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A Probabilistic Theory of the Coincidence Method. II. Non-centrosymmetric Space Groups

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A theory is given for non-centrosymmetric space groups which permits, for each specially related pair of phases of type $(\varphi_{\mathbf{H}_1+\mathbf{H}_2}, \varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q})$, the calculation of the expected value of a seminvariant cosine. In order to make full use of the symmetry, the mathematical device of the joint probability distribution functions has been suitably combined with the space-group algebra. The expected value of $(\varphi_{\mathbf{H}_1+\mathbf{H}_2} - \varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q})$ is given by means of two mathematical approaches. The first uses the Gram–Charlier expansion of the characteristic function, the second calculates directly the Fourier transform of the exponential expression of the characteristic function.

Introduction

In part I of this paper (Giacovazzo, 1977) a theory of sign coincidences has been described valid in all the centrosymmetric space groups. In order to estimate the strength of the various coincidences, the mathematical device of joint probability distribution functions was used. In this section of the paper a general probabilistic theory of phase coincidences in all non-centrosymmetric space groups is described. In our calculations atomic positions are the primitive random variables while the reciprocal vectors are assumed to be fixed. As in the centrosymmetric cases, the method requires, for the estimation of non-vanishing cumulants, the use of space-group algebra. Appendices *A* and *B* will help the reader carry out this algebraic analysis.

The mathematical approach

The method requires the derivation of a variety of conditional probability distributions. We denote by $P(A_1,A_2,\ldots,A_n,B_1,B_2,\ldots,B_n)$ the joint probability distribution function of n normalized structure factors; A_i and B_i represent the real and imaginary parts, respectively, of the ith factor. The characteristic function of the distribution is

$$C(u_1,u_2,\ldots,u_n,v_1,\ldots,v_n)=\exp\left[\sum_{2}^{\infty}{}_{v}\frac{S_v}{t^{v/2}}\right], \quad (1)$$

where $u_j, v_j, j=1,\ldots,n$, are carrying variables associated with A_j and B_j respectively, t is the number of independent atoms in the unit cell,

$$S_v=t\sum_{r+s+\ldots+w=v}\frac{1}{2^{v/2}}\frac{\lambda_{rs\ldots w}}{r!s!\ldots w!}\times(iu_1)^r(iu_2)^s\ldots(iv_n)^w,$$

and

$$\lambda_{rs\ldots w}=\frac{K_{rs\ldots w}}{K_{200}^{r/2}\ldots K_{020}^{s/2}\ldots K_{0\ldots2}^{w/2}}.$$

$K_{rs\ldots w}$ are the cumulants of the distribution.

The probability distribution is obtained by calculating the Fourier transform of (1). After suitable change of variables we obtain

$$P(R_1,\ldots,R_n,\varphi_1,\ldots,\varphi_n)=\frac{1}{(2\pi)^{2n}}\times\int_0^\infty\ldots\int_0^\infty\int_0^{2\pi}\ldots\int_0^{2\pi}\exp\{-i[\sqrt{2}\varrho_1R_1\cos(\psi_1-\varphi_1)+\ldots+\sqrt{2}\varrho_nR_n\cos(\psi_n-\varphi_n)]\}\times\exp[-\tfrac{1}{2}(\varrho_1^2+\ldots+\varrho_n^2)]2^n\left[\sum_{3}^{\infty}{}_{v}\frac{S'_v}{t^{v/2}}\right]\times R_1R_2\ldots R_n\varrho_1\varrho_2\ldots\varrho_n\mathrm{d}\varrho_1\ldots\mathrm{d}\varrho_n\mathrm{d}\psi_1\ldots\mathrm{d}\psi_n, \quad (2)$$

where

$$S'_v=t\sum_{r+s+\ldots+w=v}\frac{\lambda_{rs\ldots w}}{r!s!\ldots w!}\times(i\varrho_1\cos\psi_1)^r(i\varrho_2\cos\psi_2)^s\ldots(i\varrho_n\sin\psi_n)^w.$$

We will use two different methods to derive the probability densities. The first uses a Gram–Charlier expansion of (1), about which the reader will find exhaustive information in earlier papers (Giacovazzo, 1974*a*, 1976). The second method calculates (2) directly: a general account is given in Appendix *C*.

Both methods involve a number of integral formulas which we list for convenient reference (Watson, 1958):

$$\frac{i^m}{\pi}\int_0^\pi\exp(-iz\cos\varphi)\cos m\varphi\mathrm{d}\varphi=J_m(z); \quad (3a)$$

$$\int_0^\pi\exp(-iz\cos\varphi)\sin m\varphi\mathrm{d}\varphi=0; \quad (3b)$$

$$\int_0^\infty J_v(at)\exp(-p^2t^2)t^{\mu-1}\mathrm{d}t=\frac{(a/2p)^v\Gamma[(v+\mu)/2]}{2p^\mu\Gamma(v+1)}\,{}_1F_1\left(\frac{v+\mu}{2};v+1;-\frac{a^2}{4p^2}\right), \quad (3c)$$

where Γ represents the gamma-function and ${}_1F_1(x;y;z)$ is the generalized hypergeometric function.

Furthermore, from elementary trigonometry

$$\sum_n A_n \exp\{i(\varphi+b_n)\} = Y \exp\{i(\varphi+\xi)\}, \qquad (3d)$$

where Y and ξ are determined by

$$Y=[\sum_{\nu,n} A_\nu A_n \cos(b_\nu-b_n)]^{1/2},$$

$$Y\exp(i\xi)=\sum_n A_n \exp(ib_n).$$

The probability distribution $P(R_{\mathbf{H}_1}, R_{\mathbf{H}_2}, R_{\mathbf{H}_1+\mathbf{H}_2}, R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}, \varphi_{\mathbf{H}_1}, \varphi_{\mathbf{H}_2}, \varphi_{\mathbf{H}_1+\mathbf{H}_2}, \varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q})$ when the Gram–Charlier expansion of the characteristic function is used

As in the centrosymmetric space groups, let us study the distribution $P(E_{\mathbf{H}_1}, E_{\mathbf{H}_2}, E_{\mathbf{H}_1+\mathbf{H}_2}, E_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q})$. We introduce the abbreviations

$$E_1=R_1\exp i\varphi_1=A_1+iB_1=E_{\mathbf{H}_1}$$
$$E_2=R_2\exp i\varphi_2=\ldots=E_{\mathbf{H}_2};$$
$$E_3=R_3\exp i\varphi_3=E_{\mathbf{H}_1+\mathbf{H}_2};$$
$$E_4=R_4\exp i\varphi_4=E_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}.$$

From Appendices A and B we obtain

$$S_3'/t^{3/2}=\frac{(i)^3}{\sqrt{2N}}\{\varrho_1\varrho_2\varrho_3\cos(\psi_1+\psi_2-\psi_3)$$
$$+\varrho_1\varrho_2\varrho_4\cos[\psi_1+\psi_2-\psi_4-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)]\},$$

$$S_4'/t^2=\frac{1}{N}\{-\tfrac{1}{16}(\varrho_1^4+\varrho_2^4+\varrho_3^4+\varrho_4^4)$$
$$+\gamma(\varrho_1^2+\varrho_2^2)\varrho_3\varrho_4\cos[\psi_3-\psi_4-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)]\}.$$

γ (see Appendix B) is a number which depends on the actual symmetry class and, for a given class, on the actual operators $\mathbf{C}_p\equiv(\mathbf{R}_p,\mathbf{T}_p)$ and $\mathbf{C}_q\equiv(\mathbf{R}_q,\mathbf{T}_q)$. For example, in classes 2, m, 222 γ is always 1 whatever p and q may be. In $mm2$ $\gamma=2$ for symmetry operations corresponding to symmetry planes and $\gamma=1$ for symmetry operations involving only rotation axes.

