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A statistical interpretation of the triplet and quartet invariant in $P1$. A theoretical discussion

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We present a method that we call symbolic asymptotic development (SAD) to obtain joint probability distributions (j.p.d.'s) of phases of structure factors for general even densities of the atomic position vectors. The formula for the triplet and quartet invariant that we obtain in this way reduces to the well known classical formula for the case of a uniform density of the atomic position vectors. For the case of complete knowledge of the atomic vectors it reduces to first order to the exact probability density of the triplet (quartet) phase invariant. Applying this formula to the most general j.p.d. of the atomic vectors we obtain a statistical interpretation of Hauptman's algebraic $B_{3,0}$ and $B_{4,0}$ formulas. We also give a heuristic derivation of the SAD method. Another method that we shall discuss uses a method called linearization of the invariants that also produces formulas for the triplet phase invariant. This method is based on previous work and is also more laborious to calculate with than the SAD method. It can also give a statistical interpretation of the $B_{3,0}$ formula. We show that the formula obtained for the triplet resembles the formula obtained with SAD.

1. Introduction

Ever since the introduction of the $B_{n,0}$ formulas (see *e.g.* Karle & Hauptman, 1957) researchers have tried to establish a statistical interpretation of these algebraic formulas (see *e.g.* Brosius, 1989*b*). In this paper we derive statistical formulas for the (algebraic) $B_{3,0}$ and $B_{4,0}$ formulas. More specifically, we obtain a probabilistic formula for our $B'_{3,0}$ and $B'_{4,0}$ formulas that are equal to the former apart from a quotient factor.

In order to derive this $B'_{3,0}$ (respectively $B'_{4,0}$) formula we need to use the most general joint probability distribution (j.p.d.) of the atomic vector positions that can be obtained without additional chemical knowledge. However, this j.p.d. of the atomic vectors is not a uniform one. So we also need a method for obtaining j.p.d.'s of phases of structure factors when using a nonuniform density of the atomic position vectors. This method is called symbolic asymptotic development (SAD).

Another way to obtain statistical formulas for phase invariants is obtained by using linearized invariants. This method is based on previous work (Brosius, 1989*b*).

All formulas that we shall derive differ from the well known triplet and quartet formulas (see *e.g.* Cochran, 1955; Klug, 1958; Hauptman, 1976; Giacovazzo, 1976) obtained with a uniform density of the atomic position vectors.

In both cases we consider the space group $P1$ and a structure consisting of N *equal* (this restriction is not necessary for our derivation and is only for convenience) atoms with position vectors $\mathbf{r}_1, \ldots, \mathbf{r}_N$. The strucure factors are then given by

$$E_{\mathbf{h}} = (1/N^{1/2}) \sum_{j=1}^{N} \exp(2\pi i \mathbf{h} \cdot \mathbf{r}_j). \quad (1)$$

We also consider N random vector positions $\mathbf{x}_1, \ldots, \mathbf{x}_N$ with a general j.p.d. $f(\mathbf{x}_1, \ldots, \mathbf{x}_N)$ where every $\mathbf{x}_i$ is a random vector modeling the position vector $\mathbf{r}_i$. In this paper we use a j.p.d. $f(\mathbf{x}_1, \ldots, \mathbf{x}_N)$ that is also *even*,

$$f(\mathbf{x}_1, \ldots, \mathbf{x}_N) = f(-\mathbf{x}_1, \ldots, -\mathbf{x}_N), \quad (2)$$

and translation invariant, $f(\mathbf{x}_1 + \mathbf{a}, \ldots, \mathbf{x}_N + \mathbf{a}) = f(\mathbf{x}_1, \ldots, \mathbf{x}_N)$. Finally, we consider the random variables (r.v.'s)

$$\hat{E}_{\mathbf{h}} \equiv \hat{E}_{\mathbf{h}}(\mathbf{x}_1, \ldots, \mathbf{x}_N) = (1/N^{1/2}) \sum_{j=1}^{N} \exp(2\pi i \mathbf{h} \cdot \mathbf{x}_j). \quad (3)$$

We shall always use (to avoid possible confusion) a circumflex (ˆ) to denote an r.v. depending on the $\mathbf{x}_i$. We shall also use the traditional notation $R_{\mathbf{h}}$ to denote the absolute value of the structure factor $E_{\mathbf{h}}$, and $\hat{R}_{\mathbf{h}}$ is defined in the same way as the absolute value of $\hat{E}_{\mathbf{h}}$. Similarly $\varphi_{\mathbf{h}}$ shall denote the phase of $E_{\mathbf{h}}$ and $\hat{\varphi}_{\mathbf{h}}$ shall denote the random phase variable of $\hat{E}_{\mathbf{h}}$. For every r.v. $\hat{Z}(\mathbf{x}_1, \ldots, \mathbf{x}_N)$ of the $\mathbf{x}_i$ we define the average with respect to the j.p.d. $f(\mathbf{x}_1, \ldots, \mathbf{x}_N)$ by

$$\langle \hat{Z}(\mathbf{x}_1, \ldots, \mathbf{x}_N) \rangle_f \equiv \int \mathrm{d}\mathbf{x}_1 \ldots \mathrm{d}\mathbf{x}_N \, f(\mathbf{x}_1, \ldots, \mathbf{x}_N) \hat{Z}(\mathbf{x}_1, \ldots, \mathbf{x}_N) \quad (4)$$

or simply $\langle \hat{Z}(\mathbf{x}_1, \ldots, \mathbf{x}_N) \rangle$ if there is no risk of confusion. We also use the same symbol f to denote all marginal j.p.d.'s of the $\mathbf{x}_i$. For instance, we write

$$f(\mathbf{x}_1, \mathbf{x}_2) \equiv \int \mathrm{d}\mathbf{x}_3 \ldots \mathrm{d}\mathbf{x}_N \, f(\mathbf{x}_1, \ldots, \mathbf{x}_N). \quad (5)$$

2. The SAD method

We shall illustrate the SAD method for the calculation of the j.p.d. of the triplet phase invariant given its first neighborhood. Define $\varphi = \varphi_{\mathbf{h}} + \varphi_{\mathbf{k}} - \varphi_{\mathbf{h+k}}$. We suppose now that $h(\mathbf{x}_1, \ldots, \mathbf{x}_N)$ is the j.p.d. of the $\mathbf{x}_i$ *given* the values $\mathbf{x}_i - \mathbf{x}_j = \mathbf{r}_i - \mathbf{r}_j$, that is the whole structure is known apart from a translation. Then h is even and translation invariant. Let $P_h(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}})$ be the j.p.d. of $\hat{\varphi}_{\mathbf{h}}, \hat{\varphi}_{\mathbf{k}}, \hat{\varphi}_{\mathbf{h+k}}$ with respect to this density $h(\mathbf{x}_1, \ldots, \mathbf{x}_N)$. Since $h(\mathbf{x}_1, \ldots, \mathbf{x}_N)$ is even and translation invariant, $P_h(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}})$ is necessarily of the form

$$P_h(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}}) = \tfrac{1}{2}\delta(\bar{\varphi} - \varphi) + \tfrac{1}{2}\delta(\bar{\varphi} + \varphi), \tag{6}$$

where $\bar{\varphi} = \bar{\varphi}_{\mathbf{h}} + \bar{\varphi}_{\mathbf{k}} - \bar{\varphi}_{\mathbf{h+k}}$ and δ is a delta function (actually a periodic delta function). Since δ is here a periodic delta function we can write

$$\tfrac{1}{2}\delta(\bar{\varphi} - \varphi) + \tfrac{1}{2}\delta(\bar{\varphi} + \varphi) = 1 + \sum_{n\neq 0} 2a_n \cos(n\bar{\varphi}) + \sum_{n\neq 0} 2b_n \sin(n\bar{\varphi}). \tag{7}$$

Then $b_n = 0$ since $P_h(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}})$ is even in $\bar{\varphi}$ and

$$a_n = \textstyle\int P_h(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}}) \cos(n\bar{\varphi})\,\mathrm{d}\bar{\varphi} \equiv \langle \cos(n\hat{\varphi})\rangle_h = \cos(n\varphi), \tag{8}$$

where $\hat{\varphi} = \hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}}$. We now look for a simple algebraic relation between $\hat{E}_{\mathbf{h}}$, $\hat{E}_{\mathbf{k}}$, $\hat{E}_{\mathbf{h+k}}$ and $\langle\cos\hat{\varphi}\rangle_h$. Since $\langle \hat{R}_{\mathbf{h}}\hat{R}_{\mathbf{k}}\hat{R}_{\mathbf{h+k}} \cos(\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}})\rangle_h = R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}\cos\varphi$ and since $\langle \hat{R}^2_{\mathbf{h}}\rangle^{1/2}_h = R_{\mathbf{h}}$ *etc.*, we can write

$$\begin{aligned} P_h&(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}}) \\ &= 1 + 2\sum_{n\neq 0} \cos(n\bar{\varphi})\cos(n\varphi) \\ &\simeq 1 + 2\cos\bar{\varphi}\cos\varphi \text{ (to first order)} \\ &\simeq 1 + 2\cos\bar{\varphi}\langle\cos\hat{\varphi}\rangle_h \\ &\simeq 1 + 2\frac{R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}\langle \hat{R}_{\mathbf{h}}\hat{R}_{\mathbf{k}}\hat{R}_{\mathbf{h+k}}\cos(\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}})\rangle_h}{\langle\hat{R}^2_{\mathbf{h}}\rangle_h\langle\hat{R}^2_{\mathbf{k}}\rangle_h\langle\hat{R}^2_{\mathbf{h+k}}\rangle_h}\cos\bar{\varphi}. \end{aligned} \tag{9}$$

This equation reduces to the Cochran formula when we take $h(\mathbf{x}_1, \ldots, \mathbf{x}_N) \equiv 1$, since then $\langle\hat{R}^2_{\mathbf{h}}\rangle_h = 1, \ldots,$ $\langle \hat{R}_{\mathbf{h}}\hat{R}_{\mathbf{k}}\hat{R}_{\mathbf{h+k}}\cos(\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}})\rangle_h = 1/N^{1/2}$, and we then obtain [after inserting these formulas back into equation (9)]

$$\begin{aligned} P_h(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}}) &\simeq 1 + 2\frac{R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+h}}}{N^{1/2}}\cos\bar{\varphi} \\ &\propto \exp\left(2\frac{R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}}{N^{1/2}}\cos\bar{\varphi}\right). \end{aligned} \tag{10}$$

Let us now replace h in equation (9) by an arbitrary j.p.d. $f = f(\mathbf{x}_1, \ldots, \mathbf{x}_N)$ that is even and translation invariant. Then equation (9) becomes (to first order)

$$\begin{aligned} P_f&(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}}) \\ &\simeq 1 + 2\frac{R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}\langle \hat{R}_{\mathbf{h}}\hat{R}_{\mathbf{k}}\hat{R}_{\mathbf{h+k}}\cos(\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}})\rangle_f}{\langle\hat{R}^2_{\mathbf{h}}\rangle_f\langle\hat{R}^2_{\mathbf{k}}\rangle_f\langle\hat{R}^2_{\mathbf{h+k}}\rangle_f}\cos\bar{\varphi} \\ &\propto \exp\left(2\frac{R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}\langle \hat{R}_{\mathbf{h}}\hat{R}_{\mathbf{k}}\hat{R}_{\mathbf{h+k}}\cos(\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}})\rangle_f}{\langle\hat{R}^2_{\mathbf{h}}\rangle_f\langle\hat{R}^2_{\mathbf{k}}\rangle_f\langle\hat{R}^2_{\mathbf{h+k}}\rangle_f}\cos\bar{\varphi}\right). \end{aligned} \tag{11}$$

We would now like to have a consistent method for calculating formula (11). If we have such a method, then we could also calculate conditional probabilities of phases given *e.g.* the second neighborhood of $\hat{E}_{\mathbf{h}}, \hat{E}_{\mathbf{k}}, \hat{E}_{\mathbf{h+k}}$. Before we continue, note that equation (11) resembles the Von Mises distribution introduced by Heinerman *et al.* (1977) to calculate the triplet phase distribution given additional chemical information.

We shall now consider the three structure factors $E_{\mathbf{h}}$, $E_{\mathbf{k}}$, $E_{\mathbf{h+k}}$. Let us calculate the j.p.d. $P_f(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}})$ of $\hat{\varphi}_{\mathbf{h}}$, $\hat{\varphi}_{\mathbf{k}}$, $\hat{\varphi}_{\mathbf{h+k}}$ with respect to an arbitrary even and translation-invariant j.p.d. $f = f(\mathbf{x}_1, \ldots, \mathbf{x}_N)$. Then the j.p.d. $P_f(\bar{R}_{\mathbf{h}}, \bar{R}_{\mathbf{k}}, \bar{R}_{\mathbf{h+k}}, \bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}})$ of $\hat{E}_{\mathbf{h}}, \hat{E}_{\mathbf{k}}, \hat{E}_{\mathbf{h+k}}$ is given by the formula

$$\begin{aligned} P_f&(\bar{R}_{\mathbf{h}}, \bar{R}_{\mathbf{k}}, \bar{R}_{\mathbf{h+k}}, \bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}}) \\ &= \frac{\bar{R}_{\mathbf{h}}\bar{R}_{\mathbf{k}}\bar{R}_{\mathbf{h+k}}}{(2\pi)^6}\int_0^{2\pi}\mathrm{d}\theta_{\mathbf{h}}\ldots\int_0^{\infty}\rho_{\mathbf{h}}\,\mathrm{d}\rho_{\mathbf{h}}\ldots\exp[-i\rho_{\mathbf{h}}\bar{R}_{\mathbf{h}} \\ &\quad\times\cos(\bar{\varphi}_{\mathbf{h}} - \theta_{\mathbf{h}}) - \ldots]\phi_f(\rho_{\mathbf{h}}, \ldots, \theta_{\mathbf{h+k}}), \end{aligned} \tag{12}$$

where the characteristic function $\phi_f(\rho_{\mathbf{h}}, \ldots, \theta_{\mathbf{h+k}})$ is defined by

$$\begin{aligned} \phi_f&(\rho_{\mathbf{h}}, \ldots, \theta_{\mathbf{h+k}}) \\ &= \textstyle\int f(\mathbf{x}_1, \ldots, \mathbf{x}_N)\exp[i\rho_{\mathbf{h}}\hat{R}_{\mathbf{h}}\cos(\hat{\varphi}_{\mathbf{h}} - \theta_{\mathbf{h}}) + \ldots]\,\mathrm{d}\mathbf{x}_1\ldots\mathrm{d}\mathbf{x}_N. \end{aligned} \tag{13}$$

We shall also write

$$\begin{aligned} \phi_f(\rho_{\mathbf{h}}, \ldots, \theta_{\mathbf{h+k}}) = \langle &\exp[i\rho_{\mathbf{h}}\hat{R}_{\mathbf{h}}\cos(\hat{\varphi}_{\mathbf{h}} - \theta_{\mathbf{h}}) + \ldots \\ &+ i\rho_{\mathbf{h+k}}\hat{R}_{\mathbf{h+k}}\cos(\hat{\varphi}_{\mathbf{h+k}} - \theta_{\mathbf{h+k}})]\rangle_f. \end{aligned} \tag{14}$$

In order to calculate equation (14), we first remark that we cannot calculate expressions of the form $\langle J_k(\rho_{\mathbf{h}}\hat{R}_{\mathbf{h}}) \times \cos[k(\hat{\varphi}_{\mathbf{h}} - \theta_{\mathbf{h}})] \times \ldots \times J_s(\rho_{\mathbf{h+k}}\hat{R}_{\mathbf{h+k}})\cos(\hat{\varphi}_{\mathbf{h+k}} - \theta_{\mathbf{h+k}})\rangle_f$; we can only calculate polynomial expressions like $\langle \rho_{\mathbf{h}}\hat{R}^3_{\mathbf{h}}\cos(\hat{\varphi}_{\mathbf{h}} - \theta_{\mathbf{h}}) \times \ldots \times \rho_{\mathbf{h+k}}\hat{R}_{\mathbf{h+k}}\cos(\hat{\varphi}_{\mathbf{h+k}} - \theta_{\mathbf{h+k}})\rangle_f$. We then also have to group these forms in $\phi_f(\rho_{\mathbf{h}}, \ldots, \theta_{\mathbf{h+k}})$ from lowest order first to highest order last.

To do this we write

$$\begin{aligned} \phi_f(\rho_{\mathbf{h}}, \ldots, \theta_{\mathbf{h+k}}) = \langle &\exp[i(\rho_{\mathbf{h}}/\mu^{1/2})\hat{R}_{\mathbf{h}}\cos(\hat{\varphi}_{\mathbf{h}} - \theta_{\mathbf{h}}) + \ldots \\ &+ i(\rho_{\mathbf{h+k}}/\mu^{1/2})\hat{R}_{\mathbf{h+k}}\cos(\hat{\varphi}_{\mathbf{h+k}} - \theta_{\mathbf{h+k}})]\rangle^{\mu}_f, \end{aligned} \tag{15}$$

which is nothing other than equation (14) when $\mu = 1$. The heuristic derivation leading to equation (15) will be given in §3.

We now develop equation (15) asymptotically in μ as if μ were a large number. This way we have a prescription for how to group the terms in equation (15) from lowest order first to highest order last. We call this way of writing equation (15) as a series of terms symbolic asymptotic development (SAD). After all the calculations in equation (15) and equation (12) are done, we simply replace μ by 1. We then obtain an equation for $P_f(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}})$ which as we shall show reduces to first order to equation (11). Indeed equation (15) becomes

$$\phi_f(\rho_{\mathbf{h}}, \ldots, \theta_{\mathbf{h+k}}) = \Big\langle \underbrace{\Big\{ J_0\Big(\frac{\rho_{\mathbf{h}}}{\mu^{1/2}}\hat{R}_{\mathbf{h}}\Big) + 2\sum_{k=1}^{\infty} i^k J_k\Big(\frac{\rho_{\mathbf{h}}}{\mu^{1/2}}\hat{R}_{\mathbf{h}}\Big)\cos[k(\hat{\varphi}_{\mathbf{h}} - \theta_{\mathbf{h}})]\Big\} \times \ldots}_{\chi(\rho_{\mathbf{h}},\ldots,\theta_{\mathbf{h+k}})} \Big\rangle_f^{\mu} \tag{16}$$

and

$$\begin{aligned}\langle \chi(\rho_{\mathbf{h}}, \ldots, \theta_{\mathbf{h+k}})\rangle_f &= \Big\langle J_0\Big(\frac{\rho_{\mathbf{h}}}{\mu^{1/2}}\hat{R}_{\mathbf{h}}\Big) \times \ldots \times J_0\Big(\frac{\rho_{\mathbf{h+k}}}{\mu^{1/2}}\hat{R}_{\mathbf{h+k}}\Big)\Big\rangle_f \\ &\quad + 2^3\tfrac{1}{4}i^3\Big\langle J_1\Big(\frac{\rho_{\mathbf{h}}}{\mu^{1/2}}\hat{R}_{\mathbf{h}}\Big) \times \ldots \times J_1\Big(\frac{\rho_{\mathbf{h+k}}}{\mu^{1/2}}\hat{R}_{\mathbf{h+k}}\Big) \\ &\quad \times \cos(\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}})\Big\rangle_f \\ &\quad \times \cos(\theta_{\mathbf{h}} + \theta_{\mathbf{k}} - \theta_{\mathbf{h+k}}) + O\Big(\frac{1}{\mu}\Big).\end{aligned} \tag{17}$$

Note that $\langle \hat{R}_{\mathbf{h}} \cos\hat{\varphi}_{\mathbf{h}}\rangle_f = \ldots = \langle \hat{R}_{\mathbf{h}}^2 \hat{R}_{\mathbf{k}} \cos(2\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}})\rangle_f = 0$ because $f(\mathbf{x}_1, \ldots, \mathbf{x}_N)$ is translation invariant and $\langle \hat{R}_{\mathbf{h}}\hat{R}_{\mathbf{k}}\hat{R}_{\mathbf{h+k}} \sin(\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}})\rangle_f = 0$ because $f(\mathbf{x}_1, \ldots, \mathbf{x}_N)$ is even. Then using the formulas (149) and (150) of the Appendix, one obtains

$$\begin{aligned}\langle \chi(\rho_{\mathbf{h}}, \ldots, \theta_{\mathbf{h+k}})\rangle_f &= 1 - \frac{1}{4\mu}\rho_{\mathbf{h}}^2\langle \hat{R}_{\mathbf{h}}^2\rangle_f - \ldots - \frac{1}{4\mu}\rho_{\mathbf{h+k}}^2\langle \hat{R}_{\mathbf{h+k}}^2\rangle_f \\ &\quad - \frac{i}{4\mu\mu^{1/2}}\rho_{\mathbf{h}}\rho_{\mathbf{k}}\rho_{\mathbf{h+k}}\langle \hat{R}_{\mathbf{h}}\hat{R}_{\mathbf{k}}\hat{R}_{\mathbf{h+k}}\cos(\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}})\rangle_f \\ &\quad \times \cos(\theta_{\mathbf{h}} + \theta_{\mathbf{k}} - \theta_{\mathbf{h+k}}) + O\Big(\frac{1}{\mu^2}\Big).\end{aligned} \tag{18}$$

