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The Joint Probability Distribution of Structure Factors Incorporating Anomalous-Scattering and Isomorphous-Replacement Data

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Abstract

A method to derive joint probability distributions of structure factors is presented which incorporates anomalous-scattering and isomorphous-replacement data in a unified procedure. The structure factors F_H and F_{-H}, whose magnitudes are different due to anomalous scattering, are shown to be isomorphously related. This leads to a definition of isomorphism by means of which isomorphous-replacement and anomalous-scattering data can be handled simultaneously. The definition and calculation of the general term of the joint probability distribution for isomorphous structure factors turns out to be crucial. Its analytical form leads to an algorithm by means of which any particular joint probability distribution of structure factors can be constructed. The calculation of the general term is discussed for the case of four isomorphous structure factors in $P1$, assuming the atoms to be independently and uniformly distributed. A main result is the construction of the probability distribution of the 64 triplet phase sums present in space group $P1$ amongst four isomorphous structure factors F_H, four isomorphous F_K and four isomorphous F_{-H-K}. The procedure is readily generalized in the case where an arbitrary number of isomorphous structure factors are available for F_H, F_K and F_{-H-K}.

Abbreviations

c.f.	Characteristic function
j.p.d.('s)	Joint probability distribution(s)
c.p.d.('s)	Conditional probability distribution(s)
p.r.v.('s)	Primitive random variable(s)
r.v.('s)	Random variable(s)
s.f.('s)	Structure factor(s)
s.i.('s)	Structure invariant(s)
(S)IR	(Single-derivative) isomorphous replacement
(S)AS	(Single-wavelength) anomalous scattering

1. Introduction

Most small structures can be solved routinely by direct methods nowadays but if the structural size exceeds approximately 100 independent non-H atoms the conventional approach tends to be inadequate. Traditionally, direct methods processes a single non-anomalous data set. After the normalization, the phases related to the largest $|E|$'s are determined and refined successively *via* a tangent phase expression (Karle & Karle, 1966). This technique relies heavily on the quality of the phase-sum estimates. Their decrease in reliability for larger structures leads readily to phase-error accumulation which may obstruct a successful structure determination. On the other hand, it is well known that macromolecules can be solved by means of the isomorphous-replacement (IR) technique, often supplemented by anomalous-scattering (AS) data. The former technique (*e.g.* Srinivasan & Parthasarathy, 1976) requires data collection of at least two isomorphous structures, *e.g.* the data of a native protein and a heavy-atom derivative. Although in practice isomorphism tends to get lost at higher $(\sin\theta)/\lambda$, the number of available intensities increases markedly while the number of variables, the atomic positions, increases only marginally. A similar result can be achieved with AS (*e.g.* Ramaseshan & Abrahams, 1975). If the X-ray wavelength is selected near an atomic absorption edge, Friedel's law breaks down and the number of available intensities doubles. The wavelength dependence of AS suggests the collection of several data sets, each at a different wavelength near an absorption edge.

Traditionally, IR and/or AS data are expressed algebraically in the phase differences and magnitudes of substructures. Their implementation in a phasing procedure requires an initial phasing model for which purpose the heavy-atom substructure usually must be solved first.

More recently, attempts have been made to fuse the algebraic approach of IR and AS data with direct methods in order to circumvent the solvation of the heavy-atom substructure. Kroon, Spek & Krabbendam (1977) were the first to combine the conventional Bijvoet difference technique for anomalous-dispersion data with estimates of double Patterson quantities. After a scaling procedure, they obtained triplet estimates unique on the interval $(0, \pi)$. This procedure could be improved by estimating the double

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Patterson quantities probabilistically (Heinerman, Krabbendam, Kroon & Spek, 1978). Karle (1980) showed that an algebraic analysis of multiwavelength data leads to simultaneous equations of wavelength-independent quantities. In order to get phase estimates the triplet phase sums of the heavy-atom substructure are assumed to be concentrated near zero (Karle, 1984). Recently, Klop, Krabbendam & Kroon (1989) combined algebraic analysis results of two-wavelength anomalous-dispersive data of a single structure with the joint probability distribution (j.p.d.) of Hauptman (1982*b*) in order to estimate triplet invariants.

Alternatively, the j.p.d. of the structure factors (s.f.'s) involved may be derived which does not require necessarily the prior knowledge of the heavy-atom substructure nor the introduction of non-measurable substructural quantities. Hauptman (1982*a*) and more recently Giacovazzo, Cascarano & Zheng Chao-de (1988) obtained expressions for the triplet phase sum which are valid for single-isomorphous-replacement (SIR) data. Distributions for the triplet invariant in the single-wavelength anomalous-scattering (SAS) case were derived by Hauptman (1982*b*) and Giacovazzo (1983*a*). Fortier & Nigam (1989) showed that the SIR and SAS expressions and formulae for the partial and the complete structure, in which AS is neglected (Giacovazzo, 1983*b*), are analogous if the data sets are considered to be isomorphous. They suggested that a particular j.p.d. needs to be derived only once for a single combination of isomorphous data sets.

None of the above procedures combines AS data simultaneously with IR data in a general probabilistic derivation procedure. In this paper a technique is presented which does realise that very incorporation. The derivation procedure is based upon a previously described technique to derive automatically j.p.d.'s [Peschar & Schenk (1987); from now on P&S] but it is adapted in order to cope with complex-valued atomic scattering factors and isomorphism. It is demonstrated that F_H and F_{-H}, different due to AS, can be handled effectively as isomorphous s.f.'s, each corresponding with a different isomorphous structure. This concept enables the incorporation of IR and AS data in a single mechanism. It is argued that only the general term of the distribution of isomorphous s.f.'s needs to be defined and calculated. Once its analytical form is known, any j.p.d. can be constructed by combining the expression for the general term with the derivation mechanism described in P&S. The calculation of the general term is discussed in detail for four isomorphous s.f.'s in space group $P1$ but the resulting formulae are readily generalized to an arbitrary number of isomorphous s.f.'s. A main object of this paper is to derive the j.p.d. of four isomorphous s.f.'s F_H, four isomorphous F_K and four isomorphous F_{-H-K}. This leads to a j.p.d. of twelve s.f.'s comprising 18 two-phase and 64 unique three-phase invariants. The final expression has been constructed from the separate general terms with the help of a computer program and turns out to be expressible in such a way that the generalization to an arbitrary number of isomorphous s.f.'s is apparent. Finally, concise formulae are given for the conditional probability distribution (c.p.d.) of the two- and three-phase s.i.'s which encompass the SIR expression of Giacovazzo, Cascarano & Zheng Chao-de (1988) and the SAS expressions of Hauptman (1982*b*) and Giacovazzo (1983*a*) as marginal distributions.

2. The joint probability distribution of structure factors. Inclusion of anomalous dispersion

In the recently developed method for deriving automatically j.p.d.'s of s.f.'s in any space group (P&S), the atomic scattering factors were considered to be real-valued. In order to cope with anomalous-scattering data the allowance for complex-valued scattering factors should be introduced.

The curve of the complex-valued scattering factor $f_j(H)$, including anomalous-dispersion corrections, *versus* $(\sin\theta)/\lambda$ differs for the various atom types. The dependence of f_j on the atom type j and the reflection H can therefore not be omitted,

$$f_j(H) \equiv f_{jH} = |f_{jH}| \exp[i\delta_{jH}]$$

or, equivalently, (1)

$$f_{jH} = f^n_{jH} + f'_{jH} + if''_{jH}$$

with f^n_{jH}, f'_{jH} and f''_{jH} the non-anomalous scattering factor, the real and the imaginary anomalous correction factors, respectively.

In analogy to P&S, the general-valued s.f. is defined as a sum of n $(=N/m)$ contributions involving N atoms in the unit cell and m symmetry operations. Each operation consists of a 3×3 rotational matrix R and a 3×1 translational matrix T. H denotes the 1×3 matrix of reciprocal coordinates and X_j the 3×1 coordinate matrix of r_j,

$$F_H = \sum_{j=1}^{n} \xi_{jH} f_{jH} \tag{2}$$

with

$$\xi_{jH} = m_H \sum_{s=1}^{\tau_H} \exp[2\pi i(HR_sX_j + HT_s)] \tag{3}$$

and multiplicity m_H (≥ 1) $(m = \tau_H m_H)$.

For s.f.'s with a phase restriction to Δ and $\Delta+\pi$ with $0 \leq \Delta < \pi$ a more convenient expression is (* means complex conjugate)

$$\xi_{jH} = m_H\{\xi'_{jH} + \xi'^{*}_{jH}\} \tag{4}$$

with

$$\xi'_{jH} = \exp(i\Delta) \sum_{s=1}^{\tau'_H} \exp[2\pi i(HR_sX_j + HT_s)]. \quad (5)$$

τ'_H operations can always be selected from the total τ_H such that (4) is real (see P&S).

In our derivation technique the reflections are fixed while the atomic positions r_j act as the p.r.v.'s. As a result, the magnitude and phase of the general-valued s.f.'s of H_ν, being functions of the r_j, are themselves r.v.'s, denoted R_ν and Φ_ν respectively. Similarly, the phase-restricted s.f. of H_μ is a r.v., denoted F_μ. Henceforth, all variables dependent on or associated with H_ν or H_μ are referred to *via* the subscripts ν and μ.