The estimation of (2) may be carried out by repeated application of (3a), (3b) and (3c). We obtain

$$P(R_1,R_2,R_3,R_4,\varphi_1,\varphi_2,\varphi_3,\varphi_4)$$
$$=\frac{1}{\pi^4}R_1R_2R_3R_4\exp(-R_1^2-R_2^2-R_3^2-R_4^2)$$
$$\times\left\{1+\frac{2}{\sqrt{N}}R_1R_2R_3\cos(\varphi_1+\varphi_2-\varphi_3)\right.$$
$$+\frac{2}{\sqrt{N}}R_1R_2R_4 \times\cos[\varphi_1+\varphi_2-\varphi_4-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)]$$
$$+\frac{1}{N}[+q+R_1^2R_2^2R_3^2\cos 2(\varphi_1+\varphi_2-\varphi_3)]$$
$$+\frac{1}{N}R_1^2R_2^2R_4^2 \times\cos 2[(\varphi_1+\varphi_2-\varphi_4-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)]$$
$$+\frac{2}{N}R_1^2R_2^2R_3R_4 \times\cos[2\varphi_1+2\varphi_2-\varphi_3-\varphi_4-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)]$$
$$+\frac{2}{N}(1-R_1^2)(1-R_2^2)R_3R_4 \times\cos[\varphi_3-\varphi_4-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)]$$
$$+\frac{\gamma}{N}R_3R_4(R_1^2+R_2^2-2) \left.\times\cos[\varphi_3-\varphi_4-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)]+\ldots\right\},$$

where

$$q=(R_1^2-1)(R_2^2-1)(R_3^2-1)+(R_1^2-1)(R_2^2-1)(R_4^2-1)$$
$$-\tfrac{1}{4}(R_1^4+R_2^4+R_3^4+R_4^4)+R_1^2+R_2^2+R_3^2+R_4^2-2.$$

Let us calculate the marginal probability density

$$P(R_1,R_2,R_3,R_4,\varphi_3,\varphi_4)$$
$$=\int_{-\pi}^{\pi}P(R_1,R_2,R_3,R_4,\varphi_1,\varphi_2,\varphi_3,\varphi_4)\mathrm{d}\varphi_1\mathrm{d}\varphi_2$$
$$=\frac{4}{\pi^2}R_1R_2R_3R_4\exp(-R_1^2-R_2^2-R_3^2-R_4^2)\left\{1+\frac{q}{N}\right.$$
$$+\frac{1}{N}R_3R_4[2(1-R_1^2)(1-R_2^2)+\gamma(R_1^2+R_2^2-2)]$$
$$\left.\times\cos[\varphi_3-\varphi_4-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)]\right\}. \qquad (5)$$

If we define

$$\Phi=\varphi_3-\varphi_4-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q),$$

the desired conditional distribution $P(\Phi|R_1,R_2,R_3,R_4)$ is obtained from (5) by fixing the values of R_1,R_2,R_3,R_4 and renormalizing. By transforming (5) in exponential form (Bertaut, 1960a,b; Karle, 1972) we obtain finally

$$P(\Phi|R_1,R_2,R_3,R_4)\simeq\frac{1}{2\pi I_0(G)}\exp(G\cos\Phi), \qquad (6)$$

where

$$G=N^{-1}R_3R_4[2(1-R_1^2)(1-R_2^2)+\gamma(R_1^2+R_2^2-2)]. \qquad (7)$$

There is no problem in calculating from (6) the following functions (Hauptman, 1972a):

$$\langle\cos\Phi|G\rangle=\frac{I_1(G)}{I_0(G)}, \qquad (8)$$

$$\mathrm{var}[\cos\Phi|G]=1-\frac{I_1(G)}{GI_0(G)}-\frac{I_1^2(G)}{I_0^2(G)}, \qquad (9)$$

$$\langle\sin\Phi|G\rangle=0, \qquad (10)$$

$$\mathrm{var}[\sin\Phi|G]=\frac{I_1(G)}{GI_0(G)}. \qquad (11)$$

The expected value of $\cos\Phi$ is positive if $G\geq0$, negative if $G<0$: large values of $R_{\mathbf{H}_1+\mathbf{H}_2}R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}$ strengthen the sign indication.

Probabilistic considerations on $\Phi_e=\varphi_{\mathbf{H}_1+\mathbf{H}_2}-\varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}$

As shown in part I of this paper, given a specially related pair of phases (s.r.p.p. from now on) with indices $\mathbf{U}=\mathbf{H}_1+\mathbf{H}_2$ and $\mathbf{V}=\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q$, several pairs $(\mathbf{H}_1,\mathbf{H}_2)$ may be found each capable of giving an estimate of $\Phi=\varphi_{\mathbf{U}}-\varphi_{\mathbf{V}}-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)$. As the value of $\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q$, with fixed $\mathbf{R}_p$ and $\mathbf{R}_q$, is a function of the actual pair $(\mathbf{H}_1,\mathbf{H}_2)$, it is useful to obtain, instead of Φ, an estimate of $\Phi_e=\varphi_{\mathbf{U}}-\varphi_{\mathbf{V}}$. From (5) we derive

$$P(\Phi_e)=\frac{1}{2\pi I_0(G)}\exp\{G\cos[\Phi_e-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)]\}, \tag{12}$$

$$\langle\cos\Phi_e\rangle=\cos 2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)\frac{I_1(G)}{I_0(G)}, \tag{13}$$

$$\begin{aligned}\operatorname{var}[\cos\Phi_e]=\tfrac{1}{2}+\left[\tfrac{1}{2}-\frac{I_1(G)}{GI_0(G)}\right]\cos 4\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)\\ -\tfrac{1}{2}\frac{I_1^2(G)}{I_0^2(G)}-\tfrac{1}{2}\frac{I_1^2(G)}{I_0^2(G)}\cos 4\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q),\end{aligned}$$

$$\langle\sin\Phi_e\rangle=\sin 2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)\frac{I_1(G)}{I_0(G)},$$

$$\begin{aligned}\operatorname{var}[\sin\Phi_e]=\tfrac{1}{2}-\left[\tfrac{1}{2}-\frac{I_1(G)}{GI_0(G)}\right]\cos 4\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)\\ -\tfrac{1}{2}\frac{I_1^2(G)}{I_0^2(G)}+\tfrac{1}{2}\frac{I_1^2(G)}{I_0^2(G)}\cos 4\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q).\end{aligned}$$

The value of $(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)$ plays a critical role in assigning the average and the variance values. In particular, we emphasize that the variance of $\cos\Phi_e$ is not always smaller than that of $\sin\Phi_e$. For example, if $\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q=(2n+1)/4$, it turns out that

$$\langle\cos\Phi_e\rangle=0,$$

$$\langle\sin\Phi_e\rangle=\pm I_1(G)/I_0(G),$$

$$\operatorname{var}[\cos\Phi_e]=\frac{I_1(G)}{GI_0(G)},$$

$$\operatorname{var}[\sin\Phi_e]=1-\frac{I_1(G)}{GI_0(G)}-\frac{I_1^2(G)}{I_0^2(G)}.$$

As we see, the variance of the cosine in this case is larger than that of the sine.

We note now that

$$\operatorname{var}[\cos\Phi_e]+\operatorname{var}[\sin\Phi_e]=1-\frac{I_1^2(G)}{I_0^2(G)},$$

which is a quantity that does not vary with the symmetry operations and depends on the value of $|G|$ alone. This suggests that the value of Φ_e should be more precisely determined the larger is $|G|$. In accordance with this conclusion let us show that the variance of Φ_e does not depend on the actual value of $2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)$ but on $|G|$ alone. From

$$\langle\Phi_e\rangle=\frac{1}{2\pi I_0(G)}\times\int_{-\pi}^{\pi}\Phi_e\exp\{G\cos[\Phi_e-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)]\}\mathrm{d}\Phi_e,$$

we obtain, if $G>0$,

$$\langle\Phi_e\rangle=2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q); \tag{14a}$$

if $G<0$,

$$\langle\Phi_e\rangle=\pi+2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q). \tag{14b}$$

In view of (14*b*), when G is negative the variance equals

$$\begin{aligned}\langle\Phi_e^2\rangle-\langle\Phi_e\rangle^2=\frac{1}{2\pi I_0(G)}\\ \times\int_{-\pi}^{\pi}\Phi_e^2\exp\{-|G|\cos[\Phi_e-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)]\}\mathrm{d}\Phi_e\\ -[\pi+2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)]^2.\end{aligned}$$

After the change of variable $\Phi_e=-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)=\psi+\pi$, the above expression becomes

$$\frac{1}{2\pi I_0(G)}\int_{-\pi}^{\pi}\Phi_e^2\exp(|G|\cos\Phi_e)\mathrm{d}\Phi_e, \tag{15}$$

which is the variance expression when $G>0$. As is well known (Karle & Karle, 1966), (15) leads to

$$\operatorname{var}[\Phi_e]=\frac{\pi^2}{3}+[I_0(|G|)]^{-1}\sum_{1n}^{\infty}\left[\frac{I_{2n}(|G|)}{n^2}\right]-4[I_0(|G|)]^{-1}\frac{\sum_{0n}^{\infty}[I_{2n+1}(|G|)]}{(2n+1)^2}.$$

The infinite series converges quite rapidly for the values of G usually encountered in practice.

