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Estimation of Quartet Phase Sums from a New Joint Probability Distribution of Normalized Structure Factors

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Abstract

A new joint probability distribution of normalized structure factors is derived for equal-atom structures in space group $P1$. From this general distribution, a series expansion, the conditional joint probability distribution of the quartet phase sum is obtained, when restrictive conditions among the reciprocal vectors are imposed. The main difference from existing quartet distributions is the possibility of enclosing higher-order terms to any given order of N, although an approximation employed in the derivation limits the number of them considerably. The higher-order terms present are easily employed in the series since the determination of their explicit appearance has been automated: a computer program derives the terms up to a desired order and stores them in a coded form. In general, the incorporation of selective terms up to order N^{-3} appears to yield sufficient convergence. Only high $|E|$ values or a low N value may necessitate the use of higher-order terms. Test results show that, in contrast to results from the quartet distributions of Hauptman and Giacovazzo, systematic estimation errors are hardly present, while absolute estimation errors are reduced as well.

1. Introduction

Results of Simerska (1956) and Hauptman & Karle (1953) indicated that the four-phase structure invariant ψ_4,

$$\psi_4 = \varphi_{H_1} + \varphi_{H_2} + \varphi_{H_3} - \varphi_{H_1+H_2+H_3}, \qquad (1)$$

also called the quartet phase sum or simply quartet, lies more probably near zero for larger values of

$$E_4 = |E_{H_1} E_{H_2} E_{H_3} E_{H_1+H_2+H_3}| N^{-1}. \qquad (2)$$

However, in general the triplet relationship

$$\psi_3 = \varphi_{H_1} + \varphi_{H_2} - \varphi_{H_1+H_2} \qquad (3)$$

will be estimated more reliably because the E_3 values, which determine the reliability of the triplet estimation, are in general larger than the E_4 values since they depend on $N^{-1/2}$ only. Therefore, quartets were not used as such for practical purposes. This changed when Schenk (1973*a*) pointed out that quartets can also be formed by summing two triplets with one phase in common and he showed in this way that quartet (1) depends not only on $|E_{H_1}|$, $|E_{H_2}|$, $|E_{H_3}|$ and $|E_{H_1+H_2+H_3}|$ but also on the so-called cross terms $|E_{H_1+H_2}|$, $|E_{H_1+H_3}|$ and $|E_{H_2+H_3}|$. He argued that the larger the E_4 and cross-term magnitudes the more probably ψ_4 lies near zero. Another important result of the introduction of this cross-term principle was that quartets with small cross-term magnitudes could be predicted to lie near π (Schenk & De Jong, 1973; Schenk, 1973*a*, *b*; Hauptman, 1974; Schenk, 1974). This renewed interest in quartets and the cross-term principle led to the development of improved joint probability distributions (j.p.d.'s) for estimating the quartet phase sum (Hauptman, 1975*a*, *b*, 1976; Giacovazzo, 1976*a*, *b*) and initiated the development of the neighbourhood principle (Hauptman, 1975*b*) and the representation theory (Giacovazzo, 1977). The latter theories identify structure factors upon which the phase sum of a structure (sem)invariant most sensitively depends.

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In practice the application of quartets in direct methods has proved to be successful, in particular in starting-set procedures and figures of merit (Schenk, 1973*a*; Schenk & De Jong, 1973; Schenk, 1974; De Titta, Edmonds, Langs & Hauptman, 1975; van der Putten & Schenk, 1979; Gilmore, 1977; Freer & Gilmore, 1980). In non-centrosymmetric structure determination, the mode as obtained from the Hauptman (1975*b*) distribution [see also (I.5) and (I.9) in Appendix I]* is employed for the estimation of $|\psi_4|$, although this seems to have some drawbacks. van der Putten & Schenk (1979) found that the mode tends to overestimate quartets to be either zero or π. In the procedure they adopted, modes predicted to be zero or π were set away from these values. This appeared to work better than the use of the predicted zero and π values and also better than the use of the probabilistic means $\langle|\psi_4|\rangle$ as calculated from the Hauptman distribution.

From comparing $|\psi_4|$'s as calculated from the atomic coordinates with estimated ones (some test results can be found in this paper), it appears that the use of both the Hauptman expression and the exponential quartet expression of Giacovazzo [(1976*a*); see also (I.10) and (I.11) in Appendix I] results in systematic differences between calculated and estimated $|\psi_4|$'s. This applies to the use of both the mode and the probabilistic mean for the estimation of $|\psi_4|$. This might of course hamper solving the crystal structure with quartets.

Both the exponential quartet distributions mentioned are approximations. In the course of their derivation only those terms of order N^{-1} have been included which are dependent on the random variables for the phases, with the result that these expressions are correct up to order $N^{-1/2}$ only. An improvement of the estimates might therefore be gained by including first these missing higher-order terms of order N^{-1} in the exponential expressions and, secondly, terms of higher order than N^{-1}. However, this procedure is not straightforward, so another approach has been adopted.

Recently it has been shown that the inclusion of higher-order terms in a new series-expansion form of a joint probability distribution of the three normalized structure factors (n.s.f.'s) whose phases form a triplet invariant ψ_3 can reduce systematic differences when estimating $|\psi_3|$'s (Peschar & Schenk, 1986). In the present paper the method of deriving this new j.p.d. of three n.s.f.'s will be generalized in order to obtain a j.p.d. of an arbitrary number of structure factors. The derivation will be performed for the case of equal-atom structures in space group $P1$. Then, in the next step, the j.p.d. of the seven n.s.f.'s E_{H_1}, E_{H_2}, E_{H_3}, $E_{H_1+H_2+H_3}$, $E_{H_1+H_2}$, $E_{H_1+H_3}$ and $E_{H_2+H_3}$ and in particular the joint probability distribution of ψ_4, given the seven magnitudes, are obtained from the general expression. Finally, on the basis of test results it is shown that the new conditional j.p.d. of ψ_4, a series expansion, compares favourably with the quartet distributions of Hauptman and Giacovazzo.

