + v_{\mathbf{q}}^2K_{02}(\mathbf{q}) + 2u_{\mathbf{p}}v_{\mathbf{p}}K_{12}(\mathbf{p}) \\ &\quad + 2u_{\mathbf{p}}u_{\mathbf{q}}K_{13}(\mathbf{p}, \mathbf{q}) + 2u_{\mathbf{p}}v_{\mathbf{q}}K_{14}(\mathbf{p}, \mathbf{q}) \\ &\quad + 2v_{\mathbf{p}}u_{\mathbf{q}}K_{23}(\mathbf{p}, \mathbf{q}) + 2v_{\mathbf{p}}v_{\mathbf{q}}K_{24}(\mathbf{p}, \mathbf{q}) \\ &\quad + 2u_{\mathbf{q}}v_{\mathbf{q}}K_{34}(\mathbf{q})]\big\}\, \mathrm{d}u_{\mathbf{p}}\, \mathrm{d}v_{\mathbf{p}}\, \mathrm{d}u_{\mathbf{q}}\, \mathrm{d}v_{\mathbf{q}} \\ &= (2\pi)^{-4} \int_{-\infty}^{\infty} \ldots \int_{-\infty}^{\infty} \exp(i\bar{\mathbf{T}}\mathbf{U}) \exp[-\tfrac{1}{2}\bar{\mathbf{U}}\mathbf{K}\mathbf{U}]\, \mathrm{d}\mathbf{U}, \end{aligned} \tag{1}$$

where

$$\bar{\mathbf{U}} = (u_{\mathbf{p}}, v_{\mathbf{p}}, u_{\mathbf{q}}, v_{\mathbf{q}}),$$

$$\bar{\mathbf{T}} = [d_{10}(\mathbf{p}), d_{01}(\mathbf{p}), d_{10}(\mathbf{q}), d_{01}(\mathbf{q})] = [d_1, d_2, d_3, d_4],$$

$$d_{10}(\mathbf{p}) = [K_{10}(\mathbf{p}) - A_{\mathbf{p}}], \quad d_{01}(\mathbf{p}) = [K_{01}(\mathbf{p}) - B_{\mathbf{p}}], \ldots,$$

$$\mathbf{K} = \begin{vmatrix} K_{20}(\mathbf{p}) & K_{12}(\mathbf{p}) & K_{13}(\mathbf{p},\mathbf{q}) & K_{14}(\mathbf{p},\mathbf{q}) \\ K_{12}(\mathbf{p}) & K_{02}(\mathbf{p}) & K_{23}(\mathbf{p},\mathbf{q}) & K_{24}(\mathbf{p},\mathbf{q}) \\ K_{13}(\mathbf{p},\mathbf{q}) & K_{23}(\mathbf{p},\mathbf{q}) & K_{20}(\mathbf{q}) & K_{34}(\mathbf{q}) \\ K_{14}(\mathbf{p},\mathbf{q}) & K_{24}(\mathbf{p},\mathbf{q}) & K_{34}(\mathbf{q}) & K_{02}(\mathbf{q}) \end{vmatrix}.$$

$\mathbf{K}$ is a variance–covariance matrix, which, by definition, must have $\det \mathbf{K} \geq 0$. After some calculations, (1) reduces to

$$P(A_{\mathbf{p}}, B_{\mathbf{p}}, A_{\mathbf{q}}, B_{\mathbf{q}}) = (2\pi)^{-2} (\det \boldsymbol{\lambda})^{1/2} \exp[-\tfrac{1}{2}\bar{\mathbf{T}}\boldsymbol{\lambda}\mathbf{T}], \quad (2)$$

where

$$\boldsymbol{\lambda} = \mathbf{K}^{-1}. \quad (3)$$

In a more explicit form, (2) may be written as

$$P(A_{\mathbf{p}}, B_{\mathbf{p}}, A_{\mathbf{q}}, B_{\mathbf{q}}) = (2\pi)^{-2} (\det \boldsymbol{\lambda})^{1/2} \exp\left[-\tfrac{1}{2}\sum_{j=1}^{4} \lambda_{jj} d_j^2 - \sum_{j=2}^{4} \lambda_{1j} d_1 d_j - \sum_{j_1 > j_2 = 2}^{4} \lambda_{j_1 j_2} d_{j_1} d_{j_2} \right]. \quad (4)$$

From (4), two useful conditional distributions may be derived. Firstly,

$$P(A_{\mathbf{p}} | B_{\mathbf{p}}, A_{\mathbf{q}}, B_{\mathbf{q}}) = (\lambda_{11}/2\pi)^{1/2} \exp[-\tfrac{1}{2}\lambda_{11}(A_{\mathbf{p}} - \langle A_{\mathbf{p}} \rangle)^2], \quad (5)$$

where

$$\langle A_{\mathbf{p}} \rangle = K_{10}(\mathbf{p}) + \lambda_{11}^{-1}[\lambda_{12} d_{01}(\mathbf{p}) + \lambda_{13} d_{10}(\mathbf{q}) + \lambda_{14} d_{01}(\mathbf{q})] \quad (6)$$

is the conditional expected value of $A_{\mathbf{p}}$ and λ_{11}^{-1} is the variance. Secondly,

$$P(B_{\mathbf{p}} | A_{\mathbf{p}}, A_{\mathbf{q}}, B_{\mathbf{q}}) = (\lambda_{22}/2\pi)^{1/2} \exp[-\tfrac{1}{2}\lambda_{22}(B_{\mathbf{p}} - \langle B_{\mathbf{p}} \rangle)^2], \quad (7)$$

where

$$\langle B_{\mathbf{p}} \rangle = K_{01}(\mathbf{p}) + \lambda_{22}^{-1}[\lambda_{12} d_{10}(\mathbf{p}) + \lambda_{23} d_{10}(\mathbf{q}) + \lambda_{24} d_{01}(\mathbf{q})] \quad (8)$$

is the conditional expected value of $B_{\mathbf{p}}$ and λ_{22}^{-1} is the variance.

The distributions (5) and (7) suggest that $A_{\mathbf{p}}$ and $B_{\mathbf{p}}$ may be estimated independently: in particular, the estimate of $A_{\mathbf{p}}$ may profit by the prior knowledge of $B_{\mathbf{p}}$, $A_{\mathbf{q}}$ and $B_{\mathbf{q}}$, and the estimate of $B_{\mathbf{p}}$ by the prior knowledge of $A_{\mathbf{p}}$, $A_{\mathbf{q}}$ and $B_{\mathbf{q}}$.

4. The joint probability distribution $P(|F_{\mathbf{p}}|, |F_{\mathbf{q}}|, \varphi_{\mathbf{p}}, \varphi_{\mathbf{q}})$

In general, distributions like $P(A_1, \ldots, A_n, B_1, \ldots, B_n)$ are an intermediate step towards the calculation of the distributions $P(|F_1|, \ldots, |F_n|, \varphi_1, \ldots, \varphi_n)$. There is a basic reason for this: the $|F|$'s are the observables and therefore prior knowledge of them may be used for deriving useful conditional distributions for the phases. In the specific case we are treating here, $|F_{\mathbf{p}}|$ is an observable only when $\mathbf{p}$ is an integral component vector. Therefore, there are tentative reasons for preferring $P(|F_1|, \ldots, |F_n|, \varphi_1, \ldots, \varphi_n)$ to $P(A_1, \ldots, A_n, B_1, \ldots, B_n)$. In order to examine the possible role of such distributions, we derive, in this section, the distribution $P(|F_{\mathbf{p}}|, |F_{\mathbf{q}}|, \varphi_{\mathbf{p}}, \varphi_{\mathbf{q}})$ and, in §5, some conditional distributions. We have

$$\begin{aligned} P(|F_{\mathbf{p}}|, |F_{\mathbf{q}}|, \varphi_{\mathbf{p}}, \varphi_{\mathbf{q}}) \simeq{}& (2\pi)^{-2} (\det \boldsymbol{\lambda})^{1/2} |F_{\mathbf{p}} F_{\mathbf{q}}| L \\ &\times \exp\big\{ -\tfrac{1}{2}|F_{\mathbf{p}}|^2 (\lambda_{11} \cos^2 \varphi_{\mathbf{p}} + \lambda_{22} \sin^2 \varphi_{\mathbf{p}} \\ &+ 2\lambda_{12} \sin \varphi_{\mathbf{p}} \cos \varphi_{\mathbf{p}}) \\ &- \tfrac{1}{2}|F_{\mathbf{q}}|^2 (\lambda_{33} \cos^2 \varphi_{\mathbf{q}} + \lambda_{44} \sin^2 \varphi_{\mathbf{q}} \\ &+ 2\lambda_{34} \sin \varphi_{\mathbf{q}} \cos \varphi_{\mathbf{q}}) \\ &- |F_{\mathbf{p}} F_{\mathbf{q}}| (\lambda_{13} \cos \varphi_{\mathbf{p}} \cos \varphi_{\mathbf{q}} + \lambda_{14} \cos \varphi_{\mathbf{p}} \sin \varphi_{\mathbf{q}} \\ &+ \lambda_{23} \sin \varphi_{\mathbf{p}} \cos \varphi_{\mathbf{q}} + \lambda_{24} \sin \varphi_{\mathbf{p}} \sin \varphi_{\mathbf{q}}) \\ &+ |F_{\mathbf{p}}| [\lambda_{11} K_{10}(\mathbf{p}) \cos \varphi_{\mathbf{p}} + \lambda_{22} K_{01}(\mathbf{p}) \sin \varphi_{\mathbf{p}} \\ &+ \lambda_{12} K_{10}(\mathbf{p}) \sin \varphi_{\mathbf{p}} + \lambda_{12} K_{01}(\mathbf{p}) \cos \varphi_{\mathbf{p}} \\ &+ \lambda_{13} K_{10}(\mathbf{q}) \cos \varphi_{\mathbf{p}} + \lambda_{14} K_{01}(\mathbf{q}) \cos \varphi_{\mathbf{p}} \\ &+ \lambda_{23} K_{10}(\mathbf{q}) \sin \varphi_{\mathbf{p}} + \lambda_{24} K_{01}(\mathbf{q}) \sin \varphi_{\mathbf{p}}] \\ &+ |F_{\mathbf{q}}| [\lambda_{33} K_{10}(\mathbf{q}) \cos \varphi_{\mathbf{q}} + \lambda_{44} K_{01}(\mathbf{q}) \sin \varphi_{\mathbf{q}} \\ &+ \lambda_{13} K_{10}(\mathbf{p}) \cos \varphi_{\mathbf{q}} + \lambda_{14} K_{10}(\mathbf{p}) \sin \varphi_{\mathbf{q}} \\ &+ \lambda_{23} K_{01}(\mathbf{p}) \cos \varphi_{\mathbf{q}} + \lambda_{24} K_{01}(\mathbf{p}) \sin \varphi_{\mathbf{q}} \\ &+ \lambda_{34} K_{10}(\mathbf{q}) \sin \varphi_{\mathbf{q}} + \lambda_{34} K_{01}(\mathbf{q}) \cos \varphi_{\mathbf{q}}] \big\}, \end{aligned} \quad (9)$$