(14) is a fundamental result of the present paper. It proves that the condition

$$\langle\Phi_e\rangle\simeq\pi \tag{16}$$

can exist for symmorphic space groups. The ability to obtain such results in these space groups is one of the characteristics which distinguishes this theory from that of Debaerdemaeker & Woolfson (1972).

Comparison with a central-limit-theorem approach

A formal theory of coincidence phase relationships has already been given by Debaerdemaeker & Woolfson (1972). In our notation, these authors made direct use of the probability distributions

$$P(\varphi_{\mathbf{H}_1+\mathbf{H}_2})\simeq[2\pi I_0(A_1)]^{-1}\times\exp[A_1\cos(\varphi_{\mathbf{H}_1}+\varphi_{\mathbf{H}_2}-\varphi_{\mathbf{H}_1+\mathbf{H}_2})], \tag{17}$$

$$P_2(\varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q})\simeq[2\pi I_0(A_2)]^{-1}\exp\{A_2\cos[\varphi_{\mathbf{H}_1}+\varphi_{\mathbf{H}_2}-\varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)]\}, \quad (18)$$

where

$$A_1=2R_{\mathbf{H}_1}R_{\mathbf{H}_2}R_{\mathbf{H}_1+\mathbf{H}_2}/\sqrt{N},$$
$$A_2=2R_{\mathbf{H}_1}R_{\mathbf{H}_2}R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}/\sqrt{N},$$

in order to calculate the probability distribution of Φ_e.

On the assumption that $P_1(\varphi_{\mathbf{H}_1+\mathbf{H}_2}|\varphi_{\mathbf{H}_1},\varphi_{\mathbf{H}_2},R_{\mathbf{H}_1}, R_{\mathbf{H}_2}, R_{\mathbf{H}_1+\mathbf{H}_2})$ and $P_2(\varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}|\varphi_{\mathbf{H}_1}, \varphi_{\mathbf{H}_2}, R_{\mathbf{H}_1}, R_{\mathbf{H}_2}, R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q})$ are statistically independent of one another, Debaerdemaeker & Woolfson (1972) obtained the relevant result

$$P_{A_1,A_2}(\Phi_e)=\int_{-\pi}^{\pi}P_1(\varphi_{\mathbf{H}_1+\mathbf{H}_2})P_2(\varphi_{\mathbf{H}_1+\mathbf{H}_2}-\Phi_e)\mathrm{d}\varphi_{\mathbf{H}_1+\mathbf{H}_2}$$
$$=[2\pi I_0(A_1)I_0(A_2)]^{-1}I_0(\{A_1^2+2A_2^2+2A_1A_2\cos[\Phi_e-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)]\}^{1/2}). \quad (19)$$

From a formal point of view, (12), (17) and (18) are von Mises distributions but (19) is not. In order to compare (19) with our (12) we approximate (19) by a suitable von Mises distribution.

As is well known (Stephens, 1963), (17) and (18) may be approximated by wrapped normal distributions of type

$$P(\varphi)=\left(1+2\sum_{1\,r}^{\infty}\varrho^{r^2}\cos r\varphi\right)\Big/2\pi, \quad (20)$$

where $\varrho=\exp(-\frac{1}{2}\sigma^2)$. In (20) σ^2 assumes the values σ_1^2 and σ_2^2 respectively, defined by

$$\exp(-\sigma_1^2/2)=I_1(A_1)/I_0(A_1),$$
$$\exp(-\sigma_2^2/2)=I_1(A_2)/I_0(A_2).$$

In view of the additive property of the wrapped normal distributions, the convolution of these two distributions is again a wrapped normal distribution with parameter $\sigma_3^2=\sigma_1^2+\sigma_2^2$, which in turn may be approximated by a von Mises distribution. Then a satisfactory approximation of (19) is

$$P'(\Phi_e)=[2\pi I_0(G')]^{-1}\times\exp\{G'\cos[\Phi_e-2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)]\} \quad (21)$$

where G' is the solution of

$$\frac{I_1(G')}{I_0(G')}=\frac{I_1(A_1)}{I_0(A_1)}\,\frac{I_1(A_2)}{I_0(A_2)}.$$

As A_1 and A_2 are always positive, G' is always positive, whereas our G in (12) may in principle be negative. In conclusion, (12) and (19) suggest a different use of the information contained in the small $R_{\mathbf{H}_1}$ and $R_{\mathbf{H}_2}$ values. The postulate of statistical independence between (17) and (18) assumed in Debaerdemaeker & Woolfson's (1972) theory leads to a formula [*i.e.* (19) or (21)] which is not able in symmorphic space groups to indicate s.r.p.p.'s for which $\langle\Phi_e\rangle\neq0$. On the other hand, in the theory developed here the contribution of weak reflexions $\mathbf{H}_1$ and $\mathbf{H}_2$ is 'opposite' to that of the strong reflexions having the same parity. In practice, the presence of a large percentage of weak reflexions $\mathbf{H}_1$ and $\mathbf{H}_2$ should lead to values of $\langle\Phi_e\rangle$ and of the variance remarkably different from those obtained by the use of the strongest reflexions alone.

The distribution $P(R_{\mathbf{H}_1},R_{\mathbf{H}_2},R_{\mathbf{H}_1+\mathbf{H}_2},R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q},R_{\mathbf{H}_1+\mathbf{K}_1},R_{\mathbf{H}_2-\mathbf{K}_1},\ldots, \varphi_{\mathbf{H}_1},\varphi_{\mathbf{H}_2},\varphi_{\mathbf{H}_1+\mathbf{H}_2},\varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q},\varphi_{\mathbf{H}_1+\mathbf{K}_1},\varphi_{\mathbf{H}_2-\mathbf{K}_1},\ldots)$ when $\mathbf{K}_j(\mathbf{R}_p-\mathbf{R}_q)=0$

The study of this distribution is suggested by the algebraic evidence that several pairs of normalized structure factors with indices $(\mathbf{H}_1+\mathbf{K}_1,\ \mathbf{H}_2-\mathbf{K}_1)$, $(\mathbf{H}_1+\mathbf{K}_2,\ \mathbf{H}_2-\mathbf{K}_2)$, ..., may contribute to defining the expectation value of $\varphi_{\mathbf{H}_1+\mathbf{H}_2}-\varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}$: the fundamental condition for each $\mathbf{K}_j$ vector is $\mathbf{K}_j(\mathbf{R}_p-\mathbf{R}_q)=0$.