The j.p.d. of l general-valued s.f.'s and r-value-restricted s.f.'s can be set up in a standard way (Naya, Nitta & Oda, 1964, 1965; P&S),

$$\begin{aligned} P(R_1, \ldots, R_l, \Phi_1, \ldots, \Phi_l, F_1, \ldots, F_r) &= R_1 \ldots R_l (2\pi)^{-2l-r} \\ &\times \int_{-\infty}^{\infty} \ldots \int_{-\infty}^{\infty} \ldots \int_0^{2\pi} \ldots \int_0^{2\pi} \ldots \int_0^{\infty} \ldots \int_0^{\infty} \rho_1 \ldots \rho_l \\ &\times \exp\left[-i\left\{\sum_{\nu=1}^{l} [\rho_\nu R_\nu \cos(\theta_\nu - \Phi_\nu)] + \sum_{\mu=1}^{r} F_\mu u_\mu\right\}\right] \\ &\times C(\rho_1, \ldots, \rho_l, \theta_1, \ldots, \theta_l, u_1, \ldots, u_r) \\ &\times d\rho_1 \ldots d\rho_l \, d\theta_1 \ldots d\theta_l \, du_1 \ldots du_r. \end{aligned} \quad (6)$$

With the c.f. C in (6) being

$$\begin{aligned} C = \exp\left\{\sum_{j=1}^{n} \left[\ln\left\langle \exp\left(i\left\{\sum_{\nu=1}^{l} \tfrac{1}{2}[\rho_\nu \exp(-i\theta_\nu) f_{j\nu}\xi_{j\nu}\right.\right.\right.\right.\right. \\ + \rho_\nu \exp(i\theta_\nu) f^*_{j\nu}\xi^*_{j\nu}] \\ \left.\left.\left.\left.\left. + \sum_{\mu=1}^{r} f_{j\mu}[\xi'_{j\mu} + \xi'^*_{j\mu}]u_\mu\right\}\right)\right\rangle_{r_j}\right]\right\}. \end{aligned} \quad (7)$$

A Taylor-series expansion of the exponential argument of the logarithm in (7) yields

$$C = \exp\left(\sum_{j=1}^{n} \ln\left\{\sum_{n\max=0}^{\infty} U_{n\max}[m^{\beta_1,\ldots,\beta_l}_{\alpha_1,\ldots,\alpha_l,\gamma_1,\ldots,\gamma_r}]\right\}\right). \quad (8)$$

$U_{n\max}$ can be expressed [combining equations (16) and (24) of P&S] as

$$\begin{aligned} U_{n\max} = & \sum_{\substack{\alpha_1,\ldots,\alpha_l,\beta_1,\ldots,\beta_l,\gamma_1,\ldots,\gamma_r=0 \\ \alpha_1+\ldots+\alpha_l+\beta_1+\ldots+\beta_l+\gamma_1+\ldots+\gamma_r=n\max}}^{n\max} \cdots \sum^{n\max} \\ & \times \prod_{\nu=1}^{l} \left\{\frac{(i\rho_\nu)^{\alpha_\nu+\beta_\nu}}{2^{\alpha_\nu+\beta_\nu}\alpha_\nu!\beta_\nu!} \exp[i\theta_\nu(\beta_\nu - \alpha_\nu)]\right\} \\ & \times \prod_{\mu=1}^{r} \left\{\frac{(iu_\mu)^{\gamma_\mu}}{\gamma_\mu!}\right\} m^{\beta_1,\ldots,\beta_l}_{\alpha_1,\ldots,\alpha_l,\gamma_1,\ldots,\gamma_r} \end{aligned} \quad (9)$$

with

$$\begin{aligned} m^{\beta_1,\ldots,\beta_l}_{\alpha_1,\ldots,\alpha_l,\gamma_1,\ldots,\gamma_r} &= \left\{\prod_{\nu=1}^{l} |f_{j\nu}|^{\alpha_\nu+\beta_\nu} \exp[i\delta_{j\nu}(\alpha_\nu - \beta_\nu)]\right\} \\ &\times \left\{\prod_{\mu=1}^{r} |f_{j\mu}|^{\gamma_\mu} \exp[i\delta_{j\mu}\gamma_\mu]\right\} \\ &\times \sum_{\substack{\alpha_{l+1},\beta_{l+1}=0 \\ \alpha_{l+1}+\beta_{l+1}=\gamma_1}}^{\gamma_1} \cdots \sum_{\substack{\alpha_{l+r},\beta_{l+r}=0 \\ \alpha_{l+r}+\beta_{l+r}=\gamma_r}}^{\gamma_r} \\ &\times \frac{\gamma_1! \ldots \gamma_r!}{\alpha_{l+1}!\beta_{l+1}! \ldots \alpha_r!\beta_r!} m^{\beta_1,\ldots,\beta_l,\beta_{l+1},\ldots,\beta_{l+r}}_{\alpha_1,\ldots,\alpha_l,\alpha_{l+1},\ldots,\alpha_{l+r}} \end{aligned} \quad (10)$$

and

$$\begin{aligned} m^{\beta_1,\ldots,\beta_{l+r}}_{\alpha_1,\ldots,\alpha_{l+r}} &= \sum_{\substack{\alpha_{11}=0 \\ \alpha_{11}+\cdots+\alpha_{1\tau_1}=\alpha_1}}^{\alpha_1} \cdots \sum_{\alpha_{1\tau_1}=0}^{\alpha_1} \cdots \sum_{\substack{\beta_{l1}=0 \\ \beta_{l1}+\cdots+\beta_{l\tau_{l+r}}=\beta_{l+r}}}^{\beta_{l+r}} \cdots \sum_{\beta_{l\tau_{l+r}}=0}^{\beta_{l+r}} \\ &\times \left[\prod_{\nu=1}^{l+r} \alpha_\nu!\beta_\nu! \Big/ \prod_{\nu=1}^{l+r}\prod_{s=1}^{\tau_\nu} \alpha_{\nu s}!\beta_{\nu s}!\right] \\ &\times \exp\left\{2\pi i\left[\sum_{\nu=1}^{l+r}\sum_{s=1}^{\tau_\nu} (\alpha_{\nu s} - \beta_{\nu s}) H_\nu T_s\right]\right\} \\ &\times \left\langle \exp\left\{2\pi i\left[\sum_{\nu=1}^{l+r}\sum_{s=1}^{\tau_\nu} (\alpha_{\nu s} - \beta_{\nu s}) H_\nu R_s\right] X_j\right\}\right\rangle_{r_j}. \end{aligned} \quad (11)$$

Note that for $\nu = l+1, \ldots, l+r$ half of the symmetry operations should be used, in accordance with (4)-(5).

The scattering-factor product in (9)-(11) does not change the moments calculation as defined in P&S. Hence the average in (11) can be executed as in P&S, assuming the atomic coordinates to be uniformly and independently distributed. As a consequence, the argument in P&S concerning the moments-cumulants transformation and the taking of the logarithm in (8) also carries over completely. Only the last step to the c.f., adding the contributions of the n p.r.v.'s in (8), leads now to a different result. The dependence on both the reflections and atom types leads to a sum of scattering factors, defined in (13), which may be different for each cumulant,

$$C = \exp\left\{\sum_{n\max=2}^{\infty} U_{n\max}[Z^{\beta_1,\ldots,\beta_l}_{\alpha_1,\ldots,\alpha_l,\gamma_1,\ldots,\gamma_r} K^{\beta_1,\ldots,\beta_l}_{\alpha_1,\ldots,\alpha_l,\gamma_1,\ldots,\gamma_r}]\right\} \quad (12)$$

with

$$\begin{aligned} Z^{\beta_1,\ldots,\beta_l}_{\alpha_1,\ldots,\alpha_l,\gamma_1,\ldots,\gamma_r} &= \sum_{j=1}^{n} \left(\left\{\prod_{\nu=1}^{l} |f_{j\nu}|^{\alpha_\nu+\beta_\nu} \exp[i\delta_{j\nu}(\alpha_\nu - \beta_\nu)]\right\}\right. \\ &\times \left.\left\{\prod_{\mu=1}^{r} |f_{j\mu}|^{\gamma_\mu} \exp[i\delta_{j\mu}\gamma_\mu]\right\}\right). \end{aligned} \quad (13)$$

The Taylor-series expansion terms are arranged such that $U_{n\max}$ contains only terms of order $N^{-(n\max/2-1)}$.

The final step to the j.p.d. is the Fourier transform of (12). In order to end up with an integrable expression (12) is split into an exponential containing all terms with $n\max = 2$ (order N^0) and an exponential consisting of all terms of at least order $N^{-0.5}$ ($n\max \geq 3$). The latter is expanded with a Taylor-series expansion and the resulting series rearranged once again according to $n\max$. In order to perform the integrations term by term most efficiently only the general term of the distribution needs to be identified and integrated. The general term of a j.p.d. is defined as the exponential containing all the $n\max = 2$ terms times a general product of all integration variables involved. Since the latter is always separable, the exponential defines the functional form of the integrated general term. After the integration of the general term the complete series expansion can be constructed simply from the separate terms. In a final step, this series expansion may be transformed back into exponential form using $x = \exp[\ln(x)]$. It has been shown for numerous cases that this transformation leads to identical distributions as if they were derived by hand under the same premises (Peschar, 1987).

Both AS and IR data influence the general term *via* $n\max = 2$ terms. It turns out to be convenient to treat them simultaneously with the following isomorphism model.

3. Isomorphic structures and isomorphic data sets

In spite of their physical difference, IR and AS play a similar practical role: both techniques increase data without increasing the number of unknowns appreciably. This suggests that isomorphism should be advantageous in treating both type of data simultaneously. Recently, Fortier & Nigam (1989) elucidated the similarity between distributions in the SIR and SAS cases by introducing the concept of isomorphous data sets. They separated the anomalous-dispersion data into two distinct data sets, the F_H and the F_{-H}, which were handled as being isomorphous.