where

$$\begin{aligned} L = \exp\big\{ &-\tfrac{1}{2}[\lambda_{11} K_{10}^2(\mathbf{p}) + \lambda_{22} K_{01}^2(\mathbf{p}) \\ &+ \lambda_{33} K_{10}^2(\mathbf{q}) + \lambda_{44} K_{01}^2(\mathbf{q}) \\ &+ 2\lambda_{12} K_{10}(\mathbf{p}) K_{01}(\mathbf{p}) + 2\lambda_{13} K_{10}(\mathbf{p}) K_{10}(\mathbf{q}) \\ &+ 2\lambda_{14} K_{10}(\mathbf{p}) K_{01}(\mathbf{q}) + 2\lambda_{23} K_{01}(\mathbf{p}) K_{10}(\mathbf{q}) \\ &+ 2\lambda_{24} K_{01}(\mathbf{p}) K_{01}(\mathbf{q}) + 2\lambda_{34} K_{10}(\mathbf{q}) K_{01}(\mathbf{q})] \big\}. \end{aligned}$$

Equation (9) is a rather entangled distribution, which becomes much more simple only when both $\mathbf{p}$ and $\mathbf{q}$ are standard indices. Then

$$K_{10}(\mathbf{p}) = K_{01}(\mathbf{p}) = K_{10}(\mathbf{q}) = K_{01}(\mathbf{q}),$$

$$\mathbf{K} = \begin{vmatrix} \sum_2(\mathbf{p})/2 & 0 & 0 & 0 \\ 0 & \sum_2(\mathbf{p})/2 & 0 & 0 \\ 0 & 0 & \sum_2(\mathbf{q})/2 & 0 \\ 0 & 0 & 0 & \sum_2(\mathbf{q})/2 \end{vmatrix},$$

$$(\det \lambda) = 16/\big[\textstyle\sum_2^2(\mathbf{p})\sum_2^2(\mathbf{q})\big],$$

and the distribution (9) reduces to the product of two Wilson distributions:

$$P(|F_{\mathbf{p}}|, |F_{\mathbf{q}}|, \varphi_{\mathbf{p}}, \varphi_{\mathbf{q}}) \simeq (\pi^2)^{-1}|F_{\mathbf{p}}F_{\mathbf{q}}|\big[\textstyle\sum_2(\mathbf{p})\sum_2(\mathbf{q})\big]^{-1} \times \exp\big\{-|F_{\mathbf{p}}|^2/\big[\sum_2(\mathbf{p})\big] - |F_{\mathbf{q}}|^2/\big[\sum_2(\mathbf{q})\big]\big\}. \quad (10)$$

5. The conditional distributions $P(\varphi_{\mathbf{p}}||F_{\mathbf{p}}|, |F_{\mathbf{q}}|, \varphi_{\mathbf{q}})$ and $P(|F_{\mathbf{p}}|\,||F_{\mathbf{q}}|, \varphi_{\mathbf{q}})$

From (9), several conditional distributions can be obtained. Here, we derive only two of them, as examples for the other cases. By application of standard techniques,

$$P(\varphi_{\mathbf{p}}\big||F_{\mathbf{p}}|, |F_{\mathbf{q}}|, \varphi_{\mathbf{q}}) \simeq g^{-1}\exp[-|F_{\mathbf{p}}|^2 Z_2\cos 2(\varphi_{\mathbf{p}} - \theta_2) + |F_{\mathbf{p}}|Z_1\cos(\varphi_{\mathbf{p}} - \theta_1)]$$

is obtained, where

$$g = \int\limits_0^{2\pi}\exp[-|F_{\mathbf{p}}|^2 Z_2\cos 2(\varphi_{\mathbf{p}} - \theta_2) + |F_{\mathbf{p}}|Z_1\cos(\varphi_{\mathbf{p}} - \theta_1)]\,\mathrm{d}\varphi_{\mathbf{p}}, \quad (11)$$

$$Z_2^2 = (\lambda_{12}/2)^2 + [(\lambda_{11} - \lambda_{22})/4]^2,$$

$$\theta_2 = \tfrac{1}{2}\tan^{-1}[2\lambda_{12}/(\lambda_{11} - \lambda_{22})],$$

$$Z_1^2 = a_1^2 + a_2^2,$$

$$a_1 = \lambda_{11}K_{10}(\mathbf{p}) + \lambda_{12}K_{01}(\mathbf{p}) + \lambda_{13}[K_{10}(\mathbf{q}) - |F_{\mathbf{q}}|\cos\varphi_{\mathbf{q}}] + \lambda_{14}[K_{01}(\mathbf{q}) - |F_{\mathbf{q}}|\sin\varphi_{\mathbf{q}}],$$

$$a_2 = \lambda_{22}K_{01}(\mathbf{p}) + \lambda_{12}K_{10}(\mathbf{p}) + \lambda_{23}[K_{10}(\mathbf{q}) - |F_{\mathbf{q}}|\cos\varphi_{\mathbf{q}}] + \lambda_{24}[K_{01}(\mathbf{q}) - |F_{\mathbf{q}}|\sin\varphi_{\mathbf{q}}],$$

$$\theta_1 = \tan^{-1}(a_2/a_1).$$

The integral on the right-hand side of (11) may be calculated by expanding the exponential term in a series of Bessel functions according to (Abramowitz & Stegun, 1972)

$$\exp[-|F_{\mathbf{p}}|^2 Z_2\cos 2(\varphi_{\mathbf{p}} - \theta_2)] = I_0(|F_{\mathbf{p}}|^2 Z_2) + 2\sum_{n=1}^{\infty}\big[I_n(-|F_{\mathbf{p}}|^2 Z_2)\cos 2n(\varphi_{\mathbf{p}} - \theta_2)\big]. \quad (12)$$

The application of the relations

$$\int\limits_0^{2\pi}\cos(n\varphi)\exp(-Z\cos\varphi)\,\mathrm{d}\varphi = 2\pi I_n(Z),$$

$$\int\limits_0^{2\pi}\sin(n\varphi)\exp(-Z\cos\varphi)\,\mathrm{d}\varphi = 0$$

gives

$$g = 2\pi\big[I_0(|F_{\mathbf{p}}|^2 Z_2)I_0(|F_{\mathbf{p}}|Z_1) + 2\sum_{n=1}^{\infty}\cos 2n(\theta_1 - \theta_2) \times I_n(-|F_{\mathbf{p}}|^2 Z_2)I_{2n}(-|F_{\mathbf{p}}|Z_1)\big].$$

If $\mathbf{p}$ and $\mathbf{q}$ are integral indices, then

$$a_1 = a_2 = Z_1 = Z_2 = \lambda_{12} = 0, \quad \lambda_{11} = \lambda_{22} = 2/\textstyle\sum_2(\mathbf{h}), \quad (13)$$

θ_1 and θ_2 are unpredictable, and $g = 2\pi$. Therefore, in accordance with Wilson statistics,

$$P(\varphi_{\mathbf{h}}\big||F_{\mathbf{h}}|, |F_{\mathbf{k}}|, \varphi_{\mathbf{k}}) \simeq (2\pi)^{-1}.$$

The conditional distribution $P(|F_{\mathbf{p}}|\big||F_{\mathbf{q}}|, \varphi_{\mathbf{q}})$ is defined as

$$P(|F_{\mathbf{p}}|\big||F_{\mathbf{q}}|, \varphi_{\mathbf{q}}) = \frac{P(|F_{\mathbf{p}}|, |F_{\mathbf{q}}|, \varphi_{\mathbf{q}})}{\int_0^\infty P(|F_{\mathbf{p}}|, |F_{\mathbf{q}}|, \varphi_{\mathbf{q}})\,\mathrm{d}|F_{\mathbf{p}}|} = L^{-1}|F_{\mathbf{p}}|\exp[-|F_{\mathbf{p}}|^2(\lambda_{11} + \lambda_{22})/4] \times \int\limits_0^{2\pi}\exp[-|F_{\mathbf{p}}|^2 Z_2\cos 2(\varphi_{\mathbf{p}} - \theta_2) + |F_{\mathbf{p}}|Z_1\cos(\varphi_{\mathbf{p}} - \theta_1)]\,\mathrm{d}\varphi_{\mathbf{p}},$$

where L is a normalization constant that does not depend on $|F_{\mathbf{p}}|$. According to (12), we have

$$P(|F_{\mathbf{p}}|\big||F_{\mathbf{q}}|, \varphi_{\mathbf{q}}) \approx L^{-1}|F_{\mathbf{p}}|\exp[-|F_{\mathbf{p}}|^2(\lambda_{11} + \lambda_{22})/4] \times \Big[I_0(|F_{\mathbf{p}}|^2 Z_2)I_0(|F_{\mathbf{p}}|Z_1) + 2\sum_{n=1}^{\infty}\cos 2n(\theta_1 - \theta_2)I_n(-|F_{\mathbf{p}}|^2 Z_2) \times I_{2n}(-|F_{\mathbf{p}}|Z_1)\Big]. \quad (14)$$

The value of the normalizing function L^{-1} can be obtained by direct integration *via* formula 6.633 of Gradshteyn & Ryzhik (1965), but it is more practical to derive it by numerical methods.

The conditional distributions derived in this section (*i.e.* in terms of moduli and phases) are more entangled than those derived in §3 (*i.e.* in terms of real and imaginary parts of structure factors). If more $\mathbf{q}$ reflections are involved in the calculations, the level of complexity can rapidly increase if moduli and phases are employed. Therefore, we decided to study distributions of a general form in terms of A_i and B_i rather than of $|F_i|$ and φ_i.