Developing asymptotically $\phi_f(\rho_{\mathbf{h}}, \ldots, \theta_{\mathbf{h+k}}) = \langle \chi(\rho_{\mathbf{h}}, \ldots, \theta_{\mathbf{h+k}})\rangle_f^{\mu}$ [and using $\ln(1+x) = x - x^2/2 + x^3/3 - \ldots$ for $|x| < 1$], $\phi_f(\rho_{\mathbf{h}}, \ldots, \theta_{\mathbf{h+k}})$ reads

$$\begin{aligned}\phi_f(\rho_{\mathbf{h}}, \ldots, \theta_{\mathbf{h+k}}) &= \exp(-\tfrac{1}{4}\rho_{\mathbf{h}}^2\langle \hat{R}_{\mathbf{h}}^2\rangle_f - \ldots) \\ &\quad \times \Big[1 - \frac{i}{4\mu^{1/2}}\rho_{\mathbf{h}}\rho_{\mathbf{k}}\rho_{\mathbf{h+k}}\langle \hat{R}_{\mathbf{h}}\hat{R}_{\mathbf{k}}\hat{R}_{\mathbf{h+k}}\cos(\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}})\rangle_f \\ &\quad \times \cos(\theta_{\mathbf{h}} + \theta_{\mathbf{k}} - \theta_{\mathbf{h+k}}) + O\Big(\frac{1}{\mu}\Big)\Big].\end{aligned} \tag{19}$$

Next we define

$$P_f(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}}) \equiv P_f(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}} | \bar{R}_{\mathbf{h}} = R_{\mathbf{h}}, \bar{R}_{\mathbf{k}} = R_{\mathbf{k}}, \bar{R}_{\mathbf{h+k}} = R_{\mathbf{h+k}}). \tag{20}$$

Putting the form of $\phi_f(\rho_{\mathbf{h}}, \ldots, \theta_{\mathbf{h+k}})$ given by equation (19) into equation (12) we get

$$\begin{aligned}P_f(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}}) &= \frac{R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}}{(2\pi)^6}\int_0^{2\pi} \mathrm{d}\theta_{\mathbf{h}} \ldots \int_0^{\infty} \rho_{\mathbf{h}}\,\mathrm{d}\rho_{\mathbf{h}} \ldots \exp[-i\rho_{\mathbf{h}}R_{\mathbf{h}} \\ &\quad \times \cos(\bar{\varphi}_{\mathbf{h}} - \theta_{\mathbf{h}}) - \ldots]\exp(-\tfrac{1}{4}\rho_{\mathbf{h}}^2\langle \hat{R}_{\mathbf{h}}^2\rangle_f - \ldots) \\ &\quad \times \Big[1 - \frac{i}{4\mu^{1/2}}\rho_{\mathbf{h}}\rho_{\mathbf{k}}\rho_{\mathbf{h+k}}\langle \hat{R}_{\mathbf{h}}\hat{R}_{\mathbf{k}}\hat{R}_{\mathbf{h+k}}\cos(\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}})\rangle_f \\ &\quad \times \cos(\theta_{\mathbf{h}} + \theta_{\mathbf{k}} - \theta_{\mathbf{h+k}}) + O\Big(\frac{1}{\mu}\Big)\Big].\end{aligned} \tag{21}$$

We first perform the θ integrations in equation (21) by using formula (136). We then obtain

$$\begin{aligned}P_f(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}}) &= \frac{(2\pi)^3 R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}}{(2\pi)^6}\int_0^{\infty} \rho_{\mathbf{h}}\,\mathrm{d}\rho_{\mathbf{h}} \ldots \exp(-\tfrac{1}{4}\rho_{\mathbf{h}}^2\langle \hat{R}_{\mathbf{h}}^2\rangle_f - \ldots) \\ &\quad \times \Big[J_0(\rho_{\mathbf{h}}R_{\mathbf{h}}) \ldots J_0(\rho_{\mathbf{h+k}}R_{\mathbf{h+k}}) \\ &\quad + \frac{1}{4\mu^{1/2}}\rho_{\mathbf{h}}J_1(\rho_{\mathbf{h}}R_{\mathbf{h}})\rho_{\mathbf{k}}J_1(\rho_{\mathbf{h}}R_{\mathbf{k}})\rho_{\mathbf{h+k}}J_1(\rho_{\mathbf{h+k}}R_{\mathbf{h+k}}) \\ &\quad \times \langle \hat{R}_{\mathbf{h}}\hat{R}_{\mathbf{k}}\hat{R}_{\mathbf{h+k}}\cos(\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}})\rangle_f \\ &\quad \times \cos(\bar{\varphi}_{\mathbf{h}} + \bar{\varphi}_{\mathbf{k}} - \bar{\varphi}_{\mathbf{h+k}}) + O\Big(\frac{1}{\mu}\Big)\Big].\end{aligned} \tag{22}$$

Applying the transformations $\rho_{\mathbf{h}} \to \rho_{\mathbf{h}}/\langle \hat{R}_{\mathbf{h}}^2\rangle_f^{1/2}$, we obtain

$$\begin{aligned}P_f(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}}) &= \frac{R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}}{(2\pi)^3}\int_0^{\infty} \frac{\rho_{\mathbf{h}}}{\langle \hat{R}_{\mathbf{h}}^2\rangle_f}\,\mathrm{d}\rho_{\mathbf{h}} \ldots \exp(-\tfrac{1}{4}\rho_{\mathbf{h}}^2 - \ldots) \\ &\quad \times \Big[J_0\Big(\rho_{\mathbf{h}}\frac{R_{\mathbf{h}}}{\langle \hat{R}_{\mathbf{h}}^2\rangle_f^{1/2}}\Big) \ldots J_0\Big(\rho_{\mathbf{h+k}}\frac{R_{\mathbf{h+k}}}{\langle \hat{R}_{\mathbf{h+k}}^2\rangle_f^{1/2}}\Big) \\ &\quad + \frac{1}{4\mu^{1/2}}\frac{\rho_{\mathbf{h}}}{\langle \hat{R}_{\mathbf{h}}^2\rangle_f^{1/2}}J_1\Big(\rho_{\mathbf{h}}\frac{R_{\mathbf{h}}}{\langle \hat{R}_{\mathbf{h}}^2\rangle_f^{1/2}}\Big) \times \ldots \\ &\quad \times \frac{\rho_{\mathbf{h+k}}}{\langle \hat{R}_{\mathbf{h+k}}^2\rangle_f^{1/2}}J_1\Big(\rho_{\mathbf{h+k}}\frac{R_{\mathbf{h+k}}}{\langle \hat{R}_{\mathbf{h+k}}^2\rangle_f^{1/2}}\Big) \\ &\quad \times \langle \hat{R}_{\mathbf{h}}\hat{R}_{\mathbf{k}}\hat{R}_{\mathbf{h+k}}\cos(\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}})\rangle_f \cos(\bar{\varphi}_{\mathbf{h}} + \bar{\varphi}_{\mathbf{k}} - \bar{\varphi}_{\mathbf{h+k}}) \\ &\quad + O\Big(\frac{1}{\mu}\Big)\Big].\end{aligned} \tag{23}$$

Then applying formula (137) of the Appendix we get

$$
\begin{aligned}
&P_f(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}}) \\
&\quad = \frac{2^3 R_{\mathbf{h}} R_{\mathbf{k}} R_{\mathbf{h+k}}}{(2\pi)^3 \langle \hat{R}^2_{\mathbf{h}} \rangle_f \langle \hat{R}^2_{\mathbf{k}} \rangle_f \langle \hat{R}^2_{\mathbf{h+k}} \rangle_f} \\
&\qquad \times \exp\left(- \frac{R^2_{\mathbf{h}}}{\langle \hat{R}^2_{\mathbf{h}} \rangle_f} \ldots - \frac{R^2_{\mathbf{h+k}}}{\langle \hat{R}^2_{\mathbf{h+k}} \rangle_f} \right) \\
&\qquad \times \left[1 + \frac{2}{\mu^{1/2}} \frac{R_{\mathbf{h}} R_{\mathbf{k}} R_{\mathbf{h+k}}}{\langle \hat{R}^2_{\mathbf{h}} \rangle_f \langle \hat{R}^2_{\mathbf{k}} \rangle_f \langle \hat{R}^2_{\mathbf{h+k}} \rangle_f} \right. \\
&\qquad \times \langle \hat{R}_{\mathbf{h}} \hat{R}_{\mathbf{k}} \hat{R}_{\mathbf{h+k}} \cos(\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}}) \rangle_f \\
&\qquad \left. \times \cos(\bar{\varphi}_{\mathbf{h}} + \bar{\varphi}_{\mathbf{k}} - \bar{\varphi}_{\mathbf{h+k}}) + O\left(\frac{1}{\mu}\right) \right]. \qquad (24)
\end{aligned}
$$

This becomes (again using SAD)

$$
\begin{aligned}
&P_f(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}}) \\
&\quad \propto \exp\left[\frac{2}{\mu^{1/2}} \frac{R_{\mathbf{h}} R_{\mathbf{k}} R_{\mathbf{h+k}}}{\langle \hat{R}^2_{\mathbf{h}} \rangle_f \langle \hat{R}^2_{\mathbf{k}} \rangle_f \langle \hat{R}^2_{\mathbf{h+k}} \rangle_f} \right. \\
&\qquad \left. \times \langle \hat{R}_{\mathbf{h}} \hat{R}_{\mathbf{k}} \hat{R}_{\mathbf{h+k}} \cos(\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}}) \rangle_f \cos \bar{\varphi} \right] \\
&\qquad \times \left[1 + O\left(\frac{1}{\mu}\right) \right], \qquad (25)
\end{aligned}
$$

where $\bar{\varphi} = \bar{\varphi}_{\mathbf{h}} + \bar{\varphi}_{\mathbf{k}} - \bar{\varphi}_{\mathbf{h+k}}$. Putting $\mu = 1$ into equation (25) we finally obtain

$$
\begin{aligned}
&P_f(\bar{\varphi}_{\mathbf{h}}, \bar{\varphi}_{\mathbf{k}}, \bar{\varphi}_{\mathbf{h+k}}) \\
&\quad \propto \exp\left[2 \frac{R_{\mathbf{h}} R_{\mathbf{k}} R_{\mathbf{h+k}}}{\langle \hat{R}^2_{\mathbf{h}} \rangle_f \langle \hat{R}^2_{\mathbf{k}} \rangle_f \langle \hat{R}^2_{\mathbf{h+k}} \rangle_f} \right. \\
&\qquad \left. \times \langle \hat{R}_{\mathbf{h}} \hat{R}_{\mathbf{k}} \hat{R}_{\mathbf{h+k}} \cos(\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}}) \rangle_f \cos \bar{\varphi} \right] \qquad (26)
\end{aligned}
$$

for an even and translation-invariant j.p.d. $f = f(\mathbf{x}_1, \ldots, \mathbf{x}_N)$.

3. A heuristic derivation of the SAD method

For simplicity we shall describe the method for the case of real r.v.'s only.

What follows is a generalization of the approach given in Brosius (1989*a*). Suppose we have s r.v.'s $\hat{X}_j(\mathbf{x}_1, \ldots, \mathbf{x}_N)$ $(1 \le j \le s)$ and that every $\hat{X}_j(\mathbf{x}_1, \ldots, \mathbf{x}_N)$ can be written as a sum

$$\hat{X}_j = (1/N^{1/2}) \sum_{k=1}^{N} \hat{Z}_j(\mathbf{x}_k). \qquad (27)$$

Let $f(\mathbf{x}_1, \ldots, \mathbf{x}_N)$ be the j.p.d. of the r.v. $\mathbf{x}_1, \ldots, \mathbf{x}_N$.

Let μ be a divisor of N (this assumption is not necessary for the following, but it makes the reasoning simpler). Define $n = N/\mu$. We then partition the set of the $\mathbf{x}_1, \ldots, \mathbf{x}_N$ into μ subgroups:

$$
\begin{aligned}
&\{\mathbf{x}_1, \ldots, \mathbf{x}_N\} \\
&\quad \to \{\mathbf{x}_1, \ldots, \mathbf{x}_n\}, \{\mathbf{x}_{n+1}, \ldots\}, \ldots, \{\mathbf{x}_{n(\mu-1)+1}, \ldots, \mathbf{x}_N\}. \qquad (28)
\end{aligned}
$$

Every $\hat{X}_j(\mathbf{x}_1, \ldots, \mathbf{x}_N)$ can then be written as

$$\hat{X}_j = (1/\mu^{1/2}) \sum_{k=1}^{\mu} \hat{Y}_{j,k}, \qquad (29)$$

where

$$\hat{Y}_{j,k} \equiv \sum_{s=1}^{n} (\mu/N)^{1/2} \hat{Z}_j(\mathbf{x}_{n(k-1)+s}). \qquad (30)$$

We define a joint density $\bar{f}$ for the $\hat{Y}_{j,k}$ $(1 \le k \le \mu)$ as follows:

$$\bar{f}(\mathbf{x}_1, \ldots, \mathbf{x}_N) = \prod_{k=1}^{\mu} f_k(\mathbf{x}_{(k-1)n+1}, \ldots, \mathbf{x}_{kn}), \qquad (31)$$

where $f_k(\mathbf{x}_{(k-1)n+1}, \ldots, \mathbf{x}_{kn})$ is the *marginal* density of the j.p.d. $f(\mathbf{x}_1, \ldots, \mathbf{x}_N)$ for the r.v.'s $\mathbf{x}_{(k-1)n+1}, \ldots, \mathbf{x}_{kn}$. Now all $\hat{Y}_{j,k}$ $(1 \le k \le \mu)$ are independent r.v.'s with respect to the density $\bar{f}(\mathbf{x}_1, \ldots, \mathbf{x}_N)$. To keep things simple, suppose also that all $\hat{Z}_j$ are identical and that all f_k are equal [that is, $f(\mathbf{x}_1, \ldots, \mathbf{x}_N)$ is symmetric in the $\mathbf{x}_i$]. Then all $\hat{Y}_{j,k}$ $(1 \le k \le \mu)$ are independent and identical r.v.'s with respect to the density $\bar{f}(\mathbf{x}_1, \ldots, \mathbf{x}_N)$. Let $\bar{P}(X_1, \ldots, X_s)$ be the j.p.d. of the $\hat{X}_j(\mathbf{x}_1, \ldots, \mathbf{x}_N)$ $(1 \le j \le s)$ with respect to the j.p.d. $\bar{f}(\mathbf{x}_1, \ldots, \mathbf{x}_N)$, that is

$$\bar{P}(X_1, \ldots, X_s) = (1/2\pi)^s \textstyle\int \mathrm{d}u_1 \ldots \mathrm{d}u_s \exp(-iu_j X_j) \bar{\phi}(u_1, \ldots, u_s), \qquad (32)$$

where $\bar{\phi}(u_1, \ldots, u_s)$ is the characteristic function of the $\hat{X}_j(\mathbf{x}_1, \ldots, \mathbf{x}_N)$,

$$\bar{\phi}(u_1, \ldots, u_s) = \langle \exp(iu_j \hat{X}_j) \rangle_{\bar{f}} \equiv \textstyle\int \bar{f}(\mathbf{x}_1, \ldots, \mathbf{x}_N) \exp(iu_j \hat{X}_j). \qquad (33)$$

From the definitions given above it follows that the characteristic function $\bar{\phi}(u_1, \ldots, u_s) \equiv \langle \exp(iu_j \hat{X}_j) \rangle_{\bar{f}}$ can be written as

$$\bar{\phi}(u_1, \ldots, u_s) = \phi_1(u_1, \ldots, u_s; \mu)^{\mu}, \qquad (34)$$

where

$$\phi_1(u_1, \ldots, u_s; \mu) = \langle \exp[(1/\mu^{1/2}) iu_j \hat{Y}_{j,1}] \rangle_{f_1} \qquad (35)$$

is the characteristic function of the $\hat{Y}_{j,1}(\mathbf{x}_1, \ldots, \mathbf{x}_n)$. For all (high) μ we *always* get the *same* prescription for grouping the respective moments. Now let $\mu \to 1$. Then $f_1 \to f$, $u_j \hat{Y}_{j,1} \to u_j \hat{X}_j$ and thus $\bar{\phi} \to \phi$ for $\mu \to 1$, *i.e.*

$$\phi(u_1, \ldots, u_s) = \langle \exp[(1/\mu^{1/2}) iu_j \hat{X}_j] \rangle_f^{\mu} \quad \text{for } \mu \to 1. \qquad (36)$$

We conjecture that this formula will give the j.p.d. of the phases to first order (low exponents of $1/\mu^{1/2}$).

4. The statistical interpretation of the $B_{3,0}$ formula using SAD

Let us first recall the $B_{3,0}$ formula (Karle & Hauptman, 1957):

$$
\begin{aligned}
&R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}\cos(\varphi_{\mathbf{h}}+\varphi_{\mathbf{k}}-\varphi_{\mathbf{h+k}})\\
&\quad=\frac{1}{N^{1/2}}[(R_{\mathbf{h}}^2-1)+(R_{\mathbf{h+k}}^2-1)+(R_{\mathbf{k}}^2-1)+1]\\
&\qquad+\frac{(N-1)(N-2)}{N^{1/2}}\bigg[\sum_{\mathbf{q}}\left(\frac{R_{\mathbf{q}}^2-1}{N-1}\right)\left(\frac{R_{\mathbf{q+h}}^2-1}{N-1}\right)\\
&\qquad\times\left(\frac{R_{\mathbf{q+h+k}}^2-1}{N-1}\right)\bigg]\bigg/M,
\end{aligned}
\tag{37}
$$

where M is the number of available structure factors. We define the $B'_{3,0}$ formula by

$$
\begin{aligned}
&R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}\cos(\varphi_{\mathbf{h}}+\varphi_{\mathbf{k}}-\varphi_{\mathbf{h+k}})\\
&\quad=\frac{1}{N^{1/2}}[(R_{\mathbf{h}}^2-1)+(R_{\mathbf{h+k}}^2-1)+(R_{\mathbf{k}}^2-1)+1]\\
&\qquad+\frac{(N-1)(N-2)}{N^{1/2}}\bigg[\sum_{\mathbf{q}}\left(\frac{R_{\mathbf{q}}^2-1}{N-1}\right)\left(\frac{R_{\mathbf{q+h}}^2-1}{N-1}\right)\\
&\qquad\times\left(\frac{R_{\mathbf{q+h+k}}^2-1}{N-1}\right)\bigg]\bigg/\sum_{\mathbf{q}}\left(\frac{R_{\mathbf{q}}^2-1}{N-1}\right)^3.
\end{aligned}
\tag{38}
$$

Let

$$
Q(\mathbf{u})=\sum_{\mathbf{q}}\frac{(R_{\mathbf{q}}^2-1)}{N-1}\cos(2\pi\mathbf{q}\cdot\mathbf{u}). \tag{39}
$$

$Q(\mathbf{u})$ is the origin-removed Patterson function: it is 0 unless $\mathbf{u}$ equals some $\mathbf{r}_i-\mathbf{r}_j$. We can use the information stored in $Q(\mathbf{u})$ by requiring that the random vector variables $\mathbf{x}_i$, $\mathbf{x}_j$ range uniformly over the unit cell subject to the condition that $\mathbf{x}_i-\mathbf{x}_j$ equals some $\mathbf{r}_\alpha-\mathbf{r}_\beta$ (where α,β are different). This can be done easily by defining the j.p.d. $f(\mathbf{x}_1,\ldots,\mathbf{x}_N)$ of the r.v. $\mathbf{x}_1,\ldots,\mathbf{x}_N$ as

$$
f(\mathbf{x}_1,\ldots,\mathbf{x}_N)=\mathrm{Cte}\prod_{i=1}^{N-1}\prod_{\substack{j=2\\ i<j}}^{N}Q(\mathbf{x}_i-\mathbf{x}_j), \tag{40}
$$

where Cte is a constant defined by the condition $\int f(\mathbf{x}_1,\ldots,\mathbf{x}_N)\,\mathrm{d}\mathbf{x}_1\ldots\mathrm{d}\mathbf{x}_N=1$. Clearly the above $f(\mathbf{x}_1,\ldots,\mathbf{x}_N)$ is a good joint density: $f(\mathbf{x}_1,\ldots,\mathbf{x}_N)=0$ everywhere except when for all i,j $\mathbf{x}_i-\mathbf{x}_j=\mathbf{r}_\alpha-\mathbf{r}_\beta$ for some (different) α,β, and it is translation invariant and even. Putting it differently, $f(\mathbf{x}_1,\ldots,\mathbf{x}_N)$ is the *uniform* (in the case of no overlap of interatomic vectors $\mathbf{x}_\alpha-\mathbf{x}_\beta$) *density* where the $\mathbf{x}_i-\mathbf{x}_j$ range *only* over the interatomic position vectors. Let us now investigate the different joint distributions $f(\mathbf{x}_i,\mathbf{x}_j)$, $f(\mathbf{x}_i,\mathbf{x}_j,\mathbf{x}_l)$, $f(\mathbf{x}_i,\mathbf{x}_j,\mathbf{x}_l,\mathbf{x}_m)$ *etc.* By definition,

$$
f(\mathbf{x}_i,\mathbf{x}_j)\equiv\int\prod_{\substack{m\neq i\\ m\neq j}}\mathrm{d}\mathbf{x}_m\,f(\mathbf{x}_1,\ldots,\mathbf{x}_N), \tag{41}
$$

$$
f(\mathbf{x}_i,\mathbf{x}_j,\mathbf{x}_l)\equiv\int\prod_{\substack{m\neq i\\ m\neq j\\ m\neq l}}\mathrm{d}\mathbf{x}_m\,f(\mathbf{x}_1,\ldots,\mathbf{x}_N). \tag{42}
$$

To calculate these joint densities is a formidable task (for a computer). However, one does not need to calculate *e.g.* $\int\prod_{m\neq i,m\neq j}\mathrm{d}\mathbf{x}_m\,f(\mathbf{x}_1,\ldots,\mathbf{x}_N)$. Indeed $f(\mathbf{x}_i,\mathbf{x}_j)$ is zero everywhere except when $\mathbf{x}_i-\mathbf{x}_j=\mathbf{r}_\alpha-\mathbf{r}_\beta$ (for some different α,β). Since $Q(\mathbf{x}_i-\mathbf{x}_j)$ is also zero everywhere except when $\mathbf{x}_i-\mathbf{x}_j=\mathbf{r}_\alpha-\mathbf{r}_\beta$ (for some α,β), it is quite plausible to put

$$
f(\mathbf{x}_i,\mathbf{x}_j)=C_1Q(\mathbf{x}_i-\mathbf{x}_j), \tag{43}
$$

where C_1 is determined by the condition $\int\mathrm{d}\mathbf{x}_i\,\mathrm{d}\mathbf{x}_j\,f(\mathbf{x}_i,\mathbf{x}_j)=1$, thus $C_1=1$.