As Fortier & Nigam acknowledge, intensity differences between isomorphous s.f.'s result from differences in scattering factors. This suggests a direct-space isomorphism as a suitable starting point. In this paper structures are considered to be isomorphous if the cell constants, space-group symmetry and atomic positions are identical. The only difference between isomorphic structures being the atomic scattering factors which are allowed to be complex-valued.

Hence, a s.f. G_H will be isomorphous with F_H if the s.f. expression is the same as for F_H in (2) except for the scattering factors,

$$G_H = \sum_{j=1}^{n} \xi_{jH} g_{jH}. \quad (14)$$

Consider now the s.f. F^*_{-H} which can be expressed as

$$F^*_{-H} = \sum_{j=1}^{n} \xi^*_{j\bar{H}} f^*_{j\bar{H}} = \sum_{j=1}^{n} \xi_{jH} f^*_{jH}. \quad (15)$$

Obviously, F^*_{-H} and F_H are isomorphously related. Moreover, a distribution derived for F^*_{-H} instantly yields the corresponding expression involving F_{-H} because $|F^*_{-H}| = |F_{-H}|$ and the phase of F^*_{-H} is minus the phase of F_{-H}.

Consequently, the j.p.d. derivation for isomorphous structures is equivalent to the derivation of a j.p.d. of isomorphous s.f.'s provided the F_{-H} are entered in either of the following ways:

(*a*) as F_{-H}, taking $-H$ in (11) and f_{jH} as scattering factors;

or

(*b*) as F^*_{-H}, taking H in (11) and f^*_{jH} as scattering factors.

4. The general term of the joint probability distribution of isomorphous structure factors in space group *P*1

The definition of the general term of the j.p.d. of an arbitrary (l) number of isomorphous s.f.'s in space group $P1$ requires the identification of the exponential contributions for which $n\max = 2$.

The following abbreviation is adopted,

$$F_n = F_{H_n}$$

with (16)

$$H_n = s_n H \quad \text{and} \quad s_n = \pm 1 \quad n \in (1, \ldots, l).$$

The r.v.'s for the phases φ_n and the magnitudes $|F_n|$ are referred to as Φ_n and R_n.

The average in (11) evaluated for (16) in space group $P1$ leads to the following condition for the non-zero terms with $n\max = 2$:

$$(\alpha_m - \beta_m) H_m + (\alpha_n - \beta_n) H_n = 0$$

for (17)

$$m, n \in \{1, \ldots, l\} \quad \text{and} \quad m \leq n.$$

The only contributors are

1. $\alpha_m = \alpha_n = 1$ or $\beta_m = \beta_n = 1$ if $H_m = -H_n$ with $m \neq n$;
2. $\alpha_m = \beta_n = 1$ or $\beta_m = \alpha_n = 1$ if $H_m = H_n$ with $m \neq n$;
3. $\alpha_n = \beta_n = 1$ for all H_n.

This reduces expression (13) to a simpler formula. For $m \le n$ with $m, n \in \{1, \ldots, l\}$;

$$z_{mn} = |z_{mn}| \exp[i\Delta_{mn}] = \sum_{j=1}^{N} |f_{jm}||f_{jn}| \exp[-i(\delta_{jm} + s_{mn}\delta_{jn})] \quad (18)$$

with

$$s_{mn} = \begin{cases} -1 & \text{if } H_m = H_n \\ +1 & \text{if } H_m = -H_n. \end{cases} \quad (19)$$

Expression (19) is equivalent to the condition $H_m + s_{mn}H_n = 0$. It can simply be shown that from three of these conditions, $H_i = s_i H$, $H_m = s_m H$ and $H_n = s_n H$, the following relations can be derived:

$$s_{im}s_{in} = -s_{mn} \quad \text{for } i, m, n \in \{1, \ldots, l\}$$

and (20)

$$s_i s_m = -s_{im} \quad \text{for } i, m \in \{1, \ldots, l\}.$$

The general term $\mathrm{GT}(H_1, \ldots, H_l)$ can now be expressed as

$$\mathrm{GT} = \frac{R_1 \ldots R_l}{(2\pi)^{2l}} \int_0^\infty \ldots \int_0^{2\pi} \rho_1 \ldots \rho_l \times \exp\Bigg[-\sum_{n=1}^{l} [i\rho_n R_n \cos(\theta_n - \Phi_n)] - \sum_{n=1}^{l} \tfrac{1}{4}\rho_n^2 z_{nn} - \sum_{\substack{m=1 \\ m<n}}^{l-1} \sum_{n=2}^{l} [\tfrac{1}{2}\rho_m\rho_n |z_{mn}| \cos(\theta_m + s_{mn}\theta_n + \Delta_{mn})] + \sum_{n=1}^{l} i\theta_n(\beta_n - \alpha_n) \Bigg] [\tfrac{1}{2}i\rho_1]^{\alpha_1+\beta_1} \ldots [\tfrac{1}{2}i\rho_l]^{\alpha_l+\beta_l} \times d\theta_1 \ldots d\theta_l \, d\rho_1 \ldots d\rho_l. \quad (21)$$

If $\alpha_1 = \beta_1 = \ldots = \alpha_l = \beta_l = 0$, the GT reduces to the j.p.d. of $R_1, \ldots, R_l, \Phi_1, \ldots, \Phi_l$ correct up to $0(N^0)$.

5. The integration of the general term

The general integration procedure of (21), executed in successive steps, will be discussed first (i). Subsequently, this scheme will be applied to the case of four isomorphous s.f.'s resulting in an expression for the general term (ii). Finally, a generalization to the case of l isomorphous s.f.'s is given (iii).

(i) *General integration procedure*

The integration of (21) can be executed in l successive steps, each step consisting of both a ρ and the corresponding θ integration. In each step the integral will be rearranged into the form

$$I = \tfrac{1}{4}\pi^{-2}[i/2]^{M'+M} \int_0^\infty \int_0^{2\pi} \rho^{M+M'+1} \times \exp\Bigg[-\tfrac{1}{4}\rho^2 - i\rho \sum_k c_k \cos(\theta + \zeta_k) + i\theta(M' - M) \Bigg] d\theta \, d\rho. \quad (22)$$

The c_k are in general complex-valued and dependent on the remaining integration variables.

With $c_k = a_k + ib_k$ (a_k and b_k real) and the functions X, Y defined as

$$X = \sum_k a_k \exp[i\zeta_k] + i \sum_k b_k \exp[i\zeta_k]$$

and (23)

$$Y = \sum_k a_k \exp[-i\zeta_k] + i \sum_k b_k \exp[-i\zeta_k],$$

(22) can be shown to result in (see Appendix I)

$$I = \pi^{-1} \exp[-XY] K_{M,M'}(X, Y)$$

with (24)

$$K_{M,M'}(X, Y) = M'! \sum_{k=0}^{M'} \begin{bmatrix} M \\ k \end{bmatrix} \frac{(-1)^k}{(M'-k)!} X^{M-k} Y^{M'-k}.$$

In each integration step m the following scheme is employed.

(*a*) A variable transformation is used in order to express the quadratic exponential term as shown in (22).

(*b*) All exponential terms which depend on $\cos(\theta_m)$ are collected which enables the definition of the functions X_m and Y_m as in (23).

(*c*) The identification of M and M' using the powers of ρ and $\exp(i\theta)$.

(*d*) The calculation of $X_m Y_m$ in (24) and the expansion of $K_{M,M'}$ using (I.18)–(I.19).

The functions X_m and Y_m to be defined in step m each consist of a sum of l terms, $X_m = x_{m1} + x_{m2} + \ldots + x_{ml}$ and $Y_m = y_{m1} + y_{m2} + \ldots + y_{ml}$ if l s.f.'s are involved.

The multinomial expansion of X_m and Y_m in (24) leads to a series in the terms x_{mn} and y_{mn}, the powers of which are denoted r_{mn} and t_{mn} respectively $[n \in (1, \ldots, l)]$.

This multinomial expansion and the dependence of X_m and Y_m on the integration variables leads to a nested sequence of series expansions. The summation limits M_m and M'_m in each step m are determined by previous expansions but determine themselves in turn in successive expansions. The nested series $\mathrm{NS}(H_1, \ldots, H_l)$ which is generated in this way can be expressed in a recursive form

$$\mathrm{NS}(H_1,\ldots,H_l)=K_{M_m,M'_m}[X_m, Y_m, K_{M_{m+1},M'_{m+1}}] \quad \text{for } m=1,\ldots,l \quad (25)$$

with

$$K_{M_m,M'_m}[X_m, Y_m, K_{M_{m+1},M'_{m+1}}] = M'_m! \sum_{k=0}^{M'_m} \begin{bmatrix} M_m \\ k \end{bmatrix} \frac{(-1)^k}{(M'_m-k)!}[X_m]^{M_m-k} \times [Y_m]^{M'_m-k} K_{M_{m+1},M'_{m+1}}$$

and

$$K_{M_{l+1},M'_{l+1}}=K_{0,0}=1.$$

(ii) *The calculation of the general term for four isomorphous s.f.'s*

The procedure described in (i) has been worked out in detail for four isomorphous s.f.'s as defined in (16). The integrations for ρ_m and θ_m are executed starting with $m=1$, 2, 3 and finally $m=4$. In each step additional variables are introduced in order to relieve the notational complexity. The variables are referred to *via* subscripts. A single subscript number denotes both the r.v. it is associated with and the step in which the definition of the variable takes place. Two subscripts, as in x_{mn}, for example, refer both to the r.v.'s and in addition the first (m) denotes the step in which the variable is defined. The following functions are used throughout all derivation steps:

$$a_{mn}=|a_{mn}|\exp(i\alpha_{mn}) \quad \text{for } m, n\in\{1,\ldots,l\} \quad (26)$$

$$\tau_{mn}=\begin{cases} t_{mn} & \text{if } s_{mn}=1 \\ r_{mn} & \text{if } s_{mn}=-1 \end{cases} \quad \text{with } m, n\in\{1,\ldots,l\}$$

and (27)

$$\tau'_{mn}=\begin{cases} t_{mn} & \text{if } s_{mn}=-1 \\ r_{mn} & \text{if } s_{mn}=+1 \end{cases} \quad \text{with } m, n\in\{1,\ldots,l\}$$

in which s_{mn} follows definition (19).