6. The distribution $P(A_{\mathbf{p}}, B_{\mathbf{p}}, A_{\mathbf{q}_1}, B_{\mathbf{q}_1}, \ldots, A_{\mathbf{q}_n}, B_{\mathbf{q}_n})$

The distributions (5) and (7) answer questions like 'how may $A_{\mathbf{p}}$ and $B_{\mathbf{p}}$ can be estimated if $A_{\mathbf{q}}$ and $B_{\mathbf{q}}$ are known?'. If more pairs ($A_{\mathbf{q}}$, $B_{\mathbf{q}}$) are *a priori* known, this question becomes 'how may $A_{\mathbf{p}}$ and $B_{\mathbf{p}}$ can be estimated when ($A_{\mathbf{q}_j}$, $B_{\mathbf{q}_j}$), $j = 1, \ldots, n$, are *a priori* known?'. The problem may be solved if the conditional distributions

$$P(A_{\mathbf{p}} \big| \{A_{\mathbf{q}_j}, B_{\mathbf{q}_j}, j = 1, \ldots, n\})$$

and

$$P(B_{\mathbf{p}} \big| \{A_{\mathbf{q}_j}, B_{\mathbf{q}_j}, j = 1, \ldots, n\})$$

are derived. This is the aim of this section. In order to make simpler the reading of the conclusive formulae, the notation is simplified as follows.

(*a*) The joint probability distribution

$$P(A_{\mathbf{p}}, B_{\mathbf{p}}, A_{\mathbf{q}_1}, B_{\mathbf{q}_1}, \ldots, A_{\mathbf{q}_n}, B_{\mathbf{q}_n})$$

is denoted by

$$P(X_1, X_2, \ldots, X_{2n+1}, X_{2n+2}), \tag{15}$$

where the variable X_j may represent $A_{\mathbf{p}}$, $B_{\mathbf{p}}$, $A_{\mathbf{q}}$, $B_{\mathbf{q}}$ according to the value of j. In particular,

$$X_1 = A_{\mathbf{p}}, \quad X_2 = B_{\mathbf{p}}$$

will always denote the variables we want to estimate. Then,

$$X_3 = A_{\mathbf{q}_1}, X_4 = B_{\mathbf{q}_1}, \ldots, X_{2n+1} = A_{\mathbf{q}_n}, X_{2n+2} = B_{\mathbf{q}_n}.$$

In accordance with the above notation, odd and even values of j correspond to A and B variables, respectively.

(*b*) The characteristic function of (15), say

$$C(u_1, u_2, \ldots, u_{2n+1}, u_{2n+2}),$$

is expressed in terms of the carrying variable u_j: each u_j is associated with the corresponding X_j in (15).

(*c*) The first-order cumulants of the distribution (15) is denoted by K_j: they represent K_{10} or K_{01} cumulants according to the value of j (*e.g.* K_j will represent a K_{10} cumulant if j is odd, a K_{01} cumulant if j is even).

(*d*) The second-order cumulants of the distribution (15) are denoted by the general symbol K_{ij} (remember that $\mathbf{K}$ is a symmetric matrix):

$$K_{ij} = \langle X_i X_j \rangle - \langle X_i \rangle \langle X_j \rangle.$$

In particular, $K_{jj} = \langle X_j^2 \rangle - \langle X_j \rangle^2$ denotes cumulants of type K_{20} or K_{02} according to whether X_j represents an A or B variable.

The use of the above notation gives

$$\begin{aligned} C(u_1, u_2, \ldots, u_{2n+1}, u_{2n+2}) &= \exp\Bigg[i \sum_{j=1}^{2n+2} K_j u_j - \tfrac{1}{2} \sum_{j=1}^{2n+2} K_{jj} u_j^2 - \sum_{j=2}^{2n+2} K_{1j} u_1 u_j \\ &\quad - \sum_{j=3}^{2n+2} K_{2j} u_2 u_j - \sum_{j_1 > j_2 = 3}^{2n+2} K_{j_1 j_2} u_{j_1} u_{j_2} \Bigg]. \end{aligned}$$

In the above expression, we have regrouped the cumulants in which $A_{\mathbf{p}}$ and $B_{\mathbf{p}}$ are involved. Then,

$$\begin{aligned} P(\mathbf{X}) &= (2\pi)^{-(2n+2)} \int_{-\infty}^{\infty} \ldots \int_{-\infty}^{\infty} \exp(i \bar{\mathbf{T}} \mathbf{U}) \\ &\quad \times \exp[-\tfrac{1}{2} \bar{\mathbf{U}} \mathbf{K} \mathbf{U}] \, \mathrm{d}\mathbf{U}, \end{aligned}$$

where

$$\begin{aligned} \bar{\mathbf{X}} &= (X_1, X_2, \ldots, X_{2n+2}), \\ \bar{\mathbf{U}} &= (u_1, u_2, \ldots, u_{2n+2}), \\ \bar{\mathbf{T}} &= (d_1, d_2, \ldots, d_{2n+2}), \end{aligned}$$

$$d_j = K_j - X_j,$$

$$\mathbf{K} = \begin{vmatrix} K_{11} & K_{12} & \ldots & K_{1,2n+2} \\ K_{21} & K_{22} & \ldots & K_{2,2n+2} \\ \vdots & \vdots & \ddots & \vdots \\ K_{2n+2,1} & K_{2n+2,2} & \ldots & K_{2n+2,2n+2} \end{vmatrix}.$$

$\mathbf{K}$ is the (symmetric by definition) variance–covariance matrix: by definition, $(\det \mathbf{K}) \geq 0$. By application of standard techniques, we obtain

$$P(\mathbf{X}) = (2\pi)^{-(n+1)} (\det \boldsymbol{\lambda})^{1/2} \exp(-\tfrac{1}{2} \bar{\mathbf{T}} \boldsymbol{\lambda} \mathbf{T}), \tag{16}$$

where

$$\boldsymbol{\lambda} = \mathbf{K}^{-1} \tag{17}$$

is again a symmetric matrix.

The distribution (16) may be rewritten in a more useful form which emphasizes the terms involving λ_{1j} and λ_{2j} elements:

$$\begin{aligned} P(\mathbf{X}) &= (2\pi)^{-(n+1)} (\det \boldsymbol{\lambda})^{1/2} \exp\Bigg(-\tfrac{1}{2} \sum_{j=1}^{2n+2} \lambda_{jj} d_j^2 \\ &\quad - \sum_{j=2}^{2n+2} \lambda_{1j} d_1 d_j - \sum_{j=3}^{2n+2} \lambda_{2j} d_2 d_j - \sum_{j_1 > j_2 = 3}^{2n+2} \lambda_{j_1 j_2} d_{j_1} d_{j_2} \Bigg). \end{aligned} \tag{18}$$

Several types of conditional distributions may be derived from (18). We have

$$\begin{aligned} P(X_1 | X_2, \ldots, X_{2n+2}) &= (\lambda_{11}/2\pi)^{1/2} \exp[-\tfrac{1}{2} \lambda_{11} (X_1 - M_1)^2], \end{aligned} \tag{19}$$

where

$$M_1 = K_1 + \lambda_{11}^{-1} \sum_{j=2}^{2n+2} \lambda_{1j} d_j \qquad \text{(GPR1)}$$

is the conditional expected value of X_1, and

$$V_1 = \lambda_{11}^{-1} \qquad \text{(GPR2)}$$

is the relative variance. Also,

$$\langle X_1^2 | X_2, \ldots, X_{2n+2} \rangle = M_1^2 + V_1.$$

It may be noted that $\langle X_1^2 | X_2, \ldots, X_{2n+2} \rangle = M_1^2$ if $V_1 \equiv 0$. By analogy,

$$P(X_2 | X_1, \ldots, X_{2n+2}) = (\lambda_{22}/2\pi)^{1/2} \exp[-\tfrac{1}{2}\lambda_{22}(X_2 - M_2)^2], \qquad (20)$$

where

$$M_2 = K_2 + \lambda_{22}^{-1} \sum_{\substack{j=1\\ j\neq 2}}^{2n+2} \lambda_{2j} d_j \qquad \text{(GPR3)}$$

is the expected value of X_2 and

$$V_2 = \lambda_{22}^{-1} \qquad \text{(GPR4)}$$

is the relative variance. Again,

$$\langle X_2^2 | X_1, X_3, \ldots, X_{2n+2} \rangle = M_2^2 + V_2.$$

The distribution (19) aims at estimating X_1 assuming that X_2 is known (*i.e.* $A_{\mathbf{p}}$ is estimated assuming that $B_{\mathbf{p}}$ is known). Similarly, (20) provides the estimate of X_2 (say $B_{\mathbf{p}}$) when X_1 (*i.e.* $A_{\mathbf{p}}$) is known. Such information is not always available. Therefore, it may be useful to derive also the distribution

$$P(X_1 | X_3, X_4, \ldots, X_{2n+2}) = \frac{\int_{-\infty}^{\infty} P(X_1, X_2, X_3, \ldots, X_{2n+2})\,\mathrm{d}X_2}{\int_{-\infty}^{\infty} P(X_1, X_3, X_4, \ldots, X_{2n+2})\,\mathrm{d}X_1} = (2\pi V_{c1})^{-1/2} \exp[-\tfrac{1}{2}(X_1 - M_{c1})^2/V_{c1}], \qquad (21)$$

where

$$M_{c1} = K_1 + (\lambda_{11}\lambda_{22} - \lambda_{12}^2)^{-1} \sum_{j=3}^{2n+2} (\lambda_{22}\lambda_{1j} - \lambda_{21}\lambda_{2j}) d_j \qquad \text{(GPR5)}$$

is the expected value of X_1 and

$$V_{c1} = \lambda_{22}/(\lambda_{11}\lambda_{22} - \lambda_{12}^2) \qquad \text{(GPR6)}$$

is the variance. Again,

$$\langle X_1^2 | X_2, \ldots, X_{2n+2} \rangle = M_{c1}^2 + V_{c1}.$$

By analogy,

$$P(X_2 | X_3, X_4, \ldots, X_{2n+2}) = (2\pi V_{c2})^{-1/2} \exp[-\tfrac{1}{2}(X_2 - M_{c2})^2/V_{c2}], \qquad (22)$$

where

$$M_{c2} = K_2 + (\lambda_{11}\lambda_{22} - \lambda_{12}^2)^{-1} \sum_{j=3}^{2n+2} (\lambda_{11}\lambda_{2j} - \lambda_{21}\lambda_{1j}) d_j \qquad \text{(GPR7)}$$

is the expected value of X_2, and

$$V_{c2} = \lambda_{11}/(\lambda_{11}\lambda_{22} - \lambda_{12}^2) \qquad \text{(GPR8)}$$

is the variance. Also,

$$\langle X_2^2 | X_3, \ldots, X_{2n+2} \rangle = M_{c2}^2 + V_{c2}.$$

It should be explicitly noted that the prior knowledge of $F_{000} = \sum_{j=1}^{N} Z_j$ is automatically included in the distribution (18) [this information is intrinsically contained in the structure-factor expression]. Therefore, the vector $\mathbf{q} = (0, 0, 0)$ cannot be introduced into the set of $\mathbf{q}$ vectors.