Let us find $f(\mathbf{x}_i,\mathbf{x}_j,\mathbf{x}_l)$. We want $\mathbf{x}_i$, $\mathbf{x}_j$ and $\mathbf{x}_l$ to range uniformly over the unit cell subject to the constraints

$$
\mathbf{x}_i-\mathbf{x}_j=\mathbf{r}_{i_1}-\mathbf{r}_{i_2},\quad \mathbf{x}_j-\mathbf{x}_l=\mathbf{r}_{i_3}-\mathbf{r}_{i_4},\quad \mathbf{x}_l-\mathbf{x}_i=\mathbf{r}_{i_5}-\mathbf{r}_{i_6} \tag{44}
$$

for some $\mathbf{r}_{i_1},\mathbf{r}_{i_2},\mathbf{r}_{i_3},\mathbf{r}_{i_4},\mathbf{r}_{i_5},\mathbf{r}_{i_6}$ (where $i_1\neq i_2$, $i_3\neq i_4$, $i_5\neq i_6$). But $Q(\mathbf{x}_i-\mathbf{x}_j)Q(\mathbf{x}_j-\mathbf{x}_l)Q(\mathbf{x}_l-\mathbf{x}_i)$ is also zero except when $\mathbf{x}_i-\mathbf{x}_j=\mathbf{r}_{i_1}-\mathbf{r}_{i_2}$, $\mathbf{x}_j-\mathbf{x}_l=\mathbf{r}_{i_3}-\mathbf{r}_{i_4}$, $\mathbf{x}_l-\mathbf{x}_i=\mathbf{r}_{i_5}-\mathbf{r}_{i_6}$. So it is quite plausible to put

$$
f(\mathbf{x}_i,\mathbf{x}_j,\mathbf{x}_l)=C_2Q(\mathbf{x}_i-\mathbf{x}_j)Q(\mathbf{x}_j-\mathbf{x}_l)Q(\mathbf{x}_l-\mathbf{x}_i). \tag{45}
$$

The constant of proportionality C_2 is determined by the condition $\int\mathrm{d}\mathbf{x}_i\,\mathrm{d}\mathbf{x}_j\,\mathrm{d}\mathbf{x}_l\,f(\mathbf{x}_i,\mathbf{x}_j,\mathbf{x}_l)=1$, thus

$$
C_2^{-1}=\sum_{\mathbf{q}}\left(\frac{R_{\mathbf{q}}^2-1}{N-1}\right)^3. \tag{46}
$$

We also get the moment

$$
\begin{aligned}
&\int\mathrm{d}\mathbf{x}_i\,\mathrm{d}\mathbf{x}_j\,\mathrm{d}\mathbf{x}_l\,f(\mathbf{x}_i,\mathbf{x}_j,\mathbf{x}_l)\cos[2\pi\mathbf{h}\cdot(\mathbf{x}_i-\mathbf{x}_l)+2\pi\mathbf{k}\cdot(\mathbf{x}_j-\mathbf{x}_l)]\\
&\quad=\left[\sum_{\mathbf{q}}\frac{(R_{\mathbf{q}}^2-1)}{(N-1)}\frac{(R_{\mathbf{q+h}}^2-1)}{(N-1)}\frac{(R_{\mathbf{q+h+k}}^2-1)}{(N-1)}\right]\bigg/\sum_{\mathbf{q}}\frac{(R_{\mathbf{q}}^2-1)^3}{(N-1)}.
\end{aligned}
\tag{47}
$$

If there is no overlap of interatomic vectors $f(\mathbf{x}_i,\mathbf{x}_j,\mathbf{x}_l)$ [see equation (45)] is zero except when there exist vectors $\mathbf{r}_\alpha,\mathbf{r}_\beta,\mathbf{r}_\gamma$ (α,β,γ different) such that $\mathbf{x}_i-\mathbf{x}_j=\mathbf{r}_\alpha-\mathbf{r}_\beta$, $\mathbf{x}_j-\mathbf{x}_l=\mathbf{r}_\beta-\mathbf{r}_\gamma$, $\mathbf{x}_l-\mathbf{x}_i=\mathbf{r}_\gamma-\mathbf{r}_\alpha$. It then follows that [since $f(\mathbf{x}_i,\mathbf{x}_j,\mathbf{x}_l)$ only has peaks at $\mathbf{x}_i-\mathbf{x}_j=\mathbf{r}_\alpha-\mathbf{r}_\beta$ *etc.*]

$$
\begin{aligned}
&\int\mathrm{d}\mathbf{x}_i\,\mathrm{d}\mathbf{x}_j\,\mathrm{d}\mathbf{x}_l\,f(\mathbf{x}_i,\mathbf{x}_j,\mathbf{x}_l)\cos[2\pi\mathbf{h}\cdot(\mathbf{x}_i-\mathbf{x}_l)+2\pi\mathbf{k}\cdot(\mathbf{x}_j-\mathbf{x}_l)]\\
&\quad=\frac{1}{N(N-1)(N-2)}\sum_{(ijl)}\cos[2\pi\mathbf{h}\cdot(\mathbf{r}_i-\mathbf{r}_l)+2\pi\mathbf{k}\cdot(\mathbf{r}_j-\mathbf{r}_l)],
\end{aligned}
\tag{48}
$$

where $\sum_{(ijl)}\equiv\sum_{i\neq j,j\neq k,i\neq k}$ (and when there is no overlap of interatomic vectors). We can go on in this way to determine j.p.d.'s of a higher (more than 3) number of r.v.'s $\mathbf{x}_i$. Since we are only interested in calculating $P_f(\bar{\varphi}_{\mathbf{h}},\bar{\varphi}_{\mathbf{k}},\bar{\varphi}_{\mathbf{h+k}})$ to order $1/\mu^{1/2}$ [see equation (25)] in this paragraph we shall not discuss it here.

Next let us calculate equation (26) for our j.p.d. $f(\mathbf{x}_1,\ldots,\mathbf{x}_N)$. First calculate $\langle\hat{R}_{\mathbf{h}}^2\rangle_f$:

$$
\begin{aligned}
\langle \hat{R}_{\mathbf{h}}^2 \rangle_f &= \left\langle 1 + \frac{1}{N} \sum_{(jk)} \exp[2\pi i \mathbf{h} \cdot (\mathbf{x}_j - \mathbf{x}_k)] \right\rangle_f \\
&= 1 + \frac{1}{N} \sum_{(jk)} \langle \cos[2\pi \mathbf{h} \cdot (\mathbf{x}_j - \mathbf{x}_k)] \rangle_f \\
&= 1 + \frac{N(N-1)}{N} \left(\frac{R_{\mathbf{h}}^2 - 1}{N-1} \right) \\
&= R_{\mathbf{h}}^2. \qquad (49)
\end{aligned}
$$

Finally

$$
\begin{aligned}
&\langle \hat{R}_{\mathbf{h}} \hat{R}_{\mathbf{k}} \hat{R}_{\mathbf{h+k}} \cos(\hat{\varphi}_{\mathbf{h}} + \hat{\varphi}_{\mathbf{k}} - \hat{\varphi}_{\mathbf{h+k}}) \rangle \\
&= \frac{1}{NN^{1/2}} \mathcal{R}e \langle \hat{E}_{\mathbf{h}} \hat{E}_{\mathbf{k}} \hat{E}_{-(\mathbf{h+k})} \rangle \\
&= \frac{1}{NN^{1/2}} \sum_{i,j,l} \langle \cos[2\pi \mathbf{h} \cdot (\mathbf{x}_i - \mathbf{x}_l) + 2\pi \mathbf{k} \cdot (\mathbf{x}_j - \mathbf{x}_l)] \rangle \\
&= \frac{1}{NN^{1/2}} \Bigg\{ \sum_i 1 + \sum_{i \neq j = l} \langle \cos[2\pi \mathbf{h} \cdot (\mathbf{x}_i - \mathbf{x}_j)] \rangle \\
&\quad + \sum_{i = j \neq l} \langle \cos[2\pi (\mathbf{h} + \mathbf{k}) \cdot (\mathbf{x}_j - \mathbf{x}_l)] \rangle \\
&\quad + \sum_{i = l \neq j} \langle \cos[2\pi \mathbf{k} \cdot (\mathbf{x}_j - \mathbf{x}_l)] \rangle \\
&\quad + \sum_{(ijl)} \langle \cos[2\pi \mathbf{h} \cdot (\mathbf{x}_i - \mathbf{x}_l) + 2\pi \mathbf{k} \cdot (\mathbf{x}_j - \mathbf{x}_l)] \rangle \Bigg\} \\
&= \frac{1}{N^{1/2}} + \frac{N(N-1)}{NN^{1/2}(N-1)} [(R_{\mathbf{h}}^2 - 1) + (R_{\mathbf{h+k}}^2 - 1) + (R_{\mathbf{k}}^2 - 1)] \\
&\quad + \frac{N(N-1)(N-2)}{NN^{1/2}} \mu(\mathbf{h}, \mathbf{k}), \qquad (50)
\end{aligned}
$$

where $\mathcal{R}e\, z$ is the real part of z, $\sum_{(ijl)}$ means $\sum_{i \neq j \neq l, i \neq l}$ and where

$$
\mu(\mathbf{h}, \mathbf{k}) = \left[\sum_{\mathbf{q}} \frac{(R_{\mathbf{q}}^2 - 1)}{(N-1)} \frac{(R_{\mathbf{q+h}}^2 - 1)}{(N-1)} \times \frac{(R_{\mathbf{q+h+k}}^2 - 1)}{(N-1)} \right] \Bigg/ \sum_{\mathbf{q}} \frac{(R_{\mathbf{q}}^2 - 1)^3}{(N-1)}. \qquad (51)
$$

Inserting these formulas into equation (26) we get

$$
P_f(\bar{\varphi}) \propto \exp\{2[m(\mathbf{h}, \mathbf{k}) / R_{\mathbf{h}} R_{\mathbf{k}} R_{\mathbf{h+k}}] \cos \bar{\varphi}\}, \qquad (52)
$$

where

$$
m(\mathbf{h}, \mathbf{k}) = \frac{1}{N^{1/2}} (R_{\mathbf{h}}^2 + R_{\mathbf{k}}^2 + R_{\mathbf{h+k}}^2 - 2) + \frac{(N-1)(N-2)}{N^{1/2}} \mu(\mathbf{h}, \mathbf{k}) \qquad (53)
$$

and where $\bar{\varphi} = \bar{\varphi}_{\mathbf{h}} + \bar{\varphi}_{\mathbf{k}} - \bar{\varphi}_{\mathbf{h+k}}$.

Developing $[(N-1)(N-2)/N^{1/2}]\mu(\mathbf{h}, \mathbf{k})$ asymptotically according to inverse powers of $1/N^{1/2}$ we get

$$
\begin{aligned}
&\frac{(N-1)(N-2)}{N^{1/2}} \mu(\mathbf{h}, \mathbf{k}) \\
&= \frac{1}{N^{1/2}} [(R_{\mathbf{h}}^2 - 1)(R_{\mathbf{h+k}}^2 - 1) + (R_{\mathbf{h}}^2 - 1)(R_{\mathbf{k}}^2 - 1) \\
&\quad + (R_{\mathbf{k}}^2 - 1)(R_{\mathbf{h+k}}^2 - 1)] + O\left(\frac{1}{NN^{1/2}} \right), \qquad (54)
\end{aligned}
$$

that is

$$
\begin{aligned}
m(\mathbf{h}, \mathbf{k}) \simeq \frac{1}{N^{1/2}} [&R_{\mathbf{h}}^2 + R_{\mathbf{k}}^2 + R_{\mathbf{h+k}}^2 - 2 + (R_{\mathbf{h}}^2 - 1)(R_{\mathbf{k}}^2 - 1) \\
&+ (R_{\mathbf{h}}^2 - 1)(R_{\mathbf{h+k}}^2 - 1) + (R_{\mathbf{k}}^2 - 1)(R_{\mathbf{h+k}}^2 - 1)] \\
&+ O\left(\frac{1}{NN^{1/2}} \right). \qquad (55)
\end{aligned}
$$

5. The linearized invariants method

We shall now show by another method that the form (a Von Mises-like distribution) of equation (52) is acceptable. To this end we write

$$
\begin{aligned}
&\hat{E}_{\mathbf{h}} \hat{E}_{\mathbf{k}} \hat{E}_{-\mathbf{h}-\mathbf{k}} \\
&= (1/N^{1/2})(R_{\mathbf{h}}^2 + R_{\mathbf{k}}^2 + R_{\mathbf{h+k}}^2 - 2) \\
&\quad + N^{-3/2} \sum_{\substack{i \neq j \neq l \\ i \neq l}} \exp\{2\pi i[\mathbf{h} \cdot (\mathbf{x}_i - \mathbf{x}_l) + \mathbf{k} \cdot (\mathbf{x}_j - \mathbf{x}_l)]\} \\
&= (1/N^{1/2})(R_{\mathbf{h}}^2 + R_{\mathbf{k}}^2 + R_{\mathbf{h+k}}^2 - 2) \\
&\quad + N^{-3/2} \sum_{i<j<l} [S_{ijl}(\mathbf{h}, \mathbf{k}) + S_{ijl}(\mathbf{k}, \mathbf{h}) + S_{ijl}(\mathbf{h}, -\mathbf{h}-\mathbf{k}) \\
&\quad + S_{ijl}(-\mathbf{h}-\mathbf{k}, \mathbf{h}) + S_{ijl}(\mathbf{k}, -\mathbf{h}-\mathbf{k}) + S_{ijl}(-\mathbf{h}-\mathbf{k}, \mathbf{h})], \qquad (56)
\end{aligned}
$$

where

$$
S_{ijl}(\mathbf{h}, \mathbf{k}) = \exp\{2\pi i[\mathbf{h} \cdot (\mathbf{x}_i - \mathbf{x}_l) + \mathbf{k} \cdot (\mathbf{x}_j - \mathbf{x}_l)]\}. \qquad (57)
$$

We shall now 'linearize' $\hat{E}_{\mathbf{h}} \hat{E}_{\mathbf{k}} \hat{E}_{-\mathbf{h}-\mathbf{k}}$; that is we define an r.v.

$$
\begin{aligned}
\hat{Z}(\mathbf{h}, \mathbf{k}) \equiv\ &(1/N^{1/2})(R_{\mathbf{h}}^2 + R_{\mathbf{k}}^2 + R_{\mathbf{h+k}}^2 - 2) \\
&+ N^{-3/2} \sum_{\alpha} [S_{\alpha}(\mathbf{h}, \mathbf{k}) + S_{\alpha}(\mathbf{k}, \mathbf{h}) + S_{\alpha}(\mathbf{h}, -\mathbf{h}-\mathbf{k}) \\
&+ S_{\alpha}(-\mathbf{h}-\mathbf{k}, \mathbf{h}) + S_{\alpha}(\mathbf{k}, -\mathbf{h}-\mathbf{k}) + S_{\alpha}(-\mathbf{h}-\mathbf{k}, \mathbf{h})], \qquad (58)
\end{aligned}
$$

where

$$
S_{\alpha}(\mathbf{h}, \mathbf{k}) = \exp[2\pi i(\mathbf{h} \cdot \mathbf{u}_{\alpha} + \mathbf{k} \cdot \mathbf{v}_{\alpha})] \qquad (59)
$$

and where $\mathbf{u}_{\alpha}$ and $\mathbf{v}_{\alpha}$ are independent random vectors ranging over the unit cell and $\alpha = (ijl)$ with $i < j < l$. There are exactly $N(N-1)(N-2)/6$ such α's. It follows from equations (56) and (57) that *every value* of $\hat{E}_{\mathbf{h}} \hat{E}_{\mathbf{k}} \hat{E}_{-\mathbf{h}-\mathbf{k}}$ is also attained by $\hat{Z}(\mathbf{h}, \mathbf{k})$. We shall now impose the condition that every couple $(\mathbf{u}_{\alpha}, \mathbf{v}_{\alpha})$ is of the form $(\mathbf{r}_i - \mathbf{r}_l, \mathbf{r}_j - \mathbf{r}_l)$. We do this by using a density

$$
f(\mathbf{u}_{\alpha}, \mathbf{v}_{\alpha}) = \mathrm{Cte}\, Q(\mathbf{u}_{\alpha}) Q(\mathbf{u}_{\alpha} - \mathbf{v}_{\alpha}) Q(\mathbf{v}_{\alpha}), \qquad (60)
$$

where $Q(\mathbf{u})$ is given by equation (39). The first neighborhood of the r.v. $\hat{Z}(\mathbf{h}, \mathbf{k})$ is $\{\hat{Z}(\mathbf{h}, \mathbf{k})\}$. We shall consider the second neighborhood $\{\hat{Z}(\mathbf{h}, \mathbf{k}), \hat{U}, \hat{V}, \hat{W}\}$, where $\hat{U} \equiv \hat{Z}(\mathbf{h}, \mathbf{0})$, $\hat{V} \equiv \hat{Z}(\mathbf{0}, \mathbf{k})$ and $\hat{W} \equiv \hat{Z}(\mathbf{h}+\mathbf{k}, \mathbf{0})$, and remark that these r.v.'s are real.