The four steps will now be discussed in some detail.

Step 1. Inspection of (20) suggests a variable transformation, applicable to all four ρ's,

$$y_n=\rho_n(z_{nn})^{1/2} \quad \text{for } n=1,\ldots,4, \quad (28)$$

which leads to the introduction of the following variables:

$$d_{mn}=z_{mn}(z_{mm})^{-1/2}(z_{nn})^{-1/2}=|d_{mn}|\exp(i\Delta_{mn}) \quad \text{for } m, n\in\{1,\ldots,4\} \text{ and } m\le n \quad (29)$$

$$E_n=R_n(z_{nn})^{-1/2} \quad \text{for } n=1,\ldots,4. \quad (30)$$

After collecting all terms in the exponential which depend on $\cos(\theta_1)$, X_1 is defined as [Y_1 can be simply constructed from X_1 with (23)]

$$X_1=G_1\exp(-i\Phi_1) -\tfrac{1}{2}i[y_2d_{12}\exp(is_{12}\theta_2)+y_3d_{13}\exp(is_{13}\theta_3) +y_4d_{14}\exp(is_{14}\theta_4)]. \quad (31)$$

For notational reasons $G_1\equiv E_1$ has been introduced. By definition, $C_{11}\equiv 1\cdot 0$. The powers of y_1 and $\exp(i\theta_1)$ define M and M' as

$$M_1=\alpha_1;\; M'_1=\beta_1. \quad (32)$$

Step 2. After collecting all y_2 terms in the exponential the following variable transformation leads to the required exponential quadratic expression:

$$u_2=y_2C_{22}^{-1} \quad \text{with } C_{22}=[1-|d_{12}|^2]^{-1/2}. \quad (33)$$

Subsequently, the $\cos(\theta_2)$-dependent terms in the exponential define the variables

$$a_{21}=C_{22}|d_{12}|\exp(is_{12}\Delta_{12})$$
$$a_{23}=C_{22}\{|d_{12}||d_{13}|\exp[is_{12}(\Delta_{12}-\Delta_{13})]-d_{23}\} \quad (34)$$
$$a_{24}=C_{22}\{|d_{12}||d_{14}|\exp[is_{12}(\Delta_{12}-\Delta_{14})]-d_{24}\}.$$

As a result X_2 becomes

$$X_2=-G_1a_{21}\exp(is_{12}\Phi_1)+G_2\exp(-i\Phi_2) +\tfrac{1}{2}i[y_3a_{23}\exp(is_{23}\theta_3)+y_4a_{24}\exp(is_{24}\theta_4)] \quad (35)$$

with $$G_2=C_{22}E_2. \quad (36)$$

Collection of the powers of u_2 and $\exp(i\theta_2)$ leads to

$$M_2=\alpha_2+\tau_{12};\; M'_2=\beta_2+\tau'_{12}. \quad (37)$$

In steps 3 and 4 the same procedure is followed as in step 2, resulting in the following definitions.

Step 3.

$$u_3=y_3C_{33}^{-1} \quad \text{with } C_{33}=[1-|d_{13}|^2-|a_{23}|^2]^{-1/2} \quad (38)$$

$$a_{31}=C_{33}\{|d_{13}|\exp(is_{13}\Delta_{13}) +|a_{23}||a_{21}|\exp[is_{23}(\alpha_{23}-\alpha_{21})]\}$$
$$a_{32}=C_{33}[|a_{23}|\exp(is_{23}\alpha_{23})]$$
$$a_{34}=C_{33}\{|d_{13}||d_{14}|\exp[is_{13}(\Delta_{13}-\Delta_{14})] +|a_{23}||a_{24}|\exp[is_{23}(\alpha_{23}-\alpha_{24})]-d_{34}\} \quad (39)$$

$$X_3=-G_1a_{31}\exp(is_{13}\Phi_1) +G_2a_{32}\exp(is_{23}\Phi_2)+G_3\exp(-i\Phi_3) +\tfrac{1}{2}iy_4a_{34}\exp(is_{34}\theta_4) \quad (40)$$

$$G_3=C_{33}E_3 \quad (41)$$

$$M_3=\alpha_3+\tau_{13}+\tau_{23};\; M'_3=\beta_3+\tau'_{13}+\tau'_{23}. \quad (42)$$

Step 4.

$$u_4 = y_4 C_{44}^{-1} \text{ with } C_{44} = [1 - |d_{14}|^2 - |a_{24}|^2 - |a_{34}|^2]^{-1/2} \quad (43)$$

$$\begin{aligned} a_{41} &= C_{44}\{|d_{14}| \exp(is_{14}\Delta_{14}) \\ &\quad + |a_{24}||a_{21}| \exp[is_{24}(\alpha_{24} - \alpha_{21})] \\ &\quad + |a_{34}||a_{31}| \exp[is_{34}(\alpha_{34} - \alpha_{31})]\} \\ a_{42} &= C_{44}\{|a_{24}| \exp(is_{24}\alpha_{24}) \\ &\quad + |a_{34}||a_{32}| \exp[is_{34}(\alpha_{34} - \alpha_{32})]\} \\ a_{43} &= C_{44}[|a_{34}| \exp(is_{34}\alpha_{34})] \end{aligned} \quad (44)$$

$$\begin{aligned} X_4 &= -G_1 a_{41} \exp(is_{14}\Phi_1) + G_2 a_{42} \exp(is_{24}\Phi_2) \\ &\quad + G_3 a_{43} \exp(is_{34}\Phi_3) + G_4 \exp(-i\Phi_4) \end{aligned} \quad (45)$$

$$G_4 = C_{44} E_4 \quad (46)$$

$$M_4 = \alpha_4 + \tau_{14} + \tau_{24} + \tau_{34};$$
$$M'_4 = \beta_4 + \tau'_{14} + \tau'_{24} + \tau'_{34}. \quad (47)$$

The following shorthand is introduced:

$$A_{mn} = a_{mn} C_{nn} \quad \text{for } m < n \quad \text{and} \quad m, n \in \{1, \ldots, 4\} \quad (48)$$

as well as the functions $L_{mn} = |L_{mn}| \exp(i\lambda_{mn})$ with $m \le n$. By definition, $L_{mn} \equiv L_{nm}$ with $m \le n$. For $m, n \in \{1, \ldots, 4\}$ the L_{mn} are specified as

$$\begin{aligned} |L_{11}| &= -\tfrac{1}{2}[1 + |a_{21}|^2 + |a_{31}|^2 + |a_{41}|^2]; \ \lambda_{11} = 0 \\ L_{12} &= |a_{21}| \exp(is_{12}\alpha_{21}) \\ &\quad + |a_{31}||a_{32}| \exp[is_{13}(\alpha_{31} - \alpha_{32})] \\ &\quad + |a_{41}||a_{42}| \exp[is_{14}(\alpha_{41} - \alpha_{42})] \\ L_{13} &= |a_{31}| \exp(is_{13}\alpha_{31}) \\ &\quad + |a_{41}||a_{43}| \exp[is_{14}(\alpha_{41} - \alpha_{43})] \\ L_{14} &= |a_{41}| \exp(is_{14}\alpha_{41}); \\ |L_{22}| &= -\tfrac{1}{2}[1 + |a_{32}|^2 + |a_{42}|^2]; \ \lambda_{22} = 0 \\ L_{23} &= -|a_{32}| \exp(is_{23}\alpha_{32}) \\ &\quad - |a_{42}||a_{43}| \exp[is_{24}(\alpha_{42} - \alpha_{43})] \\ L_{24} &= -|a_{42}| \exp(is_{24}\alpha_{42}); \\ |L_{33}| &= -\tfrac{1}{2}[1 + |a_{43}|^2]; \ \lambda_{33} = 0 \\ L_{34} &= -|a_{43}| \exp(is_{34}\alpha_{43}); \ |L_{44}| = -\tfrac{1}{2}; \ \lambda_{44} = 0. \end{aligned} \quad (49)$$

The integrated expression for $\mathrm{GT}(H_1, H_2, H_3, H_4)$ can now be noted:

$$\begin{aligned} \mathrm{GT} &= \pi^{-4}\left[\prod_{n=1}^{4} G_n(z_{nn})^{-1/2(\alpha_n+\beta_n+1)} C_{nn}^{(\alpha_n+\beta_n+1)}\right] \\ &\quad \times \exp\left[\sum_{\substack{m=1 \\ m \le n}}^{4} \sum_{n=1}^{4} 2G_m G_n |L_{mn}| \right. \\ &\quad \left. \times \cos(\Phi_m + s_{mn}\Phi_n + \lambda_{mn})\right] Q(H_1, \ldots, H_4). \end{aligned} \quad (50)$$