We emphasize here only the terms of the probability distribution function which are the most significant for defining $\langle\Phi_e\rangle$ and var $\langle\Phi_e\rangle$. The reader will surely be able to derive all the distribution terms up to order N^{-1} from the more complete analysis presented in paper I. Under these limitations and when n pairs $(\mathbf{H}_1+\mathbf{K}_j,\ \mathbf{H}_2-\mathbf{K}_j)$ are involved in the distribution we obtain

$$\begin{aligned}P(R_{\mathbf{H}_1},\ldots,\varphi_{\mathbf{H}_2-\mathbf{K}_n})&\\
\simeq\frac{1}{\pi^{2n+2}}&R_{\mathbf{H}_1+\mathbf{H}_2}R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}R_{\mathbf{H}_1+\mathbf{K}_1}\ldots R_{\mathbf{H}_2-\mathbf{K}_n}\\
&\times\exp[-R_{\mathbf{H}_1}^2-R_{\mathbf{H}_2}^2-\ldots-R_{\mathbf{H}_2-\mathbf{K}_n}^2]\\
&\times\Big\{1+\frac{2}{\sqrt{N}}\sum_j[R_{\mathbf{H}_1+\mathbf{K}_j}R_{\mathbf{H}_2-\mathbf{K}_j}R_{\mathbf{H}_1+\mathbf{H}_2}\\
&\times\cos(\varphi_{\mathbf{H}_1+\mathbf{K}_j}+\varphi_{\mathbf{H}_2-\mathbf{K}_j}-\varphi_{\mathbf{H}_1+\mathbf{H}_2})]\\
&+\frac{2}{\sqrt{N}}\sum_j[R_{\mathbf{H}_1+\mathbf{K}_j}R_{\mathbf{H}_2-\mathbf{K}_j}R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}\\
&\times\cos(\varphi_{\mathbf{H}_1+\mathbf{K}_j}+\varphi_{\mathbf{H}_2-\mathbf{K}_j}-\varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}-\Delta_j)]+\ldots\\
&+\frac{1}{N}R_{\mathbf{H}_1+\mathbf{H}_2}R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}\sum_j[2(1-R_{\mathbf{H}_1+\mathbf{K}_j}^2)(1-R_{\mathbf{H}_2-\mathbf{K}_j}^2)\\
&+\gamma(R_{\mathbf{H}_1+\mathbf{K}_j}^2+R_{\mathbf{H}_2-\mathbf{K}_j}^2-2)]\cos(\Phi_e-\Delta_j)\Big\}, \quad (22)\end{aligned}$$

where $\Delta_j=2\pi[(\mathbf{H}_1+\mathbf{K}_j)\mathbf{T}_p+(\mathbf{H}_2-\mathbf{K}_j)\mathbf{T}_q]$.

From (22) we obtain

$$P(\Phi_e|R_{\mathbf{H}_1},R_{\mathbf{H}_2},\ldots,R_{\mathbf{H}_1+\mathbf{K}_n},R_{\mathbf{H}_2-\mathbf{K}_n})\simeq\frac{\exp[\sum_j G_j\cos(\Phi_e-\Delta_j)]}{\int_{-\pi}^{\pi}\exp\{\sum_j G_j\cos(\Phi_e-\Delta_j)\}\mathrm{d}\Phi_e}, \quad (23)$$

where

$$G_j=N^{-1}R_{\mathbf{H}_1+\mathbf{H}_2}R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}[2(1-R_{\mathbf{H}_1+\mathbf{K}_j}^2)(1-R_{\mathbf{H}_2-\mathbf{K}_j}^2)+\gamma(R_{\mathbf{H}_1+\mathbf{K}_j}^2+R_{\mathbf{H}_2-\mathbf{K}_j}^2-2)].$$

On substituting

$$\sum_j G_j \cos \Delta_j = A \cos \theta$$
$$\sum_j G_j \sin \Delta_j = A \sin \theta,$$

(23) becomes

$$P(\Phi_e|\ldots) \simeq [2\pi I_0(A)]^{-1} \exp [A \cos (\Phi_{3,4} - \theta)], \quad (24)$$

where

$$A = [(\sum_j G_j \cos \Delta_j)^2 + (\sum_j G_j \sin \Delta_j)^2]^{1/2}, \quad (25)$$

$$\tan \theta = \frac{\sum_j G_j \sin \Delta_j}{\sum_j G_j \cos \Delta_j}. \quad (26)$$

(24) has a maximum when $\Phi_e = \theta$, and the larger the value of A, the higher this maximum will be. The variance of Φ_e for a fixed set of G_j factors is given by

$$\text{var}\,[\Phi_e] = \frac{\pi^2}{3} + [I_0(A)]^{-1} \sum_{1\,n}^{\infty} \frac{I_{2n}(A)}{n^2} - 4[I_0(A)]^{-1} \sum_{0\,n}^{\infty} \frac{I_{2n+1}(A)}{(2n+1)^2}. \quad (27)$$

We note explicitly that (24) is formally different from (12). The value of A in (24), in fact, is assumed always positive whereas G in (12) may be positive or negative. The two equations nevertheless, give equivalent results when $n=1$. In this case, in fact, $\theta = 2\pi(\mathbf{H}_1\mathbf{T}_p + \mathbf{H}_2\mathbf{T}_q)$ if $G>0$; $\theta = \pi + 2\pi(\mathbf{H}_1\mathbf{T}_p + \mathbf{H}_2\mathbf{T}_q)$ if $G<0$. (26) and (27) are basic results of the present paper.

Cosine and sine expected values

From (24) we obtain the expected values of $\cos \Phi_e$ and $\sin \Phi_e$ for a fixed value of A:

$$\langle \cos \Phi_e \rangle = \frac{I_1(A)}{I_0(A)} \cos \theta,$$

$$\langle \sin \Phi_e \rangle = \frac{I_1(A)}{I_0(A)} \sin \theta.$$

The variance values are:

$$\text{var}\,[\cos \Phi_e] = \left(\frac{1+\cos 2\theta}{2}\right)\left[1 - \frac{I_1^2(A)}{I_0^2(A)}\right] - \frac{I_1(A)}{AI_0(A)} \cos 2\theta,$$

$$\text{var}\,[\sin \Phi_e] = \left(\frac{1-\cos 2\theta}{2}\right)\left[1 - \frac{I_1^2(A)}{I_0^2(A)}\right] + \frac{I_1(A)}{AI_0(A)} \cos 2\theta.$$

If A is large enough the expected values of $\cos \Phi_e$ and $\sin \Phi_e$ are very close to $\cos \theta$ and $\sin \theta$ respectively. As in these conditions small variance values occur, θ should be a reliable estimate of Φ_e.

A comparison with the cosine seminvariant method

Formulae for linear combinations of two phases which are structure seminvariants have been derived in some space groups by Hauptman (1972*b*) by means of an algebraic approach. The derivation was presented in the space group $P2$ and useful formulae were fixed in $P2_1$ and $P2_12_12_1$. In this paragraph we show that our theory is able to generalize Hauptman's results and gives them a new probabilistic interpretation.

In order to facilitate comparison with Hauptman's formulation, let us assume, without any loss of generality, that $E_{\mathbf{H}_1+\mathbf{H}_2}$ and $E_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}$ are centrosymmetric reflexions. From (4) we derive the marginal conditional density

$$P(R_{\mathbf{H}_1+\mathbf{K}_1},\ldots,R_{\mathbf{H}_2-\mathbf{K}_n}|E_{\mathbf{H}_1+\mathbf{H}_2},E_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}) \simeq 2^{2n} R_{\mathbf{H}_1+\mathbf{K}_1}\ldots R_{\mathbf{H}_2-\mathbf{K}_n} \exp(-R^2_{\mathbf{H}_1+\mathbf{K}_1} - \ldots - R^2_{\mathbf{H}_2-\mathbf{K}_n}) \times \{1 + \sum_j G_j(-1)^{2[(\mathbf{H}_1+\mathbf{K}_j)\mathbf{T}_p + (\mathbf{H}_2-\mathbf{K}_j)\mathbf{T}_q]}\}$$

from which the mean value of the quantity

$$(1 - R^2_{\mathbf{H}_1+\mathbf{K}_j})(1 - R^2_{\mathbf{H}_2-\mathbf{K}_j})(-1)^{2[(\mathbf{H}_1+\mathbf{K}_j)\mathbf{T}_p + (\mathbf{H}_2-\mathbf{K}_j)\mathbf{T}_q]}$$

is derived:

$$E_{\mathbf{H}_1+\mathbf{H}_2} E_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q} \simeq \frac{N}{2} \langle (1 - R^2_{\mathbf{H}_1+\mathbf{K}_j})(1 - R^2_{\mathbf{H}_2-\mathbf{K}_j}) \times (-1)^{2[(\mathbf{H}_1+\mathbf{K}_j)\mathbf{T}_p + (\mathbf{H}_2-\mathbf{K}_j)\mathbf{T}_q]} \rangle. \quad (28)$$

In order to describe a practical application of (28) let us consider in space group $P2_12_12_1$ the case in which $\mathbf{R}_p = \mathbf{I}$,

$$\mathbf{R}_q = \begin{vmatrix} \bar{1} & 0 & 0 \\ 0 & \bar{1} & 0 \\ 0 & 0 & 1 \end{vmatrix}.$$