We now calculate the j.p.d. $P(\psi, |Z|, U, V, W)$ of the r.v. $\hat{\psi}, |\hat{Z}|$ $[\hat{Z} \equiv |\hat{Z}| \exp(i\hat{\psi})]$, $\hat{U}, \ldots, \hat{W}$.

$$
\begin{aligned}
&P(\psi, |Z|, \ldots, W) \\
&\quad \equiv [|Z|/(2\pi)^5] \int_0^{2\pi} \mathrm{d}\theta \int_0^{\infty} z\,\mathrm{d}z \int_{-\infty}^{\infty} \mathrm{d}u \int_{-\infty}^{\infty} \mathrm{d}v \int_{-\infty}^{\infty} \mathrm{d}w \exp[-iz|Z| \\
&\qquad \times \cos(\psi - \theta) - iuU - ivV - iwW] \\
&\qquad \times \varphi(z, \theta, u, v, w)^K \exp[izN^{-1/2} \cos\theta (R_{\mathbf{h}}^2 + R_{\mathbf{k}}^2 + R_{\mathbf{h+k}}^2 - 2)] \\
&\qquad \times \exp\{iu[N^{1/2} + 2N^{-1/2}(R_{\mathbf{h}}^2 - 1)] \\
&\qquad + iv[N^{1/2} + 2N^{-1/2}(R_{\mathbf{k}}^2 - 1)] \\
&\qquad + iw[N^{1/2} + 2N^{-1/2}(R_{\mathbf{h+k}}^2 - 1)]\},
\end{aligned} \tag{61}
$$

where $K = N(N-1)(N-2)/6$ and where

$$
\begin{aligned}
&\varphi(z, \theta, u, v, w) \\
&\quad = \Big\langle \exp\Big[izN^{-3/2}\Big(\cos[2\pi(\mathbf{h}\cdot\mathbf{u}_\alpha + \mathbf{k}\cdot\mathbf{v}_\alpha) - \theta] \\
&\qquad + \cos[2\pi(\mathbf{k}\cdot\mathbf{u}_\alpha + \mathbf{h}\cdot\mathbf{v}_\alpha) - \theta] \\
&\qquad + \cos\{2\pi[\mathbf{h}\cdot\mathbf{u}_\alpha - (\mathbf{h}+\mathbf{k})\cdot\mathbf{v}_\alpha] - \theta\} \\
&\qquad + \cos\{2\pi[-(\mathbf{h}+\mathbf{k})\cdot\mathbf{u}_\alpha + \mathbf{h}\cdot\mathbf{v}_\alpha] - \theta\} \\
&\qquad + \cos\{2\pi[\mathbf{k}\cdot\mathbf{u}_\alpha - (\mathbf{h}+\mathbf{k})\cdot\mathbf{v}_\alpha] - \theta\} \\
&\qquad + \cos\{2\pi[(-\mathbf{h}-\mathbf{k})\cdot\mathbf{u}_\alpha + \mathbf{h}\cdot\mathbf{v}_\alpha] - \theta\}\Big)\Big] \\
&\qquad \times \exp\Big(2iuN^{-3/2}\{\cos(2\pi\mathbf{h}\cdot\mathbf{u}_\alpha) + \cos(2\pi\mathbf{h}\cdot\mathbf{v}_\alpha) \\
&\qquad + \cos[2\pi(\mathbf{h}\cdot\mathbf{u}_\alpha - \mathbf{h}\cdot\mathbf{v}_\alpha)]\}\Big) \\
&\qquad \times \exp\Big(2ivN^{-3/2}\{\cos(2\pi\mathbf{k}\cdot\mathbf{u}_\alpha) + \cos(2\pi\mathbf{k}\cdot\mathbf{v}_\alpha) \\
&\qquad + \cos[2\pi(\mathbf{k}\cdot\mathbf{u}_\alpha - \mathbf{k}\cdot\mathbf{v}_\alpha)]\}\Big) \\
&\qquad \times \exp\Big[2iwN^{-3/2}\Big(\cos[2\pi(\mathbf{h}+\mathbf{k})\cdot\mathbf{u}_\alpha] \\
&\qquad + \cos[2\pi(\mathbf{h}+\mathbf{k})\cdot\mathbf{v}_\alpha] \\
&\qquad + \cos\{2\pi[(\mathbf{h}+\mathbf{k})\cdot\mathbf{u}_\alpha - (\mathbf{h}+\mathbf{k})\cdot\mathbf{v}_\alpha]\}\Big)\Big]\Big\rangle.
\end{aligned} \tag{62}
$$

Next define

$$
\begin{aligned}
\mu(\mathbf{h}, \mathbf{k}) &\equiv \langle \cos[2\pi(\mathbf{h}\cdot\mathbf{u}_\alpha + \mathbf{k}\cdot\mathbf{v}_\alpha)] \rangle \\
&\equiv \int \mathrm{d}\mathbf{u}_\alpha\,\mathrm{d}\mathbf{v}_\alpha\, f(\mathbf{u}_\alpha, \mathbf{v}_\alpha) \cos[2\pi(\mathbf{h}\cdot\mathbf{u}_\alpha + \mathbf{k}\cdot\mathbf{v}_\alpha)] \\
&= \Big[\sum_{\mathbf{q}} \Big(\frac{R_{\mathbf{q}}^2 - 1}{N-1}\Big)\Big(\frac{R_{\mathbf{q+h}}^2 - 1}{N-1}\Big) \\
&\qquad \times \Big(\frac{R_{\mathbf{q+h+k}}^2 - 1}{N-1}\Big)\Big] \Big/ \sum_{\mathbf{q}} \Big(\frac{R_{\mathbf{q}}^2 - 1}{N-1}\Big)^3, \\
\mu(\mathbf{h}) &\equiv \langle \cos(2\pi\mathbf{h}\cdot\mathbf{u}_\alpha) \rangle = \langle \cos(2\pi\mathbf{h}\cdot\mathbf{v}_\alpha) \rangle = \mu(\mathbf{h}, \mathbf{0}) \\
&= \Big[\sum_{\mathbf{q}} \Big(\frac{R_{\mathbf{q}}^2 - 1}{N-1}\Big)^2 \Big(\frac{R_{\mathbf{q+h}}^2 - 1}{N-1}\Big)\Big] \Big/ \sum_{\mathbf{q}} \Big(\frac{R_{\mathbf{q}}^2 - 1}{N-1}\Big)^3.
\end{aligned} \tag{63}
$$

Remark that theoretically [see equations (50) or (38)]

$$
\begin{aligned}
\mu(\mathbf{h}, \mathbf{k}) &= \frac{1}{NN^{1/2}} R_{\mathbf{h}} R_{\mathbf{k}} R_{\mathbf{h+k}} \cos(\varphi_{\mathbf{h}} + \varphi_{\mathbf{k}} - \varphi_{\mathbf{h+k}}) - O\Big(\frac{1}{N^2}\Big) \\
&= O\Big(\frac{1}{NN^{1/2}}\Big) \\
\mu(\mathbf{h}) &= \frac{N^{1/2}}{NN^{1/2}} R_{\mathbf{h}}^2 - \frac{N^{1/2}}{NN^{1/2}} + \frac{1}{N^2}(2R_{\mathbf{h}}^2 - 2) \\
&= \frac{1}{N}(R_{\mathbf{h}}^2 - 1) + O\Big(\frac{1}{N^2}\Big) = O\Big(\frac{1}{N}\Big).
\end{aligned} \tag{64}
$$

We assume that in practice equation (64) is also true. We now calculate $\varphi(z, \theta, u, v, w)$ up to and including terms of order $O(1/KK^{1/2}) = O(1/N^4N^{1/2})$. But first we need some additional definitions:

$$
\begin{aligned}
\hat{Z}_\alpha &= S_\alpha(\mathbf{h}, \mathbf{k}) + S_\alpha(\mathbf{k}, \mathbf{h}) + S_\alpha(\mathbf{h}, -\mathbf{h}-\mathbf{k}) + S_\alpha(-\mathbf{h}-\mathbf{k}, \mathbf{h}) \\
&\quad + S_\alpha(\mathbf{k}, -\mathbf{h}-\mathbf{k}) + S_\alpha(-\mathbf{h}-\mathbf{k}, \mathbf{h}), \\
\hat{U}_\alpha &= \cos(2\pi\mathbf{h}\cdot\mathbf{u}_\alpha) + \cos(2\pi\mathbf{h}\cdot\mathbf{v}_\alpha) + \cos[2\pi(\mathbf{h}\cdot\mathbf{u}_\alpha - \mathbf{h}\cdot\mathbf{v}_\alpha)], \\
\hat{V}_\alpha &= \cos(2\pi\mathbf{k}\cdot\mathbf{u}_\alpha) + \cos(2\pi\mathbf{k}\cdot\mathbf{v}_\alpha) + \cos[2\pi(\mathbf{k}\cdot\mathbf{u}_\alpha - \mathbf{k}\cdot\mathbf{v}_\alpha)], \\
\hat{W}_\alpha &= \cos[2\pi(\mathbf{h}+\mathbf{k})\cdot\mathbf{u}_\alpha] + \cos[2\pi(\mathbf{h}+\mathbf{k})\cdot\mathbf{v}_\alpha] \\
&\quad + \cos\{2\pi[(\mathbf{h}+\mathbf{k})\cdot\mathbf{u}_\alpha - (\mathbf{h}+\mathbf{k})\cdot\mathbf{v}_\alpha]\}.
\end{aligned} \tag{65}
$$

We then get

$$
\begin{aligned}
\varphi(z, \theta, u, v, w) &= 1 + izN^{-3/2}6\mu(\mathbf{h}, \mathbf{k})\cos\theta + 2iuN^{-3/2}3\mu(\mathbf{h}) \\
&\quad + 2ivN^{-3/2}3\mu(\mathbf{k}) + 2iwN^{-3/2}3\mu(\mathbf{h}+\mathbf{k}) \\
&\quad - \tfrac{1}{2}z^2N^{-3}[\tfrac{6}{2} + O(1/N)] - \tfrac{1}{2}4u^2N^{-3}[\tfrac{3}{2} + O(1/N)] \\
&\quad - \tfrac{1}{2}4v^2N^{-3}[\tfrac{3}{2} + O(1/N)] - \tfrac{1}{2}4w^2N^{-3}[\tfrac{3}{2} + O(1/N)] \\
&\quad - \tfrac{1}{2}2(1/N^3)z2u\langle \hat{Z}_\alpha \hat{U}_\alpha \rangle - (1/N^3)2zv\langle \hat{Z}_\alpha \hat{V}_\alpha \rangle \\
&\quad - (1/N^3)2zw\langle \hat{Z}_\alpha \hat{W}_\alpha \rangle - (i/3!)3!z2u2v(1/N^4N^{1/2})\tfrac{1}{4}6 \\
&\quad - (6/N^4N^{1/2})izuw - (6i/N^4N^{1/2})zvw \\
&\quad + \text{h.o.},
\end{aligned} \tag{66}
$$

where h.o. means terms of order $1/N^4$ that do not contribute to the phase determination. Notice that *e.g.* $\langle \hat{Z}_\alpha \hat{U}_\alpha \rangle$ is of order $1/N$ as it contains terms of the form $\mu(\mathbf{k})$ and thus the terms in equation (66) of the form $-(2/N^3)zu\langle \hat{Z}_\alpha \hat{U}_\alpha \rangle$ are actually of order $1/N^4$.

Then

$$
\begin{aligned}
&\ln\varphi(z, \theta, u, v, w) \\
&\quad = 6izN^{-3/2}\mu(\mathbf{h}, \mathbf{k})\cos\theta + 6iuN^{-3/2}\mu(\mathbf{h}) + 6ivN^{-3/2}\mu(\mathbf{k}) \\
&\qquad + 6iwN^{-3/2}\mu(\mathbf{h}+\mathbf{k}) - \tfrac{6}{4}z^2N^{-3} - \tfrac{6}{2}u^2N^{-3} - \tfrac{6}{2}v^2N^{-3} \\
&\qquad - \tfrac{6}{2}w^2N^{-3} - (2/N^3)zu\langle \hat{Z}_\alpha \hat{U}_\alpha \rangle - (2/N^3)zv\langle \hat{Z}_\alpha \hat{V}_\alpha \rangle \\
&\qquad - (2/N^3)zw\langle \hat{Z}_\alpha \hat{W}_\alpha \rangle - (6i/N^4N^{1/2})zuv \\
&\qquad - (6i/N^4N^{1/2})zuw - (6i/N^4N^{1/2})zvw + \text{h.o.}
\end{aligned} \tag{67}
$$

It then follows from equation (67) [and recall that $K = N(N-1)(N-2)/6$]

$$
\begin{aligned}
&\varphi(z,\theta,u,v,w)^K\\
&\quad= \exp\bigg[\frac{(N-1)(N-2)}{N^{1/2}} iz\mu(\mathbf{h},\mathbf{k})\cos\theta\\
&\qquad+\frac{(N-1)(N-2)}{N^{1/2}} iu\mu(\mathbf{h})\\
&\qquad+\frac{(N-1)(N-2)}{N^{1/2}} iv\mu(\mathbf{k})+\frac{(N-1)(N-2)}{N^{1/2}} iw\mu(\mathbf{h}+\mathbf{k})\\
&\qquad-\frac{(N-1)(N-2)}{N^2}\tfrac{1}{4}z^2-\frac{(N-1)(N-2)}{N^2}\tfrac{1}{2}u^2\\
&\qquad-\frac{(N-1)(N-2)}{N^2}\tfrac{1}{2}v^2-\frac{(N-1)(N-2)}{N^2}\tfrac{1}{2}w^2\\
&\qquad-\tfrac{1}{3}zu\langle\hat{Z}_\alpha\hat{U}_\alpha\rangle-\tfrac{1}{3}zv\langle\hat{Z}_\alpha\hat{V}_\alpha\rangle-\tfrac{1}{3}zw\langle\hat{Z}_\alpha\hat{W}_\alpha\rangle\\
&\qquad-\frac{i}{NN^{1/2}}zuv\cos\theta-\frac{i}{NN^{1/2}}zuw\cos\theta\\
&\qquad-\frac{i}{NN^{1/2}}zvw\cos\theta+\text{h.o.}\bigg]\\
&\quad= \exp\bigg[\frac{(N-1)(N-2)}{N^{1/2}} iz\mu(\mathbf{h},\mathbf{k})\cos\theta+NN^{1/2}iu\mu(\mathbf{h})\\
&\qquad+NN^{1/2}iv\mu(\mathbf{k})+NN^{1/2}iw\mu(\mathbf{h}+\mathbf{k})\\
&\qquad-\tfrac{1}{4}z^2-\tfrac{1}{2}u^2-\tfrac{1}{2}v^2-\tfrac{1}{2}w^2\\
&\qquad-\tfrac{1}{3}zu\langle\hat{Z}_\alpha\hat{U}_\alpha\rangle-\tfrac{1}{3}zv\langle\hat{Z}_\alpha\hat{V}_\alpha\rangle-\tfrac{1}{3}zw\langle\hat{Z}_\alpha\hat{W}_\alpha\rangle\\
&\qquad-\frac{i}{NN^{1/2}}zuv\cos\theta-\frac{i}{NN^{1/2}}zuw\cos\theta\\
&\qquad-\frac{i}{NN^{1/2}}zvw\cos\theta+\text{h.o.}\bigg],
\end{aligned}\tag{68}
$$

where here h.o. denotes terms of order $1/N$ or higher that do not contribute to the phase determination. Substituting equation (68) into equation (61) yields

$$
\begin{aligned}
&P(\psi,|Z|,\ldots,W)\\
&\quad\propto\int\limits_0^{2\pi}\mathrm{d}\theta\int\limits_0^{\infty}z\,\mathrm{d}z\int\limits_{-\infty}^{\infty}\mathrm{d}u\int\limits_{-\infty}^{\infty}\mathrm{d}v\int\limits_{-\infty}^{\infty}\mathrm{d}w\,\exp\{-iz[|Z|\cos\psi\\
&\qquad-m(\mathbf{h},\mathbf{k})]\cos\theta-iz|Z|\sin\psi\sin\theta-iu[U-m(\mathbf{h})]\\
&\qquad-iv[V-m(\mathbf{k})]-iw[W-m(\mathbf{h}+\mathbf{k})]\\
&\qquad-\tfrac{1}{4}z^2-\tfrac{1}{2}u^2-\tfrac{1}{2}v^2-\tfrac{1}{2}w^2\}\\
&\qquad\times\bigg(1-\tfrac{1}{3}zu\langle\hat{Z}_\alpha\hat{U}_\alpha\rangle-\tfrac{1}{3}zv\langle\hat{Z}_\alpha\hat{V}_\alpha\rangle-\tfrac{1}{3}zw\langle\hat{Z}_\alpha\hat{W}_\alpha\rangle\\
&\qquad-\frac{i}{NN^{1/2}}zuv\cos\theta-\frac{i}{NN^{1/2}}zuw\cos\theta\\
&\qquad-\frac{i}{NN^{1/2}}zvw\cos\theta+\text{h.o.}\bigg),
\end{aligned}\tag{69}
$$

where $m(\mathbf{h},\mathbf{k})$ and $m(\mathbf{h})$ are given by

$$
\begin{aligned}
m(\mathbf{h},\mathbf{k})&=\frac{1}{N^{1/2}}(R_\mathbf{h}^2+R_\mathbf{k}^2+R_{\mathbf{h}+\mathbf{k}}^2-2)\\
&\qquad+\frac{(N-1)(N-2)}{N^{1/2}}\mu(\mathbf{h},\mathbf{k})\quad\text{[see equation (63)]}\\
&\simeq\frac{1}{N^{1/2}}(R_\mathbf{h}^2+R_\mathbf{k}^2+R_{\mathbf{h}+\mathbf{k}}^2-2)+NN^{1/2}\mu(\mathbf{h},\mathbf{k})\\
m(\mathbf{h})&=N^{1/2}+\frac{2}{N^{1/2}}(R_\mathbf{h}^2-1)+\frac{(N-1)(N-2)}{N^{1/2}}\mu(\mathbf{h})\\
&\qquad\text{[see equation (63)]}\\
&\simeq N^{1/2}+\frac{2}{N^{1/2}}(R_\mathbf{h}^2-1)+NN^{1/2}\mu(\mathbf{h}).
\end{aligned}\tag{70}
$$

Next we calculate the conditional j.p.d. $P(\psi)\equiv P(\psi\big||Z|=R_\mathbf{h}R_\mathbf{k}R_{\mathbf{h}+\mathbf{k}},\ U=N^{1/2}R_\mathbf{h}^2,\ V=N^{1/2}R_\mathbf{k}^2,\ W=N^{1/2}R_{\mathbf{h}+\mathbf{k}}^2)$ and we define

$$
\begin{aligned}
\delta_\mathbf{h}&=N^{1/2}[R_\mathbf{h}^2-1-N\mu(\mathbf{h})-(2/N)(R_\mathbf{h}^2-1)],\\
\delta_\mathbf{k}&=N^{1/2}[R_\mathbf{k}^2-1-N\mu(\mathbf{k})-(2/N)(R_\mathbf{k}^2-1)],\\
\delta_{\mathbf{h}+\mathbf{k}}&=N^{1/2}[R_{\mathbf{h}+\mathbf{k}}^2-1-N\mu(\mathbf{h}+\mathbf{k})-(2/N)(R_{\mathbf{h}+\mathbf{k}}^2-1)],\\
a&=R_\mathbf{h}R_\mathbf{k}R_{\mathbf{h}+\mathbf{k}}\cos\psi-m(\mathbf{h},\mathbf{k}),\\
b&=R_\mathbf{h}R_\mathbf{k}R_{\mathbf{h}+\mathbf{k}}\sin\psi,\\
\tan\alpha&=b/a,\\
Q&=(a^2+b^2)^{1/2}.
\end{aligned}\tag{71}
$$

Because of equation (64) $\delta_\mathbf{h}$, $\delta_\mathbf{k}$ and $\delta_{\mathbf{h}+\mathbf{k}}$ are of order $1/N^{1/2}$. Then

$$
\begin{aligned}
P(\psi)&\propto\int\limits_0^{2\pi}\mathrm{d}\theta\int\limits_0^{\infty}z\,\mathrm{d}z\int\limits_{-\infty}^{\infty}\mathrm{d}u\int\limits_{-\infty}^{\infty}\mathrm{d}v\int\limits_{-\infty}^{\infty}\mathrm{d}w\,\exp[-izQ\cos(\theta-\alpha)]\\
&\quad\times\exp(-iu\delta_\mathbf{h})\exp(-iv\delta_\mathbf{k})\exp(-iw\delta_{\mathbf{h}+\mathbf{k}})\\
&\quad\times\exp(-\tfrac{1}{4}z^2-\tfrac{1}{2}u^2-\tfrac{1}{2}v^2-\tfrac{1}{2}w^2)\\
&\quad\times[1-\tfrac{1}{3}zu\langle\hat{Z}_\alpha\hat{U}_\alpha\rangle-\tfrac{1}{3}zv\langle\hat{Z}_\alpha\hat{V}_\alpha\rangle-\tfrac{1}{3}zw\langle\hat{Z}_\alpha\hat{W}_\alpha\rangle\\
&\quad-(i/NN^{1/2})zuv\cos\theta-(i/NN^{1/2})zuw\cos\theta\\
&\quad-(i/NN^{1/2})zvw\cos\theta+\text{h.o.}].
\end{aligned}\tag{72}
$$