* Appendix I has been deposited with the British Library Document Supply Centre as Supplementary Publication No. 8UP 43430 (2pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

2. The joint probability distribution of the normalized structure factors $E_{H_1}, E_{H_2}, \ldots, E_{H_W}$, $W \geq 3$

For structures consisting of N identical atoms in the space group $P1$, the normalized structure factor is given by

$$E_H = |E_H| \exp(i\varphi_H) = N^{-1/2} \sum_{j=1}^{N} \exp(2\pi i \mathbf{H} \cdot \mathbf{r}_j). \tag{4}$$

Suppose that W reciprocal vectors $\mathbf{H}_1, \ldots, \mathbf{H}_W$ are fixed, while subject to some restrictive relations among them; these relations need not be specified yet. The atomic position vectors $\mathbf{r}_j$ are assumed to be independent primitive random variables (p.r.v.'s). Hence, the n.s.f.'s, being functions of $\mathbf{r}_j$, can be considered to be continuous random variables themselves. Denote by R_H and Φ_H the continuous random variables for the magnitude $|E_H|$ and the phase φ_H of an n.s.f. E_H respectively.

By analogy with Hauptman (1975*a*), the j.p.d. of the absolute values and phases of the W n.s.f.'s, is written as a $2W$-fold integral:

$$\begin{aligned} P &= P(\Phi_1, \ldots, \Phi_W, R_1, \ldots, R_W) \\ &= \frac{R_1 \ldots R_W}{(2\pi)^{2W}} \int_{\rho_1,\ldots,\rho_W=0}^{\infty} \int_{\theta_1,\ldots,\theta_W=0}^{2\pi} \rho_1 \ldots \rho_W \\ &\quad \times \exp\left[-i \sum_{\nu=1}^{W} \rho_\nu R_\nu \cos(\theta_\nu - \Phi_\nu)\right] \\ &\quad \times C(\theta_1, \ldots, \theta_W, \rho_1, \ldots, \rho_W) \\ &\quad \times \mathrm{d}\theta_1 \ldots \mathrm{d}\theta_W \, \mathrm{d}\rho_1 \ldots \mathrm{d}\rho_W, \end{aligned} \tag{5}$$

with the characteristic function (c.f.) being

$$C = C(\theta_1, \ldots, \theta_W, \rho_1, \ldots, \rho_W) = \prod_{j=1}^{N} c_j \tag{6}$$

and

$$c_j = \left\langle \exp\left[iN^{-1/2} \sum_{\nu=1}^{W} \rho_\nu \cos(2\pi \mathbf{H}_\nu \cdot \mathbf{r}_j - \theta_\nu)\right]\right\rangle_{\mathbf{r}_j}. \tag{7}$$

However, in contrast to the method of Hauptman (*e.g.* 1975), the average in (7) is taken over all possible

positions of the atomic position vector $\mathbf{r}_j$. Expression (7) can be evaluated by applying (I.1) of Appendix I and taking the average over the $\mathbf{r}_j$. Assuming the independence of those variables as well as uniform distributions, the integrations yield zero unless

$$\sum_{i=1}^{W} n_{ij}\mathbf{H}_i = \mathbf{0} \tag{8}$$

in which $n_{ij}, \ldots, n_{Wj}$ are integers. From this it can be concluded that only those numerical $(n_{1j}, \ldots, n_{Wj})$ combinations contribute to the c.f., and hence to the j.p.d., for which

$$\sum_{i=1}^{W} n_{ij}\varphi_i \qquad \text{is a structure invariant.} \tag{9}$$

Next, denote by

$$\sum_{\mathbf{n}_j} \text{with } \mathbf{n}_j = (n_{1j}, \ldots, n_{Wj}) \tag{10}$$

a summation over all $(n_{1j}, \ldots, n_{Wj})$ combinations which are in accordance with (8). Thus, (7) can be rewritten

$$c_j = \sum_{\mathbf{n}_j} \prod_{\nu=1}^{W} (i)^{n_{\nu j}} J_{n_{\nu j}}(N^{-1/2}\rho_\nu) \exp(-i\theta_\nu n_{\nu j}) \tag{11}$$

and the c.f. (6) consequently as an N-fold product of these summations:

$$C = \sum_{\mathbf{n}_1,\ldots,\mathbf{n}_N} \prod_{k=1}^{N} \prod_{\nu=1}^{W} (i)^{m_\nu} J_{n_{\nu k}}(N^{-1/2}\rho_\nu) \times \exp(-i\theta_\nu m_\nu) \tag{12}$$

with

$$m_\nu = \sum_{k=1}^{N} n_{\nu k} \quad \text{for } \nu = 1, \ldots, W. \tag{13}$$

Since there are no restrictions on the integer m_ν values, (12) can be reordered such that a summation over the m_ν values is performed first while the subsequent summations over $(\mathbf{n}_i, \ldots, \mathbf{n}_N)$ combinations have to be carried out under the condition (13):

$$C = \sum_{m_1,\ldots,m_W=-\infty}^{\infty} \left[\prod_{\mu=1}^{W} i^{m_\mu} \exp(-im_\mu\theta_\mu)\right] \times \sum_{\mathbf{n}_1,\ldots,\mathbf{n}_N} \prod_{k=1}^{N} \prod_{\nu=1}^{W} J_{n_{\nu k}}(N^{-1/2}\rho_\nu) \tag{14}$$

with

$$n_{\nu 1} + \ldots + n_{\nu N} = m_\nu \quad \text{for } \nu = 1, \ldots, W.$$

From (5) and (4) and after performing all θ integrations using (I.2) of Appendix I, the expression for the j.p.d. is obtained as

$$P = (R_1 \ldots R_W)/(2\pi)^W \times \sum_{m_1,\ldots,m_W=-\infty}^{\infty} \left[\prod_{\mu=1}^{W} \exp(-im_\mu\Phi_\mu)\right] \times h_{m_1,\ldots,m_W}(R_1, \ldots, R_W) \tag{15}$$

with

$$h_{m_1,\ldots,m_W}(R_1, \ldots, R_W) = \sum_{\mathbf{n}_1,\ldots,\mathbf{n}_N} \prod_{\nu=1}^{W} \left[\int_0^\infty \rho_\nu J_{m_\nu}(\rho_\nu R_\nu) \times \prod_{k=1}^{N} J_{n_{\nu k}}(N^{-1/2}\rho_\nu)\, d\rho_\nu\right] \tag{16}$$

under condition (13).