$Q(H_1, \ldots, H_4)$ in (50) is defined as in (25) (with $l = 4$) but with T_m instead of X_m and T_m^* instead of Y_m,

$$Q(H_1, \ldots, H_4) = K_{M_m, M'_m}[T_m, T_m^*, K_{M_{m+1}, M'_{m+1}}] \quad \text{for } m = 1, \ldots, 4$$

with

$$\begin{aligned} &K_{M_m, M'_m}[T_m, T_m^*, K_{M_{m+1}, M'_{m+1}}] \\ &\quad = M'_m! \sum_{k=0}^{M'_m} \begin{bmatrix} M_m \\ k \end{bmatrix} \frac{(-1)^k}{(M'_m - k)!} [T_m]^{M_m - k} \\ &\quad \times [T_m^*]^{M'_m - k} K_{M_{m+1}, M'_{m+1}} \\ &K_{M_{l+1}, M'_{l+1}} = K_{0,0} = 1 \end{aligned}$$

and

$$\begin{aligned} T_1 &= G_1 \exp(-i\Phi_1) - G_2 A_{12} \exp(is_{12}\Phi_2) \\ &\quad - G_3 A_{13} \exp(is_{13}\Phi_3) - G_4 A_{14} \exp(is_{14}\Phi_4) \\ T_2 &= -G_1 a_{21} \exp(is_{21}\Phi_1) + G_2 \exp(-i\Phi_2) \\ &\quad + G_3 A_{23} \exp(is_{23}\Phi_3) + G_4 A_{24} \exp(is_{24}\Phi_4) \\ T_3 &= -G_1 a_{31} \exp(is_{31}\Phi_1) + G_2 a_{32} \exp(is_{32}\Phi_2) \\ &\quad + G_3 \exp(-i\Phi_3) + G_4 A_{34} \exp(is_{34}\Phi_4) \\ T_4 &= -G_1 a_{41} \exp(is_{41}\Phi_1) + G_2 a_{42} \exp(is_{42}\Phi_2) \\ &\quad + G_3 a_{43} \exp(is_{43}\Phi_3) + G_4 \exp(-i\Phi_4). \end{aligned} \quad (51)$$

Comparison of the definitions of X_n and Y_n with T_n leads to the conclusion that the integrations change $\frac{1}{2} i y_n a_{mn}$ into $G_n A_{mn}$ and $\frac{1}{2} i y_n a_{mn} \exp(is_{mn}\theta_n)$ into $G_n a_{mn} \exp(is_{mn}\Phi_n)$ which comes down to changing X_n into T_n and Y_n into T_n^*.

(iii) *Generalization to l isomorphous s.f.'s*

A detailed analysis of the above definitions and additional calculations with more s.f.'s, not shown here for briefness, indicates a generalization to the case of l isomorphous s.f.'s. The variables defined in a particular integration step n (>1) turn out to be

Step n, $1 < n \le l$.

$$u_n = y_n C_{nn}^{-1} \quad \text{with } C_{nn} = \left[1 - |d_{1n}|^2 - \sum_{k=2}^{n-1} |a_{kn}|^2\right]^{-1/2} \quad (52)$$

$$G_n = E_n C_{nn} \tag{53}$$

$$a_{n1} = C_{nn}\Big\{|d_{1n}|\exp(is_{1n}\Delta_{1n}) + \sum_{k=2}^{n-1}|a_{kn}||a_{k1}|\exp[is_{kn}(\alpha_{kn}-\alpha_{k1})]\Big\}.$$

For $1<j<n$

$$a_{nj} = C_{nn}\Big\{|a_{jn}|\exp(is_{jn}\alpha_{jn}) + \sum_{k=j+1}^{n-1}|a_{kn}||a_{kj}|\exp[is_{kn}(\alpha_{kn}-\alpha_{kj})]\Big\}$$

and for $n<j\le l$

$$a_{nj} = C_{nn}\Big\{|d_{1n}||d_{1j}|\exp[is_{1n}(\Delta_{1n}-\Delta_{1j})] - d_{nj} + \sum_{k=2}^{n-1}|a_{kn}||a_{kj}|\exp[is_{kn}(\alpha_{kn}-\alpha_{kj})]\Big\} \tag{54}$$

$$X_n = -G_1 a_{n1}\exp(is_{1n}\Phi_1) + \sum_{k=2}^{n-1} G_k a_{nk}\exp(is_{kn}\Phi_k) + G_n\exp(-i\Phi_n) + \sum_{k=n+1}^{l}\tfrac{1}{2}i y_k a_{nk}\exp(is_{nk}\theta_k) \tag{55}$$

$$M_n = \alpha_n + \sum_{k=1}^{n-1}\tau_{kn};\; M'_n = \beta_n + \sum_{k=1}^{n-1}\tau'_{kn}. \tag{56}$$

The integrated expression for the general term $\mathrm{GT}(H_1,\ldots,H_l)$ becomes

$$\mathrm{GT} = \pi^{-l}\Big[\prod_{n=1}^{l} G_n(z_{nn})^{-1/2(\alpha_n+\beta_n+1)} C_{nn}^{(\alpha_n+\beta_n+1)}\Big] \times\exp\Big[\sum_{\substack{m=1\\ m\le n}}^{l}\sum_{n=1}^{l} 2G_mG_n|L_{mn}| \times\cos(\Phi_m + s_{mn}\Phi_n + \lambda_{mn})\Big] Q(H_1,\ldots,H_l). \tag{57}$$

$Q(H_1,\ldots,H_l)$ is defined as in (51) with

$$T_1 = G_1\exp[-i\Phi_1] - \sum_{k=2}^{l} A_{1n}G_k\exp[is_{1k}\Phi_k]$$

and

$$T_n = -G_1 a_{n1}\exp[is_{n1}\Phi_1] + \sum_{k=2}^{n-1} G_k a_{nk}\exp[is_{nk}\Phi_k] + G_n\exp[-i\Phi_n] + \sum_{k=n+1}^{l} G_k A_{nk}\exp[is_{nk}\Phi_k]. \tag{58}$$

The definitions of the L_{mn} functions are

$$|L_{nn}| = -\tfrac{1}{2}\Big[1 + \sum_{k=n+1}^{l}|a_{kn}|^2\Big] \quad\text{and}\quad \lambda_{nn}=0 \quad \text{for } n=1,\ldots,l$$

and for $1\le m<n\le l$ ($L_{nm}\equiv L_{mn}$)

$$L_{mn} = q_m\Big\{|a_{nm}|\exp(is_{mn}\alpha_{nm}) + \sum_{k=n+1}^{l}|a_{km}||a_{kn}|\exp[is_{mk}(\alpha_{km}-\alpha_{kn})]\Big\} \tag{59}$$

in which $q_m=+1$ if $m=1$ and $q_m=-1$ if $m\ne 1$.

6. The joint probability distribution and the conditional probability distribution for the two-phase invariants

The j.p.d. of the phases and magnitudes of l isomorphous s.f.'s correct up to $0(N^0)$ is directly obtained from (57) since up to this order $Q(H_1,\ldots,H_4)=1$. Hence,

$$P(R_1,\Phi_1,\ldots,R_l,\Phi_l) = C^{-1}\exp\Big[2\sum_{\substack{m=1\\ m\le n}}^{l}\sum_{n=1}^{l} G_mG_n|L_{mn}| \times\cos(\Phi_m + s_{mn}\Phi_n + \lambda_{mn})\Big] \tag{60}$$

with C^{-1} a normalization constant.

Expression (60) contains $l(l-1)/2$ two-phase s.i.'s,

$$\Psi_{mn} = \Phi_m + s_{mn}\Phi_n \quad\text{with } m<n\in\{1,\ldots,l\}. \tag{61}$$

The c.p.d. for each of the two-phase s.i.'s may be calculated from (60) in the usual way by fixing the magnitudes and integrating out the phases which do not take part in the two-phase s.i. This procedure does not lead to a convenient analytical expression but to a complicated summation.

Alternatively, a phase which forms no part of the invariant under investigation can be expressed *via* a marginal distribution in a phase which does form part of the invariant (*e.g.* Hauptman, 1982*a*; Peschar, 1987). From the marginal distribution of (61) it follows that

$$\langle\exp(i\Phi_m)\rangle = B_{mn}\exp[-i(s_{mn}\Phi_n + \lambda_{mn}g_{mn})] \quad\text{with } m,n\in\{1,\ldots,l\}$$

in which s_{mn} has been defined in (19), L_{mn} in (49)

$$B_{mn} = \frac{I_1[2G_mG_n|L_{mn}|]}{I_0[2G_mG_n|L_{mn}|]} \quad\text{for } m\ne n \text{ and } B_{mm}\equiv 1\cdot 0$$

and

$$g_{mn} = \begin{cases} 1\cdot 0 & \text{for } m\le n \\ s_{mn} & \text{for } m>n, \end{cases} \tag{62}$$

I_1 and I_0 are modified Bessel functions.

Each Φ_i can be expressed *via* (62) in both Φ's of a Ψ_{mn} ($i \neq m, i \neq n$) equally likely so a Ψ_{uv} with $u < v \in \{1, \ldots, l\}$ can be expressed as

$$P(\Psi_{uv} | R_1, \ldots, R_l) = C' \exp \Bigg[\sum_{\substack{m=1 \\ m \neq n \\ m \neq v; n \neq u}}^{l-1} \sum_{n=2}^{l} G_m G_n |L_{mn}| B_{mu} B_{nv} \times \cos(\Phi_u + s_{uv}\Phi_v + s_{um} g_{um} \lambda_{um} - s_{un} g_{vn} \lambda_{vn} - s_{um} g_{mn} \lambda_{mn}) \Bigg] \quad (63)$$

with C' a normalization constant.