Because of trivial algebraic considerations, the parities of the reciprocal vectors involved in (28) are so fixed:

(*a*) $\mathbf{H}_1 + \mathbf{H}_2 \equiv (h_1, k_1, 0)$, $\mathbf{H}_1 + \mathbf{H}_2\mathbf{R}_q = (h_2, k_2, 0)$,

where $h_1 \pm h_2$ and $k_1 \pm k_2$ are even. The condition $l=0$ is fixed by the chosen centrosymmetric nature of the s.r.p.p. ($E_{\mathbf{H}_1+\mathbf{H}_2}$, $E_{\mathbf{H}_1+\mathbf{H}_2\mathbf{R}_q}$);

(*b*) $$\mathbf{H}_1 + \mathbf{K}_j = \left(\frac{h_1+h_2}{2}, \frac{k_1+k_2}{2}, l\right),$$

$$\mathbf{H}_2 - \mathbf{K}_j = \left(\frac{h_1-h_2}{2}, \frac{k_1-k_2}{2}, l\right).$$

Then (28) reduces to

$$E_{h_1k_10} E_{h_2k_20} \simeq \frac{N}{2} \langle (-1)^{l+(h_1+h_2)/2} (|E_{(h_1+h_2)/2\ (k_1+k_2)/2\ l}|^2 - 1) \times (|E_{(h_1-h_2)/2\ (k_1-k_2)/2\ l}|^2 - 1) \rangle_l,$$

which coincides with (4.5) in Hauptman's (1972*b*) paper. One can show that all Hauptman's formulae may be derived in a similar way.

Further remarks about Hauptman's and our methods could be useful. (1) Hauptman's method, as well as our approach, is able to take full advantage of the space-group symmetry, but needs to know the algebraic form of the structure factor. The derivation of the phase information in any space group thus requires an *ad hoc* mathematical treatment. Our approach however involves the symmetry operators alone so that the expected values of the seminvariant phases are easily derivable whatever the space group. (2) Hauptman's method obtains the value of $\langle\cos\Phi_e\rangle$ by averaging the quantity (in our notation)

$$\sum_j (R^2_{\mathbf{H}_1+\mathbf{K}_j}-1)(R^2_{\mathbf{H}_2-\mathbf{K}_j}-1)\cos\Delta_j. \qquad (29)$$

Our probabilistic approach advises us to average the quantity

$$\sum_j G_j\cos\Delta_j$$

of which (29) is a part. In fact, the mean value of the term

$$\sum\gamma(R^2_{\mathbf{H}_1+\mathbf{K}}+R^2_{\mathbf{H}_2-\mathbf{K}}-2)\cos\Delta_j$$

does not vanish and its estimate may give a further contribution to knowledge of $\cos\Phi_e$. (3) Hauptman's formulae are formally able to give the exact value of the seminvariant cosines when the structure consists of N identical point atoms and when no rational dependence of atomic coordinates occurs. A fundamental postulate is that the vector $\mathbf{K}$ ranges uniformly throughout reciprocal space on condition that $\mathbf{K}(\mathbf{R}_p-\mathbf{R}_q)=0$. However, all the computed averages are of necessity only estimates of the true averages and are based on the finite number of available diffraction data. Our formulation, on the other hand, is able to take account of the sampling effect and to provide expected values of the seminvariant cosines as well as the variance values.

$$+\frac{2}{\sqrt{N}}R_{\mathbf{H}_1}R_{\mathbf{H}_2}R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}$$
$$\times\cos(\varphi_{\mathbf{H}_1}+\varphi_{\mathbf{H}_2}-\varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}-\Delta)$$
$$+\frac{2}{N}[(1-\gamma)+\frac{\gamma-2}{2}(R^2_{\mathbf{H}_1}+R^2_{\mathbf{H}_2})]R_{\mathbf{H}_1+\mathbf{H}_2}R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}$$
$$\times\cos(\varphi_{\mathbf{H}_1+\mathbf{H}_2}-\varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}-\Delta)\}, \qquad (30)$$

where $\Delta=\cos 2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)$.

$P(\Phi_e|R_{\mathbf{H}_1},\ldots,R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q})$ is found by (30) by fixing $R_{\mathbf{H}_1}$, ..., $R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}$, integrating with respect to $\varphi_{\mathbf{H}_1}$, $\varphi_{\mathbf{H}_2}$ from 0 to 2π and multiplying the result by a suitable normalizing constant. Thus,

$$P(\Phi_e|R_{\mathbf{H}_1},\ldots,R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q})\simeq\frac{1}{2\pi L}I_0(A)\exp[2B\cos(\Phi_e-\Delta)], \qquad (31)$$

where

$$A=\frac{2}{\sqrt{N}}R_{\mathbf{H}_1}R_{\mathbf{H}_2}[R^2_{\mathbf{H}_1+\mathbf{H}_2}+R^2_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}+2R_{\mathbf{H}_1+\mathbf{H}_2}R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}\cos(\Phi_e-\Delta)]^{1/2},$$

$$B=\frac{1}{N}\left[(1-\gamma)+\left(\frac{\gamma-2}{2}\right)(R^2_{\mathbf{H}_1}+R^2_{\mathbf{H}_2})\right]R_{\mathbf{H}_1+\mathbf{H}_2}R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q},$$

$$L=\sum_{-\infty\, m}^{-\infty}I_m\left(\frac{2R_{\mathbf{H}_1}R_{\mathbf{H}_2}R_{\mathbf{H}_1+\mathbf{H}_2}}{\sqrt{N}}\right)\times I_m\left(\frac{2R_{\mathbf{H}_1}R_{\mathbf{H}_2}R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}}{\sqrt{N}}\right)I_m(2B);$$

I_m is the modified Bessel function of order m.

In the same way, the conditional expected value of $\cos\Phi_e$ is found from (30):

$$\langle\cos\Phi_e|R_{\mathbf{H}_1},\ldots,R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}\rangle\simeq\cos\Delta\frac{\sum\limits_{-\infty\, m}^{+\infty}I_m\left(\frac{2R_{\mathbf{H}_1}R_{\mathbf{H}_2}R_{\mathbf{H}_1+\mathbf{H}_2}}{\sqrt{N}}\right)I_m\left(\frac{2R_{\mathbf{H}_1}R_{\mathbf{H}_2}R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}}{\sqrt{N}}\right)I_{m+1}(2B)}{\sum\limits_{-\infty\, m}^{+\infty}I_m\left(\frac{2R_{\mathbf{H}_1}R_{\mathbf{H}_2}R_{\mathbf{H}_1+\mathbf{H}_2}}{\sqrt{N}}\right)I_m\left(\frac{2R_{\mathbf{H}_1}R_{\mathbf{H}_2}R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}}{\sqrt{N}}\right)I_m(2B)}. \qquad (32)$$

The probabilistic theory of the seminvariant cosines when the exponential form of the characteristic function is used

Appendix C gives

$$P(R_{\mathbf{H}_1},R_{\mathbf{H}_2},R_{\mathbf{H}_1+\mathbf{H}_2},R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q},\varphi_{\mathbf{H}_1},\ldots,\varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q})$$
$$\simeq\frac{1}{\pi^4}R_{\mathbf{H}_1}\ldots R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}$$
$$\times\exp\{-(R^2_{\mathbf{H}_1}+\ldots+R^2_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q})$$
$$+\frac{2}{\sqrt{N}}R_{\mathbf{H}_1}R_{\mathbf{H}_2}R_{\mathbf{H}_1+\mathbf{H}_2}\cos(\varphi_{\mathbf{H}_1}+\varphi_{\mathbf{H}_2}-\varphi_{\mathbf{H}_1+\mathbf{H}_2})$$