At this point it appears to be helpful to introduce different summation variables [see Peschar & Schenk (1986) for a similar procedure] in order to arrive at a reordered expression. Suppose that in a certain $(\mathbf{n}_1, \ldots, \mathbf{n}_N)$ combination the number of different $\mathbf{n}$ sets is k. Denote these k different sets $\mathbf{q}_\lambda$, $\lambda \in [1, k]$ and $\mathbf{q}_\lambda = (n_{1\lambda}, \ldots, n_{W\lambda})$. Denote further by l_λ the frequency of the $\mathbf{q}_\lambda$ set in the $(\mathbf{n}_1, \ldots, \mathbf{n}_N)$ combination. So

$$\sum_{\lambda=1}^{k} l_\lambda = N \tag{17}$$

and

$$\sum_{\lambda=1}^{k} l_\lambda n_{\nu\lambda} = m_\nu. \tag{18}$$

As a consequence, the summations in (16) are changed into summations over (i) the total number of different $\mathbf{q}_\lambda$ sets; (ii) the numerical values of the $\mathbf{q}_\lambda = (\mathbf{n}_{1\lambda}, \ldots, \mathbf{n}_{W\lambda})$ sets, under conditions (18); and (iii) the frequency l_λ of the $\mathbf{q}_\lambda$ sets, under condition (17). For a certain $(\mathbf{q}_1, \ldots, \mathbf{q}_k, l_1, \ldots, l_k)$ choice, the total frequency $\text{com}_{\mathbf{q},l}$ in the $(\mathbf{n}_1, \ldots, \mathbf{n}_N)$ combination is

$$\text{com}_{\mathbf{q},l} = N! \Big/ \prod_{\lambda=1}^{k} l_\lambda! \tag{19}$$

Hence, (16) can be written as

$$h_{m_1,\ldots,m_W}(R_1, \ldots, R_W) = \sum_{\lambda=1}^{\infty} \sum_{\mathbf{q}_1,\ldots,\mathbf{q}_\lambda} \sum_{l_1,\ldots,l_\lambda} \left(\text{com}_{\mathbf{q},l} \prod_{\nu=1}^{W} \left\{\int_0^\infty \rho_\nu J_{m_\nu}(\rho_\nu R_\nu) \times \prod_{\lambda=1}^{k} [J_{n_{\nu\lambda}}(N^{-1/2}\rho_\nu)]^{l_\lambda}\, d\rho_\nu\right\}\right). \tag{20}$$

Since the $n_{\nu\lambda}$ values in the product

$$\sum_{\lambda=1}^{k} [J_{n_{\nu\lambda}}(N^{-1/2}\rho_\nu)]^{l_\lambda} \tag{21}$$

are not necessarily all different, other variables will facilitate the calculation of (20).

Suppose that in (21), for a fixed ν value, s different $n_{\nu\lambda}$ values are found. Denote these values $r_{\nu t}$, $t \in [1, s]$ and $s \leq k$. Further, denote the frequency of $r_{\nu t}$ among the $n_{\nu\lambda}$ values by $\alpha_{\nu t}$. Thus,

$$\sum_{t=1}^{s} r_{\nu t}\alpha_{\nu t} = m_\nu. \tag{22}$$

Hence, by introducing the new variables, (21) can be changed into

$$\prod_{t=1}^{s} [J_{r_{\nu t}}(N^{-1/2}\rho_\nu)]^{\alpha_{\nu t}}. \tag{23}$$

The procedure to perform the integrations in (20), combined with (23), has already been dealt with extensively for similar expressions elsewhere (Peschar & Schenk, 1986). So it suffices here to indicate that the approximation expressions (I.3) and (I.4) of Appendix I have been used.

The final expression obtained for the j.p.d. of the phases and the absolute values of the W n.s.f.'s can now be written:

$$\begin{aligned} P(\Phi_1, \ldots, \Phi_W, R_1, \ldots, R_W) &= \pi^{-W} \prod_{i=1}^{W} R_i \exp[-R_i^2] \\ &\times \sum_{m_1,\ldots,m_W} \exp\left[-i \sum_{j=1}^{W} (m_j \Phi_j)\right] \\ &\times g_{m_1,\ldots,m_W}(R_1, \ldots, R_W) \end{aligned} \tag{24}$$

with

$$\begin{aligned} g_{m_1,\ldots,m_W}(R_1, \ldots, R_W) &= \sum_{\lambda=1}^{k} \sum_{\mathbf{q}_1,\ldots,\mathbf{q}_\lambda} \sum_{l_1,\ldots,l_\lambda} \mathrm{com}_{\mathbf{q},l} \\ &\times \prod_{\nu=1}^{W} \left\{ \frac{\exp[R_\nu^2(1-D_\nu^{-2})] P_{\mu_\nu,\mu_\nu^*}(R_\nu D_\nu^{-1})}{N^{(\mu_\nu+\mu_\nu^*)/2} D_\nu^{\mu_\nu+\mu_\nu^*+2} \prod_{t=1}^{s} (|r_{\nu t}|!)^{\alpha_{\nu t}}} \right\}. \end{aligned} \tag{25}$$

The variables μ_ν, μ_ν^*, D_ν and P_{μ_ν,μ_ν^*} are defined by

$$\begin{aligned} \mu_\nu &= \left[\sum_{t=1}^{s} |r_{\nu t}|\alpha_{\nu t} + m_\nu\right] \Big/ 2 \\ \mu_\nu^* &= \left[\sum_{t=1}^{s} |r_{\nu t}|\alpha_{\nu t} - m_\nu\right] \Big/ 2 \end{aligned} \tag{26}$$

$$D_\nu^2 = N^{-1} \sum_{t=1}^{s} \alpha_{\nu t}/(|r_{\nu t}|+1) \tag{27}$$

$$P_{\mu_\nu,\mu_\nu^*}\left(\frac{R_\nu}{D_\nu}\right) = \sum_{\tau=0}^{\mu^*} (-1)^\tau \tau! \binom{\mu_\nu}{\tau} \binom{\mu_\nu^*}{\tau} \left(\frac{R_\nu}{D_\nu}\right)^{\mu_\nu+\mu_\nu^*-2\tau} \tag{28}$$

for $\mu_\nu \geq \mu_\nu^*$ and $P_{\mu_\nu,\mu_\nu^*} = P_{\mu_\nu^*,\mu_\nu}$. Note that the way in which (25) is expressed follows as closely as possible the way in which Naya, Nitta & Oda (1965) express their formulae.