7. The joint probability distribution of three sets of four isomorphous structure factors in space group *P*1

A j.p.d. involving non-isomorphous s.f.'s can be constructed in a simple way from the expression for the general term for isomorphous s.f.'s. In this paper the distribution for the triplet invariant will be considered with H and K linearly independent

$$\psi = \varphi_H + \varphi_K + \varphi_{-H-K}. \quad (64)$$

For each of the s.f.'s F_H, F_K and F_{-H-K}, four isomorphous alternatives are included which comes down to constructing a j.p.d. of 12 s.f.'s.

The following s.f. notations will be employed:

$$F_a = F_{H_a}, \qquad F_b = F_{H_b} \quad \text{and} \quad F_c = F_{H_c}$$

with

$$H_a = s_a H, \qquad H_b = s_b K$$

and

$$H_c = s_c(-H - K) \quad \{a, b, c\}$$

with $\{a, b, c\}$ meaning $a = 1, \ldots, 4$; $b = 5, \ldots, 8$ and $c = 9, \ldots, 12$ and

$$s_a = \pm 1 \quad \text{if } H_a = \pm H, \qquad s_b = \pm 1 \quad \text{if } H_b = \pm K,$$
$$s_c = \pm 1 \quad \text{if } H_c = \pm(-H - K)$$

or, equivalently,

$$F_n = F_{H_n} \quad \{n\} \quad (65)$$

with $\{n\}$ meaning $n = 1, \ldots, 12$ and H_n and s_n as given above.

The subscript a is used exclusively for the isomorphous F_H, b for the isomorphous F_K and c for the isomorphous F_{-H-K}.

The r.v.'s for the s.f. magnitude and phase are referred to as R_n and Φ_n with the same conventions $\{n\}$ and $\{a, b, c\}$.

The general term of the j.p.d. of $R_1, \ldots, R_{12}$ and $\Phi_1, \ldots, \Phi_{12}$ can be set up similar to (21). However, no combination of two reflections (H_a, H_b), (H_a, H_c) or (H_b, H_c) with $\{a, b, c\}$ satisfies (17). As a result, the integrations for $(H_1, \ldots, H_4)$ on one hand, those concerning $(H_5, \ldots, H_8)$ on the other hand and finally those for $(H_9, \ldots, H_{12})$ can be executed independently so the general term $\mathrm{GT}(H_1, \ldots, H_{12})$ becomes the product of three distinct general terms, one for each of the isomorphous sets

$$\mathrm{GT}(H_1, \ldots, H_{12}) = \mathrm{GT}(H_1, \ldots, H_4) \times \mathrm{GT}(H_5, \ldots, H_8)\, \mathrm{GT}(H_9, \ldots, H_{12}). \quad (66)$$

The j.p.d. correct up to $0(N^{-1/2})$ requires an expansion of the exponential including only terms for which $n\max = 3$. Hence $\exp(x) \simeq 1 + x$ with x all terms for which $n\max = 3$ suffices. From (11) the existence condition for these terms can be inferred to be

$$(\alpha_a - \beta_a)\mathbf{H}_a + (\alpha_b - \beta_b)\mathbf{H}_b + (\alpha_c - \beta_c)\mathbf{H}_c = \mathbf{0} \quad \{a, b, c\}$$

with (67)

$$\alpha_1 + \beta_1 + \alpha_2 + \beta_2 + \ldots + \alpha_{12} + \beta_{12} = 3.$$

Defining

$$S_a = \alpha_a - \beta_a, \quad S_b = \alpha_b - \beta_b$$

and (68)

$$S_c = \alpha_c - \beta_c \quad \{a, b, c\},$$

two complementary sets of values turn out to contribute.

Set I.

$$S_a s_a = S_b s_b = S_c s_c = 1 \quad (69)$$

and all other α's and β's zero $\{a, b, c\}$.

For given H_a, H_b and H_c the s_a, s_b and s_c are fixed so that S_a, S_b and S_c must be selected such that (69) and (70) hold. Obviously, 64 unique contributing (a, b, c) combinations exist in $\{a, b, c\}$.

Set II.

$$S_a s_a = S_b s_b = S_c s_c = -1 \quad \{a, b, c\}. \quad (70)$$

The α and β values of set II equal the β and α values, respectively, of set I.

The values of set I lead to the existence of 64 unique triplets

$$\psi_{abc} = S_a\varphi_a + S_b\varphi_b + S_c\varphi_c \quad \{a, b, c\}. \quad (71)$$

Those in set II correspond then with the $-\psi_{abc}$.

The $(\alpha_1, \beta_1, \ldots, \alpha_{12}, \beta_{12})$ combination of each of the 64 unique terms can be separated into three sets of eight summation indices $(\alpha_1, \ldots, \beta_4)$, $(\alpha_5, \ldots, \beta_8)$

and $(\alpha_9, \ldots, \beta_{12})$. The latter define the three (in general different) nested series expansions, $Q(H_1, \ldots, H_4)$, $Q(H_5, \ldots, H_8)$ and $Q(H_9, \ldots, H_{12})$, respectively. The complete series is the product of these three nested series. In addition, for each of the 64 triplets in set I a scattering-factor product is present which can be noted as

$$\begin{aligned} Z_{abc} &= |Z_{abc}| \exp(i\Delta_{abc})| \\ &= z_{aa}^{-\frac{1}{2}(\alpha_a+\beta_a)} z_{bb}^{-\frac{1}{2}(\alpha_b+\beta_b)} z_{cc}^{-\frac{1}{2}(\alpha_c+\beta_c)} \\ &\quad \times \sum_{j=1}^{N} (|f_{ja}|^{\alpha_a+\beta_a} |f_{jb}|^{\alpha_b+\beta_b} |f_{jc}|^{\alpha_c+\beta_c} \\ &\quad \times \exp\{-i[(\alpha_a-\beta_a)\delta_{ja} + (\alpha_b-\beta_b)\delta_{jb} \\ &\quad + (\alpha_c-\beta_c)\delta_{jc}]\}) \quad \{a, b, c\}. \end{aligned} \tag{72}$$

Z^*_{abc} is the corresponding term for a triplet from set II.

After transforming the series back to exponential form with $1+x \simeq \exp(x)$, the j.p.d. correct up to $0(N^{-1/2})$ can be expressed as

$$\begin{aligned} &P(\Phi_1, R_1, \ldots, \Phi_{12}, R_{12}) \\ &= \pi^{-12}\left[\prod_{n=1}^{12} G_n z_{nn}^{-1} C_{nn}\right] \\ &\times \exp\left\{2 \sum_{a=1}^{4} \sum_{\substack{a'=1 \\ a<a'}}^{4} [G_a G_{a'} |L_{aa'}| \right. \\ &\times \cos(\Phi_a + s_{aa'}\Phi_{a'} + \lambda_{aa})] \\ &+ 2 \sum_{b=5}^{8} \sum_{\substack{b'=5 \\ b<b'}}^{8} [G_b G_{b'} |L_{bb'}| \cos(\Phi_b + s_{bb'}\Phi_{b'} + \lambda_{bb})] \\ &+ 2 \sum_{c=9}^{12} \sum_{\substack{c'=9 \\ c\le c'}}^{12} [G_c G_{c'} |L_{cc'}| \cos(\Phi_c + s_{cc'}\Phi_{c'} + \lambda_{cc})] \\ &+ \sum_{a=1}^{4} \sum_{b=5}^{8} \sum_{c=9}^{12} C_{aa}^{(\alpha_a+\beta_a)} C_{bb}^{(\alpha_b+\beta_b)} C_{cc}^{(\alpha_c+\beta_c)} \\ &\times [Z_{abc} Q(H_1, \ldots, H_4) Q(H_5, \ldots, H_8) \\ &\times Q(H_9, \ldots, H_{12}) + Z^*_{abc} Q^*(H_1, \ldots, H_4) \\ &\left.\times Q^*(H_5, \ldots, H_8) Q^*(H_9, \ldots, H_{12})]\right\}. \end{aligned} \tag{73}$$

For most applications (73) will be cumbersome to use. More conveniently, the j.p.d. should be expressed directly in terms of the 64 triplets. For that purpose the product of the series $Q(H_1, \ldots, H_4)$, $Q(H_5, \ldots, H_8)$ and $Q(H_9, \ldots, H_{12})$ must be evaluated for each of the contributions in (73). This extremely time-consuming work has been carried out by a suitable computer program, taking approximately 15 s CPU on a Cyber 750. A manual analysis of the results shows that a contributor of set I with an (a, b, c) combination can be expressed concisely as

$$\begin{aligned} &Q(H_1, \ldots, H_4) Q(H_5, \ldots, H_8) Q(H_9, \ldots, H_{12}) \\ &= V_a V_b V_c. \end{aligned} \tag{74}$$

The function V_a is completely expressible in the variables of $H_1, \ldots, H_4$, V_b in the variables of $H_5, \ldots, H_8$ and V_c in those of $H_9, \ldots, H_{12}$:

$$V_a = \sum_{a'=1}^{4} G_{a'} e_{aa'} \exp[-is_{a'}\Phi_{a'}] \quad \text{for } a = 1, \ldots, 4 \tag{75}$$

with $s_{a'}$ as defined in (65).