We now calculate the probability density function when several pairs of normalized structure factors with indices $(\mathbf{H}_1+\mathbf{K}_1,\ \mathbf{H}_2-\mathbf{K}_1)$, $(\mathbf{H}_1+\mathbf{K}_2,\ \mathbf{H}_2-\mathbf{K}_2)$, ... contribute to defining the expectation value of Φ_e. Of course, $\mathbf{K}_j(\mathbf{R}_p-\mathbf{R}_q)$ must be zero for each $\mathbf{K}_j$ vector. If n pairs of type $(\mathbf{H}_1+\mathbf{K}_j,\ \mathbf{H}_2-\mathbf{K}_j)$ contribute to Φ_e, we obtain after lengthy calculations,

$$P(\Phi_e|R_{\mathbf{H}_1},R_{\mathbf{H}_2},R_{\mathbf{H}_1+\mathbf{K}_1},R_{\mathbf{H}_1+\mathbf{K}_2},\ldots,R_{\mathbf{U}},R_{\mathbf{V}})\simeq\frac{1}{2\pi L}\left\{\prod_{1\,j}^{n}I_0(A_j)\exp[2B\cos(\Phi_e-\beta)]\right\}, \qquad (33)$$

where

$$\mathbf{U}=\mathbf{H}_1+\mathbf{H}_2,\quad \mathbf{V}=\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q,$$

$$A_j=\frac{2}{\sqrt{N}}R_{\mathbf{H}_1+\mathbf{K}_j}R_{\mathbf{H}_2-\mathbf{K}_j}[R_{\mathbf{U}}^2+R_{\mathbf{V}}^2+2R_{\mathbf{U}}R_{\mathbf{V}}\cos(\Phi_e-\Delta_j)]^{1/2},$$

$$B=\left[\left(\sum_{1j}^{n}B_j\cos\Delta_j\right)^2+\left(\sum_{1j}^{n}B_j\sin\Delta_j\right)^2\right]^{1/2},$$

$$B_j=\frac{1}{N}\left[(1-\gamma)+\left(\frac{\gamma-2}{2}\right)(R_{\mathbf{H}_1+\mathbf{K}_j}^2+R_{\mathbf{H}_2-\mathbf{K}_j}^2)\right]R_{\mathbf{U}}R_{\mathbf{V}},$$

$$\tan\beta=\frac{\sum_{1j}^{n}B_j\sin\Delta_j}{\sum_{1j}^{n}B_j\cos\Delta_j},$$

$$\Delta_j=2\pi[(\mathbf{H}_1+\mathbf{K}_j)\mathbf{T}_p+(\mathbf{H}_2-\mathbf{K}_j)\mathbf{T}_q],$$

$$L=\sum_{-\infty\, m,\nu,\ldots,\varrho}^{+\infty}I_m\left(\frac{2R_{\mathbf{H}_1+\mathbf{K}_1}R_{\mathbf{H}_2-\mathbf{K}_1}R_{\mathbf{U}}}{\sqrt{N}}\right)\times I_m\left(\frac{2R_{\mathbf{H}_1+\mathbf{K}_1}R_{\mathbf{H}_2-\mathbf{K}_1}R_{\mathbf{V}}}{\sqrt{N}}\right)\ldots\times I_\varrho\left(\frac{2R_{\mathbf{H}_1+\mathbf{K}_n}R_{\mathbf{H}_2-\mathbf{K}_n}R_{\mathbf{U}}}{\sqrt{N}}\right)I_\varrho\left(\frac{2R_{\mathbf{H}_1+\mathbf{K}_n}R_{\mathbf{H}_2-\mathbf{K}_n}R_{\mathbf{V}}}{\sqrt{N}}\right)\times I_{m+\nu+\ldots+\varrho}(2B)\cos[m\Delta_1+\nu\Delta_2+\ldots+\varrho\Delta_n-(m+\nu+\ldots+\varrho)\beta].$$

From (33) the conditional expected value of $\cos\Phi_e$ is

$$\langle\cos\Phi_e|\ldots\rangle\simeq\frac{1}{L}\left\{\sum_{-\infty\, m,\nu,\ldots,\varrho}^{+\infty}I_{m,\nu,\ldots,\varrho}\right\}\cos\beta,\quad(34)$$

where

$$I_{m,\nu,\ldots,\varrho}=I_m\left(\frac{2R_{\mathbf{H}_1+\mathbf{K}_1}R_{\mathbf{H}_2-\mathbf{K}_1}R_{\mathbf{U}}}{\sqrt{N}}\right)\times I_m\left(\frac{2R_{\mathbf{H}_1+\mathbf{K}_1}R_{\mathbf{H}_2-\mathbf{K}_1}R_{\mathbf{V}}}{\sqrt{N}}\right)\ldots\times I_\varrho\left(\frac{2R_{\mathbf{H}_1+\mathbf{K}_n}R_{\mathbf{H}_2-\mathbf{K}_n}R_{\mathbf{U}}}{\sqrt{N}}\right)I_\varrho\left(\frac{2R_{\mathbf{H}_1+\mathbf{K}_n}R_{\mathbf{H}_2-\mathbf{K}_n}R_{\mathbf{V}}}{\sqrt{N}}\right)\times I_{m+\nu+\ldots+\varrho+1}(2B)\cos[m\Delta_1+\ldots+\varrho\Delta_n-(m+\nu+\ldots+\varrho)\beta].$$

(33) and (34) are the major results of this paper.

As further details would make this paper dull reading, the practical use of (33) and (34) and their connexions with (24) and (26) will be described in a later paper.

Concluding remarks

A theory has been described which is capable of deriving in each space group the expected value of the seminvariant cosine $\cos(\varphi_{\mathbf{H}_1+\mathbf{H}_2}-\varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q})$ given one or more pairs of magnitudes $(R_{\mathbf{H}_1+\mathbf{K}_j}, R_{\mathbf{H}_2-\mathbf{K}_j})$, on condition that $\mathbf{K}_j(\mathbf{R}_p-\mathbf{R}_q)=0$. The final formulae are derived by means of two different techniques: the first uses a Gram–Charlier expansion of the characteristic function, the second uses directly its exponential function. The mathematical approach seems quite general: its application in the automatic procedures for phase determination is made easier by the fact that the method requires knowledge of only the symmetry operators.

Coincidence information may be used in several ways to improve and speed up the multisolution procedures of solving crystal structures. Debaerdemaeker & Woolfson (1972), Hauptman (1972*c*) and Viterbo (1974) suggested their usefulness in enlarging the starting set or in reducing the number of ambiguities which must be introduced at the beginning of the phasing procedure. However, no one seems to have used coincidence information to select the correct phase set from all the sets produced by a multisolution procedure. This use is supported by the evidence that the theory described here is able to define in the symmorphic space groups the s.r.p.p.'s for which

$$\varphi_{\mathbf{H}_1+\mathbf{H}_2}-\varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}\simeq\pi.$$

So, new figures of merit are suggested by this theory which, together with that described by De Titta, Edmonds, Lang & Hauptman (1975) and Giacovazzo (1976), can help to select the correct phase set in the multisolution procedures.

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APPENDIX *A*

Let the symmetry number of the actual space group be denoted by m, and ζ and η are the real and imaginary parts of the trigonometric structure factor ξ: then

$$\xi(\mathbf{h})=\zeta(\mathbf{h})+i\eta(\mathbf{h})=\sum_{1p}^{m}\exp 2\pi i\mathbf{h}(\mathbf{R}_p\mathbf{x}+\mathbf{T}_p).$$

In particular

$$\zeta(\mathbf{h})=\sum_{1p}^{m}\cos 2\pi\mathbf{h}(\mathbf{R}_p\mathbf{x}+\mathbf{T}_p),$$

$$\eta(\mathbf{h})=\sum_{1p}^{m}\sin 2\pi\mathbf{h}(\mathbf{R}_p\mathbf{x}+\mathbf{T}_p).$$

With a view to deriving the multivariate standardized cumulants, the linearization theory (Bertaut, 1959) will be used. For the sake of simplicity, we consider here only the reciprocal vectors $\mathbf{H}_1$, $\mathbf{H}_2$, $\mathbf{H}_1+\mathbf{H}_2$, $\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q$ whose statistical weights equal unity. Special vector covariances, in fact, would require further, additional, algebraic considerations [see Giacovazzo (1974*b*,*c*) for cumulants of order three] which would make this paper too dull reading.