3. Conditional j.p.d. of the quartet phase sum Ψ_4 given the seven magnitudes $|E_{H_1}|$, $|E_{H_2}|$, $|E_{H_3}|$, $|E_{H_1+H_2+H_3}|$, $|E_{H_1+H_2}|$, $|E_{H_1+H_3}|$ and $|E_{H_2+H_3}|$

From (24), the j.p.d. of the phases and magnitudes of the seven n.s.f.'s E_{H_1}, E_{H_2}, $E_{H_1+H_2+H_3}$, $E_{H_1+H_2}$, $E_{H_1+H_3}$ and $E_{H_2+H_3}$ can be obtained by taking $W=7$ and specifying four restrictive conditions among the reciprocal vectors:

$$\begin{aligned} \mathbf{H}_4 &= -\mathbf{H}_1 - \mathbf{H}_2 - \mathbf{H}_3; & \mathbf{H}_5 &= \mathbf{H}_1 + \mathbf{H}_2; \\ \mathbf{H}_6 &= \mathbf{H}_1 + \mathbf{H}_3; & \mathbf{H}_7 &= \mathbf{H}_2 + \mathbf{H}_3. \end{aligned} \tag{29}$$

Assume further the extra condition that $\mathbf{H}_1$, $\mathbf{H}_2$ and $\mathbf{H}_3$ are linearly independent. The conditional j.p.d. of Ψ_4 given the seven magnitudes can be obtained from the j.p.d. of the phases and magnitudes by integrating with respect to the random variables $\Phi_1, \ldots, \Phi_7$ under the condition

$$\Psi_4 = \Phi_1 + \Phi_2 + \Phi_3 + \Phi_4 \tag{30}$$

in which Ψ_4 is the random variable for the quartet phase sum. Only those $(m_1, \ldots, m_7)$ combinations in (24) (with $W=7$) yield non-zero contributions to the conditional distribution which satisfy the conditions

$$m_1 = m_2 = m_3 = m_4 \equiv M, \qquad M \text{ integer} \tag{31a}$$

and

$$m_5 = m_6 = m_7 = 0. \tag{31b}$$

The function $g_{m_1,\ldots,m_7}(R_1, \ldots, R_7)$ will therefore be abbreviated to $g_M(R_1, \ldots, R_7)$. This results in the following expression for the conditional j.p.d. of Ψ_4 given the seven magnitudes:

$$P(\Psi_4|R_1, \ldots, R_7) = L^{-1} \sum_{M=-\infty}^{\infty} \exp(-iM\Psi_4) g_M(R_1, \ldots, R_7) \tag{32}$$

with L^{-1} = a normalization constant.

It should be noted that because (I.3) of Appendix I has been used, (32) does not contain all possible terms up to a certain order but, owing to the approximation, only a selective number of them, which is nevertheless vast, as can be judged from Table 1.

4. The evaluation of expression (32)

The calculation of (32) seems to be impracticable because of the infinite summation. However, for practical applications, (32) should contain only those terms which control its shape, and therefore it is necessary to have an indicator for the relative importance of the individual terms. The N dependence serves this purpose because it can be shown that for terms of increasing order of N their contribution to the sum decreases.

From (25) with $W=7$ it follows that the order depends on the frequency $\mathrm{com}_{\mathbf{q},l}$ (19) and on

$$\prod_{\nu=1}^{7} N^{(1/2)(\mu_\nu+\mu_\nu^*)}. \tag{33}$$

Although (19) and (33) cannot be expressed in the

Table 1. *Cumulative number of different* $\mathbf{q} = (n_1, \ldots, n_7)$ *combinations and the cumulative number of terms in equation* (32) *with* $M \geq 0$ *up to order* N^{-4}

Order: N^{-s}, s varies from 0 to 4.

s	Cumulative number of $\mathbf{q}$	Cumulative number of terms in equation (32) with $M \geq 0$
0·0	1	1
0·5	13	1
1·0	27	11
1·5	55	11
2·0	77	103
2·5	145	103
3·0	207	1010
3·5	319	1010
4·0	483	9133

same variables, for a given $(\mathbf{q}_1, \ldots, \mathbf{q}_\lambda, l_1, \ldots, l_\lambda)$ combination in (25), the order is calculated easily. Then the derivation of (32) resolves itself into the determination of explicit expressions for all $g_M(R_1, \ldots, R_7)$. This involves the following steps:

(*a*) The selection of the allowed $\mathbf{q}$ combinations in accordance with (8) and (29).

(*b*) The selection of $(\mathbf{q}_1, \ldots, \mathbf{q}_\lambda, l_1, \ldots, l_\lambda)$ combinations under the restrictive conditions (17), (18) and (31).

(*c*) For each selected $(\mathbf{q}_1, \ldots, l_\lambda)$ combination, the calculation and coded storage of the variables and constants of (25)-(27) without using numerical values for N or the $|E|$'s.