The relations (GPR1) to (GPR8) are the probabilistic relationships we wished to obtain and constitute the most general result of this paper.

7. The canonical case: the basic relations

The joint probability distribution (16) and the basic relationships (GPR1) to (GPR8) are valid under quite general conditions. Indeed, (*a*) the indices of the reflections can be arbitrarily chosen in the set of the rational indices. In particular, they are not restricted to integral or half-integral values. (*b*) The value of n can be arbitrarily fixed in the interval $(1, \infty)$.

Even if the conclusive formulae are formally simple, their practical use is critical when n is large: in this case, high-order $\mathbf{K}$ and $\boldsymbol{\lambda}$ matrices are involved in the calculations. Since the cumulant K_{ij} with $i \neq j$ are often non-negligible, the matrices $\mathbf{K}$ are not diagonal, and their inversion can be critical and time consuming for large n values. A remarkable simplification of the process may be obtained in the canonical case (see paper III), *e.g.* when $F_{\mathbf{p}}$ is a half-integral index reflection and the $F_{\mathbf{q}_j}$'s, $j = 1, \ldots, n$, are standard reflections (this is the *first option*), or *vice versa*, when $F_{\mathbf{p}}$ is a standard reflection and the $F_{\mathbf{q}_j}$'s, $j = 1, \ldots, n$, are half-integral index reflections (this is the *second option*).

Let us consider the first option when $n = 1$ [*i.e.* for the case $P(A_{\mathbf{p}}, B_{\mathbf{p}}, A_{\mathbf{q}}, B_{\mathbf{q}})$]: for the sake of clearness, we will adopt for the cumulants the notation of the §3. Then,

$$\begin{gathered} c_{\mathbf{p}} \equiv c_{\mathbf{q}} \equiv s_{\mathbf{q}} \equiv 0, \\ K_{10}(\mathbf{p}) \equiv K_{10}(\mathbf{q}) \equiv K_{01}(\mathbf{q}) \equiv 0, \\ K_{20}(\mathbf{p}) = \textstyle\sum_2(\mathbf{p})/2, \quad K_{02}(\mathbf{p}) = \sum_2(\mathbf{p})(1 - 2s_{\mathbf{p}}^2)/2, \\ K_{20}(\mathbf{q}) = K_{02}(\mathbf{q}) = \textstyle\sum_2(\mathbf{q})/2, \\ K_{12}(\mathbf{p}) \equiv K_{34}(\mathbf{q}) \equiv K_{13}(\mathbf{p}, \mathbf{q}) \equiv K_{24}(\mathbf{p}, \mathbf{q}) \equiv 0, \\ K_{14}(\mathbf{p}, \mathbf{q}) = \textstyle\sum_{11}(\mathbf{p}, \mathbf{q})(s_{\mathbf{p}+\mathbf{q}} + s_{\mathbf{q}-\mathbf{p}})/2, \\ K_{23}(\mathbf{p}, \mathbf{q}) = \textstyle\sum_{11}(\mathbf{p}, \mathbf{q})(s_{\mathbf{p}+\mathbf{q}} + s_{\mathbf{p}-\mathbf{q}})/2. \end{gathered} \qquad (23)$$

Accordingly,

$$\mathbf{K}=\begin{vmatrix} K_{20}(\mathbf{p}) & 0 & 0 & K_{14}(\mathbf{p},\mathbf{q}) \\ 0 & K_{02}(\mathbf{p}) & K_{23}(\mathbf{p},\mathbf{q}) & 0 \\ 0 & K_{23}(\mathbf{p},\mathbf{q}) & K_{20}(\mathbf{q}) & 0 \\ K_{14}(\mathbf{p},\mathbf{q}) & 0 & 0 & K_{02}(\mathbf{q}) \end{vmatrix} \tag{24}$$

reduces to a matrix with non-vanishing elements only on the two main diagonals. If we now consider the matrix $\mathbf{K}$ for $n > 1$: all the cumulants relative to the pairs $(\mathbf{q}_i, \mathbf{q}_j)$ vanish and $\mathbf{K}$ assumes, in the notation adopted in §6, the form

$$\mathbf{K}=\begin{vmatrix} K_{11} & 0 & 0 & K_{14} & 0 & K_{16} & 0 & K_{18} & \dots & K_{1,2n+2} \\ 0 & K_{22} & K_{23} & 0 & K_{25} & 0 & K_{27} & 0 & \dots & 0 \\ 0 & K_{23} & K_{33} & 0 & 0 & 0 & 0 & 0 & \dots & 0 \\ K_{14} & 0 & 0 & K_{44} & 0 & 0 & 0 & 0 & \dots & 0 \\ 0 & K_{25} & 0 & 0 & K_{55} & 0 & 0 & 0 & \dots & 0 \\ K_{16} & 0 & 0 & 0 & 0 & K_{66} & 0 & 0 & \dots & 0 \\ 0 & K_{27} & 0 & 0 & 0 & 0 & K_{77} & 0 & \dots & 0 \\ K_{18} & 0 & 0 & 0 & 0 & 0 & 0 & K_{88} & \dots & 0 \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \ddots & \vdots \\ K_{1,2n+2} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \dots & K_{2n+2,2n+2} \end{vmatrix}. \tag{25}$$

Let us now consider the second option in the case $n = 1$ [*i.e.* for the case $P(A_{\mathbf{p}}, B_{\mathbf{p}}, A_{\mathbf{q}}, B_{\mathbf{q}})$]. Then,

$$\begin{gathered} c_{\mathbf{p}} \equiv s_{\mathbf{p}} \equiv c_{\mathbf{q}} \equiv 0, \\ K_{10}(\mathbf{p}) \equiv K_{01}(\mathbf{p}) \equiv K_{10}(\mathbf{q}) \equiv 0, \\ K_{20}(\mathbf{p}) = K_{02}(\mathbf{p}) = \textstyle\sum_2(\mathbf{p})/2, \\ K_{20}(\mathbf{q}) = \textstyle\sum_2(\mathbf{q})/2, \quad K_{02}(\mathbf{q}) = \textstyle\sum_2(\mathbf{q})(1-2s_{\mathbf{q}}^2)/2, \\ K_{12}(\mathbf{p}) \equiv K_{34}(\mathbf{q}) \equiv K_{13}(\mathbf{p},\mathbf{q}) \equiv K_{24}(\mathbf{p},\mathbf{q}) \equiv 0, \\ K_{14}(\mathbf{p},\mathbf{q}) = \textstyle\sum_{11}(\mathbf{p},\mathbf{q})(s_{\mathbf{p}+\mathbf{q}} + s_{\mathbf{q}-\mathbf{p}})/2, \\ K_{23}(\mathbf{p},\mathbf{q}) = \textstyle\sum_{11}(\mathbf{p},\mathbf{q})(s_{\mathbf{p}+\mathbf{q}} + s_{\mathbf{p}-\mathbf{q}})/2. \end{gathered} \tag{26}$$

Again, $\mathbf{K}$ reduces to the form (24). However, if $n > 1$, all the cumulants relative to the pairs $(\mathbf{q}_i, \mathbf{q}_j)$ vanish except

$$K_{24}(\mathbf{q}_i, \mathbf{q}_j) = -\textstyle\sum_{11}(\mathbf{q}_i, \mathbf{q}_j) s_{\mathbf{q}_i} s_{\mathbf{q}_j}.$$

Thus, for this second option, $\mathbf{K}$ assumes the form

$$\mathbf{K}=\begin{vmatrix} K_{11} & 0 & 0 & K_{14} & 0 & K_{16} & 0 & K_{18} & \dots & K_{1,2n+2} \\ 0 & K_{22} & K_{23} & 0 & K_{25} & 0 & K_{27} & 0 & \dots & 0 \\ 0 & K_{23} & K_{33} & 0 & 0 & 0 & 0 & 0 & \dots & 0 \\ K_{14} & 0 & 0 & K_{44} & 0 & K_{46} & 0 & K_{48} & \dots & K_{4,2n+2} \\ 0 & K_{25} & 0 & 0 & K_{55} & 0 & 0 & 0 & \dots & 0 \\ K_{16} & 0 & 0 & K_{46} & 0 & K_{66} & 0 & K_{68} & \dots & K_{6,2n+2} \\ 0 & K_{27} & 0 & 0 & 0 & 0 & K_{77} & 0 & \dots & 0 \\ K_{18} & 0 & 0 & K_{48} & 0 & K_{68} & 0 & K_{88} & \dots & K_{8,2n+2} \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \ddots & \vdots \\ K_{1,2n+2} & 0 & 0 & K_{4,2n+2} & 0 & K_{6,2n+2} & 0 & K_{8,2n+2} & \dots & K_{2n+2,2n+2} \end{vmatrix}.$$

However, the maximum value of $|K_{24}(\mathbf{q}_i, \mathbf{q}_j)|$ is

$$\textstyle\sum_{11}(\mathbf{q}_i, \mathbf{q}_j)[(\sin\pi/2)/(\pi/2)]^6 = 0.066\textstyle\sum_{11}(\mathbf{q}_i, \mathbf{q}_j),$$

attained when $\mathbf{q}_i = (1/2, 1/2, 1/2)$, $\mathbf{q}_j = (\pm 1/2, \pm 1/2, \pm 1/2)$. As soon as some of the six components increase, $K_{24}(\mathbf{q}_i, \mathbf{q}_j)$ rapidly decreases. Accordingly, as a first approximation, $K_{24}(\mathbf{q}_i, \mathbf{q}_j)$ contributions can be neglected; then $\mathbf{K}$ reduces again to (25).