As is well known the only non-vanishing moments in S_3' which involve the reciprocal vectors $\mathbf{H}_1, \mathbf{H}_2, \mathbf{H}_3=\mathbf{H}_1+\mathbf{H}_2$, are

$$\langle\zeta(\mathbf{H}_1)\zeta(\mathbf{H}_2)\zeta(\mathbf{H}_3)\rangle,\quad \langle\zeta(\mathbf{H}_1)\eta(\mathbf{H}_2)\eta(\mathbf{H}_3)\rangle,$$
$$\langle\eta(\mathbf{H}_1)\eta(\mathbf{H}_2)\zeta(\mathbf{H}_3)\rangle,\quad \langle\eta(\mathbf{H}_1)\zeta(\mathbf{H}_2)\eta(\mathbf{H}_3)\rangle. \qquad (A.1)$$

Their estimation leads to the probability function

$$P(\Phi_{\mathbf{H}_1+\mathbf{H}_2})=[2\pi I_0(G)]^{-1} \times\exp\left[G\cos\left(\varphi_{\mathbf{H}_1}+\varphi_{\mathbf{H}_2}-\varphi_{\mathbf{H}_1+\mathbf{H}_2}\right)\right]. \qquad (A.2)$$

When the $\mathbf{H}_1$, $\mathbf{H}_2$, $\mathbf{H}_4=\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q$ vectors are considered, a larger number of moments must be tested. For example, if $\mathbf{R}_p=\mathbf{I}$, $\mathbf{R}_q=\mathbf{R}_s$, in addition to those in $(A.1)$, the moments

$$\langle\zeta(\mathbf{H}_1)\zeta(\mathbf{H}_2)\eta(\mathbf{H}_4)\rangle,\quad \langle\eta(\mathbf{H}_1)\eta(\mathbf{H}_2)\eta(\mathbf{H}_4)\rangle,$$
$$\langle\eta(\mathbf{H}_1)\zeta(\mathbf{H}_2)\zeta(\mathbf{H}_4)\rangle,\quad \langle\zeta(\mathbf{H}_1)\eta(\mathbf{H}_2)\zeta(\mathbf{H}_4)\rangle, \qquad (A.3)$$

should not vanish. The mathematical device of linearization theory offers a useful tool for estimating the moments. For example, the quantity

$$\begin{aligned}&\langle\zeta(\mathbf{H}_1)\zeta(\mathbf{H}_2)\zeta(\mathbf{H}_4)\rangle\\ &=\tfrac{1}{2}\Big\langle\sum_{1\ p,q,n}^{m}\{\cos 2\pi[(\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q)\mathbf{x}+\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q]\\ &\quad+\cos 2\pi[(\mathbf{H}_1\mathbf{R}_p-\mathbf{H}_2\mathbf{R}_q)\mathbf{x}+\mathbf{H}_1\mathbf{T}_p-\mathbf{H}_2\mathbf{T}_q]\}\\ &\quad\times\cos 2\pi[(\mathbf{H}_1\mathbf{R}_n+\mathbf{H}_2\mathbf{R}_s\mathbf{R}_n)\mathbf{x}+\mathbf{H}_1\mathbf{T}_n+\mathbf{H}_2\mathbf{R}_s\mathbf{T}_n]\Big\rangle,\end{aligned}$$

does not vanish when $\mathbf{R}_p=\mathbf{R}_n$, $\mathbf{R}_q=\mathbf{R}_s\mathbf{R}_n$. Then

$$\langle\zeta(\mathbf{H}_1)\zeta(\mathbf{H}_2)\zeta(\mathbf{H}_1+\mathbf{H}_2\mathbf{R}_s)\rangle=\frac{m}{4}\cos 2\pi\mathbf{H}_2\mathbf{T}_s.$$

In general one obtains the probability function

$$P(\varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q})=[2\pi I_0(G)]^{-1} \times\exp\left[G\cos\left(\varphi_{\mathbf{H}_1}+\varphi_{\mathbf{H}_2}-\varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}-\Delta\right)\right], \qquad (A.4)$$

where $\Delta=2\pi(\mathbf{H}_1\mathbf{T}_p+\mathbf{H}_2\mathbf{T}_q)$. $(A.4)$ generalizes $(A.2)$.

APPENDIX *B*

The expression of S_4'/t^2 in (2) derives from an approximate estimate of

$$\begin{aligned}S_4'/t^2=\frac{1}{t}\Big\{&\frac{\lambda_{40000000}}{4!}\varrho_1^4\cos^4\psi_1+\ldots\\ &+\frac{\lambda_{00000004}}{4!}\varrho_4^4\sin^4\psi_4\\ &+\frac{\lambda_{20002000}}{2!2!}\varrho_1^4\cos^2\psi_1\sin^2\psi_1+\ldots\\ &+\frac{\lambda_{00020002}}{2!2!}\varrho_4^4\cos^2\psi_4\sin^2\psi_4\\ &+\frac{\lambda_{20110000}}{2!1!1!}\varrho_1^2\varrho_3\varrho_4\cos^2\psi_1\cos\psi_3\cos\psi_4\\ &+\frac{\lambda_{02110000}}{2!1!1!}\varrho_2^2\varrho_3\varrho_4\cos^2\psi_2\cos\psi_3\cos\psi_4\\ &+\frac{\lambda_{00112000}}{1!1!2!}\varrho_1^2\varrho_3\varrho_4\sin^2\psi_1\cos\psi_3\cos\psi_4\\ &+\frac{\lambda_{00000211}}{2!1!1!}\varrho_2^2\varrho_3\varrho_4\sin^2\psi_2\sin\psi_3\sin\psi_4\\ &+\frac{\lambda_{00002011}}{2!1!1!}\varrho_1^2\varrho_3\varrho_4\sin^2\psi_1\sin\psi_3\sin\psi_4\\ &+\frac{\lambda_{00110200}}{1!1!2!}\varrho_2^2\varrho_3\varrho_4\sin^2\psi_2\cos\psi_3\cos\psi_4\\ &+\frac{\lambda_{20000011}}{2!1!1!}\varrho_1^2\varrho_3\varrho_4\cos^2\psi_1\sin\psi_3\sin\psi_4\\ &+\frac{\lambda_{02000011}}{2!1!1!}\varrho_2^2\varrho_3\varrho_4\cos^2\psi_2\sin\psi_3\sin\psi_4\Big\}.\end{aligned} \qquad (B.1)$$

The approximation is because in accordance with the algebraic results described in the Appendices of part I of this paper, some cumulants in $(B.1)$ assume values which vary with the space-group symmetry. Fortunately the modifications introduced by the symmetry for the monovariate and bivariate standardized cumulants in $(B.1)$ turn into weak perturbations of $\cos(\varphi_{\mathbf{H}_1+\mathbf{H}_2}-\varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q})$. Therefore, we will not estimate them here. We limit ourselves to showing how the values of the most important cumulants in S_4', the trivariate cumulants, vary with the space-group symmetry.

Without loss of generality, let us consider, when $\mathbf{R}_p=\mathbf{I}$, $\mathbf{R}_q=\mathbf{R}_s$, the first of the trivariate cumulants which appear in $(B.1)$. As

$$\lambda_{20110000}=m_{20110000}/(m^2/4),$$

we derive first the expression

$$\begin{aligned}m_{20110000}&=\langle\zeta^2(\mathbf{H}_1)\zeta(\mathbf{H}_1+\mathbf{H}_2)\zeta(\mathbf{H}_1+\mathbf{H}_2\mathbf{R}_s)\rangle\\ &=\tfrac{1}{4}\Big\langle\Big\{\sum_{1\ p,q}^{m}\cos 2\pi\mathbf{H}_1[(\mathbf{R}_p+\mathbf{R}_q)\mathbf{x}+\mathbf{T}_p+\mathbf{T}_q]\\ &\quad+\cos 2\pi[\mathbf{H}_1(\mathbf{R}_p-\mathbf{R}_q)\mathbf{x}+\mathbf{T}_p-\mathbf{T}_q]\Big\}\\ &\quad\times\Big\{\sum_{1\ n,v}^{m}\cos 2\pi[\mathbf{H}_1(\mathbf{R}_n\mathbf{x}+\mathbf{R}_v\mathbf{x}+\mathbf{T}_n+\mathbf{T}_v)\\ &\quad+\mathbf{H}_2(\mathbf{R}_n\mathbf{x}+\mathbf{R}_s\mathbf{R}_v\mathbf{x}+\mathbf{T}_n+\mathbf{R}_s\mathbf{T}_v)]\\ &\quad+\cos 2\pi[\mathbf{H}_1(\mathbf{R}_n\mathbf{x}-\mathbf{R}_v\mathbf{x}+\mathbf{T}_n-\mathbf{T}_v)\\ &\quad+\mathbf{H}_2(\mathbf{R}_n\mathbf{x}-\mathbf{R}_s\mathbf{R}_v\mathbf{x}+\mathbf{T}_n-\mathbf{R}_s\mathbf{T}_v)]\Big\}\Big\rangle\\ &=(L_1+L_2)(L_3+L_4).\end{aligned}$$