Although this procedure can be carried out by hand, it is rather cumbersome and time consuming to do so. Instead, a computer program has been written in Fortran V in which the steps are performed, resulting in the j.p.d. (32) in the form of a number of coded terms written on an internal computer device. Thus, the whole procedure to arrive at the explicit form of the joint probability distribution amounts to a computer-aided derivation. In Table 1, the number of terms arising from the development of (32) is listed up to order N^{-4}. The second column shows the cumulative number of different $\mathbf{q} = (n_1, \ldots, n_7)$ combinations, calculated according to (8) and (32). The third column lists the cumulative number of allowed $(\mathbf{q}_1, \ldots, \mathbf{q}_\lambda, l_1, \ldots, l_\lambda)$ combinations in accordance with (17), (18) and (31), although it should be noted that, in view of $g_{-M}(R_1, \ldots, R_7) = g_M(R_1, \ldots, R_7)$, only those terms with $M \geq 0$ need to be considered. The computer time needed to generate the terms listed in Table 1 varies from 1·5 to 16 central-processor seconds on a Cyber 750, depending on the inclusion of terms up to and including order N^{-3} or order N^{-4}. The number of terms listed in Table 1, comprising the conditional j.p.d. (32), form the basis of subsequent calculations, *e.g.* the calculation of expectation values of Ψ_4. In view of the foregoing, these calculations are reduced to simple summations employing the stored information as well as numerical values for N and $|E|$'s. Therefore, the computer time needed to calculate these expectation values is almost the same as the time needed when using the Hauptman distribution [(I.5)-(I.9) of Appendix I] and only about five times as much as the time needed for the Giacovazzo distribution [(I.10)-(I.11)].

5. Convergence of expression (32)

The convergence properties of (32) have been investigated by calculating expectation values and variances of $|\Psi_4|$ for several combinations of N and $|E_H|$ values, while the order up to which the higher-order terms are included in (32) has been varied as well. Hence, the changes in the estimated $|\Psi_4|$ values inform us about the achieved convergence. Some representative test results are shown in Table 2. For comparison the calculations have also been done with the expressions of Hauptman (I.5)-(I.9) and Giacovazzo (I.10)-(I.11). From this table it can be seen that for most quartets a reasonable convergence is achieved by including terms up to order N^{-4} or even N^{-3}. However, for quartets with a large E_4 value in combination with either all large cross magnitudes $|E_{H_5}|$, $|E_{H_6}|$ and $|E_{H_7}|$ or small ones, the estimates, and, more particularly, the variances may not be sufficiently reliable. For these quartets the inclusion of still higher-order terms seems to be advisable, but at present this has not been investigated further. If the data obtained with (32) are compared with those of the Hauptman and Giacovazzo distributions, it appears that for almost all quartets, except those predicted to be clearly negative, the expectation values $\langle|\Psi_4|\rangle$ from (32) lie in between those obtained from the Hauptman and Giacovazzo distributions. The same observation has been made for quartets of which only two cross-term magnitudes were available, although for brevity these results are not given in tabulated form.

6. Results and discussion

Remarks concerning the testing procedures

In order to investigate the quality of the phase sum estimates obtained from (32) for several equal-atom structures, in artificial as well as realistic models, an overall comparison between actual and estimated $|\Psi_4|$'s has been made. From the atomic coordinates, E_H values were calculated. After that, in the group of strongest reflections, the quartet relations were generated while employing a lower limit for E_4.

For the estimation of $|\Psi_4|$ two options have been tested: (i) the probabilistic expectation value, the mean of $|\Psi_4|$, denoted by $\langle|\Psi_4|\rangle$; and (ii) the mode of the distribution, *i.e.* the most probable value of $|\Psi_4|$, denoted by $|\Psi_4|_{\text{mode}}$.

For the overall comparison of true and estimated $|\Psi_4|$'s, two cumulative averages have been calculated:

$$\mathrm{AV1} = \langle |\Psi_4|_{\mathrm{true}} - |\Psi_4|_{\mathrm{est}} \rangle \tag{34}$$

and

$$\mathrm{AV2} = \langle | \, |\Psi_4|_{\mathrm{true}} - |\Psi_4|_{\mathrm{est}} | \rangle. \tag{35}$$

AV2 is the mean absolute difference between the actual and the estimated $|\Psi_4|$'s. The other average, AV1, is the overall systematic error of the estimated $|\Psi_4|$'s with respect to the true $|\Psi_4|$'s. For the cumulative presentation, the quartets should be ranked according to a weight which indicates the reliability of the estimation. For this ranking two weights have been employed: (i) the quartet product E_4, and (ii) the inverse variance of the estimated $|\Psi_4|$. The variance associated with the mean $\langle |\Psi_4| \rangle$ is given by

$$\sigma^2(|\Psi_4|) = \langle |\Psi_4|^2 \rangle - (\langle |\Psi_4| \rangle)^2, \tag{36}$$

while the variance for $|\Psi_4|_{\mathrm{mode}}$ is defined by

$$\sigma^2(|\Psi_4|_{\mathrm{mode}}) = \int_0^\pi (|\Psi_4|_{\mathrm{mode}} - \Psi_4)^2 \, \mathrm{d}\Psi_4. \tag{37}$$

Apart from cumulative distributions, non-cumulative distributions have been calculated as well. For comparison of these distributions the averages

$$M1 = \langle |\Psi_4|_{\mathrm{true}} - |\Psi_4|_{\mathrm{est}} \rangle \tag{38}$$

and

$$M2 = \langle | \, |\Psi_4|_{\mathrm{true}} - |\Psi_4|_{\mathrm{est}} | \rangle \tag{39}$$

have been calculated. These averages include quartets with $|\Psi_4|$ estimated in a certain $|\Psi_4|$ range only. Hence, $M1$ is an indicator for the systematic errors of the $|\Psi_4|$ estimates which lie in a certain $|\Psi_4|$ range. Likewise, $M2$ indicates the absolute errors for the estimates in the $|\Psi_4|$ range.

Expression (32) has been tested for four structures: three randomly generated structures in space group $P1$ with code names P1A25, P1A50 and P1A100, consisting of 25, 50 and 100 equal atoms respectively, and a 30-atom realistic model structure with codename KANTER [the structure of Kanters & van Veen (1973) changed to an equal-atom structure]. The calculations have been performed for quartets with at least two cross-term magnitudes present. In these calculations (32) included terms up to order N^{-3} since for structure P1A25 preliminary results with (32), including the terms of order N^{-4}, showed no notable improvement of the overall estimates. Occasionally, some variances were estimated to be negative. These quartets were included, but for practical reasons the negative values were reset to positive ones. The discussion of the test results will concentrate on the data for P1A25, since the conclusions to be drawn from these data are applicable to the other structures tested as well. Therefore, only the results for P1A25 are listed completely in Table 3. In Table 6 a summary is given of the most important results for all four structures, from which the representativity can be judged.