The determinant of the matrix (25), calculated *via* the Laplace technique, is given by

$$\begin{aligned} \det(\mathbf{K}) = {} & \prod_{j=1}^{2n+2} K_{jj} - \sum_{r=2}^{n+1}\Bigg(K_{1,2r}^2 \prod_{\substack{j=2\\ j\neq 2r}}^{2n+2} K_{jj}\Bigg) \\ & - \sum_{r=2}^{n+1}\Bigg(K_{2,2r-1}^2 \prod_{\substack{j=1\\ j\neq 2r-1\\ j\neq 2}}^{2n+2} K_{jj}\Bigg) \\ & + \sum_{r=2}^{n+1} K_{1,2r}^2 \Bigg[\sum_{s=2}^{n+1}\Bigg(K_{2,2s-1}^2 \prod_{\substack{j=3\\ j\neq 2r\\ j\neq 2s-1}}^{2n+2} K_{jj}\Bigg)\Bigg]. \end{aligned}$$

As an example, for $n = 2$,

$$\begin{aligned} \det(\mathbf{K}) = {} & K_{11}K_{22}\dots K_{66} - K_{14}^2 K_{22}K_{33}K_{55}K_{66} \\ & - K_{16}^2 K_{22}K_{33}K_{44}K_{55} - K_{23}^2 K_{11}K_{44}K_{55}K_{66} \\ & - K_{25}^2 K_{11}K_{33}K_{44}K_{66} + K_{14}^2 K_{23}^2 K_{55}K_{66} \\ & + K_{14}^2 K_{25}^2 K_{33}K_{66} + K_{16}^2 K_{23}^2 K_{44}K_{55} \\ & + K_{16}^2 K_{25}^2 K_{33}K_{44}. \end{aligned}$$

Let us now calculate L_{11}, the cofactor of K_{11} in (25):

$$\begin{aligned} L_{11} &= \prod_{j=2}^{2n+2} K_{jj} - \sum_{r=2}^{n+1}\Bigg(K_{2,2r-1}^2 \prod_{\substack{j=3\\ j\neq 2r-1}}^{2n+2} K_{jj}\Bigg) \\ &= K_{11}^{-1}\Bigg[\prod_{j=1}^{2n+2} K_{jj} - \sum_{r=2}^{n+1}\Bigg(K_{2,2r-1}^2 \prod_{\substack{j=1\\ j\neq 2\\ j\neq 2r-1}}^{2n+2} K_{jj}\Bigg)\Bigg]. \end{aligned}$$

For example, for $n = 2$,

$$\begin{aligned} L_{11} = K_{11}^{-1}(& K_{11}K_{22}\dots K_{66} - K_{23}^2 K_{11}K_{44}K_{55}K_{66} \\ & - K_{25}^2 K_{11}K_{33}K_{44}K_{66}). \end{aligned}$$

Now,

$$\begin{aligned} \lambda_{11}^{-1} &= \det(\mathbf{K})/L_{11} \\ &= K_{11} - \Bigg\{\sum_{r=2}^{n+1} K_{1,2r}^2 \Bigg[\prod_{\substack{j=1\\ j\neq 2r}}^{2n+2} K_{jj} \\ &\qquad - \sum_{s=2}^{n+1}\Bigg(K_{2,2s-1}^2 \prod_{\substack{j=1\\ j\neq 2r\\ j\neq 2s-1\\ j\neq 2}}^{2n+2} K_{jj}\Bigg)\Bigg]\Bigg\} \\ &\qquad \times \Bigg[\prod_{j=1}^{2n+2} K_{jj} - \sum_{r=2}^{n+1}\Bigg(K_{2,2r-1}^2 \prod_{\substack{j=1\\ j\neq 2\\ j\neq 2r-1}}^{2n+2} K_{jj}\Bigg)\Bigg]^{-1} \\ &= K_{11} - \sum_{r=2}^{n+1}(K_{1,2r}^2/K_{2r,2r}). \end{aligned}$$

Let us now derive a simple expression for the elements λ_{1r}. We observe (a) that the cofactors of K_{1r}, say L_{1r}, are vanishing for odd values of r, and (b) that for even values of r

$$L_{1r} = K_{1r}\left[\sum_{s=2}^{n+1}\left(K^2_{2,2s-1}\prod_{\substack{j=3\\ j\neq 2s-1\\ j\neq r}}^{2n+2}K_{jj}\right) - \prod_{\substack{j=2\\ j\neq r}}^{2n+2}K_{jj}\right]$$

$$= \frac{K_{1r}}{K_{11}K_{rr}}\left[\sum_{s=2}^{n+1}\left(K^2_{2,2s-1}\prod_{\substack{j=1\\ j\neq 2s-1\\ j\neq 2}}^{2n+2}K_{jj}\right) - \prod_{j=1}^{2n+2}K_{jj}\right].$$

For example, for $n = 2$,

$$L_{14} = K_{14}(K^2_{23}K_{55}K_{66} + K^2_{25}K_{33}K_{66} - K_{22}K_{33}K_{55}K_{66}),$$
$$L_{16} = K_{16}(K^2_{23}K_{44}K_{55} + K^2_{25}K_{33}K_{44} - K_{22}K_{33}K_{44}K_{55}).$$

As a consequence, (a) for odd values of r (provided $r \neq 1$),

$$\lambda_{1r} \equiv 0,$$

and (b), for even values of r,

$$\lambda_{1r}/\lambda_{11} = L_{1r}/L_{11} = -K_{1r}/K_{rr}. \qquad (27a)$$

It may also be shown that

$$L_{22} = K^{-1}_{22}\left[\prod_{j=1}^{2n+2}K_{jj} - \sum_{r=2}^{n+1}\left(K^2_{1,2r}\prod_{\substack{j=2\\ j\neq 2r}}^{2n+2}K_{jj}\right)\right],$$

$$\lambda^{-1}_{22} = K_{22} - \sum_{r=2}^{n+1}(K^2_{2,2r-1}/K_{2r-1,2r-1}),$$

$$L_{2r} = K_{2r}\left[\sum_{s=2}^{n+1}\left(K^2_{1,2s}\prod_{\substack{j=3\\ j\neq 2s\\ j\neq r}}^{2n+2}K_{jj}\right) - \prod_{\substack{j=1\\ j\neq 2\\ j\neq r}}^{2n+2}K_{jj}\right]$$

$$= \frac{K_{2r}}{K_{22}K_{rr}}\left[\sum_{s=2}^{n+1}\left(K^2_{1,2s}\prod_{\substack{j=2\\ j\neq 2s}}^{2n+2}K_{jj}\right) - \prod_{j=1}^{2n+2}K_{jj}\right],$$

$$\lambda_{2r}/\lambda_{22} = -K_{2r}/K_{rr}, \qquad (27b)$$

with $\lambda_{2r} \equiv 0$ for even values of r (provided $r \neq 2$).

8. The canonical case: the conclusive formulae

The relationships obtained in §7 allow, for the canonical case, a strong simplification of the formulae for the estimation of $A_{\mathbf{p}}$ and $B_{\mathbf{p}}$; indeed, the inversion of the matrix $\mathbf{K}$ is no longer necessary. In particular, the general relationships (GPR1) and (GPR2) may be replaced by

$$M_1 = K_1 - \sum_{j=2}^{2n+2}(K_{1j}/K_{jj})d_j$$

and

$$V_1 = K_{11} - \sum_{j=2}^{n+1}(K^2_{1,2j}/K_{2j,2j}),$$

respectively. Since $\lambda_{1j} \equiv 0$ for odd values of j, we have

$$M_1 = K_1 - \sum_{j=1}^{n+1}(K_{1,2j}/K_{2j,2j})d_{2j}$$

and, since $K_{12} \equiv 0$,

$$M_1 = K_1 - \sum_{j=2}^{n+1}(K_{1,2j}/K_{2j,2j})d_{2j}.$$

In a more explicit form,

$$\langle A_{\mathbf{p}}|B_{\mathbf{p}}, \{A_{\mathbf{q}}, B_{\mathbf{q}}\}\rangle = K_{10}(\mathbf{p}) + \sum_{\mathbf{q}}[K_{14}(\mathbf{p},\mathbf{q})/K_{02}(\mathbf{q})] \times [B_{\mathbf{q}} - K_{01}(\mathbf{q})] \qquad \text{(CPR1)}$$

and

$$V_{A_{\mathbf{p}}} = K_{10}(\mathbf{p}) - \sum_{\mathbf{q}}[K^2_{14}(\mathbf{p},\mathbf{q})/K_{02}(\mathbf{q})]. \qquad \text{(CPR2)}$$

By analogy, the relations (GPR3) and (GPR4) may be replaced by

$$M_2 = K_2 - \sum_{j=1}^{n+1}(K_{2,2j-1}/K_{2j-1,2j-1})d_{2j-1}$$

and

$$V_2 = K_{22} - \sum_{j=1}^{n+1}(K^2_{2,2j-1}/K_{2j-1,2j-1}),$$

respectively. In a more explicit form,

$$\langle B_{\mathbf{p}}|A_{\mathbf{p}}, \{A_{\mathbf{q}}, B_{\mathbf{q}}\}\rangle = K_{01}(\mathbf{p}) + \sum_{\mathbf{q}}[K_{23}(\mathbf{p},\mathbf{q})/K_{20}(\mathbf{q})] \times [A_{\mathbf{q}} - K_{10}(\mathbf{q})] \qquad \text{(CPR3)}$$

and

$$V_{B_{\mathbf{p}}} = K_{02}(\mathbf{p}) - \sum_{\mathbf{q}}[K^2_{23}(\mathbf{p},\mathbf{q})/K_{20}(\mathbf{q})]. \qquad \text{(CPR4)}$$

It may be observed that the additional prior knowledge of anyone of $A_{\mathbf{q}}$ and $B_{\mathbf{q}}$ always reduces $V_{B_{\mathbf{p}}}$ or $V_{A_{\mathbf{p}}}$, respectively. Accordingly, the estimates are expected to be accurate if a sufficiently large number of $\mathbf{q}$ terms are *a priori* known.