Whereas the products L_1L_3, L_2L_3, L_1L_4 always vanish, L_2L_4 is non-zero when

$$\mathbf{H}_1[(\mathbf{R}_p-\mathbf{R}_q)\mathbf{x}+\mathbf{T}_p-\mathbf{T}_q]=\mathbf{H}_1[(\mathbf{R}_n-\mathbf{R}_v)\mathbf{x}+\mathbf{T}_n-\mathbf{T}_v],$$
$$(\mathbf{R}_n-\mathbf{R}_s\mathbf{R}_v)\mathbf{x}+\mathbf{T}_n-\mathbf{R}_s\mathbf{T}_v\equiv 0\ (\mathrm{mod.}\ 1). \qquad (B.2)$$

The conditions

(*a*) $\mathbf{R}_s\mathbf{R}_v=\mathbf{R}_n$, $\mathbf{R}_p=\mathbf{R}_n$, $\mathbf{R}_q=\mathbf{R}_v$,

(*b*) $\mathbf{R}_s\mathbf{R}_v=\mathbf{R}_n$, $\mathbf{R}_p=\mathbf{R}_v$, $\mathbf{R}_q=\mathbf{R}_n$,

satisfy $(B.2)$. As $\mathbf{R}_s\mathbf{T}_v=\mathbf{T}_n-\mathbf{T}_s$ when $\mathbf{R}_s\mathbf{R}_v=\mathbf{R}_n$, then

$$m_{20110000} = \frac{m}{4}\cos 2\pi\mathbf{H}_2\mathbf{T}_s,$$

$$\lambda_{20110000} = \frac{1}{m}\cos 2\pi\mathbf{H}_2\mathbf{T}_s. \qquad (B.3)$$

While in the symmetry classes 1, 2, m, 222 the trivariate cumulants equal $(1/m)\cos 2\pi\mathbf{H}_2\mathbf{T}_s$, in classes with higher symmetry that is not always true. In $mm2$, for example, in which the rotation components of the symmetry operations are

$$\mathbf{R}_1 = \begin{vmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{vmatrix}; \quad \mathbf{R}_2 = \begin{vmatrix} \bar{1} & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{vmatrix};$$

$$\mathbf{R}_3 = \begin{vmatrix} 1 & 0 & 0 \\ 0 & \bar{1} & 0 \\ 0 & 0 & 1 \end{vmatrix}; \quad \mathbf{R}_4 = \begin{vmatrix} \bar{1} & 0 & 0 \\ 0 & \bar{1} & 0 \\ 0 & 0 & 1 \end{vmatrix},$$

the standardized trivariate cumulants equal $\cos 2\pi\mathbf{H}_2\mathbf{T}_s/m$ when $s=4$; $2\cos 2\pi\mathbf{H}_2\mathbf{T}_s/m$ when $s=2,3$. The additional contribution in the cases $s=2,3$ derives from the combinations

(1) $\mathbf{R}_p=\mathbf{R}_1$, $\mathbf{R}_q=\mathbf{R}_2$, $\mathbf{R}_n=\mathbf{R}_3$, $\mathbf{R}_\nu=\mathbf{R}_4$,

(2) $\mathbf{R}_p=\mathbf{R}_1$, $\mathbf{R}_q=\mathbf{R}_2$, $\mathbf{R}_n=\mathbf{R}_4$, $\mathbf{R}_\nu=\mathbf{R}_3$,

(3) $\mathbf{R}_p=\mathbf{R}_2$, $\mathbf{R}_q=\mathbf{R}_1$, $\mathbf{R}_n=\mathbf{R}_3$, $\mathbf{R}_\nu=\mathbf{R}_4$,

(4) $\mathbf{R}_p=\mathbf{R}_2$, $\mathbf{R}_q=\mathbf{R}_1$, $\mathbf{R}_n=\mathbf{R}_4$, $\mathbf{R}_\nu=\mathbf{R}_3$.

APPENDIX C

Let

$$R_1=R_{\mathbf{H}_1},\ R_2=R_{\mathbf{H}_2},\ R_3=R_{\mathbf{H}_3},\ R_4=R_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q},$$

$$\varphi_1=\varphi_{\mathbf{H}_1},\ \ldots\ldots\ldots\ldots,\ \varphi_4=\varphi_{\mathbf{H}_1\mathbf{R}_p+\mathbf{H}_2\mathbf{R}_q}.$$

If the exponential form of the characteristic function is used,

$$P(R_1,R_2,R_3,R_4,\varphi_1,\ldots,\varphi_4) \simeq \frac{R_1\ldots R_4}{(2\pi)^8}$$
$$\times \int_0^\infty\ldots\int_0^\infty\int_0^{2\pi}\ldots\int_0^{2\pi} \varrho_1\varrho_2\varrho_3\varrho_4 \mathrm{d}\varrho_1\ldots\mathrm{d}\varrho_4$$
$$\times \exp\{-i[\varrho_1R_1\cos(\psi_1-\varphi_1)+\ldots$$
$$+\varrho_4R_4\cos(\psi_4-\varphi_4)]-\tfrac{1}{4}(\varrho_1^2+\ldots+\varrho_4^2)$$
$$-\frac{i}{4\sqrt{N}}[\varrho_1\varrho_2\varrho_3\cos(\psi_1+\psi_2-\psi_3)$$
$$+\varrho_1\varrho_2\varrho_4\cos(\psi_1+\psi_2-\psi_4-\Delta)]$$
$$+\frac{1}{16N}\gamma\varrho_3\varrho_4(\varrho_1^2+\varrho_2^2)$$
$$\times\cos(\psi_3-\psi_4-\Delta)\}\mathrm{d}\psi_1\ldots\mathrm{d}\psi_4. \qquad (C.1)$$

In view of (3d) none of the ψ integrations presents any difficulty. In fact, in order to carry out the ψ_j integration, one collects the terms involving ψ_j in the exponent of (C.1) and obtains an expression of the type

$$Y_j\cos(\xi_j-\psi_j),$$

where Y_j and ξ_j are independent of ψ_j. The integration is then easily done by means of (3a) and (3b).

Some remarks should be made concerning the ϱ_1 and ϱ_2 integrations. If the ψ_1 and ψ_2 integrations have already been carried out, one needs to calculate expressions of type

$$\int_0^\infty \varrho_j \exp\left[-\tfrac{1}{4}\varrho_j^2-\frac{\gamma}{4N}\varrho_j^2R_3R_4\cos(\varphi_3-\varphi_4-\Delta)\right.$$
$$\left.\times 2\pi J_0(\varrho_jA_j)\right]\mathrm{d}\varrho_j, \qquad (C.2)$$

where A_j is a factor independent of ϱ_j. The direct integration of (C.2) is feasible but leads to a complicated expression of the probability density function. We prefer to introduce into (C.2) the Taylor expansion

$$\exp\left[-\frac{\gamma}{4N}\varrho_j^2R_3R_4\cos(\varphi_3-\varphi_4-\Delta)\right]$$
$$\simeq 1-\frac{\gamma}{4N}\varrho_j^2R_3R_4\cos(\varphi_3-\varphi_4-\Delta). \qquad (C.3)$$

In view of (C.3), (C.2) then becomes

$$4\pi\exp(-A_j^2)\left[1-\frac{\gamma}{4N}R_3R_4(1-R_j^2)\cos(\varphi_3-\varphi_4-\Delta)\right],$$

which, by means of (C.3), may be written

$$4\pi\exp\left[-A_j^2-\frac{\gamma}{4N}R_3R_4(1-R_j^2)\cos(\varphi_3-\varphi_4-\Delta)\right].$$

This procedure leads to the probability density function (30).

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