Table 2. *Some expectation values and variances of* $|\Psi_4|$ *as obtained from* (32), *while* (32) *includes terms up to order* $N^{-s_{\max}}$

$s_{\max}$ varies from 1 to 4. For comparison the values as obtained from the quartet expressions of Hauptman (I.5) and Giacovazzo (I.10) are listed as well.

$N = 25 \quad |E_{H_1}| = |E_{H_2}| = |E_{H_3}| = |E_{H_1+H_2+H_3}| = 2{\cdot}0$

	A	*B*	*C*	*D*	*E*	*F*	*G*	*H*	*I*	*J*
$\lvert E_{H_1+H_2}\rvert$	2·0	2·0	2·0	1·5	1·5	1·5	1·0	1·0	1·0	0·5
$\lvert E_{H_1+H_3}\rvert$	2·0	2·0	1·5	1·5	1·5	1·0	1·0	1·0	0·5	0·5
$\lvert E_{H_2+H_3}\rvert$	2·0	1·5	1·5	1·5	1·0	1·0	1·0	0·5	0·5	0·5

	$s_{\max}$	1	2	3	4	Hauptman	Giacovazzo
A	$\langle \lvert\Psi_4\rvert \rangle$*	95	89	54	42	89	36
	$\sigma^2(\lvert\Psi_4\rvert)$†	−19	32	−10	−19	28	5
B	$\langle \lvert\Psi_4\rvert \rangle$	97	90	62	56	99	40
	$\sigma^2(\lvert\Psi_4\rvert)$	−17	32	0	−1	34	6
C	$\langle \lvert\Psi_4\rvert \rangle$	99	92	71	71	111	45
	$\sigma^2(\lvert\Psi_4\rvert)$	−13	33	11	14	42	8
D	$\langle \lvert\Psi_4\rvert \rangle$	102	96	83	85	129	54
	$\sigma^2(\lvert\Psi_4\rvert)$	−6	34	22	24	55	11
E	$\langle \lvert\Psi_4\rvert \rangle$	107	103	103	106	155	64
	$\sigma^2(\lvert\Psi_4\rvert)$	2	40	39	39	71	16
F	$\langle \lvert\Psi_4\rvert \rangle$	115	123	134	135	194	83
	$\sigma^2(\lvert\Psi_4\rvert)$	6	55	58	58	97	30
G	$\langle \lvert\Psi_4\rvert \rangle$	136	176	183	182	247	140
	$\sigma^2(\lvert\Psi_4\rvert)$	40	83	80	80	115	81
H	$\langle \lvert\Psi_4\rvert \rangle$	185	320	258	255	300	218
	$\sigma^2(\lvert\Psi_4\rvert)$	104	17	91	97	109	126
I	$\langle \lvert\Psi_4\rvert \rangle$	‡	399	311	312	346	312
	$\sigma^2(\lvert\Psi_4\rvert)$	‡	−61	113	109	86	115
J	$\langle \lvert\Psi_4\rvert \rangle$	−8	218	365	382	381	377
	$\sigma^2(\lvert\Psi_4\rvert)$	−288	176	87	53	61	66

* $\langle |\Psi_4| \rangle$ in mc (=rad × 1000/2π), rounded off.
† $\sigma^2(|\Psi_4|)$ in rad² × 1000/2π, rounded off.
‡: out of range.

Ranking criterion for the estimation: E_4 *versus inverse variance*

In Table 3 results of (32) for the structure P1A25 are listed. In the upper section of this table, the data are ranked according to σ^{-2}, the inverse variance of the estimates, while in the middle section the E_4 value is the ranking criterion. The results for P1A25 obtained with the Hauptman and Giacovazzo distributions, ordered in the same way, can be found in Tables 4 and 5 respectively. Comparison of the data ranked according to σ^{-2} and E_4 leads to the conclusion that, irrespective of the type of estimation, mode or mean, or the number of cross-term magnitudes present, for all three distributions ordering according to σ^{-2} is superior in view of the lower overall systematic and absolute estimation errors (AV1 and AV2 respectively). This confirms clearly the importance of including information on the cross terms. Therefore, only the ranking according to the inverse variance will be considered further for the cumulative averages.

Table 3. *Comparison of true and estimated* $|\Psi_4|$'s

Estimates from the new quartet distribution, equation (32) for P1A25. Quartets calculated from the 150 strongest reflections. $E_4 \geq 0{\cdot}5$. $|\Psi_4|$ estimated with $\langle|\Psi_4|\rangle$ and $|\Psi_4|_{mode}$. Ranking of quartets according to inverse variance $\sigma^{-2}(|\Psi_4|)$, E_4 and estimated $|\Psi_4|$. Cumulative averages AV1 and AV2 and the averages for the estimates in the $|\Psi_4|$ ranges, $M1$ and $M2$, all in millicycles (mc).

	Three cross magnitudes known						Two cross magnitudes known					
		$\langle\|\Psi_4\|\rangle$			$\|\Psi_4\|_{mode}$			$\langle\|\Psi_4\|\rangle$			$\|\Psi_4\|_{mode}$	
σ^{-2}	CNΨ_4*	AV1	AV2	CNΨ_4	AV1	AV2	CNΨ_4	AV1	AV2	CNΨ_4	AV1	AV2
	81	6	42	100	65	73	84	3	43	75	71	76
	262	2	46	210	70	80	269	−3	47	201	82	82
	331	−3	49	315	66	87	385	−6	47	329	79	81
	470	0	56	503	61	91	652	−2	53	542	83	88
	584	−1	59	652	70	97	933	−2	58	1054	76	100
	655	0	63	825	78	104	1511	−3	68	1530	86	108
	825	2	71				2075	−5	78	2075	89	120
E_4	CNΨ_4	AV1	AV2	CNΨ_4	AV1	AV2	CNΨ_4	AV1	AV2	CNΨ_4	AV1	AV2
	90	22	65	90	92	102	50	−14	56	50	70	90
	265	6	67	265	79	101	203	−10	66	203	76	96
	825	2	71	825	78	104	665	−3	75	665	87	112
							2075	−5	78	2075	89	120
$\|\Psi_4\|$ range	NΨ_4†	$M1$	$M2$	NΨ_4	$M1$	$M2$	NΨ_4	$M1$	$M2$	NΨ_4	$M1$	$M2$
0-100	265	−2	46	624	102	104	413	−3	47	1675	118	119
100-200	432	−4	78	91	42	103	1277	−6	79	152	41	107
200-300	89	−7	109	53	5	120	328	−4	112	111	4	119
300-400	39	16	81	35	−41	91	57	16	96	51	−79	131
400-500	0	—	—	22	−107	111	0	—	—	86	−166	171