Let us now describe how in the canonical case the relationships (GPR5) to (GPR8) may be simplified. Since $\lambda_{12} \equiv 0$ and $\lambda_{1j} \equiv 0$ for odd values of j,

$$M_{c1} = K_1 + \sum_{j=3}^{2n+2} (\lambda_{1j}/\lambda_{11}) d_j$$
$$= K_1 - \sum_{j=3}^{2n+2} (K_{1j}/K_{jj}) d_j$$
$$= K_1 - \sum_{j=2}^{n+1} (K_{1,2j}/K_{2j,2j}) d_{2j}$$

and

$$V_{c1} = K_{11} - \sum_{j=2}^{n+1} (K^2_{1,2j}/K_{2j,2j}).$$

After comparison of the above relations with those obtained for M_1 and V_1, the following statement may be made: the relations (CPR1) to (CPR4) estimate $A_{\mathbf{p}}$ and $B_{\mathbf{p}}$ no matter if $B_{\mathbf{p}}$ and $A_{\mathbf{p}}$, respectively, are *a priori* known.

9. Rational indices and space-group symmetry

The probabilistic results obtained in the preceding sections of this paper hold provided that (*a*) the space group is $P1$ and (*b*) all the atomic coordinates satisfy the condition

$$0 \le x_j \le 1 \quad 0 \le y_j \le 1 \quad 0 \le z_j \le 1, \quad j = 1, \ldots, N. \tag{28}$$

What should we expect if different assumptions are made? We will first analyse the effects of the translation operation over reflections with rational indices and then we will examine the most relevant symmetry aspects. Let $P1$ be the space group; then,

$$F_{\mathbf{p}} = \sum_{j=1}^{N} f_j \exp(2\pi i \mathbf{p} \cdot \mathbf{r}_j)$$

is the structure factor when (28) is satisfied. Suppose we want to replace the condition (28) by the condition

$$-1/2 \le x_j \le 1/2, \quad -1/2 \le y_j \le 1/2, \quad -1/2 \le z_j \le 1/2, \qquad j = 1, \ldots, N. \tag{29}$$

This may be accomplished in two ways. Firstly, by applying the same fixed shift $\boldsymbol{\tau} = (-1/2, -1/2, -1/2)$ to all positional vectors. Then, as already observed in paper I (see I, §13), the new structure factor will be

$$F'_{\mathbf{p}} = \exp(2\pi i \mathbf{p} \cdot \boldsymbol{\tau}) F_{\mathbf{p}} \quad \text{and} \quad |F_{\mathbf{p}}| = |F'_{\mathbf{p}}|.$$

If we consider two crystal structures with the same structure-factor moduli to be identical, it may be concluded that two structures in $P1$, related by a fixed shift vector, are identical even if 'observed' through reflections with rational indices.

Secondly, by applying a proper lattice shift $\boldsymbol{\tau}_j$ to the jth atom when its coordinates do not satisfy (29) {*e.g.* if the jth atom has coordinates $(x_j, y_j, z_j) = (0.8, 0.3, 0.7)$, the shift $\boldsymbol{\tau}_j$ will be $[\bar{1}, 0, \bar{1}]$}. Then,

$$F'_{\mathbf{p}} = \sum_{j=1}^{N} f_j \exp[2\pi i \mathbf{p} \cdot (\mathbf{r}_j + \boldsymbol{\tau}_j)] \quad \text{and} \quad |F_{\mathbf{p}}| \ne |F'_{\mathbf{p}}|. \tag{30}$$

The following result arises: two $P1$ structures, one obtained from the other by shifting some atoms by proper lattice vectors, are different when 'observed' through rational indices reflections.

Let us now consider a crystal structure with space-group symmetry higher than $P1$. Let the atoms in the asymmetric unit have coordinates satisfying (28), while the equivalent positions are obtained by application of the symmetry operators [these values, therefore, can violate condition (28)]. Do the reflections with rational indices satisfy the space-group symmetry? Since

$$F_{\mathbf{p}} = \sum_{s=1}^{m} \sum_{j=1}^{t} f_j \exp(2\pi i \mathbf{p} \mathbf{C}_s \mathbf{r}_j), \tag{31}$$

the relation

$$F_{\mathbf{p}\mathbf{R}_\mathbf{q}} = F_{\mathbf{p}} \exp(-2\pi i \mathbf{h} \mathbf{T}_{\mathbf{q}}) \tag{32}$$

does not hold for reflections with rational indices unless the space group is symmorphic. Indeed,

$$F_{\mathbf{p}\mathbf{R}_\mathbf{q}} \exp(2\pi i \mathbf{p} \mathbf{T}_{\mathbf{q}}) = \sum_{s=1}^{m} \sum_{j=1}^{t} f_j \exp[2\pi i \mathbf{p}(\mathbf{R}_{\mathbf{q}} \mathbf{R}_{\mathbf{s}} \mathbf{r}_j + \mathbf{R}_{\mathbf{q}} \mathbf{T}_{\mathbf{s}} + \mathbf{T}_{\mathbf{q}})]. \tag{33}$$

If $\mathbf{R}_{\mathbf{n}} = \mathbf{R}_{\mathbf{q}} \mathbf{R}_{\mathbf{s}}$, it is easily seen that $\mathbf{T}'_{\mathbf{n}} = \mathbf{R}_{\mathbf{q}} \mathbf{T}_{\mathbf{s}} + \mathbf{T}_{\mathbf{q}}$ is different from $\mathbf{T}_{\mathbf{n}}$ by a vector $\boldsymbol{\tau}(\mathbf{q}, \mathbf{s})$. For example, in $P2_1$, $\mathbf{I} = \mathbf{R}_2 \mathbf{R}_2$ and $\mathbf{R}_2 \mathbf{T}_2 + \mathbf{T}_2 = [010]$. Therefore, (32) becomes

$$F_{\mathbf{p}\mathbf{R}_\mathbf{q}} \exp(2\pi i \mathbf{p} \mathbf{T}_{\mathbf{q}}) = \sum_{s=1}^{m} \sum_{j=1}^{t} f_j \exp\{2\pi i \mathbf{p}[\mathbf{R}_{\mathbf{n}} \mathbf{r}_j + \mathbf{T}_{\mathbf{n}} + \boldsymbol{\tau}(\mathbf{q}, \mathbf{s})]\},$$

which, according to (30), is not equivalent to $F_{\mathbf{p}}$ (*i.e.* $|F_{\mathbf{p}\mathbf{R}_\mathbf{q}}| \ne |\mathbf{F}_{\mathbf{p}}|$).

Let us now consider the case in which the atomic coordinates in the asymmetric unit satisfy (28), while proper shifts are applied to the equivalent atomic positions (as obtained by application of symmetry operators) to relocate these in the unit cell [so as to satisfy (28)]. Then, according to the first result described in this section, an additional loss of symmetry can occur even in symmorphic space groups. For example, half-integral index reflections satisfy the space-group symmetry for $P2$ and $P422$ but not for $P2_1$, $P4_1$ and $P3$. Similarly, the rules for the symmetry phase restriction which hold for the standard reflections can be violated for rational index reflections. For example, the phases of the reflections $(p_1, 0, p_3)$, with half-integral values of p_1 and p_3, are restricted to $(0, \pi)$, but reflections $(p_1, 0, p_3)$,

$(0, p_2, p_3)$, $(p_1, p_2, 0)$, with p_1, p_2, p_3 half-integers, are not restricted in $P2_12_12_1$ to $(0, \pi)$ or to $\pm\pi/2$. It is then clear that the space groups $P1$ and $P\bar{1}$ play a special role when reflections with rational indices are used; indeed, most of the calculations have to be made with reference to these groups.

10. The Hilbert-transform method

Let $F(z^*)$ be a complex function that approaches zero as z^* approaches infinity. Let x^* be the real part of z^*; then the real part [say $A(x^*)$] and the imaginary part [say $B(x^*)$] of $F(z^*)$ can be related by (Toll, 1956; London, 1973)

$$A(x^*) = \pi^{-1} P \int_{-\infty}^{\infty} \frac{B(x'^*)}{(x'^* - x^*)} \, dx'^* \tag{34}$$

and

$$B(x^*) = -\pi^{-1} P \int_{-\infty}^{\infty} \frac{A(x'^*)}{(x'^* - x^*)} \, dx'^*. \tag{35}$$

P denotes the Cauchy principal value, *i.e.*

$$P\int_a^b \frac{C(x^*)}{(x^* - x_0^*)} \, dx^* = \lim_{\delta \to 0} \left(\int_a^{x_0 - \delta} + \int_{x_0+\delta}^b \right) \frac{C(x^*)}{(x^* - x_0^*)} \, dx^*,$$

where a and b are the lower and upper bounds, respectively. The contributions of (34) and (35) give

$$F(x^*) = -i\pi^{-1} P \int_{-\infty}^{\infty} \frac{F(x'^*)}{(x'^* - x^*)} \, dx'^*. \tag{36}$$

Relationships (34) and (35) are known in mathematics as a Hilbert transform (HT), but are widely used in optics as Kramers–Kronig relations. In order to familiarize the reader with the Hilbert transform, we directly derive (34) and (35) in Appendix *B*, on the assumption that $F(x^*) = A(x^*) + iB(x^*)$ represents the general structure-factor expression.

Ramachandran (1969) first conjectured about the possible use of the HT to solve the phase problem in crystallography. He made the following assumptions (treated here in one dimension, for the sake of simplicity): (*a*) $z^* = x^*$ defines a generic point in the reciprocal space; (*b*) $F(x^*)$ represents the structure factor. It follows from (34) and (35) that if $B(x^*)$ [or $A(x^*)$] is known for the whole range $(-\infty, \infty)$, then $A(x^*)$ [or $B(x^*)$] may be calculated. The main obstacle to single-crystal experiments is that $F(x^*)$ vanishes for all values of x^*, except for the lattice points; as a consequence, the integral on the right-hand side of each of (34) and (35) cannot be calculated in practice. To overcome this difficulty, Ramachandran proposed a set of equations, which, however, involved unknown derivatives. Thus, the equations proved of little use in practice.

The problem was partially overcome by Mishnev (1993), who applied the Shannon sampling theorem (Shannon, 1949) to reconstruct $F(x^*)$ from the values sampled at the reciprocal-lattice points. It may be of use to the reader to recall such a basic theorem:

'Let $f(t)$ be a band-limited function with Fourier transform $\rho(x)$:

$$\rho(x) = 0 \quad \text{for} \quad |x| > a$$

$$f(t) = (2\pi)^{-1} \int_{-a}^{a} \rho(x) \exp(ixt) \, dx.$$

Then, $f(t)$ can be determined from its values $f(nT)$ at a sequence of equidistant points $t = nT$, provided $T \le \pi/a$, *via* the relation

$$f(t) = \sum_{n=-\infty}^{\infty} f(nt) \frac{\sin a'(t - nT)}{a'(t - nT)},$$

where $a' = \pi/T$.'