Then doing the u, v, w integrations using formulas (151) and (152) we get

$$
\begin{aligned}
P(\psi)&\propto\int\limits_0^{2\pi}\mathrm{d}\theta\int\limits_0^{\infty}z\,\mathrm{d}z\exp[-izQ\cos(\theta-\alpha)]\exp(-\tfrac{1}{4}z^2)\\
&\quad\times\exp(-\tfrac{1}{2}\delta_\mathbf{h}^2-\tfrac{1}{2}\delta_\mathbf{k}^2-\tfrac{1}{2}\delta_{\mathbf{h}+\mathbf{k}}^2)\\
&\quad\times[1+\tfrac{1}{3}iz\cos\theta(\delta_\mathbf{h}\langle\hat{Z}_\alpha\hat{U}_\alpha\rangle+\delta_\mathbf{k}\langle\hat{Z}_\alpha\hat{V}_\alpha\rangle+\delta_{\mathbf{h}+\mathbf{k}}\langle\hat{Z}_\alpha\hat{W}_\alpha\rangle)\\
&\quad+(1/NN^{1/2})iz(\delta_\mathbf{h}\delta_\mathbf{k}+\delta_\mathbf{h}\delta_{\mathbf{h}+\mathbf{k}}+\delta_\mathbf{k}\delta_{\mathbf{h}+\mathbf{k}})\cos\theta+\text{h.o.}].
\end{aligned}\tag{73}
$$

Doing now the θ, z integrations in equation (73) we get

$$
\begin{aligned}
P(\psi)&\propto\exp(-Q^2)\bigg\{1+Q\cos\alpha\bigg[\tfrac{2}{3}(\delta_\mathbf{h}\langle\hat{Z}_\alpha\hat{U}_\alpha\rangle+\delta_\mathbf{k}\langle\hat{Z}_\alpha\hat{V}_\alpha\rangle\\
&\qquad+\delta_{\mathbf{h}+\mathbf{k}}\langle\hat{Z}_\alpha\hat{W}_\alpha\rangle)+\frac{2}{NN^{1/2}}(\delta_\mathbf{h}\delta_\mathbf{k}+\delta_\mathbf{h}\delta_{\mathbf{h}+\mathbf{k}}+\delta_\mathbf{k}\delta_{\mathbf{h}+\mathbf{k}})\bigg]\\
&\qquad+O\Big(\frac{1}{N^2}\Big)\bigg\}
\end{aligned}
$$

$$\propto \exp\Big\{-Q^2 + Q\cos\alpha\Big[\tfrac{2}{3}(\delta_{\mathbf{h}}\langle\hat{Z}_\alpha\hat{U}_\alpha\rangle + \delta_{\mathbf{k}}\langle\hat{Z}_\alpha\hat{V}_\alpha\rangle + \delta_{\mathbf{h+k}}\langle\hat{Z}_\alpha\hat{W}_\alpha\rangle) + \frac{2}{NN^{1/2}}(\delta_{\mathbf{h}}\delta_{\mathbf{k}} + \delta_{\mathbf{h}}\delta_{\mathbf{h+k}} + \delta_{\mathbf{k}}\delta_{\mathbf{h+k}})\Big] + O\Big(\frac{1}{N^2}\Big)\Big\}$$

$$\propto \exp\Big[2R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}A(\mathbf{h},\mathbf{k})\cos\psi + O\Big(\frac{1}{N^2}\Big)\Big], \qquad (74)$$

where

$$A(\mathbf{h},\mathbf{k}) = m(\mathbf{h},\mathbf{k}) + \tfrac{1}{3}(\delta_{\mathbf{h}}\langle\hat{Z}_\alpha\hat{U}_\alpha\rangle + \delta_{\mathbf{k}}\langle\hat{Z}_\alpha\hat{V}_\alpha\rangle + \delta_{\mathbf{h+k}}\langle\hat{Z}_\alpha\hat{W}_\alpha\rangle) + (1/NN^{1/2})(\delta_{\mathbf{h}}\delta_{\mathbf{k}} + \delta_{\mathbf{h}}\delta_{\mathbf{h+k}} + \delta_{\mathbf{k}}\delta_{\mathbf{h+k}}). \qquad (75)$$

It is now interesting to calculate $P(\psi)$ when we choose $f(\mathbf{u}_\alpha, \mathbf{v}_\alpha) \equiv 1$ [see equation (60)] instead. Then $\mu(\mathbf{h},\mathbf{k}) = 0$ and $\mu(\mathbf{h}) = 0$. It then follows from equations (70), (71) and (65) that

$$\begin{aligned} m(\mathbf{h},\mathbf{k}) &= (1/N^{1/2})(R_{\mathbf{h}}^2 + R_{\mathbf{k}}^2 + R_{\mathbf{h+k}}^2 - 2),\\ m(\mathbf{h}) &= N^{1/2} + (2/N^{1/2})(R_{\mathbf{h}}^2 - 1),\\ \delta_{\mathbf{h}} &= N^{1/2}[R_{\mathbf{h}}^2 - 1 - (2/N)(R_{\mathbf{h}}^2 - 1)],\\ &= N^{1/2}(R_{\mathbf{h}}^2 - 1) + O(1/N^{1/2}),\\ \langle\hat{Z}_\alpha\hat{U}_\alpha\rangle &= \langle\hat{Z}_\alpha\hat{V}_\alpha\rangle = \langle\hat{Z}_\alpha\hat{W}_\alpha\rangle = 0. \end{aligned} \qquad (76)$$

Then $A(\mathbf{h},\mathbf{k})$ in equation (75) becomes

$$A(\mathbf{h},\mathbf{k}) = (1/N^{1/2})[R_{\mathbf{h}}^2 + R_{\mathbf{k}}^2 + R_{\mathbf{h+k}}^2 - 2 + (R_{\mathbf{h}}^2 - 1)(R_{\mathbf{k}}^2 - 1) + (R_{\mathbf{h}}^2 - 1)(R_{\mathbf{h+k}}^2 - 1) + (R_{\mathbf{k}}^2 - 1)(R_{\mathbf{h+k}}^2 - 1)]. \qquad (77)$$

This is exactly the result for the approximate value of $m(\mathbf{h},\mathbf{k})$ given in equation (55).

6. The quartet in *P*1 using SAD

We consider the seven r.v.'s $\hat{E}_1 = \hat{E}_{\mathbf{h}}$, $\hat{E}_2 = \hat{E}_{\mathbf{k}}$, $\hat{E}_3 = \hat{E}_{\mathbf{l}}$, $\hat{E}_4 = \hat{E}_{\mathbf{h+k+l}}$, $\hat{E}_5 = \hat{E}_{\mathbf{h+k}}$, $\hat{E}_6 = \hat{E}_{\mathbf{h+l}}$ and $\hat{E}_7 = \hat{E}_{\mathbf{k+l}}$; we likewise define $\hat{\varphi}_1, \hat{\varphi}_2, \ldots, \hat{\varphi}_7$. We are interested in the calculation of the j.p.d.

$$P(\bar{R}_1, \bar{R}_2, \bar{R}_3, \bar{R}_4, \bar{R}_5, \bar{R}_6, \bar{R}_7, \bar{\varphi}_1, \bar{\varphi}_2, \bar{\varphi}_3, \bar{\varphi}_4, \bar{\varphi}_5, \bar{\varphi}_6, \bar{\varphi}_7) \equiv \frac{\bar{R}_1\ldots\bar{R}_7}{(2\pi)^{14}}\int_0^{2\pi}\mathrm{d}\theta_1\ldots\int_0^\infty \rho_1\,\mathrm{d}\rho_1\ldots\exp[-i\rho_1\bar{R}_1 \times\cos(\bar{\varphi}_1 - \theta_1) - \ldots]\phi(\rho_1,\ldots,\theta_7), \qquad (78)$$

where

$$\phi(\rho_1,\ldots,\theta_7) = \chi(\rho_1,\ldots,\theta_7)^\mu \qquad (79)$$

and

$$\chi(\rho_1,\ldots,\theta_7) = \langle\exp[(1/\mu^{1/2})i\rho_1\hat{R}_1\cos(\hat{\varphi}_1 - \theta_1) + \ldots + (1/\mu^{1/2})i\rho_7\hat{R}_7\cos(\hat{\varphi}_7 - \theta_7)]\rangle \qquad (80)$$

and where $\langle\ldots\rangle$ means $\langle\ldots\rangle_f$ with respect to an arbitrary even and translation-invariant j.p.d. $f(\mathbf{x}_1,\ldots,\mathbf{x}_N)$. Then developing $\langle\exp[(1/\mu^{1/2})i\rho_1\hat{R}_1\cos(\hat{\varphi}_1 - \theta_1) + \ldots + (1/\mu^{1/2})i\rho_7\hat{R}_7\cos(\hat{\varphi}_7 - \theta_7)]\rangle$ asymptotically according to powers of $1/\mu^{1/2}$ we can write

$$\begin{aligned}\chi(\rho_1,\ldots,\theta_7) &= \Big\langle\Big\{J_0\Big(\frac{\rho_1}{\mu^{1/2}}\hat{R}_1\Big) + 2\sum_{k=1}^\infty i^kJ_k\Big(\frac{\rho_1}{\mu^{1/2}}\hat{R}_1\Big)\times\cos[k(\hat{\varphi}_1 - \theta_1)]\Big\}\times\ldots\times\Big\{J_0\Big(\frac{\rho_7}{\mu^{1/2}}\hat{R}_7\Big) + 2\sum_{k=1}^\infty i^kJ_k\Big(\frac{\rho_7}{\mu^{1/2}}\hat{R}_7\Big)\cos[k(\hat{\varphi}_7 - \theta_7)]\Big\}\Big\rangle\\ &= \Big\langle J_0\Big(\frac{\rho_1}{\mu^{1/2}}\hat{R}_1\Big)\ldots J_0\Big(\frac{\rho_7}{\mu^{1/2}}\hat{R}_7\Big)\Big\rangle\\ &\quad + 2^4i^4\tfrac{1}{8}\Big\langle J_1\Big(\frac{\rho_1}{\mu^{1/2}}\hat{R}_1\Big)\ldots J_1\Big(\frac{\rho_4}{\mu^{1/2}}\hat{R}_4\Big)\times\cos(\hat{\varphi}_1 + \hat{\varphi}_2 + \hat{\varphi}_3 - \hat{\varphi}_4)\Big\rangle\cos(\theta_1 + \theta_2 + \theta_3 - \theta_4)\\ &\quad + 2^3i^3\tfrac{1}{4}\Big\langle J_1\Big(\frac{\rho_1}{\mu^{1/2}}\hat{R}_1\Big)J_1\Big(\frac{\rho_2}{\mu^{1/2}}\hat{R}_2\Big)J_1\Big(\frac{\rho_5}{\mu^{1/2}}\hat{R}_5\Big)\times\cos(\hat{\varphi}_1 + \hat{\varphi}_2 - \hat{\varphi}_5)\Big\rangle\cos(\theta_1 + \theta_2 - \theta_5) + \ldots\\ &\quad + 2^3i^3\tfrac{1}{4}\Big\langle J_1\Big(\frac{\rho_1}{\mu^{1/2}}\hat{R}_1\Big)J_1\Big(\frac{\rho_4}{\mu^{1/2}}\hat{R}_4\Big)J_1\Big(\frac{\rho_7}{\mu^{1/2}}\hat{R}_7\Big)\times\cos(\hat{\varphi}_1 - \hat{\varphi}_4 + \hat{\varphi}_7)\Big\rangle\cos(\theta_1 - \theta_4 + \theta_7)\\ &\quad + O\Big(\frac{1}{\mu^2\mu^{1/2}}\Big).\end{aligned} \qquad (81)$$

Then

$$\begin{aligned}\chi(\rho_1,\ldots,\theta_7) &= \Big\langle\Big(1 - \frac{1}{4\mu}\rho_1^2\hat{R}_1^2 + \frac{1}{4^3\mu^2}\rho_1^4\hat{R}_1^4 + \ldots\Big)\times\ldots\times\Big(1 - \frac{1}{4\mu}\rho_7^2\hat{R}_7^2 + \frac{1}{4^3\mu^2}\rho_7^4\hat{R}_7^4 + \ldots\Big)\Big\rangle\\ &\quad + 2^3i^3\tfrac{1}{4}\Big\langle\Big(\frac{\rho_1}{2\mu^{1/2}}\hat{R}_1\Big)\Big(\frac{\rho_2}{2\mu^{1/2}}\hat{R}_2\Big)\Big(\frac{\rho_5}{2\mu^{1/2}}\hat{R}_5\Big)\times\cos(\hat{\varphi}_1 + \hat{\varphi}_2 - \hat{\varphi}_5)\Big\rangle\cos(\theta_1 + \theta_2 - \theta_5) + \ldots\\ &\quad + 2^3i^3\tfrac{1}{4}\Big\langle\Big(\frac{\rho_1}{2\mu^{1/2}}\hat{R}_1\Big)\Big(\frac{\rho_4}{2\mu^{1/2}}\hat{R}_4\Big)\Big(\frac{\rho_7}{2\mu^{1/2}}\hat{R}_7\Big)\times\cos\Big(\hat{\varphi}_1 - \hat{\varphi}_4 + \hat{\varphi}_7\Big)\Big\rangle\cos(\theta_1 - \theta_4 + \theta_7)\\ &\quad + 2^4i^4\tfrac{1}{8}\Big\langle\Big(\frac{\rho_1}{2\mu^{1/2}}\hat{R}_1\Big)\times\ldots\times\Big(\frac{\rho_4}{2\mu^{1/2}}\hat{R}_4\Big)\times\cos(\hat{\varphi}_1 + \ldots - \hat{\varphi}_4)\Big\rangle\cos(\theta_1 + \theta_2 + \theta_3 - \theta_4)\\ &\quad + O(\mu^{-5/2}).\end{aligned} \qquad (82)$$

It then follows that

$$
\begin{aligned}
&\chi(\rho_1, \ldots, \theta_7)\\
&\quad = 1 - \frac{1}{4\mu}\rho_1^2\langle\hat{R}_1^2\rangle \ldots - \frac{1}{4\mu}\rho_7^2\langle\hat{R}_7^2\rangle\\
&\quad\quad - \frac{i}{4\mu\mu^{1/2}}\rho_1\rho_2\rho_5\langle\hat{R}_1\hat{R}_2\hat{R}_5\cos(\hat{\varphi}_1+\hat{\varphi}_2-\hat{\varphi}_5)\rangle\\
&\quad\quad \times\cos(\theta_1+\theta_2-\theta_5) + \ldots\\
&\quad\quad - \frac{i}{4\mu\mu^{1/2}}\rho_1\rho_4\rho_7\langle\hat{R}_1\hat{R}_4\hat{R}_7\cos(\hat{\varphi}_1-\hat{\varphi}_4+\hat{\varphi}_7)\rangle\\
&\quad\quad \times\cos(\theta_1-\theta_4+\theta_7)\\
&\quad\quad + \frac{1}{8\mu^2}\rho_1\rho_2\rho_3\rho_4\langle\hat{R}_1\hat{R}_2\hat{R}_3\hat{R}_4\cos(\hat{\varphi}_1+\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_4)\rangle\\
&\quad\quad \times\cos(\theta_1+\theta_2+\theta_3-\theta_4)\\
&\quad\quad + \frac{1}{16\mu^2}\rho_1^2\rho_2^2\langle\hat{R}_1^2\hat{R}_2^2\rangle + \ldots + \frac{1}{16\mu^2}\rho_6^2\rho_7^2\langle\hat{R}_6^2\hat{R}_7^2\rangle\\
&\quad\quad + \frac{1}{4^3\mu^2}\rho_1^4\langle\hat{R}_1^4\rangle + \ldots + \frac{1}{4^3\mu^2}\rho_7^4\langle\hat{R}_7^4\rangle + O(\mu^{-5/2}). \qquad (83)
\end{aligned}
$$

Developing asymptotically $\chi(\rho_1, \ldots, \theta_7)$ according to powers of $1/\mu^{1/2}$ using $\ln(1+x) = x - \frac{1}{2}x^2 + \frac{1}{3}x^3 + \ldots$, we can write

$$
\begin{aligned}
&\ln\chi(\rho_1, \ldots, \theta_7)\\
&\quad = -\frac{1}{4\mu}\rho_1^2\langle\hat{R}_1^2\rangle - \ldots - \frac{1}{4\mu}\rho_7^2\langle\hat{R}_7^2\rangle\\
&\quad\quad - \frac{i}{4\mu\mu^{1/2}}\rho_1\rho_2\rho_5\langle\hat{R}_1\hat{R}_2\hat{R}_5\cos(\hat{\varphi}_1+\hat{\varphi}_2-\hat{\varphi}_5)\rangle\\
&\quad\quad \times\cos(\theta_1+\theta_2-\theta_5) + \ldots\\
&\quad\quad - \frac{i}{4\mu\mu^{1/2}}\rho_1\rho_4\rho_7\langle\hat{R}_1\hat{R}_4\hat{R}_7\cos(\hat{\varphi}_1-\hat{\varphi}_4+\hat{\varphi}_7)\rangle\\
&\quad\quad \times\cos(\theta_1-\theta_4+\theta_7)\\
&\quad\quad + \frac{1}{8\mu^2}\rho_1\rho_2\rho_3\rho_4\langle\hat{R}_1\hat{R}_2\hat{R}_3\hat{R}_4\cos(\hat{\varphi}_1+\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_4)\rangle\\
&\quad\quad \times\cos(\theta_1+\theta_2+\theta_3-\theta_4)\\
&\quad\quad + \frac{1}{16\mu^2}\rho_1^2\rho_2^2(\langle\hat{R}_1^2\hat{R}_2^2\rangle - \langle\hat{R}_1^2\rangle\langle\hat{R}_2^2\rangle) + \ldots\\
&\quad\quad + \frac{1}{16\mu^2}\rho_6^2\rho_7^2(\langle\hat{R}_6^2\hat{R}_7^2\rangle - \langle\hat{R}_6^2\rangle\langle\hat{R}_7^2\rangle)\\
&\quad\quad + \frac{1}{4^3\mu^2}\rho_1^4(\langle\hat{R}_1^4\rangle - 2\langle\hat{R}_1^2\rangle^2) + \ldots + \frac{1}{4^3\mu^2}\rho_7^4(\langle\hat{R}_7^4\rangle - 2\langle\hat{R}_7^2\rangle^2)\\
&\quad\quad + O(\mu^{-5/2}). \qquad (84)
\end{aligned}
$$

Developing now $\exp[\mu\ln\chi(\rho_1, \ldots, \theta_7)]$ asymptotically in μ using $\exp(x) = 1 + x + \frac{1}{2}x^2 + \ldots$, we get

$$
\begin{aligned}
&\exp[\mu\ln\chi(\rho_1, \ldots, \theta_7)]\\
&\quad = \exp(-\tfrac{1}{4}\rho_1^2\langle\hat{R}_1^2\rangle - \ldots - \tfrac{1}{4}\rho_7^2\langle\hat{R}_7^2\rangle)\\
&\quad\quad \times\Big[1 - \frac{i}{4\mu^{1/2}}\rho_1\rho_2\rho_5\langle\hat{R}_1\hat{R}_2\hat{R}_5\cos(\hat{\varphi}_1+\hat{\varphi}_2-\hat{\varphi}_5)\rangle\\
&\quad\quad \times\cos(\theta_1+\theta_2-\theta_5) + \ldots\\
&\quad\quad - \frac{i}{4\mu^{1/2}}\rho_1\rho_4\rho_7\langle\hat{R}_1\hat{R}_4\hat{R}_7\cos(\hat{\varphi}_1-\hat{\varphi}_4+\hat{\varphi}_7)\rangle\\
&\quad\quad \times\cos(\theta_1-\theta_4+\theta_7)\\
&\quad\quad + \frac{1}{8\mu}\rho_1\rho_2\rho_3\rho_4\langle\hat{R}_1\hat{R}_2\hat{R}_3\hat{R}_4\cos(\hat{\varphi}_1+\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_4)\rangle\\
&\quad\quad \times\cos(\theta_1+\theta_2+\theta_3-\theta_4)\\
&\quad\quad - \frac{1}{2^5\mu}\rho_1\rho_2\rho_3\rho_4\rho_5^2\langle\hat{R}_1\hat{R}_2\hat{R}_5\cos(\hat{\varphi}_1+\hat{\varphi}_2-\hat{\varphi}_5)\rangle\\
&\quad\quad \times\langle\hat{R}_3\hat{R}_4\hat{R}_5\cos(\hat{\varphi}_3-\hat{\varphi}_4+\hat{\varphi}_5)\rangle\\
&\quad\quad \times\cos(\theta_1+\theta_2+\theta_3-\theta_4) + \ldots\\
&\quad\quad - \frac{1}{2^5\mu}\rho_1\rho_2\rho_3\rho_4\rho_7^2\langle\hat{R}_2\hat{R}_3\hat{R}_7\cos(\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_7)\rangle\\
&\quad\quad \times\langle\hat{R}_1\hat{R}_4\hat{R}_7\cos(\hat{\varphi}_1-\hat{\varphi}_4+\hat{\varphi}_7)\rangle\cos(\theta_1+\theta_2+\theta_3-\theta_4)\\
&\quad\quad + \frac{1}{4^3\mu}\rho_1^4(\langle\hat{R}_1^4\rangle - 2\langle\hat{R}_1^2\rangle^2) + \ldots + \frac{1}{4^3\mu}\rho_7^4(\langle\hat{R}_7^4\rangle - 2\langle\hat{R}_7^2\rangle^2)\\
&\quad\quad - \frac{1}{32\mu}\rho_1^2\rho_2^2\rho_5^2\langle\hat{R}_1\hat{R}_2\hat{R}_5\cos(\hat{\varphi}_1+\hat{\varphi}_2-\hat{\varphi}_5)\rangle^2\\
&\quad\quad \times\cos^2(\theta_1+\theta_2-\theta_5) + \ldots\\
&\quad\quad - \frac{1}{16\mu}\rho_1^2\rho_2\rho_3\rho_5\rho_6\langle\hat{R}_1\hat{R}_2\hat{R}_5\cos(\hat{\varphi}_1+\hat{\varphi}_2-\hat{\varphi}_5)\rangle\\
&\quad\quad \times\langle\hat{R}_1\hat{R}_3\hat{R}_6\cos(\hat{\varphi}_1+\hat{\varphi}_3-\hat{\varphi}_6)\rangle\cos(\theta_1+\theta_2-\theta_5)\\
&\quad\quad \times\cos(\theta_1+\theta_3-\theta_6) + \ldots + O(\mu^{-3/2})\Big]. \qquad (85)
\end{aligned}
$$