* CNΨ_4 = Number of quartets included in cumulative averages.
† NΨ_4 = number of quartets with $|\Psi_4|$ estimated in indicated interval.

Table 4. *Comparison of true and estimated* $|\Psi_4|$'s

Estimates from the Hauptman distribution, equations (I.5)-(I.9) of Appendix I for P1A25. Ranking of quartets according to σ^{-2}, E_4 and estimated $|\Psi_4|$. Cumulative means AV1 and AV2 and the means for the $|\Psi_4|$ ranges, $M1$ the absolute mean and $M2$ the systematic mean, all in mc.

	Three cross magnitudes known						Two cross magnitudes known					
		$\langle\|\Psi_4\|\rangle$			$\|\Psi_4\|_{mode}$			$\langle\|\Psi_4\|\rangle$			$\|\Psi_4\|_{mode}$	
σ^{-2}	CNΨ_4	AV1	AV2	CNΨ_4	AV1	AV2	CNΨ_4	AV1	AV2	CNΨ_4	AV1	AV2
	81	−40	53	86	10	83	67	−23	53	72	77	77
	253	−44	59	317	−7	89	371	−32	57	406	30	89
	329	−43	63	538	16	89	694	−33	64	873	51	96
	484	−44	69	825	30	103	1463	−36	78	1691	74	113
	825	−46	83				2075	−37	87	2075	69	124
E_4	CNΨ_4	AV1	AV2	CNΨ_4	AV1	AV2	CNΨ_4	AV1	AV2	CNΨ_4	AV1	AV2
	90	−34	68	90	32	91	50	−54	75	50	62	96
	265	−46	80	265	28	97	203	−47	79	203	61	101
	825	−46	83	825	30	103	665	−39	85	665	70	114
							2075	−37	87	2075	69	124
$\|\Psi_4\|$ range	NΨ_4	$M1$	$M2$	NΨ_4	$M1$	$M2$	NΨ_4	$M1$	$M2$	NΨ_4	$M1$	$M2$
0-100	15	−25	41	511	92	93	33	−37	52	1610	117	117
100-200	528	−50	74	112	−11	92	1347	−37	77	100	21	102
200-300	214	−48	107	86	−45	101	621	−43	110	113	−53	115
300-400	68	−15	94	46	−107	132	74	8	102	79	−86	133
400-500	0	—	—	70	−171	171	0	—	—	173	−197	199

Comparison of the use of the mean and the mode of expression (32) *for estimating* $|\Psi_4|$

From the upper left three columns in Table 3 it can be seen that for the 825 quartets with three measured cross-term magnitudes, the use of the mean, $\langle|\Psi_4|\rangle$, for estimating $|\Psi_4|$ results in a systematic estimation error of only 2 millicycles (mc) and an absolute overall estimation error of 71 mc. In contrast, employment of the mode, $|\Psi_4|_{mode}$, of (32) leads for the same quartets to a systematic error of +78 mc which means that the estimates are on the average 78 mc too low and, moreover, the absolute error is also higher: 104 mc.

This remarkable difference between the use of the mean and the mode can be elucidated further. In the lowest section of Table 3 the quartets have been ranked according to their estimated $|\Psi_4|$ value. For five $|\Psi_4|$ estimated ranges the overall systematic and absolute differences with respect to the true $|\Psi_4|$

Table 5. *Comparison of true and estimated $|\Psi_4|$'s*

Estimates from the Giacovazzo distribution, equations (I.10)-(I.11) of Appendix I P1A25. Ranking of quartets according to σ^{-2}, E_4 and estimated $|\Psi_4|$. All averages in mc

	Three cross magnitudes known						Two cross magnitudes known					
		$\langle\Psi_4\rangle$			$\lvert\Psi_4\rvert_{\text{mode}}$			$\langle\lvert\Psi_4\rvert\rangle$			$\lvert\Psi_4\rvert_{\text{mode}}$	
σ^{-2}	$CN\Psi_4$	AV1	AV2	$CN\Psi_4$	AV1	AV2	$CN\Psi_4$	AV1	AV2	$CN\Psi_4$	AV1	AV2
	168	32	45	168	78	78	358	29	46	362	79	79
	260	36	51	278	85	85	611	33	52	602	89	89
	497	40	60	503	99	99	1412	37	68	1336	111	111
	825	46	77	825	108	131	2075	29	79	2075	111	138
E_4	$CN\Psi_4$	AV1	AV2	$CN\Psi_4$	AV1	AV2	$CN\Psi_4$	AV1	AV2	$CN\Psi_4$	AV1	AV2
	90	70	81	90	124	139	50	30	55	50	88	98
	265	55	77	265	115	128	203	31	65	203	99	113
	825	46	77	825	108	131	665	35	75	665	111	129
							2075	29	79	2075	111	138
$\lvert\Psi_4\rvert$ range	$N\Psi_4$	*M*1	*M*2	$N\Psi_4$	*M*1	*M*2	$N\Psi_4$	*M*1	*M*2	$N\Psi_4$	*M*1	*M*2
0-100	607	43	65	771	128	128	1159	38	63	1934	134	134
100-200	143	64	113	0	—	—	611	22	92	0	—	—
200-300	51	34	120	0	—	—	247	4	114	0	—	—
300-400	24	34	83	0	—	—	58	14	96	0	—	—
400-500	0	—	—	54	−176	176	0	—	—	141	−201	201