The integration of the sampling theorem into the Hilbert-transform relationships (34) and (35) led Mishnev to propose equations relating structure amplitudes with half-integral indices to structure amplitudes with integral indices and *vice versa*. In the notation adopted by Zanotti *et al.* (1996), such equations are written as

$$A(\mathbf{h}/2) = (8/\pi^3) \sum_{k_1} \sum_{k_2} \sum_{k_3} B(\mathbf{k}/2) \prod_{i=1}^{3} 1/(h_i - k_i), \tag{37a}$$

$$B(\mathbf{h}/2) = -(8/\pi^3) \sum_{k_1} \sum_{k_2} \sum_{k_3} A(\mathbf{k}/2) \prod_{i=1}^{3} 1/(h_i - k_i), \tag{37b}$$

$$A(\mathbf{k}/2) = (8/\pi^3) \sum_{h_1} \sum_{h_2} \sum_{h_3} B(\mathbf{h}/2) \prod_{i=1}^{3} 1/(k_i - h_i), \tag{37c}$$

$$B(\mathbf{k}/2) = -(8/\pi^3) \sum_{h_1} \sum_{h_2} \sum_{h_3} A(\mathbf{h}/2) \prod_{i=1}^{3} 1/(k_i - h_i). \tag{37d}$$

In the above relationships (37), $\mathbf{h}$ indicates an index with even integer components, and $\mathbf{k}$ one with odd integer components. It may be worthwhile noting that, in the summations at the right-hand sides of equations (37), the vector $\mathbf{q} = 0$ has to be included.

The reader may notice that the right-hand members of the equations of Zanotti *et al.* (1996) have opposite sign with respect to those reported here. This is probably a typographic error in the Zanotti *et al.* paper.

11. The Hilbert-transform method and the probabilistic approach in the canonical case

It may be useful to compare equations (37) with the relationships (CPR1) to (CPR4). We observe the following.

(*a*) Equations (37) are strictly valid only if the $\mathbf{q}$ reflections are regularly sampled and only if all of them are used in the summations at the right-hand side. On the contrary, (CPR1) to (CPR4) hold for any arbitrary, finite or infinite, set of reflections, no matter if they are regularly sampled or not. The limiting case in which the set of $\mathbf{q}$ reflections is constituted by only one reflection has been explicitly considered in §§3 and 4.

(*b*) When equations (37) are used as a truncated sampling expansion, the truncation error can be bounded (see Papoulis, 1968), but no reliability estimate is possible. On the contrary, our probabilistic approach always provides the variance to associate with each estimate.

(*c*) If the number of $\mathbf{q}$ reflections is sufficiently large, then the expected conditional $A_{\mathbf{p}}$ and $B_{\mathbf{p}}$ values provided by (CPR1) and (CPR3) should be related to the estimates provided by (37).

Let us prove statement (*c*) for the first option. Introducing (23) into (CPR1) and (CPR3) gives

$$\langle A_{\mathbf{p}}|\ldots\rangle = \sum_{\mathbf{q}} \left\{ \left[\textstyle\sum_{11}(\mathbf{p},\mathbf{q}) / \sum_2(\mathbf{q}) \right] (s_{\mathbf{p}+\mathbf{q}} + s_{\mathbf{q}-\mathbf{p}}) \right\} B_{\mathbf{q}} \tag{38}$$

and

$$\langle B_{\mathbf{p}}|\ldots\rangle = K_{01}(\mathbf{p}) + \sum_{\mathbf{q}} \left\{ \left[\textstyle\sum_{11}(\mathbf{p},\mathbf{q}) / \sum_2(\mathbf{q}) \right] \times (s_{\mathbf{p}+\mathbf{q}} + s_{\mathbf{p}-\mathbf{q}}) \right\} A_{\mathbf{q}}. \tag{39}$$

When $F_{\mathbf{q}}$ is *a priori* known, $F_{-\mathbf{q}}$ is known too, and $P(F_{\mathbf{p}}|F_{\mathbf{q}},\ldots) \equiv P(F_{\mathbf{p}}|F_{\mathbf{q}}, F_{-\mathbf{q}},\ldots)$. Accordingly, the summations at the right-hand sides of (38) and (39) [and, obviously, of the most general relationships (GPR1) to (GPR8)] will not contain Friedel mates. If we prefer to include Friedel mates explicitly in the summations, we can replace (38) and (39) by

$$\langle A_{\mathbf{p}}|\ldots\rangle = \sum_{\mathbf{q}}{}' \left\{ \left[\textstyle\sum_{11}(\mathbf{p},\mathbf{q}) / \sum_2(\mathbf{q}) \right] s_{\mathbf{q}-\mathbf{p}} \right\} B_{\mathbf{q}} \tag{40}$$

and

$$\langle B_{\mathbf{p}}|\ldots\rangle = K_{01}(\mathbf{p}) + \sum_{\mathbf{q}}{}' \left\{ \left[\textstyle\sum_{11}(\mathbf{p},\mathbf{q}) / \sum_2(\mathbf{q}) \right] s_{\mathbf{p}-\mathbf{q}} \right\} A_{\mathbf{q}}, \tag{41}$$

where the prime to the summation symbol warns the reader that Friedel pairs may be included. It is now more clear that, in (38) and (39), $s_{\mathbf{q}-\mathbf{p}} \equiv -s_{\mathbf{p}-\mathbf{q}}$ is the term arising from the prior knowledge of the $\mathbf{q}$ reflection, while $s_{-\mathbf{q}-\mathbf{p}} \equiv -s_{\mathbf{q}+\mathbf{p}}$ is the term arising from the simultaneous knowledge of the $(-\mathbf{q})$ reflection.

The numerical aspects of the relations (40) and (41) will now be examined.

(i) According to the definition, $s_{\mathbf{p}} = \sin(\pi p_s) c_{p_1/2} c_{p_2/2} c_{p_3/2}$. Since $\mathbf{p}$ is a vector with half-integer components,

$$\sin(\pi p_s) = (-1)^{(2p_s-1)/2}$$

and

$$c_{p_i/2} = (\pi p_i)^{-1} (-1)^{(2p_i-1)/2}.$$

Then,

$$\begin{aligned} s_{\mathbf{p}} &= -\pi^{-3}(p_1 p_2 p_3)^{-1}, \\ s_{\mathbf{p}-\mathbf{q}} &= -\pi^{-3}[(p_1-q_1)(p_2-q_2)(p_3-q_3)]^{-1}. \end{aligned}$$

(ii) At the end of §6, it was explicitly noted that the contribution to the estimate of $F_{\mathbf{p}}$ arising from the prior knowledge of F_{000} is automatically included in the relationships (GRP1) to (GRP8). Therefore, (40) and (41) also must not include the vector $\mathbf{q} = (0, 0, 0)$ in the $\mathbf{q}$ set. On the contrary, $\mathbf{h} = (0, 0, 0)$ has to be explicitly included in the right-hand side of the equations (37*c*) and (37*d*). In order to make (37*c*), (37*d*), (40) and (41) homogeneous, we observe that the contribution to $\langle A_{\mathbf{p}}|\ldots\rangle$ arising from the prior knowledge of F_{000} is vanishing, while that to be associated with $\langle A_{\mathbf{p}}|\ldots\rangle$ is $K_{01}(\mathbf{p})$. An approximate value of $K_{01}(\mathbf{p})$ may be obtained by allowing $\mathbf{q}$ to assume the $\mathbf{0}$ value in (41):

$$\left[\textstyle\sum_{11}(\mathbf{p},\mathbf{0}) / \sum_2(\mathbf{0}) \right] A_{\mathbf{0}} s_{\mathbf{p}} = \left[\textstyle\sum_{11}(\mathbf{p},\mathbf{0}) / \sum_2(\mathbf{0}) \right] K_{01}(\mathbf{p}). \tag{42}$$

The value of (42) depends on the resolution of the reflection $\mathbf{p}$. If this is neglected, the following approximation holds:

$$\left[\textstyle\sum_{11}(\mathbf{p},\mathbf{0}) / \sum_2(\mathbf{0}) \right] K_{01}(\mathbf{p}) \simeq K_{01}(\mathbf{p}). \tag{43}$$

(iii) The term

$$\textstyle\sum_{11}(\mathbf{p},\mathbf{q}) / \sum_2(\mathbf{q}) \equiv \left[\displaystyle\sum_{j=1}^{N} f_j(\mathbf{p}) f_j(\mathbf{q}) \right] \Big/ \left[\displaystyle\sum_{j=1}^{N} f_j^2(\mathbf{q}) \right]$$

is a resolution-dependent function: its value changes with $\mathbf{q}$. If $\mathbf{p}$ is very close to $\mathbf{q}$ (the largest contributions occur in this case), then

$$\textstyle\sum_{11}(\mathbf{p},\mathbf{q}) / \sum_2(\mathbf{q}) \simeq 1. \tag{44}$$

If the approximations (43) and (44) are introduced into (40) and (41), one obtains

$$\langle A_{\mathbf{p}}|\ldots\rangle \approx \pi^{-3} \sum_{\mathbf{q}}{}' B_{\mathbf{q}} \prod_{j=1}^{3} (p_i - q_i)^{-1} \tag{45}$$

and

$$\langle B_{\mathbf{p}}|\ldots\rangle \approx -\pi^{-3}\sum_{\mathbf{q}}{}' A_{\mathbf{q}} \prod_{j=1}^{3}(p_i - q_i)^{-1} \qquad (46)$$

where now $\mathbf{q} = \mathbf{0}$ is included in the summations. The relations (45) and (46) are identical to (37*c*) and (37*d*) if one recalls that $\mathbf{k} = 2\mathbf{p}$, $\mathbf{h} = 2\mathbf{q}$.