Then equation (78) becomes

$$
\begin{aligned}
&P(\bar{R}_1, \ldots, \bar{\varphi}_7)\\
&\quad = \frac{\bar{R}_1\ldots\bar{R}_7}{(2\pi)^{14}}\int_0^\infty\rho_1\,\mathrm{d}\rho_1\ldots\int_0^{2\pi}\mathrm{d}\theta_1\ldots\exp[-i\rho_1\bar{R}_1\\
&\quad\quad \times\cos(\theta_1-\bar{\varphi}_1) - \ldots]\phi(\rho_1, \ldots, \theta_7)\\
&\quad \propto \bar{R}_1\ldots\bar{R}_7\int_0^\infty\rho_1\,\mathrm{d}\rho_1\ldots\int_0^{2\pi}\mathrm{d}\theta_1\ldots\exp[-i\rho_1\bar{R}_1\\
&\quad\quad \times\cos(\theta_1-\bar{\varphi}_1) - \ldots]\exp(-\tfrac{1}{4}\rho_1^2\langle\hat{R}_1^2\rangle)\times\ldots\\
&\quad\quad \times\exp(-\tfrac{1}{4}\rho_7^2\langle\hat{R}_7^2\rangle)\\
&\quad\quad \times\Big[1 - \frac{i}{4\mu^{1/2}}\rho_1\rho_2\rho_5\langle\hat{R}_1\hat{R}_2\hat{R}_5\cos(\hat{\varphi}_1+\hat{\varphi}_2-\hat{\varphi}_5)\rangle\\
&\quad\quad \times\cos(\theta_1+\theta_2-\theta_5) + \ldots\\
&\quad\quad - \frac{i}{4\mu^{1/2}}\rho_1\rho_4\rho_7\langle\hat{R}_1\hat{R}_4\hat{R}_7\cos(\hat{\varphi}_1-\hat{\varphi}_4+\hat{\varphi}_7)\rangle\\
&\quad\quad \times\cos(\theta_1-\theta_4+\theta_7)\\
&\quad\quad + \frac{1}{8\mu}\rho_1\rho_2\rho_3\rho_4\langle\hat{R}_1\hat{R}_2\hat{R}_3\hat{R}_4\cos(\hat{\varphi}_1+\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_4)\rangle\\
&\quad\quad \times\cos(\theta_1+\theta_2+\theta_3-\theta_4)\\
&\quad\quad - \frac{1}{2^5\mu}\rho_1\rho_2\rho_3\rho_4\rho_5^2\langle\hat{R}_1\hat{R}_2\hat{R}_5\cos(\hat{\varphi}_1+\hat{\varphi}_2-\hat{\varphi}_5)\rangle\\
&\quad\quad \times\langle\hat{R}_3\hat{R}_4\hat{R}_5\cos(\hat{\varphi}_3-\hat{\varphi}_4+\hat{\varphi}_5)\rangle\\
&\quad\quad \times\cos(\theta_1+\theta_2+\theta_3-\theta_4) + \ldots\\
&\quad\quad - \frac{1}{2^5\mu}\rho_1\rho_2\rho_3\rho_4\rho_7^2\langle\hat{R}_2\hat{R}_3\hat{R}_7\cos(\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_7)\rangle\\
&\quad\quad \times\langle\hat{R}_1\hat{R}_4\hat{R}_7\cos(\hat{\varphi}_1-\hat{\varphi}_4+\hat{\varphi}_7)\rangle\cos(\theta_1+\theta_2+\theta_3-\theta_4)\\
&\quad\quad + \frac{1}{16\mu}\rho_1^2\rho_2^2(\langle\hat{R}_1^2\hat{R}_2^2\rangle - \langle\hat{R}_1^2\rangle\langle\hat{R}_2^2\rangle) + \ldots
\end{aligned}
$$

$$+\ \frac{1}{4^3\mu}\rho_1^4(\langle\hat{R}_1^4\rangle-2\langle\hat{R}_1^2\rangle^2)+\ldots+\frac{1}{4^3\mu}\rho_7^4(\langle\hat{R}_7^4\rangle-2\langle\hat{R}_7^2\rangle^2)$$
$$+O'\left(\frac{1}{\mu}\right)+O\left(\frac{1}{\mu\mu^{1/2}}\right)\bigg], \quad (86)$$

where $O'(1/\mu)$ does not contain constants and only contains phases of order $1/\mu$ that do not contain $\cos(\theta_1+\theta_2+\theta_3-\theta_4)$.

Applying the transformation $\rho_i\to\rho_i/\langle R_i^2\rangle^{1/2}$ and doing the θ_i integrations in equation (86), equation (86) becomes

$$
\begin{aligned}
&P(\bar{R}_1,\ldots,\bar{\varphi}_7)\\
&\quad\propto \bar{R}_1\ldots\bar{R}_7\int_0^\infty\rho_1\,\mathrm{d}\rho_1\ldots\exp(-\tfrac{1}{4}\rho_1^2-\ldots-\tfrac{1}{4}\rho_7^2)\\
&\quad\times\bigg[J_0\left(\rho_1\frac{\bar{R}_1}{\langle\hat{R}_1^2\rangle^{1/2}}\right)\times\ldots\times J_0\left(\rho_7\frac{\bar{R}_7}{\langle\hat{R}_7^2\rangle^{1/2}}\right)\\
&\quad-\frac{i(-i)^3}{4\mu^{1/2}}\frac{\rho_1\rho_2\rho_5}{\langle\hat{R}_1^2\rangle^{1/2}\langle\hat{R}_2^2\rangle^{1/2}\langle\hat{R}_5^2\rangle^{1/2}}\\
&\quad\times\langle\hat{R}_1\hat{R}_2\hat{R}_5\cos(\hat{\varphi}_1+\hat{\varphi}_2-\hat{\varphi}_5)\rangle\\
&\quad\times J_1\left(\rho_1\frac{\bar{R}_1}{\langle\hat{R}_1^2\rangle^{1/2}}\right)J_1\left(\rho_2\frac{\bar{R}_2}{\langle\hat{R}_2^2\rangle^{1/2}}\right)J_1\left(\rho_5\frac{\bar{R}_5}{\langle\hat{R}_5^2\rangle^{1/2}}\right)\\
&\quad\times J_0\left(\rho_3\frac{\bar{R}_3}{\langle\hat{R}_3^2\rangle^{1/2}}\right)J_0\left(\rho_4\frac{\bar{R}_4}{\langle\hat{R}_4^2\rangle^{1/2}}\right)\\
&\quad\times J_0\left(\rho_6\frac{\bar{R}_6}{\langle\hat{R}_6^2\rangle^{1/2}}\right)J_0\left(\rho_7\frac{\bar{R}_7}{\langle\hat{R}_7^2\rangle^{1/2}}\right)\cos(\bar{\varphi}_1+\bar{\varphi}_2-\bar{\varphi}_5)+\ldots\\
&\quad+\frac{(-i)^4}{8\mu}\frac{\rho_1\rho_2\rho_3\rho_4}{\langle\hat{R}_1^2\rangle^{1/2}\langle\hat{R}_2^2\rangle^{1/2}\langle\hat{R}_3^2\rangle^{1/2}\langle\hat{R}_4^2\rangle^{1/2}}\\
&\quad\times\langle\hat{R}_1\hat{R}_2\hat{R}_3\hat{R}_4\cos(\hat{\varphi}_1+\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_4)\rangle\\
&\quad\times\cos(\bar{\varphi}_1+\bar{\varphi}_2+\bar{\varphi}_3-\bar{\varphi}_4)\\
&\quad\times J_1\left(\rho_1\frac{\bar{R}_1}{\langle\hat{R}_1^2\rangle^{1/2}}\right)\times\ldots\times J_1\left(\rho_4\frac{\bar{R}_4}{\langle\hat{R}_4^2\rangle^{1/2}}\right)\\
&\quad\times J_0\left(\rho_5\frac{\bar{R}_5}{\langle\hat{R}_5^2\rangle^{1/2}}\right)J_0\left(\rho_6\frac{\bar{R}_6}{\langle\hat{R}_6^2\rangle^{1/2}}\right)J_0\left(\rho_7\frac{\bar{R}_7}{\langle\hat{R}_7^2\rangle^{1/2}}\right)\\
&\quad-\frac{(-i)^4}{2^5\mu}\frac{\rho_1\rho_2\rho_3\rho_4\rho_5^2}{\langle\hat{R}_1^2\rangle^{1/2}\langle\hat{R}_2^2\rangle^{1/2}\langle\hat{R}_3^2\rangle^{1/2}\langle\hat{R}_4^2\rangle^{1/2}\langle\hat{R}_5^2\rangle}\\
&\quad\times\langle\hat{R}_1\hat{R}_2\hat{R}_5\cos(\hat{\varphi}_1+\hat{\varphi}_2-\hat{\varphi}_5)\rangle\langle\hat{R}_3\hat{R}_4\hat{R}_5\cos(\hat{\varphi}_3-\hat{\varphi}_4+\hat{\varphi}_5)\rangle\\
&\quad\times J_1\left(\rho_1\frac{\bar{R}_1}{\langle\hat{R}_1^2\rangle^{1/2}}\right)\times\ldots\times J_1\left(\rho_4\frac{\bar{R}_4}{\langle\hat{R}_4^2\rangle^{1/2}}\right)\\
&\quad\times J_0\left(\rho_5\frac{\bar{R}_5}{\langle\hat{R}_5^2\rangle^{1/2}}\right)J_0\left(\rho_6\frac{\bar{R}_6}{\langle\hat{R}_6^2\rangle^{1/2}}\right)J_0\left(\rho_7\frac{\bar{R}_7}{\langle\hat{R}_7^2\rangle^{1/2}}\right)\\
&\quad\times\cos(\bar{\varphi}_1+\bar{\varphi}_2+\bar{\varphi}_3-\bar{\varphi}_4)+\ldots+O'\left(\frac{1}{\mu}\right)\bigg]. \qquad (87)
\end{aligned}
$$

In equation (87) $O'(1/\mu)$ denotes terms of order $1/\mu$ that do not contribute to the quartet phase. Using formulas (137) and (138) of the Appendix we then obtain

$$
\begin{aligned}
&P(\bar{R}_1,\ldots,\bar{\varphi}_7)\\
&\quad\propto \bar{R}_1\ldots\bar{R}_7\exp\left(-\frac{\bar{R}_1^2}{\langle\hat{R}_1^2\rangle}\ldots-\frac{\bar{R}_7^2}{\langle\hat{R}_7^2\rangle}\right)\\
&\quad\times\bigg[1+\frac{1}{4\mu^{1/2}}\frac{2^3\bar{R}_1\bar{R}_2\bar{R}_5}{\langle\hat{R}_1^2\rangle\langle\hat{R}_2^2\rangle\langle\hat{R}_5^2\rangle}\langle\hat{R}_1\hat{R}_2\hat{R}_5\cos(\hat{\varphi}_1+\hat{\varphi}_2-\hat{\varphi}_5)\rangle\\
&\quad\times\cos(\bar{\varphi}_1+\bar{\varphi}_2-\bar{\varphi}_5)+\ldots\\
&\quad+\frac{1}{8\mu}2^4\frac{\bar{R}_1\bar{R}_2\bar{R}_3\bar{R}_4}{\langle\hat{R}_1^2\rangle\ldots\langle\hat{R}_4^2\rangle}\langle\hat{R}_1\hat{R}_2\hat{R}_3\hat{R}_4\cos(\hat{\varphi}_1+\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_4)\rangle\\
&\quad\times\cos(\bar{\varphi}_1+\bar{\varphi}_2+\bar{\varphi}_3-\bar{\varphi}_4)\\
&\quad-\frac{1}{2^5\mu}(1!)2^4 2^2\frac{\bar{R}_1\ldots\bar{R}_4(1-\bar{R}_5^2/\langle\hat{R}_5^2\rangle)}{\langle\hat{R}_1^2\rangle\ldots\langle\hat{R}_4^2\rangle\langle\hat{R}_5^2\rangle}\\
&\quad\times\langle\hat{R}_1\hat{R}_2\hat{R}_5\cos(\hat{\varphi}_1+\hat{\varphi}_2-\hat{\varphi}_5)\rangle\langle\hat{R}_3\hat{R}_4\hat{R}_5\cos(\hat{\varphi}_3-\hat{\varphi}_4+\hat{\varphi}_5)\rangle\\
&\quad\times\cos(\bar{\varphi}_1+\bar{\varphi}_2+\bar{\varphi}_3-\bar{\varphi}_4)+\ldots\\
&\quad-\frac{2}{\mu}\frac{\bar{R}_1\ldots\bar{R}_4(1-\bar{R}_7^2/\langle\hat{R}_7^2\rangle)}{\langle\hat{R}_1^2\rangle\ldots\langle\hat{R}_4^2\rangle\langle\hat{R}_7^2\rangle}\langle\hat{R}_2\hat{R}_3\hat{R}_7\cos(\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_7)\rangle\\
&\quad\times\langle\hat{R}_1\hat{R}_4\hat{R}_7\cos(\hat{\varphi}_1-\hat{\varphi}_4+\hat{\varphi}_7)\rangle\cos(\bar{\varphi}_1+\bar{\varphi}_2+\bar{\varphi}_3-\bar{\varphi}_4)\\
&\quad+O'\left(\frac{1}{\mu}\right)\bigg]. \qquad (88)
\end{aligned}
$$

We now define

$$
\begin{aligned}
m_{125}&=\frac{\bar{R}_1\bar{R}_2\bar{R}_5\langle\hat{R}_1\hat{R}_2\hat{R}_5\cos(\hat{\varphi}_1+\hat{\varphi}_2-\hat{\varphi}_5)\rangle}{\langle\hat{R}_1^2\rangle\langle\hat{R}_2^2\rangle\langle\hat{R}_5^2\rangle}\ \textit{etc.},\\
m_{1234}&=\frac{\bar{R}_1\bar{R}_2\bar{R}_3\bar{R}_4\langle\hat{R}_1\hat{R}_2\hat{R}_3\hat{R}_4\cos(\hat{\varphi}_1+\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_4)\rangle}{\langle\hat{R}_1^2\rangle\langle\hat{R}_2^2\rangle\langle\hat{R}_3^2\rangle\langle\hat{R}_4^2\rangle},\\
d_5&=\frac{\bar{R}_1\bar{R}_2\bar{R}_3\bar{R}_4(1-\bar{R}_5^2/\langle\hat{R}_5^2\rangle)}{\langle\hat{R}_1^2\rangle\langle\hat{R}_2^2\rangle\langle\hat{R}_3^2\rangle\langle\hat{R}_4^2\rangle\langle\hat{R}_5^2\rangle}\langle\hat{R}_1\hat{R}_2\hat{R}_5\cos(\hat{\varphi}_1+\hat{\varphi}_2-\hat{\varphi}_5)\rangle\\
&\quad\times\langle\hat{R}_3\hat{R}_4\hat{R}_5\cos(\hat{\varphi}_3-\hat{\varphi}_4+\hat{\varphi}_5)\rangle\ \textit{etc.} \qquad (89)
\end{aligned}
$$

Then

$$
\begin{aligned}
&P(\bar{R}_1,\ldots,\bar{\varphi}_7)\\
&\quad\propto \bar{R}_1\ldots\bar{R}_7\exp\left(-\frac{\bar{R}_1^2}{\langle\hat{R}_1^2\rangle}\ldots-\frac{\bar{R}_7^2}{\langle\hat{R}_7^2\rangle}\right)\\
&\quad\times\bigg[1+\frac{2}{\mu^{1/2}}m_{125}\cos(\bar{\varphi}_1+\bar{\varphi}_2-\bar{\varphi}_5)\\
&\quad+\frac{2}{\mu^{1/2}}m_{345}\cos(\bar{\varphi}_3-\bar{\varphi}_4+\bar{\varphi}_5)\\
&\quad+\frac{2}{\mu^{1/2}}m_{136}\cos(\bar{\varphi}_1+\bar{\varphi}_3-\bar{\varphi}_6)\\
&\quad+\frac{2}{\mu^{1/2}}m_{246}\cos(\bar{\varphi}_2-\bar{\varphi}_4+\bar{\varphi}_6)\\
&\quad+\frac{2}{\mu^{1/2}}m_{237}\cos(\bar{\varphi}_2+\bar{\varphi}_3-\bar{\varphi}_7)\\
&\quad+\frac{2}{\mu^{1/2}}m_{147}\cos(\bar{\varphi}_1-\bar{\varphi}_4+\bar{\varphi}_7)\\
&\quad+\frac{2}{\mu}m_{1234}\cos(\bar{\varphi}_1+\bar{\varphi}_2+\bar{\varphi}_3-\bar{\varphi}_4)
\end{aligned}
$$

$$
\begin{aligned}
&-\frac{2}{\mu}\frac{\bar{R}_1\ldots\bar{R}_4(1-\bar{R}_5^2/\langle\hat{R}_5^2\rangle)}{\langle\hat{R}_1^2\rangle\ldots\langle\hat{R}_4^2\rangle\langle\hat{R}_5^2\rangle}\langle\hat{R}_1\hat{R}_2\hat{R}_5\cos(\hat{\varphi}_1+\hat{\varphi}_2-\hat{\varphi}_5)\rangle\\
&\times\langle\hat{R}_3\hat{R}_4\hat{R}_5\cos(\hat{\varphi}_3-\hat{\varphi}_4+\hat{\varphi}_5)\rangle\cos(\bar{\varphi}_1+\bar{\varphi}_2+\bar{\varphi}_3-\bar{\varphi}_4)\\
&-\frac{2}{\mu}\frac{\bar{R}_1\ldots\bar{R}_4(1-\bar{R}_6^2/\langle\hat{R}_6^2\rangle)}{\langle\hat{R}_1^2\rangle\ldots\langle\hat{R}_4^2\rangle\langle\hat{R}_6^2\rangle}\langle\hat{R}_1\hat{R}_3\hat{R}_6\cos(\hat{\varphi}_1+\hat{\varphi}_3-\hat{\varphi}_6)\rangle\\
&\times\langle\hat{R}_2\hat{R}_4\hat{R}_6\cos(\hat{\varphi}_2-\hat{\varphi}_4+\hat{\varphi}_6)\rangle\cos(\bar{\varphi}_1+\bar{\varphi}_2+\bar{\varphi}_3-\bar{\varphi}_4)\\
&-\frac{2}{\mu}\frac{\bar{R}_1\ldots\bar{R}_4(1-\bar{R}_7^2/\langle\hat{R}_7^2\rangle)}{\langle\hat{R}_1^2\rangle\ldots\langle\hat{R}_4^2\rangle\langle\hat{R}_7^2\rangle}\langle\hat{R}_2\hat{R}_3\hat{R}_7\cos(\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_7)\rangle\\
&\times\langle\hat{R}_1\hat{R}_4\hat{R}_7\cos(\hat{\varphi}_1-\hat{\varphi}_4+\hat{\varphi}_7)\rangle\cos(\bar{\varphi}_1+\bar{\varphi}_2+\bar{\varphi}_3-\bar{\varphi}_4)\\
&+O'\left(\frac{1}{\mu}\right)\Bigg]. \qquad (90)
\end{aligned}
$$