values are listed. From the data obtained with the mean, it can be seen that for all five $|\Psi_4|$ estimated ranges the systematic errors are low. On the other hand, the data for the mode show large deviations. The $|\Psi_4|$ values predicted to be in the ranges 0-100 and 100-200 mc have been estimated systematically too low, respectively 102 and 42 mc. However, those predicted to lie in the regions 300-400 and 400-500 mc are estimated too high, respectively 41 and 107 mc. These results clearly illustrate the tendency of the mode to overestimate quartets to be either zero or π. These conclusions apply also to the quartets of which only two cross-term magnitudes have been measured, as can be inferred from the right half of Table 3. As mentioned before, the trends visible in Table 3 are representative for the other structures tested, so it can be concluded that for estimating $|\Psi_4|$ using expression (32), the use of the mean yields better results than the mode. Moreover, since the means result in almost an absence of systematic errors, it can be concluded that the joint probability distribution (32) in its approximate form fits the true distribution of phase sums $|\Psi_4|$ almost completely.

Estimating $|\Psi_4|$ using the distributions of Hauptman and Giacovazzo. Comparison with expression (32)

The results for structure P1A25 obtained with the Hauptman expression (I.5)-(I.9) are listed in Table 4. From these data it appears that employment of the mean leads to systematic errors: the $|\Psi_4|$ value of quartets of which two and three cross-term magnitudes have been measured is estimated on the average 46 and 37 mc too high, respectively. The lower section of this table indicates that the estimates up to 300 mc are responsible for this result. In contrast, the use of the mode from the Hauptman distribution leads to overestimates of zero and π, although these errors cancel partly in the overall systematic averages. For example, Table 4 shows that from the 825 quartets with three measured cross-term magnitudes 511 have been estimated in the $|\Psi_4|$ range 0-100 mc, though the estimates are on the average 92 mc too low, while those estimated in the $|\Psi_4|$ ranges 300-400 and 400-500 mc are on the average estimated 107 and 171 mc too high, respectively. Nevertheless, the overall systematic error for these quartets employing the mode is considerably smaller: 30 mc. The data for P1A25 obtained with the distribution of Giacovazzo (I.10)-(I.11) can be found in Table 5. Since almost all estimates are too low in comparison with the real data, it can be concluded that the Giacovazzo expression shows a clear tendency to overestimate $|\Psi_4|$ to be zero. The only exceptions are the estimates of π obtained with the mode of this distribution, but these are systematically too high.

Comparison of the results for all four structures

Finally, in Table 6 a summary is given of the overall averages for the four structures tested. It can be concluded from these data that estimating $|\Psi_4|$ with the mean $\langle|\Psi_4|\rangle$ from (32) yields the best results. Most important is the enormous reduction of the systematic errors of the estimates. For example, compare for the 807 quartets with three measured cross-term magnitudes of the structure KANTER the systematic error of 1 mc when employing the mean from (32), with the −46 and +44 mc obtained with the means from the Hauptman and Giacovazzo distributions respectively. The absolute estimation errors are also the lowest if the mean from (32) is used, *e.g.* compare

Table 6. *Survey of overall $|\Psi_4|$ estimates using expression* (32) *and the quartet expressions of Hauptman* (*H*) *and Giacovazzo* (*G*)

The overall systematic and absolute differences between the estimates and the true data, AV1 and AV2 respectively, are given in mc. Structures tested: P1A25, KANTER, P1A50 and P1A100 containing 25, 30, 50 and 100 equal atoms respectively.

Number of strongest reflections used for generating quartets with lower limit E_4			P1A25 150 0·5		KANTER 200 0·7	P1A50 200 0·4	P1A100 250 0·1
Structure	Number of quartets	Number of cross terms	AV1	AV2	AV1	AV2	Distribution
P1A25	825	3	2	71	78	104	(32)
			−46	83	30	103	*H*(1.5)
			46	77	108	121	*G*(1.10)
	2075	2	−5	78	89	120	(32)
			−37	87	69	124	*H*(1.9)
			29	79	111	138	*G*(1.11)
KANTER	807	3	1	52	74	80	(32)
			−46	66	53	74	*H*(1.5)
			44	57	86	87	*G*(1.10)
	1353	2	−6	54	78	86	(32)
			−38	64	73	87	*H*(1.9)
			36	57	90	94	*G*(1.11)
P1A50	1267	3	−1	82	69	115	(32)
			−32	89	33	120	*H*(1.5)
			46	86	119	152	*G*(1.10)
	1417	2	−2	89	101	138	(32)
			−22	93	78	139	*H*(1.9)
			34	89	119	156	*G*(1.11)
P1A100	898	3	−4	107	106	177	(32)
			−16	109	80	178	*H*(1.5)
			19	107	123	195	*G*(1.10)
	1592	2	−1	109	124	187	(32)
			−9	110	112	186	*H*(1.9)
			16	108	129	194	*G*(1.11)

for the same 807 quartets the absolute error of 52 mc for (32) with the 66 and 57 mc respectively for the Hauptman and Giacovazzo expressions. Going from the data of P1A25 to P1A100, *i.e.* to larger structures, it can be seen that the overall differences between the results of the three expressions decrease, although even for the 100-atom structure P1A100 they are significantly present.

In conclusion, it has been shown in this paper that the incorporation of a selection of higher-order terms up to and including $O(N^{-3})$ terms in a new joint probability distribution of seven normalized structure factors, expressed as a series expansion, yields an improvement of the estimation of $|\Psi_4|$. In particular the systematic differences between the real and estimated $|\Psi_4|$'s are reduced considerably when compared with the results from the quartet expressions of Hauptman (1975*b*) and Giacovazzo (1976*a*). The absolute estimation errors are also lower for the new distribution although this reduction is smaller. In particular, the low systematic errors show that, although not all terms up to $O(N^{-3})$ have been included due to the approximation (I.3) employed, the approximation discussed leads to results which seem to be hardly improvable by including all terms up to $O(N^{-3})$.

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