A similar procedure may be followed to relate, for the second option, (CPR1) and (CPR2) to (37*a*) and (37*b*). The combination of (26) with (CPR1) and (CPR2) gives

$$\langle A_{\mathbf{p}}|\ldots\rangle = \sum_{\mathbf{q}} [K_{14}(\mathbf{p},\mathbf{q})/K_{02}(\mathbf{q})][B_{\mathbf{q}} - K_{01}(\mathbf{q})] \qquad (47)$$

and

$$\langle B_{\mathbf{p}}|\ldots\rangle = \sum_{\mathbf{q}} [K_{23}(\mathbf{p},\mathbf{q})/K_{20}(\mathbf{q})]A_{\mathbf{q}}. \qquad (48)$$

$K_{01}(\mathbf{q})$ is non-negligible only for very low resolution reflections. If we adopt the approximations

$$B_{\mathbf{q}} - K_{01}(\mathbf{q}) \approx B_{\mathbf{q}}$$

and

$$K_{02}(\mathbf{q}) = \textstyle\sum_2(\mathbf{q})(1 - 2s_{\mathbf{q}}^2)/2 \approx \sum_2(\mathbf{q})/2$$

and we introduce also the other approximations used for the first option, we obtain again (45) and (46). Now, however, the contribution corresponding to $\mathbf{q} = \mathbf{0}$ has not to be included in the summations since $\mathbf{q}$ represents the set of vectors with half-integral components. Relationships (45) and (46) are identical to (37*a*) and (37*b*).

12. Conclusions

The method of joint probability distribution functions has been used to estimate structure factors with rational indices given prior knowledge of other structure factors with rational indices. A general approach was first described which allows the use of any type of reflections. Then the canonical case was studied (involving standard reflections and half-integral index reflections) and more simple formulae were derived. The final formulae were compared with those obtained by Mishnev (1993) by applying the Shannon sampling theorem to the Hilbert-transform relationships. It is shown that our probabilistic formulae (*a*) can be applied to any subset of reflections and are always able to provide the reliability of the estimates, and (*b*) encompass Mishnev relationships. For brevity, the experimental calculations were not included. A following paper will be devoted to such experimental aspects, but we anticipate that the formulae derived here will prove to be quite reliable.

APPENDIX *A*

Let us suppose that the primitive random variables x_j, y_j and z_j, $j = 1, \ldots, N$, are independently and uniformly distributed in the interval (0,1). Then,

$$\begin{aligned}\langle A_{\mathbf{p}}B_{\mathbf{q}}\rangle &= \sum_{j_1,j_2=1}^{N} f_{j_1}(\mathbf{p})f_{j_2}(\mathbf{q})\langle\cos(2\pi\mathbf{p}\cdot\mathbf{r}_{j_1})\sin(2\pi\mathbf{q}\cdot\mathbf{r}_{j_2})\rangle\\ &= \sum_{j=1}^{N} f_j(\mathbf{p})f_j(\mathbf{q})\langle\tfrac{1}{2}\{\sin[2\pi(\mathbf{p}+\mathbf{q})\cdot\mathbf{r}_j]\\ &\quad + \sin[2\pi(\mathbf{q}-\mathbf{p})\cdot\mathbf{r}_j]\}\rangle\\ &\quad + \left[\sum_{j_1\neq j_2=1}^{N} f_{j_1}(\mathbf{p})f_{j_2}(\mathbf{q})\right]c_{\mathbf{p}}s_{\mathbf{q}}\\ &= \tfrac{1}{2}\textstyle\sum_{11}(\mathbf{p},\mathbf{q})(s_{\mathbf{p}+\mathbf{q}} + s_{\mathbf{q}-\mathbf{p}})\\ &\quad + \sum_{j_1\neq j_2=1}^{N} f_{j_1}(\mathbf{p})f_{j_2}(\mathbf{q})c_{\mathbf{p}}s_{\mathbf{q}}. \end{aligned} \qquad (49)$$

Since

$$\textstyle\sum_1(\mathbf{p})\sum_1(\mathbf{q}) = \sum_{11}(\mathbf{p},\mathbf{q}) + \displaystyle\sum_{j_1\neq j_2=1}^{N} f_{j_1}(\mathbf{p})f_{j_2}(\mathbf{q}),$$

(49) may be written as

$$\begin{aligned}\langle A_{\mathbf{p}}B_{\mathbf{q}}\rangle &= \tfrac{1}{2}\textstyle\sum_{11}(\mathbf{p},\mathbf{q})(s_{\mathbf{p}+\mathbf{q}} + s_{\mathbf{q}-\mathbf{p}} - c_{\mathbf{p}}s_{\mathbf{q}})\\ &\quad + \textstyle\sum_1(\mathbf{p})\sum_1(\mathbf{q})c_{\mathbf{p}}s_{\mathbf{q}}.\end{aligned} \qquad (50)$$

Owing to the fact that

$$\langle A_{\mathbf{p}}\rangle = \textstyle\sum_1(\mathbf{p})c_{\mathbf{p}} \quad \text{and} \quad \langle B_{\mathbf{q}}\rangle = \sum_1(\mathbf{q})s_{\mathbf{q}},$$

from (50) we obtain

$$\begin{aligned}K_{14}(\mathbf{p},\mathbf{q}) &= \langle A_{\mathbf{p}}B_{\mathbf{q}}\rangle - \langle A_{\mathbf{p}}\rangle\langle B_{\mathbf{q}}\rangle\\ &= \tfrac{1}{2}\textstyle\sum_{11}(\mathbf{p},\mathbf{q})(s_{\mathbf{p}+\mathbf{q}} + s_{\mathbf{q}-\mathbf{p}} - 2c_{\mathbf{p}}s_{\mathbf{q}}).\end{aligned} \qquad (51)$$

APPENDIX *B*

Let us assume that (*a*) $\rho(x)$ is the electron density defined in the unit cell (*e.g.*, for the one-dimensional case, $0 \leq x \leq 1$), (*b*) $F(p) = A(p) + iB(p)$ is the structure factor (p denotes the coordinate of a fixed point in reciprocal space), (*c*) q is the coordinate of a generic point in reciprocal space. Consider

$$\begin{aligned}P\int_{-\infty}^{\infty}\frac{B(q)}{q-p}\,\mathrm{d}q &= P\int_{-\infty}^{\infty}\mathrm{d}q\int_0^1\rho(x)\frac{\sin(2\pi qx)}{q-p}\,\mathrm{d}x\\ &= \int_0^1 \mathrm{d}x\rho(x)P\int_{-\infty}^{\infty}\frac{\sin(2\pi qx)}{q-p}\,\mathrm{d}q,\end{aligned} \qquad (52)$$

where P denotes the Cauchy principal value. If the variable q in (52) is replaced by $\Delta = q - p$ and the relation

$$P\int_{-\infty}^{\infty}\frac{\sin(2\pi\Delta x)}{\Delta}\,\mathrm{d}\Delta = 2P\int_0^{\infty}\frac{\sin(2\pi\Delta x)}{\Delta}\,\mathrm{d}\Delta = \pi$$

is applied, then (52) reduces to

$$\pi\int_0^1 \rho(x)\cos(2\pi px)\,\mathrm{d}x = \pi A(p),$$

from which the required Hilbert transform

$$A(p) = \frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{B(q)}{q-p}\,\mathrm{d}q \tag{53}$$

is obtained. The same procedure may be applied to the integral

$$P\int_{-\infty}^{\infty} \frac{A(q)}{q-p}\,\mathrm{d}q = P\int_{-\infty}^{\infty} \mathrm{d}q \int_0^1 \rho(x)\frac{\cos(2\pi qx)}{q-p}\,\mathrm{d}x,$$

yielding

$$B(p) = -\frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{A(q)}{q-p}\,\mathrm{d}q, \tag{54}$$

so that

$$\begin{aligned} F(p) &= A(p) + iB(p) \\ &= \frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{B(q) - iA(q)}{q-p}\,\mathrm{d}q \\ &= -\frac{i}{\pi} P \int_{-\infty}^{\infty} \frac{F(q)}{q-p}\,\mathrm{d}q. \end{aligned} \tag{55}$$

Let us now explicitly consider the three-dimensional case: $\bar{\mathbf{q}} = (q_1, q_2, q_3)$ and $\bar{\mathbf{p}} = (p_1, p_2, p_3)$ are the generic points in the reciprocal space S^*. Then,

$$\begin{aligned} &P\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \frac{B(q_1,q_2,q_3)}{(q_1-p_1)(q_2-p_2)(q_3-p_3)}\,\mathrm{d}q_1\,\mathrm{d}q_2\,\mathrm{d}q_3 \\ &= \int_0^1\int_0^1\int_0^1 \rho(x,y,z)\,\mathrm{d}x\,\mathrm{d}y\,\mathrm{d}z \\ &\quad\times P\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \frac{\sin 2\pi(q_1x+q_2y+q_3z)}{(q_1-p_1)(q_2-p_2)(q_3-p_3)}\,\mathrm{d}q_1\,\mathrm{d}q_2\,\mathrm{d}q_3 \\ &= -\pi^3\int_0^1\int_0^1\int_0^1 \rho(x,y,z)\cos 2\pi(p_1x+p_2y+p_3z)\,\mathrm{d}x\,\mathrm{d}y\,\mathrm{d}z \\ &= -\pi^3 A(\mathbf{p}). \end{aligned}$$

Therefore,

$$A(\mathbf{p}) = -\pi^{-3} P \int_{S^*} \frac{B(\mathbf{q})}{(q_1-p_1)(q_2-p_2)(q_3-p_3)}\,\mathrm{d}S^*. \tag{56}$$

In the same way, one can find

$$B(\mathbf{p}) = \pi^{-3} P \int_{S^*} \frac{A(\mathbf{q})}{(q_1-p_1)(q_2-p_2)(q_3-p_3)}\,\mathrm{d}S^*, \tag{57}$$

so that

$$F(\mathbf{p}) = (-i/\pi)^{-3} P \int_{S^*} \frac{F(\mathbf{q})}{(q_1-p_1)(q_2-p_2)(q_3-p_3)}\,\mathrm{d}S^*. \tag{58}$$

It should be useful to note that the dimension of the reciprocal space defines the integrand on the right-hand side of the relationships (53), (54), (56) and (57). In particular, in the two-dimensional case,

$$A(\mathbf{p}) = -\pi^{-2} P \int_{S^*} \frac{A(\mathbf{q})}{(q_1-p_1)(q_2-p_2)}\,\mathrm{d}S^*$$

and

$$B(\mathbf{p}) = -\pi^{-2} P \int_{S^*} \frac{B(\mathbf{q})}{(q_1-p_1)(q_2-p_2)}\,\mathrm{d}S^*.$$

Accordingly, in an n-dimensional case, the general formula to apply is

$$F(\mathbf{p}) = (-i/\pi)^n P \int_{S^*} \frac{F(\mathbf{q})}{\prod_{j=1}^{n}(q_j-p_j)}\,\mathrm{d}S^*. \tag{59}$$

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