If $H_{a'} = H$ for $a' = 1, \ldots, 4$ the functions $e_{aa'}$ with $e_{aa'} = |e_{aa'}| \exp(i\varepsilon_{aa'})$ $[a, a' \in \{1, \ldots, 4\}]$ are

$$\begin{aligned} e_{11} &= 1 + D_{12}a_{21} + D_{12}A_{23}a_{31} + D_{12}A_{23}A_{34}a_{41} \\ &\quad + D_{12}A_{24}a_{41} + D_{13}a_{31} + D_{13}A_{34}a_{41} + D_{14}a_{41} \\ e_{12} &= -(D_{12} + D_{12}A_{23}a_{32} + D_{12}A_{23}A_{34}a_{42} \\ &\quad + D_{12}A_{24}a_{42} + D_{13}a_{32} + D_{13}A_{34}a_{42} + D_{14}a_{42}) \\ e_{13} &= -(D_{12}A_{23} + D_{12}A_{23}A_{34}a_{43} + D_{12}A_{24}a_{43} \\ &\quad + D_{13} + D_{13}A_{34}a_{43} + D_{14}a_{43}) \\ e_{14} &= -(D_{12}A_{23}A_{34} + D_{12}A_{24} + D_{13}A_{34} + D_{14}) \\ e_{21} &= -(a_{21} + A_{23}a_{31} + A_{23}A_{34}a_{41} + A_{24}a_{41}) \\ e_{22} &= 1 + A_{23}a_{32} + A_{23}A_{34}a_{42} + A_{24}a_{42} \\ e_{23} &= A_{23} + A_{23}A_{34}a_{43} + A_{24}a_{43} \\ e_{24} &= A_{23}A_{34} + A_{24} \\ e_{31} &= -(a_{31} + A_{34}a_{41});\ e_{32} = a_{32} + A_{34}a_{42} \\ e_{33} &= 1 + A_{34}a_{43};\ e_{34} = A_{34} \\ e_{41} &= -a_{41};\ e_{42} = a_{42};\ e_{43} = a_{43};\ e_{44} = 1 \end{aligned} \tag{76}$$

in which the following shorthand has been introduced:

$$D_{nn'} = d_{nn'}C_{n'n'} \quad \text{for } n < n' \in \{1, \ldots, 4\}. \tag{77}$$

For each $H_n = -H$ the changes in the $e_{nn'}$ are:

$$\begin{aligned} &a_{nn'} \text{ are replaced by } a^*_{nn'} \text{ and } A_{nn'} \text{ by } A^*_{nn'} \\ &\qquad \text{with } n, n' \in \{1, \ldots, 4\}; \quad (78) \\ &d_{nn'} \text{ are replaced by } d^*_{nn'} \text{ and } D_{nn'} \text{ by } D^*_{nn'} \\ &\qquad \text{with } n, n' \in \{1, \ldots, 4\}; \end{aligned}$$

V_b, V_c, e_{bb} and e_{cc} are defined completely similarly to (75)-(77) with $b' = 5, \ldots, 8$ and $c' = 9, \ldots, 12$.

As a result, the terms in (73) of $0(N^{-1/2})$ become

$$\exp\left\{\sum_{a=1}^{4} \sum_{b=5}^{8} \sum_{c=9}^{12} C_{aa}C_{bb}C_{cc} \times [Z_{abc}V_aV_bV_c + Z^*_{abc}V^*_aV^*_bV^*_c]\right\} \tag{79}$$

with

$$V_aV_bV_c=\sum_{a'=1}^{4}\sum_{b'=5}^{8}\sum_{c'=9}^{12}G_{a'}G_{b'}G_{c'}e_{aa'}e_{bb'}e_{cc'}$$
$$\times\exp\left[-i(s_{a'}\Phi_{a'}+s_{b'}\Phi_{b'}+s_{c'}\Phi_{c'})\right].$$

Inspection of (79) leads to the conclusion that each triplet in set I contributes to all 64 Z_{abc} terms. Since the summations $\{a, b, c\}$ concerning the Z_{abc} terms and the summations $\{a', b', c'\}$ over the phases are interchangeable, a convenient rearrangement leads to a readily applicable expression for the j.p.d. of the twelve s.f.'s.

$$\begin{aligned}P(\Phi_1, R_1, \ldots, \Phi_{12}, R_{12})\\ =\pi^{-12}\left[\prod_{n=1}^{12}G_nz_{nn}^{-1}C_{nn}\right]\exp\Bigg[2\\ \times\sum_{\substack{a=1\\a\le a'}}^{4}\sum_{a'=1}^{4}\left[G_aG_{a'}|L_{aa'}|\cos\left(\Phi_a+s_{aa'}\Phi_{a'}+\lambda_{aa'}\right)\right]\\ +2\sum_{\substack{b=5\\b\le b'}}^{8}\sum_{b'=5}^{8}\left[G_bG_{b'}|L_{bb'}|\cos\left(\Phi_b+s_{bb'}\Phi_{b'}+\lambda_{bb'}\right)\right]\\ +2\sum_{\substack{c=9\\c\le c'}}^{12}\sum_{c'=9}^{12}\left[G_cG_{c'}|L_{cc'}|\cos\left(\Phi_c+s_{cc'}\Phi_{c'}+\lambda_{cc'}\right)\right]\\ +2\Bigg(\sum_{a'=1}^{4}\sum_{b'=5}^{8}\sum_{c'=9}^{12}G_{a'}G_{b'}G_{c'}\\ \times\Bigg\{\sum_{a=1}^{4}\sum_{b=5}^{8}\sum_{c=9}^{12}C_{aa}C_{bb}C_{cc}|e_{aa'}||e_{bb'}||e_{cc'}|\\ \times|Z_{abc}|\cos\left[s_{a'}\Phi_{a'}+s_{b'}\Phi_{b'}+s_{c'}\Phi_{c'}\right.\\ \left.-\varepsilon_{aa'}-\varepsilon_{bb'}-\varepsilon_{cc'}+\Delta_{abc}\right]\Bigg\}\Bigg)\Bigg].\end{aligned}\tag{80}$$

8. The joint probability distribution of triplets: a generalization to an arbitrary number of isomorphous structure factors

The results presented in the previous section can be generalized to the j.p.d. of triplets which exist amongst l isomorphous s.f.'s F_H, l isomorphous s.f.'s F_K and l isomorphous s.f.'s F_{-H-K} with l an arbitrary positive number. The total number of invariants amongst these s.f.'s up to $0(N^{-1/2})$ is $l\times l\times l/2$ triplets and $3\times l\times(l-1)/2$ two-phase invariants.

An analysis of the results presented so far leads readily to the conclusion that for this j.p.d. the definition of the functions $e_{nn'}$ with $n, n'\in\{1,\ldots,l\}$ is required. Only the case $H_n=H$ needs to be discussed since if $H_n=-H$ (77) can be applied.

Expression (76) shows that all $e_{nn'}$ are constructed solely from $A_{nn'}$, $a_{nn'}$ and $D_{nn'}$. An analysis of these terms leads to the following conclusion:

Each $e_{nn'}$ consist of the sum of all possible single terms $h_{nn'}$ and all possible products of the form $h_{nn_1}h_{n_1n_2}\ldots h_{n_pn}$ which comply with two criteria:

1.
$$h_{nn'}=\begin{cases}a_{nn'} & \text{if } 1<n'<n\\ A_{nn'} & \text{if } 1<n<n'\\ -D_{1n'} & \text{if } n=1 \text{ and } n'\neq 1\\ -a_{n1} & \text{if } 1<n\\ 1 & \text{if } n=n'=1;\end{cases}\tag{81}$$

2. the product $h_{nn_1}h_{n_1n_2}\ldots h_{n_pn}$, consisting of p terms $(2<p<l)$, has subscripts $n_1,\ldots,n_p$ such that

$$n<n_1<n_2<\ldots<n_p>n'.\tag{82}$$

9. The conditional probability distributions of the triplets

The c.p.d.'s of the three-phase s.i.'s can be calculated with the technique described in § 6. Each phase $\Phi_{a'}$, $\Phi_{b'}$ and $\Phi_{c'}$ with $\{a', b', c'\}$ can be expressed in the phases Φ_u, Φ_v and Φ_w respectively of the three-phase s.i. Ψ_{uvw} with $\{u, v, w\}$. With reference to (62) for $\Phi_{a'}$ two cases are possible:

$$\begin{aligned}&1.\ a'=u\text{: } \cos\left[s_{a'}\Phi_{a'}+\ldots\right]\\ &\quad\text{becomes } \cos\left[s_u\Phi_u+\ldots\right];\\ &2.\ a'\neq u\text{: } \cos\left[s_{a'}\Phi_{a'}+\ldots\right]\\ &\quad\text{becomes } B_{a'u}\cos\left(s_u\Phi_u-s_{a'}\lambda_{a'u}g_{a'u}\right);\end{aligned}\tag{83}$$

$\Phi_{b'}$ and $\Phi_{c'}$ are handled similarly.