Using $1+x=\exp[\ln(1+x)]=\exp(x-x^2/2+x^3/3-\ldots)$, equation (90) reads

$$
\begin{aligned}
&P(\bar{R}_1,\ldots,\bar{\varphi}_7)\\
&\quad\propto\bar{R}_1\ldots\bar{R}_7\exp\left(-\frac{\bar{R}_1^2}{\langle\hat{R}_1^2\rangle}\ldots-\frac{\bar{R}_7^2}{\langle\hat{R}_7^2\rangle}\right)\\
&\quad\times\exp\Bigg[\frac{2}{\mu^{1/2}}m_{125}\cos(\bar{\varphi}_1+\bar{\varphi}_2-\bar{\varphi}_5)\\
&\quad+\frac{2}{\mu^{1/2}}m_{345}\cos(\bar{\varphi}_3-\bar{\varphi}_4+\bar{\varphi}_5)\\
&\quad+\frac{2}{\mu^{1/2}}m_{136}\cos(\bar{\varphi}_1+\bar{\varphi}_3-\bar{\varphi}_6)\\
&\quad+\frac{2}{\mu^{1/2}}m_{246}\cos(\bar{\varphi}_2-\bar{\varphi}_4+\bar{\varphi}_6)\\
&\quad+\frac{2}{\mu^{1/2}}m_{237}\cos(\bar{\varphi}_2+\bar{\varphi}_3-\bar{\varphi}_7)\\
&\quad+\frac{2}{\mu^{1/2}}m_{147}\cos(\bar{\varphi}_1-\bar{\varphi}_4+\bar{\varphi}_7)\\
&\quad+\frac{2}{\mu}m_{1234}\cos(\bar{\varphi}_1+\bar{\varphi}_2+\bar{\varphi}_3-\bar{\varphi}_4)\\
&\quad-\frac{2}{\mu}(m_{125}m_{345}+m_{136}m_{246}+m_{237}m_{147})\\
&\quad\times\cos(\bar{\varphi}_1+\bar{\varphi}_2+\bar{\varphi}_3-\bar{\varphi}_4)\\
&\quad-\frac{2}{\mu}(d_5+d_6+d_7)\cos(\bar{\varphi}_1+\bar{\varphi}_2+\bar{\varphi}_3-\bar{\varphi}_4)\\
&\quad+O'\left(\frac{1}{\mu}\right)\Bigg]. \qquad (91)
\end{aligned}
$$

We can stop here and put $\mu=1$ into equation (91). But we can also go further, since we can write for the marginal density of $\bar{\varphi}\equiv\bar{\varphi}_1+\bar{\varphi}_2+\bar{\varphi}_3-\bar{\varphi}_4$

$$
P(\bar{R}_1,\ldots,\bar{R}_7,\bar{\varphi})\equiv\int\limits_0^{2\pi}\mathrm{d}\varphi_5\int\limits_0^{2\pi}\mathrm{d}\varphi_6\int\limits_0^{2\pi}\mathrm{d}\varphi_7\,P(\bar{R}_1,\ldots,\bar{\varphi}_7), \qquad (92)
$$

$$
\begin{aligned}
&P(\bar{R}_1,\ldots,\bar{R}_7,\bar{\varphi})\\
&\quad\propto\bar{R}_1\ldots\bar{R}_7\exp(-\bar{R}_1^2/\langle\hat{R}_1^2\rangle\ldots-\bar{R}_7^2/\langle\hat{R}_7^2\rangle)\\
&\qquad\times\exp\{(2/\mu)[m_{1234}-(m_{125}m_{345}+m_{136}m_{246}+m_{237}m_{147})\\
&\qquad-(d_5+d_6+d_7)]\cos\bar{\varphi}\}\\
&\qquad\times I_0[(2/\mu^{1/2})(m_{125}^2+m_{345}^2+2m_{125}m_{345}\cos\bar{\varphi})^{1/2}]\\
&\qquad\times I_0[(2/\mu^{1/2})(m_{136}^2+m_{246}^2+2m_{136}m_{246}\cos\bar{\varphi})^{1/2}]\\
&\qquad\times I_0[(2/\mu^{1/2})(m_{147}^2+m_{237}^2+2m_{147}m_{237}\cos\bar{\varphi})^{1/2}]. \qquad (93)
\end{aligned}
$$

To obtain equation (93) we used the identity

$$
\begin{aligned}
&a\cos(\theta+\alpha)+b\cos(\beta-\theta)\\
&\quad=[a^2+b^2+2ab\cos(\alpha+\beta)]^{1/2}\cos(\theta+\gamma). \qquad (94)
\end{aligned}
$$

Using SAD and the relation $I_0(x)\simeq\exp(+\frac{1}{4}x^2)$ for small x in equation (93) we obtain

$$
\begin{aligned}
&P(\bar{R}_1,\ldots,\bar{R}_7,\bar{\varphi})\\
&\quad\propto\bar{R}_1\ldots\bar{R}_7\exp(-\bar{R}_1^2/\langle\hat{R}_1^2\rangle\ldots-\bar{R}_7^2/\langle\hat{R}_7^2\rangle)\\
&\qquad\times\exp\{(2/\mu)[m_{1234}-(m_{125}m_{345}+m_{136}m_{246}\\
&\qquad+m_{237}m_{147})-(d_5+d_6+d_7)]\cos\bar{\varphi}\}\\
&\qquad\times\exp[(2/\mu)(m_{125}m_{345}+m_{136}m_{246}+m_{237}m_{147})\cos\bar{\varphi}]\\
&\quad\propto\bar{R}_1\ldots\bar{R}_7\exp(-\bar{R}_1^2/\langle\hat{R}_1^2\rangle\ldots-\bar{R}_7^2/\langle\hat{R}_7^2\rangle)\\
&\qquad\times\exp\{(2/\mu)[m_{1234}-(d_5+d_6+d_7)]\cos\bar{\varphi}\}. \qquad (95)
\end{aligned}
$$

Defining $P(\bar{\varphi})=P(\bar{\varphi}|\bar{R}_1=R_1,\ldots,\bar{R}_7=R_7)$ and putting $\mu=1$, equation (95) becomes

$$
P(\bar{\varphi})\propto\exp\{2[m_{1234}-(d_5+d_6+d_7)]\cos\bar{\varphi}\}, \qquad (96)
$$

where one must put $\bar{R}_i=R_i$ in $[m_{1234}-(d_5+d_6+d_7)]$ in equation (96).

We shall now consider a uniform j.p.d. $f(\mathbf{x}_1,\ldots,\mathbf{x}_N)=1$ for equation (96). Then

$$
\begin{aligned}
m_{1234}&=\frac{R_1R_2R_3R_4\langle\hat{R}_1\hat{R}_2\hat{R}_3\hat{R}_4\cos(\hat{\varphi}_1+\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_4)\rangle}{\langle\hat{R}_1^2\rangle\langle\hat{R}_2^2\rangle\langle\hat{R}_3^2\rangle\langle\hat{R}_4^2\rangle}\\
&=\frac{R_1R_2R_3R_4}{N},\\
d_5&=\frac{R_1R_2R_3R_4(1-R_5^2/\langle\hat{R}_5^2\rangle)}{\langle\hat{R}_1^2\rangle\langle\hat{R}_2^2\rangle\langle\hat{R}_3^2\rangle\langle\hat{R}_4^2\rangle\langle\hat{R}_5^2\rangle}\langle\hat{R}_1\hat{R}_2\hat{R}_5\cos(\hat{\varphi}_1+\hat{\varphi}_2-\hat{\varphi}_5)\rangle\\
&\quad\times\langle\hat{R}_3\hat{R}_4\hat{R}_5\cos(\hat{\varphi}_3-\hat{\varphi}_4+\hat{\varphi}_5)\rangle\\
&=\frac{R_1R_2R_3R_4(1-R_5^2)}{N}\ \textit{etc.} \qquad (97)
\end{aligned}
$$

and equation (96) becomes in this case

$$
P(\bar{\varphi})\propto\exp\left[\frac{2R_1R_2R_3R_4}{N}(R_5^2+R_6^2+R_7^2-2)\cos\bar{\varphi}\right], \qquad (98)
$$

which is the classical formula for the quartet in $P1$.

7. A statistical interpretation of the $B_{4,0}$ formula using SAD

For the $B_{4,0}$ formula we refer to Karle & Hauptman (1957). We consider the j.p.d.

$$f(\mathbf{x}_1, \ldots, \mathbf{x}_N) = \mathrm{Cte} \prod_{i=1}^{N-1} \prod_{\substack{j=2 \\ i<j}}^{N} Q(\mathbf{x}_i - \mathbf{x}_j). \tag{99}$$

Then

$$f(\mathbf{x}_i, \mathbf{x}_j, \mathbf{x}_l, \mathbf{x}_m) = C_3 Q(\mathbf{x}_j - \mathbf{x}_i) Q(\mathbf{x}_j - \mathbf{x}_l) Q(\mathbf{x}_m - \mathbf{x}_l) \times Q(\mathbf{x}_i - \mathbf{x}_m) Q(\mathbf{x}_m - \mathbf{x}_j) Q(\mathbf{x}_l - \mathbf{x}_i). \tag{100}$$

This is the most general product of $Q(\mathbf{x}_{i_1} - \mathbf{x}_{i_2})$ containing only the random vector variables $\mathbf{x}_i, \mathbf{x}_j, \mathbf{x}_l, \mathbf{x}_m$. The constant C_3 is determined by the condition $\int \mathrm{d}\mathbf{x}_i\, \mathrm{d}\mathbf{x}_j\, \mathrm{d}\mathbf{x}_l\, \mathrm{d}\mathbf{x}_m\, f(\mathbf{x}_i, \mathbf{x}_j, \mathbf{x}_l, \mathbf{x}_m) = 1$. But then

$$C_3^{-1} = \sum_{\mathbf{k},\mathbf{l},\mathbf{m}} \left(\frac{R_{\mathbf{k}}^2 - 1}{N-1}\right)\left(\frac{R_{\mathbf{l}}^2 - 1}{N-1}\right)\left(\frac{R_{\mathbf{m}}^2 - 1}{N-1}\right)\left(\frac{R_{\mathbf{k}+\mathbf{l}-\mathbf{m}}^2 - 1}{N-1}\right) \times \left(\frac{R_{\mathbf{k}-\mathbf{m}}^2 - 1}{N-1}\right)\left(\frac{R_{\mathbf{l}-\mathbf{m}}^2 - 1}{N-1}\right). \tag{101}$$

This is pictorially represented in Fig. 1.

If we assume that there is no overlap of interatomic vectors, it follows that $f(\mathbf{x}_i, \mathbf{x}_j, \mathbf{x}_l, \mathbf{x}_m) = 0$ except when there exist different atomic vectors $\mathbf{r}_\alpha, \mathbf{r}_\beta, \mathbf{r}_\gamma, \mathbf{r}_\delta$ such that

$$\begin{aligned} &\mathbf{x}_i - \mathbf{x}_j = \mathbf{r}_\gamma - \mathbf{r}_\beta, \quad \mathbf{x}_j - \mathbf{x}_l = \mathbf{r}_\beta - \mathbf{r}_\gamma, \quad \mathbf{x}_l - \mathbf{x}_m = \mathbf{r}_\gamma - \mathbf{r}_\delta, \\ &\mathbf{x}_m - \mathbf{x}_i = \mathbf{r}_\delta - \mathbf{r}_\alpha, \quad \mathbf{x}_m - \mathbf{x}_j = \mathbf{r}_\delta - \mathbf{r}_\beta, \quad \mathbf{x}_l - \mathbf{x}_i = \mathbf{r}_\gamma - \mathbf{r}_\alpha. \end{aligned} \tag{102}$$

However, calculating moments with this density $f(\mathbf{x}_i, \mathbf{x}_j, \mathbf{x}_l, \mathbf{x}_m)$ {*e.g.* the constant $C_3^{-1} = \sum_{\mathbf{k},\mathbf{l},\mathbf{m}} [(R_{\mathbf{k}}^2 - 1)/(N-1)] \times [(R_{\mathbf{l}}^2 - 1)/(N-1)][(R_{\mathbf{m}}^2 - 1)/(N-1)][(R_{\mathbf{k}+\mathbf{l}-\mathbf{m}}^2 - 1)/(N-1)] \times [(R_{\mathbf{k}-\mathbf{m}}^2 - 1)/(N-1)][(R_{\mathbf{l}-\mathbf{m}}^2 - 1)/(N-1)]$} is too laborious (for the computer). Fortunately we can avoid such triple summations over reciprocal space. Indeed if we take for $f(\mathbf{x}_i, \mathbf{x}_j, \mathbf{x}_l, \mathbf{x}_m)$ the simpler formula

$$f(\mathbf{x}_i, \mathbf{x}_j, \mathbf{x}_l, \mathbf{x}_m) = C_4 Q(\mathbf{x}_i - \mathbf{x}_j) Q(\mathbf{x}_j - \mathbf{x}_l) Q(\mathbf{x}_l - \mathbf{x}_m) Q(\mathbf{x}_m - \mathbf{x}_i), \tag{103}$$

we shall only have one summation over reciprocal space [see equation (106)]. Equation (103) is represented by the solid lines in Fig. 1. If there is no overlap of interatomic vectors then there exist again different atomic vectors $\mathbf{r}_\alpha, \mathbf{r}_\beta, \mathbf{r}_\gamma, \mathbf{r}_\delta$ such that

$$\begin{aligned} &\mathbf{x}_i - \mathbf{x}_j = \mathbf{r}_\alpha - \mathbf{r}_\beta, \quad \mathbf{x}_j - \mathbf{x}_l = \mathbf{r}_\beta - \mathbf{r}_\gamma, \\ &\mathbf{x}_l - \mathbf{x}_m = \mathbf{r}_\gamma - \mathbf{r}_\delta, \quad \mathbf{x}_m - \mathbf{x}_i = \mathbf{r}_\delta - \mathbf{r}_\alpha. \end{aligned} \tag{104}$$

The constant C_4 is much simpler than the constant C_3 [equation (101)],

$$C_4^{-1} = \sum_{\mathbf{q}} \left(\frac{R_{\mathbf{q}}^2 - 1}{N-1}\right)^4. \tag{105}$$

We also obtain for the moment [using equation (103)]

$$\begin{aligned} &\int \mathrm{d}\mathbf{x}_i\, \mathrm{d}\mathbf{x}_j\, \mathrm{d}\mathbf{x}_l\, \mathrm{d}\mathbf{x}_m\, f(\mathbf{x}_i, \mathbf{x}_j, \mathbf{x}_l, \mathbf{x}_m) \\ &\quad \times \cos\{2\pi[\mathbf{h}\cdot(\mathbf{x}_i - \mathbf{x}_m) + \mathbf{k}\cdot(\mathbf{x}_j - \mathbf{x}_m) + \mathbf{l}(\mathbf{x}_l - \mathbf{x}_m)]\} \\ &\quad = \left[\sum_{\mathbf{q}} \left(\frac{R_{\mathbf{q}}^2 - 1}{N-1}\right)\left(\frac{R_{\mathbf{q}+\mathbf{h}}^2 - 1}{N-1}\right)\left(\frac{R_{\mathbf{q}+\mathbf{h}+\mathbf{k}}^2 - 1}{N-1}\right) \right. \\ &\quad \left. \times \left(\frac{R_{\mathbf{q}+\mathbf{h}+\mathbf{k}+\mathbf{l}}^2 - 1}{N-1}\right)\right] \Big/ \sum_{\mathbf{q}} \left(\frac{R_{\mathbf{q}}^2 - 1}{N-1}\right)^4. \end{aligned} \tag{106}$$

The numerator in equation (106) is the most important part of the $B_{4,0}$ formula (Karle & Hauptman, 1957).

Let us apply all this to formula (96) [using equation (103) instead of equation (100)]. We already know that

$$\langle \hat{R}_1^2 \rangle = \langle \hat{R}_1^2 \rangle_f = R_1^2. \tag{107}$$

Then *e.g.*

$$d_5 = 0 \text{ since } \langle \hat{R}_5^2 \rangle = R_5^2 \text{ and } \bar{R}_5^2 = R_5^2. \tag{108}$$

Let us calculate

$$\begin{aligned} m_{1234} &= \frac{\bar{R}_1 \bar{R}_2 \bar{R}_3 \bar{R}_4 \langle \hat{R}_1 \hat{R}_2 \hat{R}_3 \hat{R}_4 \cos(\hat{\varphi}_1 + \hat{\varphi}_2 + \hat{\varphi}_3 - \hat{\varphi}_4) \rangle}{\langle \hat{R}_1^2 \rangle \langle \hat{R}_2^2 \rangle \langle \hat{R}_3^2 \rangle \langle \hat{R}_4^2 \rangle} \\ &= \frac{R_1 R_2 R_3 R_4}{\langle \hat{R}_1^2 \rangle \langle \hat{R}_2^2 \rangle \langle \hat{R}_3^2 \rangle \langle \hat{R}_4^2 \rangle} \langle \hat{R}_1 \hat{R}_2 \hat{R}_3 \hat{R}_4 \cos(\hat{\varphi}_1 + \hat{\varphi}_2 + \hat{\varphi}_3 - \hat{\varphi}_4) \rangle \\ &= \frac{R_1 R_2 R_3 R_4}{(R_1 R_2 R_3 R_4)^2} \langle \hat{R}_1 \hat{R}_2 \hat{R}_3 \hat{R}_4 \cos(\hat{\varphi}_1 + \hat{\varphi}_2 + \hat{\varphi}_3 - \hat{\varphi}_4) \rangle \\ &= \frac{\langle \hat{R}_1 \hat{R}_2 \hat{R}_3 \hat{R}_4 \cos(\hat{\varphi}_1 + \hat{\varphi}_2 + \hat{\varphi}_3 - \hat{\varphi}_4) \rangle}{R_1 R_2 R_3 R_4}. \end{aligned} \tag{109}$$

Just as for the triplet case [equation (50)] we can write

$$\begin{aligned} &\langle \hat{R}_1 \hat{R}_2 \hat{R}_3 \hat{R}_4 \cos(\hat{\varphi}_1 + \hat{\varphi}_2 + \hat{\varphi}_3 - \hat{\varphi}_4) \rangle \\ &\quad = \frac{1}{N^2} \mathcal{R}e \sum_{p,q,r,s} \langle \exp\{2\pi i[\mathbf{h}\cdot(\mathbf{x}_p - \mathbf{x}_s) + \mathbf{k}\cdot(\mathbf{x}_q - \mathbf{x}_s) \\ &\quad + \mathbf{l}\cdot(\mathbf{x}_r - \mathbf{x}_s)]\}\rangle_f \\ &\quad = \frac{1}{N}(R_{\mathbf{h}}^2 + R_{\mathbf{k}}^2 + R_{\mathbf{l}}^2 + R_{\mathbf{h}+\mathbf{k}+\mathbf{l}}^2 + R_{\mathbf{h}+\mathbf{k}}^2 + R_{\mathbf{h}+\mathbf{l}}^2 + R_{\mathbf{k}+\mathbf{l}}^2 - 5) \\ &\quad + \frac{(N-1)(N-2)}{N}[\mu(\mathbf{h},\mathbf{k}) + \mu(\mathbf{h},\mathbf{l}) + \mu(\mathbf{k},\mathbf{l}) \\ &\quad + \mu(\mathbf{h}+\mathbf{k},\mathbf{l}) + \mu(\mathbf{h}+\mathbf{l},\mathbf{k}) + \mu(\mathbf{h},\mathbf{k}+\mathbf{l})] \\ &\quad + \frac{(N-1)(N-2)(N-3)}{N}\mu(\mathbf{h},\mathbf{k},\mathbf{l}), \end{aligned} \tag{110}$$

where $\mu(\mathbf{h}, \mathbf{k})$ is given by equation (51) and $\mu(\mathbf{h}, \mathbf{k}, \mathbf{l})$ is given by

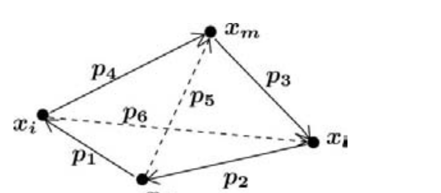

Figure 1
Pictorial representation of equation (100) ($\mathbf{p}_1, \mathbf{p}_2, \mathbf{p}_3, \mathbf{p}_4, \mathbf{p}_5, \mathbf{p}_6$ represent interatomic vectors).

$$\mu(\mathbf{h},\mathbf{k},\mathbf{l}) = \left[\sum_{\mathbf{q}} \frac{(R^2_{\mathbf{q}}-1)}{(N-1)}\frac{(R^2_{\mathbf{q+h}}-1)}{(N-1)}\frac{(R^2_{\mathbf{q+h+k}}-1)}{(N-1)} \times \frac{(R^2_{\mathbf{q+h+k+l}}-1)}{(N-1)}\right] \Big/ \sum_{\mathbf{q}}\left(\frac{(R^2_{\mathbf{q}}-1)}{(N-1)}\right)^4 \qquad (111)$$

[see equation (106))]. If we replace $\langle \hat{R}_1\hat{R}_2\hat{R}_3\hat{R}_4 \times \cos(\hat{\varphi}_1+\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_4)\rangle$ in equation (110) by $R_1R_2R_3R_4\cos(\varphi_1+\varphi_2+\varphi_3-\varphi_4)$ we obtain a formula that we shall call the $B'_{4,0}$ formula. Finally, we get the following formula for the quartet:

$$P(\bar{\varphi}) = \exp\left[2\frac{\langle \hat{R}_1\hat{R}_2\hat{R}_3\hat{R}_4\cos(\hat{\varphi}_1+\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_4)\rangle}{R_1R_2R_3R_4}\cos(\bar{\varphi})\right], \qquad (112)$$

where $\langle \hat{R}_1\hat{R}_2\hat{R}_3\hat{R}_4\cos(\hat{\varphi}_1+\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_4)\rangle$ is given by equation (110) and where $\bar{\varphi} = \bar{\varphi}_1+\bar{\varphi}_2+\bar{\varphi}_3-\bar{\varphi}_4$.

If we develop asymptotically $\mu(\mathbf{h},\mathbf{k},\mathbf{l})$ according to inverse powers of N then we get

$$\begin{aligned} N^3\mu(\mathbf{h},\mathbf{k},\mathbf{l}) \simeq{}& (R^2_{\mathbf{h}}-1)(R^2_{\mathbf{h+k}}-1)(R^2_{\mathbf{h+k+l}}-1) \\ &+(R^2_{\mathbf{h}}-1)(R^2_{\mathbf{k}}-1)(R^2_{\mathbf{k+l}}-1) \\ &+(R^2_{\mathbf{k}}-1)(R^2_{\mathbf{l}}-1)(R^2_{\mathbf{h+k}}-1) \\ &+(R^2_{\mathbf{l}}-1)(R^2_{\mathbf{k+l}}-1)(R^2_{\mathbf{h+k+l}}-1). \end{aligned} \qquad (113)$$

If we then replace $\mu(\mathbf{h},\mathbf{k})$ by its approximate value [equation (54)],

$$\begin{aligned} N^2\mu(\mathbf{h},\mathbf{k}) \simeq{}& (R^2_{\mathbf{h}}-1)(R^2_{\mathbf{h+k}}-1)+(R^2_{\mathbf{h}}-1)(R^2_{\mathbf{k}}-1) \\ &+(R^2_{\mathbf{k}}-1)(R^2_{\mathbf{h+k}}-1), \end{aligned} \qquad (114)$$

and insert these approximate expressions back into equation (110), then we can see that there is strong evidence that $\cos(\varphi_1+\varphi_2+\varphi_3-\varphi_4)$ is *probably negative when* $R_{\mathbf{h+k}} \simeq R_{\mathbf{h+l}} \simeq R_{\mathbf{k+l}} \simeq 0$ *and large* $R_{\mathbf{h}}$, $R_{\mathbf{k}}$, $R_{\mathbf{l}}$, $R_{\mathbf{h+k+l}}$, *which is in perfect agreement with* numerical tests (see *e.g.* Giacovazzo, 1976).

Note that formula (112) is also the formula for the quartet given only its first neighborhood $R_{\mathbf{h}}$, $R_{\mathbf{k}}$, $R_{\mathbf{l}}$, $R_{\mathbf{h+k+l}}$!

8. Conclusion and some numerical tests

8.1. Approximate formulas for $N^3\mu$ (h, k, l) and $N^2\mu$(h, k)

We have considered a structure with 100 atoms (N = 100). We took a random sample of 400 triplets and quartets. In almost all cases we could approximate $N^3\mu(\mathbf{h},\mathbf{k},\mathbf{l})$ by formula (113),

$$\begin{aligned} N^3\mu(\mathbf{h},\mathbf{k},\mathbf{l}) \simeq{}& (R^2_{\mathbf{h}}-1)(R^2_{\mathbf{h+k}}-1)(R^2_{\mathbf{h+k+l}}-1) \\ &+(R^2_{\mathbf{h}}-1)(R^2_{\mathbf{k}}-1)(R^2_{\mathbf{k+l}}-1) \\ &+(R^2_{\mathbf{k}}-1)(R^2_{\mathbf{l}}-1)(R^2_{\mathbf{h+k}}-1) \\ &+(R^2_{\mathbf{l}}-1)(R^2_{\mathbf{k+l}}-1)(R^2_{\mathbf{h+k+l}}-1), \end{aligned} \qquad (115)$$

and we could approximate $N^2\mu(\mathbf{h},\mathbf{k})$ by formula (54),

$$\begin{aligned} N^2\mu(\mathbf{h},\mathbf{k}) \simeq{}& (R^2_{\mathbf{h}}-1)(R^2_{\mathbf{h+k}}-1)+(R^2_{\mathbf{h}}-1)(R^2_{\mathbf{k}}-1) \\ &+(R^2_{\mathbf{k}}-1)(R^2_{\mathbf{h+k}}-1). \end{aligned} \qquad (116)$$

It is also interesting to note that the right-hand side of equation (116) is the exact result of $A(\mathbf{h},\mathbf{k})$ [equation (75)] when we take $f(\mathbf{u}_\alpha,\mathbf{v}_\alpha)=1$ instead of formula (60) (see the end of §5).

8.2. Numerical tests for the linearized invariants method

We used a struture of 100 atoms (N = 100), 8000 structure factors (M = 8000) and a random sample of 400 triplets. We tested formula (74) with $A(\mathbf{h},\mathbf{k})$ given by equation (77):

$$\begin{aligned} P(\psi) &\propto \exp[2R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}A(\mathbf{h},\mathbf{k})\cos\psi], \\ A(\mathbf{h},\mathbf{k}) &= (1/N^{1/2})[R^2_{\mathbf{h}}+R^2_{\mathbf{k}}+R^2_{\mathbf{h+k}}-2+(R^2_{\mathbf{h}}-1)(R^2_{\mathbf{k}}-1) \\ &\quad +(R^2_{\mathbf{h}}-1)(R^2_{\mathbf{h+k}}-1)+(R^2_{\mathbf{k}}-1)(R^2_{\mathbf{h+k}}-1)]. \end{aligned} \qquad (117)$$

Then

$$\langle\cos\psi\rangle = \frac{I_1[2R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}A(\mathbf{h},\mathbf{k})]}{I_0[2R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}A(\mathbf{h},\mathbf{k})]}. \qquad (118)$$

Let

$$\varphi = \varphi_{\mathbf{h}}+\varphi_{\mathbf{k}}-\varphi_{\mathbf{h+k}}. \qquad (119)$$

We define

$$\psi_{\rm calc} \equiv \cos^{-1}(\langle\cos\psi\rangle), \qquad (120)$$

where $\cos^{-1}$ is the inverse of cos with values in $[0,\pi]$. We also consider

$$\varphi' \equiv \cos^{-1}(\cos\varphi). \qquad (121)$$

Then we found the empirical probabilities for the case $|2R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}A(\mathbf{h},\mathbf{k})| \geq 6$ ($\psi_{\rm calc} \leq 24°$):

$$\begin{aligned} {\rm Prob}(|\psi_{\rm calc}-\varphi'| \leq 80°) &= 0.8, \\ {\rm Prob}(|\psi_{\rm calc}-\varphi'| \leq 30°) &= 0.5. \end{aligned} \qquad (122)$$

100% of the calculated $A(\mathbf{h},\mathbf{k})$ had a positive sign and 75% of the calculated $\langle\cos\psi\rangle$ had the same sign as $\cos\varphi$. This shows that, although $A(\mathbf{h},\mathbf{k})$ can take on negative values for large $R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}$, this situation rarely happens.

8.3. Numerical tests for the SAD method for both triplet and quartet

Here the results were disappointing, due to sign mismatches between the calculated cosine and the true cosine. Apparently the $B_{3,0}$ ($B'_{3,0}$) formula fails to predict the correct sign (as we shall see below).

8.3.1. Triplet. As already stated in §8.1 $N^2\mu(\mathbf{h},\mathbf{k})$ can be approximated by the right-hand side of equation (116). We tested equation (52),

$$P_f(\bar{\varphi}) \propto \exp\left[2\frac{m(\mathbf{h},\mathbf{k})}{R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}}\cos\bar{\varphi}\right], \qquad (123)$$

where

$$\begin{aligned} m(\mathbf{h},\mathbf{k}) ={}& (1/N^{1/2})[R^2_{\mathbf{h}}+R^2_{\mathbf{k}}+R^2_{\mathbf{h+k}}-2+(R^2_{\mathbf{h}}-1)(R^2_{\mathbf{k}}-1) \\ &+(R^2_{\mathbf{h}}-1)(R^2_{\mathbf{h+k}}-1)+(R^2_{\mathbf{k}}-1)(R^2_{\mathbf{h+k}}-1)]. \end{aligned} \qquad (124)$$

Let

$$\bar{\varphi}_{\rm calc} = \cos^{-1}(\langle\cos\bar{\varphi}\rangle) = \cos^{-1}\left\{I_1\left[2\frac{m(\mathbf{h},\mathbf{k})}{R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}}\right]\Big/I_0\left[2\frac{m(\mathbf{h},\mathbf{k})}{R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}}\right]\right\} \quad (125)$$

and

$$\varphi' \equiv \cos^{-1}(\cos\varphi) \text{ with } \varphi = \varphi_{\mathbf{h}} + \varphi_{\mathbf{k}} - \varphi_{\mathbf{h+k}}. \quad (126)$$

$N = 100$ and $M = 8000$.

Case A (random sample of 400 triplets): $R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}} \geq 0$ and $|2[m(\mathbf{h},\mathbf{k})/R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}]| \geq 6$ ($\bar{\varphi}_{\rm calc} \leq 24°$).

$$\mathrm{Prob}(|\bar{\varphi}_{\rm calc} - \varphi'| \leq 130°) = 0.8,$$
$$\mathrm{Prob}(|\bar{\varphi}_{\rm calc} - \varphi'| \leq 70°) = 0.5. \quad (127)$$

53% of the calculated cosines had the same sign as the real cosine and 32% of the calculated cosines had a negative sign.

Case B (random sample of 400 triplets): $R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}} \geq 0$ and $|2[m(\mathbf{h},\mathbf{k})/R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}]| \geq 2$ ($\bar{\varphi}_{\rm calc} \leq 45°$).

$$\mathrm{Prob}(|\bar{\varphi}_{\rm calc} - \varphi'| \leq 110°) = 0.8,$$
$$\mathrm{Prob}(|\bar{\varphi}_{\rm calc} - \varphi'| \leq 60°) = 0.5. \quad (128)$$

54% of the calculated cosines had the same sign as the real cosine and 43% of the calculated cosines had a negative sign.

Case C (random sample of 70 triplets): $R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}} \geq 0.5$ and $|2[m(\mathbf{h},\mathbf{k})/R_{\mathbf{h}}R_{\mathbf{k}}R_{\mathbf{h+k}}]| \geq 2$ ($\bar{\varphi}_{\rm calc} \leq 45°$).

$$\mathrm{Prob}(|\bar{\varphi}_{\rm calc} - \varphi'| \leq 90°) = 0.82,$$
$$\mathrm{Prob}(|\bar{\varphi}_{\rm calc} - \varphi'| \leq 40°) = 0.54. \quad (129)$$

No negative calculated cosines. 57% of the calculated cosines had the same sign as the real cosine. These results are slightly better than Case B.

Case D (random sample of 100 triplets). We now test the formula $P_f(\bar{\varphi}) \propto \exp[2m(\mathbf{h},\mathbf{k})\cos\bar{\varphi}]$ that we obtain if we set in equation (20) $\bar{R}_{\mathbf{h}} = R_{\mathbf{h}}\langle\hat{R}_{\mathbf{h}}^2\rangle^{1/2}, \ldots$. We tested it for $|2m(\mathbf{h},\mathbf{k})| \geq 3$. We obtained

$$\mathrm{Prob}(|\bar{\varphi}_{\rm calc} - \varphi'| \leq 50°) = 0.8,$$
$$\mathrm{Prob}(|\bar{\varphi}_{\rm calc} - \varphi'| \leq 25°) = 0.5. \quad (130)$$

82% of the calculated cosines had the same sign as the real cosine, none of the calculated cosines had a negative sign. We call this $P_f(\bar{\varphi})$ a rescaled formula.

8.3.2. Quartet. We tested formula (112)

$$P(\bar{\varphi}) = \exp\left[2\frac{\langle\hat{R}_1\hat{R}_2\hat{R}_3\hat{R}_4\cos(\hat{\varphi}_1+\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_4)\rangle}{R_1R_2R_3R_4}\cos(\bar{\varphi})\right], \quad (131)$$

where [see equation (110)]

$$\begin{aligned}\langle\hat{R}_1\hat{R}_2\hat{R}_3\hat{R}_4\cos(\hat{\varphi}_1+\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_4)\rangle &= \frac{1}{N}(R_{\mathbf{h}}^2 + R_{\mathbf{k}}^2 + R_{\mathbf{k}}^2 + R_{\mathbf{h+k+l}}^2 + R_{\mathbf{h+k}}^2 + R_{\mathbf{h+l}}^2 + R_{\mathbf{k+l}}^2 - 5) \\ &+ \frac{(N-1)(N-2)}{N}[\mu(\mathbf{h},\mathbf{k}) + \mu(\mathbf{h},\mathbf{l}) + \mu(\mathbf{k},\mathbf{l}) \\ &+ \mu(\mathbf{h+k},\mathbf{l}) + \mu(\mathbf{h+l},\mathbf{k}) + \mu(\mathbf{h},\mathbf{k+l})] \\ &+ \frac{(N-1)(N-2)(N-3)}{N}\mu(\mathbf{h},\mathbf{k},\mathbf{l}),\end{aligned} \quad (132)$$

where we used equations (115) and (116). We used a structure of 100 atoms ($N = 100$) and 8000 structure factors ($M = 8000$). Let

$$\begin{aligned}\bar{\varphi}_{\rm calc} &= \cos^{-1}(\langle\cos\bar{\varphi}\rangle) \\ &= \cos^{-1}\left\{I_1\left[2\frac{\langle\hat{R}_1\hat{R}_2\hat{R}_3\hat{R}_4\cos(\hat{\varphi}_1+\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_4)\rangle}{R_1R_2R_3R_4}\right]\right. \\ &\quad \left.\times I_0\left[2\frac{\langle\hat{R}_1\hat{R}_2\hat{R}_3\hat{R}_4\cos(\hat{\varphi}_1+\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_4)\rangle}{R_1R_2R_3R_4}\right]^{-1}\right\}\end{aligned} \quad (133)$$

and

$$\varphi' \equiv \cos^{-1}(\cos\varphi) \text{ with } \varphi = \varphi_1 + \varphi_2 + \varphi_3 - \varphi_4. \quad (134)$$

Case E (400 random quartets). $|2[\langle\hat{R}_1\hat{R}_2\hat{R}_3\hat{R}_4\cos(\hat{\varphi}_1+\hat{\varphi}_2+\hat{\varphi}_3-\hat{\varphi}_4)\rangle/R_1R_2R_3R_4]| \geq 6$ ($\bar{\varphi}_{\rm calc} \leq 24°$).

$$\mathrm{Prob}(|\bar{\varphi}_{\rm calc} - \varphi'| \leq 120°) = 0.8,$$
$$\mathrm{Prob}(|\bar{\varphi}_{\rm calc} - \varphi'| \leq 70°) = 0.54. \quad (135)$$

50% of the cosines of the quartets had the same sign as the real cosine and 33% of the cosines had a negative sign.

8.4. Conclusions

(1) The method of linearized invariants gives acceptable results.

(2) The SAD method gives disappointing results (now) (but see Case C of the triplet). Clearly the $B_{3,0}$ ($B'_{3,0}$) and $B_{4,0}$ ($B'_{4,0}$) formulas fail to compute the correct sign of the cosine: about 50% of the calculated results gave the same sign. To improve the formula obtained with the SAD method we suggest calculating the j.p.d. of structure factors to higher order in inverse powers of $\mu^{1/2}$. We hope to present these formulas in the future.

(3) Modified (rescaled) formulas (see case D of the triplet) can also give acceptable results.

APPENDIX *A*
Some useful formulas

$$\int_0^{2\pi}\exp[-i\rho\cos(\theta-\varphi)]\cos[n(\theta+\alpha)]\,\mathrm{d}\theta = (-i)^n 2\pi J_n(\rho)\cos[n(\varphi+\alpha)]. \quad (136)$$

$$\int_0^{\infty}\rho^{n+1}\exp(-\tfrac{1}{4}\rho^2)J_n(\rho R)\,\mathrm{d}\rho = 2^{n+1}R^n\exp(-R^2). \quad (137)$$

$$\int_0^{\infty}\exp(-\tfrac{1}{4}\rho^2)\rho^{2n+k+1}J_k(\rho R)\,\mathrm{d}\rho = 2^{2n+k+1}n!\exp(-R^2)R^kL_n^k(R^2). \quad (138)$$

$$L_n^k(z) = (1/n!)\exp(x)x^{-k}\,(\mathrm{d}^n/\mathrm{d}x^n)\,[\exp(-x)x^{n+k}]. \quad (139)$$

$$L_n^k(z) = \sum_{m=0}^{n} (-1)^m \binom{n+k}{n-m} \frac{z^m}{m!}. \quad (140)$$

$$L_n(z) = L_n^0(z). \quad (141)$$

$$L_0(z) = 1 = L_0^k(z). \quad (142)$$

$$L_1(z) = 1 - z. \quad (143)$$

$$L_2(z) = 1 - 2z + z^2/2. \quad (144)$$

$$J_n(\rho) \simeq (2/\pi\rho)^{1/2} \cos(\rho - n\pi/2 - \pi/4) \text{ for large } \rho. \quad (145)$$

$$J_n(-\rho) = (-1)^n J_n(\rho). \quad (146)$$

$$\exp(iz\cos\varphi) = J_0(z) + 2\sum_{k=1}^{\infty} i^k J_k(z)\cos(k\varphi). \quad (147)$$

$$J_n(z) = \left(\frac{z}{2}\right)^n \sum_{k=0}^{\infty} \frac{(-1)^k}{k!(n+k)!} \left(\frac{z}{2}\right)^{2k}. \quad (148)$$

$$J_0(z) = 1 - (1/4)z^2 + (1/4^3)z^4 - \ldots. \quad (149)$$

$$J_1(z) = (z/2) - (z^3/8) + \ldots. \quad (150)$$

$$\int_{-\infty}^{\infty} \exp(ibx)\exp(-\tfrac{1}{2}x^2)\,\mathrm{d}x = (2\pi)^{1/2}\exp(-\tfrac{1}{2}b^2). \quad (151)$$

$$\int_{-\infty}^{\infty} (iu)^n \exp(-iuE)\exp(-\tfrac{1}{2}u^2)\,\mathrm{d}u = (2\pi)^{1/2}\exp(-\tfrac{1}{2}E^2)H_n(E). \quad (152)$$

$$H_0(x) = 1. \quad (153)$$

$$H_1(x) = x. \quad (154)$$

$$H_2(x) = x^2 - 1. \quad (155)$$

$$H_3(x) = x^3 - 3x. \quad (156)$$

$$H_4(x) = x^4 - 6x^2 + 3. \quad (157)$$

$$H_5(x) = x^5 - 10x^3 + 15x. \quad (158)$$

$$\int_{-\infty}^{\infty} x\exp(ibx)\exp(-\tfrac{1}{2}x^2)\,\mathrm{d}x = (2\pi)^{1/2} ib\exp(-\tfrac{1}{2}b^2). \quad (159)$$

$$\int_{-\infty}^{\infty} x^2\exp(ibx)\exp(-\tfrac{1}{2}x^2)\,\mathrm{d}x = (2\pi)^{1/2}(1-b^2)\exp(-\tfrac{1}{2}b^2). \quad (160)$$

$$\ln(1+x) = x - x^2/2 + x^3/3 - \ldots \quad \text{for } |x| < 1. \quad (161)$$

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