The c.p.d. for $\Psi_{uvw}=s_u\Phi_u+s_v\Phi_v+s_w\Phi_w$ with $\{u, v, w\}$ then becomes

$$P(\Psi_{uvw}|R_1,\ldots,R_{12})=L^{-1}\exp\left[2|W_{uvw}|\cos\left(\Psi_{uvw}-\zeta_{uvw}\right)\right]$$

with

$$\begin{aligned}|W_{uvw}|\exp\left(-i\zeta_{uvw}\right)\\ =\sum_{a'=1}^{4}\sum_{b'=5}^{8}\sum_{c'=9}^{12}G_{a'}G_{b'}G_{c'}\\ \times\Bigg\{\sum_{a=1}^{4}\sum_{b=5}^{8}\sum_{c=9}^{12}C_{aa}C_{bb}C_{cc}\\ \times|e_{aa'}||e_{bb'}||e_{cc'}||Z_{abc}|B_{a'u}B_{b'v}B_{c'w}\\ \times\exp\left[-i(\varepsilon_{aa'}+\varepsilon_{bb'}+\varepsilon_{cc'}-\Delta_{abc}\right.\\ \left.+s_{a'}\lambda_{a'u}g_{a'u}+s_{b'}\lambda_{b'v}g_{b'v}+s_{c'}\lambda_{c'w}g_{c'w})\right]\Bigg\}.\end{aligned}\tag{84}$$

10. Concluding remarks

In this paper a j.p.d. theory has been developed for three data sets $\{F_H\}$, $\{F_K\}$ and $\{F_{-H-K}\}$, each consisting of four isomorphous structure factors, by means of which the estimation of two-phase invariants and three-phase invariants present amongst them can be achieved. It has been shown that anomalous data $\{F_H\}$ and $\{F_{-H}\}$ can be considered as two isomorphous data sets, each corresponding with different isomorphous structures. As a result, anomalous-scattering and isomorphous-replacement data can be handled simultaneously. Indeed, the SAS formulae of Hauptman (1982*b*) and Giacovazzo (1983*a*) and the SIR expression of Giacovazzo, Cascarano & Zheng Chao-de (1988) are encompassed in (84).

So far, encouraging test results have been obtained for ideal data with the SIR expressions (Hauptman, Potter & Weeks, 1982) and the SAS formulae (Hauptman, 1982*b*; Giacovazzo, 1983*a*), using solely the diffraction data and the number and type of (anomalous) scatterers. It is expected that the probabilistic fusion of both techniques, as presented in this paper, may enforce the strength of direct methods appreciably. Currently procedures are being developed in order to test the quality of prediction of the new joint probability distribution.

The derivation procedure described so far requires no prior information except the quantity and type of anomalous scatterers. This may be advantageous because there is no need to solve the heavy-atom substructure first. On the other hand, it has been shown by various authors that heavy-atom phase information may improve the quality of prediction of the SIR and SAS expressions. The absolute values of the two-phase invariants can be calculated from the heavy-atom positions, leaving only their signs to be determined. Once the latter are available, the calculated two-phase estimates can be used as an alternative to (62) in getting three-phase-invariant estimates. The problem of determining these signs has been tackled in several ways. Fortier, Fraser & Moore (1986) and Fortier, Moore & Fraser (1985) advocated an analysis of the three-phase-invariant values for the eight possible sign combinations. In spite of the clustering around a few values, the sign ambiguity could not be solved uniquely. Hao Quan & Fan Hai-fu (1988) employed probabilistic sign estimates which rely on the assumption that the triplets of the heavy-atom substructure are equal to zero.

Although the above-described methods to incorporate additional structural knowledge may be combined with the newly derived j.p.d., a more appropriate method seems to be possible. Knowledge concerning the atomic positions, *e.g.* the heavy-atom substructure, directly affects the allowable distributions for the p.r.v.'s. Introduction of this knowledge in the derivation of the joint probability distribution may turn out to be more advantageous than its introduction afterwards. A paper on this subject is in preparation.

APPENDIX I

Consider the integral

$$I=\tfrac{1}{4}\pi^{-2}(i/2)^{M'+M}\int\limits_0^\infty\int\limits_0^{2\pi}\rho^{M+M'+1}\times\exp\left[-\tfrac{1}{4}\rho^2-i\rho\sum_k c_k\cos(\theta+\zeta_k)+i\theta(M'-M)\right]\mathrm{d}\theta\,\mathrm{d}\rho \quad (\mathrm{I}.1)$$

in which the c_k are complex-valued and M' and M non-negative integers.

Expressing $c_k=a_k+ib_k$ (a_k and b_k real) and defining

$$A\exp(i\alpha)=\sum_k a_k\exp(i\zeta_k)$$

and (I.2)

$$B\exp(i\beta)=\sum_k b_k\exp(i\zeta_k),$$

one gets

$$\sum_k c_k\cos(\theta+\zeta_k)=A\cos(\theta+\alpha)+iB\cos(\theta+\beta). \quad (\mathrm{I}.3)$$

If one invokes the expansions in normal and modified Bessel functions

$$\exp[-i\rho A\cos(\theta+\alpha)]=\sum_{m_1=-\infty}^{\infty}i^{m_1}J_{m_1}(-\rho A)\exp[im_1(\theta+\alpha)] \quad (\mathrm{I}.4)$$

and

$$\exp[\rho B\cos(\theta+\beta)]=\sum_{m_2=-\infty}^{\infty}I_{m_2}(\rho B)\exp[im_2(\theta+\beta)], \quad (\mathrm{I}.5)$$

the θ integration in (I.1) becomes

$$\int\limits_0^{2\pi}\exp[i\theta(M'-M+m_1+m_2)]\,\mathrm{d}\theta=\begin{cases}2\pi & \text{for } m_2+m_1+M'-M=0\\ 0 & \text{for } m_2+m_1+M'-M\neq 0.\end{cases} \quad (\mathrm{I}.6)$$

With the help of the relations amongst Bessel functions,

$$J_n(x)=(-i)^nI_n(ix) \quad (\mathrm{I}.7)$$

and

$$J_{-n}(x)=(-1)^nJ_n(x), \quad (\mathrm{I}.8)$$

and employment of (I.2)-(I.6), integral (I.1) reduces to

$$I=\frac{(-1)^M}{\pi 2^{M+M'+1}}\exp[i(M-M')\beta]$$
$$\times\int_0^\infty\sum_{m_1=-\infty}^{\infty}J_{m_1}(-\rho A)J_{m_1+M'-M}(i\rho B)$$
$$\times\exp[im_1(\alpha-\beta)]\rho^{M'+M+1}\exp[-\tfrac{1}{4}\rho^2]\,d\rho. \quad (I.9)$$

Graf's summation theorem for Bessel functions (Watson, 1952, p. 394) and (I.8) simplifies (I.9) to

$$I=\frac{(-1)^{M'}}{\pi 2^{M+M'+1}}\exp[i(M-M')(\beta-\gamma)]$$
$$\times\int_0^\infty J_{M'-M}(\rho C)\rho^{M'+M+1}\exp[-\tfrac{1}{4}\rho^2]\,d\rho \quad (I.10)$$

with

$$C^2=A^2-B^2+2iAB\cos(\alpha-\beta)$$

and (I.11)

$$\exp(2i\gamma)=\{iB+A\exp[-i(\alpha-\beta)]\}$$
$$\times\{iB+A\exp[i(\alpha-\beta)]\}^{-1}.$$

With the definition

$$X=A\exp(i\alpha)+iB\exp(i\beta)$$

and (I.12)

$$Y=A\exp(-i\alpha)+iB\exp(-i\beta),$$

(I.11) becomes

$$C^2=XY$$

with (I.13)

$$\exp(i2\gamma)=Y\exp(i\beta)[X\exp(-i\beta)]^{-1}.$$

The ρ integration in (I.10) can be shown to result in (Naya *et al.*, 1965)

$$I=[(-1)^{M'}M'!/\pi]$$
$$\times\exp(-XY)\exp[i(M-M')(\beta-\gamma)]$$
$$\times L_{M'}^{M-M'}(XY)(XY)^{(M-M')/2} \quad (I.14)$$

with $L_{M'}^{M-M'}$ the associated Laguerre polynomial.

From (I.13) and $(XY)=[X\exp(i\beta)\,Y\exp(-i\beta)]$ it is easily shown that

$$\exp[i(M-M')(\beta-\gamma)](XY)^{(M-M')/2}=X^{M-M'}. \quad (I.15)$$

By incorporation of the formula for the associated Laguerre polynomial

$$L_M^{M-M'}(x)=\sum_{k=0}^{M'}\begin{bmatrix}M\\k\end{bmatrix}\frac{(-x)^{M'-k}}{(M'-k)!}, \quad (I.16)$$

the final expression for (I.1) becomes

$$I=\pi^{-1}\exp[-XY]K_{M,M'}(X,Y) \quad (I.17)$$

with

$$K_{M,M'}(X,Y)=M'!\sum_{k=0}^{M'}\begin{bmatrix}M\\k\end{bmatrix}\frac{(-1)^k}{(M'-k)!}X^{M-k}Y^{M'-k}. \quad (I.18)$$

If X and Y each consist of a sum of p terms ($p>0$),

$$X=x_1+x_2+\ldots+x_p,\quad Y=y_1+y_2+\ldots+y_p \quad p>0,$$

each term in (I.18) can be expressed as an eightfold summation

$$X^{M-k}Y^{M'-k}=(M-k)!(M'-k)!$$
$$\times\sum_{\substack{r_1,\ldots,r_4=0\\ r_1+\ldots+r_4=M-k}}^{M-k}\frac{x_1^{r_1}\ldots x_p^{r_p}}{r_1!\ldots r_p!}$$
$$\times\sum_{\substack{t_1,\ldots,t_4=0\\ t_1+\ldots+t_4=M'-k}}^{M'-k}\frac{y_1^{t_1}\ldots y_p^{t_p}}{t_1!\ldots t_p!}. \quad (I.19)$$

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