N is a small power of two times an odd number and, further, there is also an advantage if that odd number is a product of several different prime numbers rather than a high power of a small one. A library of efficient FFT routines for a wide range of values of N is being developed (An, Lu, Prince & Tolimieri, 1992).

References

Agarwal, R. C. & Cooley, J. W. (1987). *Proc. IEEE*, **75**, 1283–1292.

An, M., Lu, C., Prince, E. & Tolimieri, R. (1992). *Acta Cryst.* A**48**, 346–349.

Blahut, R. E. (1985). *Fast Algorithms for Digital Signal Processing*. Reading, MA: Addison-Wesley.

Burrus, C. S. & Eschenbacher, P. W. (1979). *IEEE Trans. Acoust. Speech Signal Process.* **ASSP-29**, 806–817.

Cooley, J. W. & Tukey, J. W. (1965). *Math. Comput.* **19**, 297–301.

Good, I. J. (1958). *J. R. Statist. Soc. B*, **20**, 361–372.

Kolba, D. P. & Parks, T. W. (1977). *IEEE Trans. Acoust. Speech Signal Process.* **ASSP-25**, 281–294.

Press, W. H., Flannery, B. P., Teukolsky, S. A. & Vetterling, W. T. (1986). *Numerical Recipes*. Cambridge Univ. Press.

Rader, C. M. (1968). *Proc. IEEE*, **56**, 1107–1108.

Temperton, C. (1985). *J. Comput. Phys.* **58**, 283–299.

Ten Eyck, L. F. (1973). *Acta Cryst.* A**29**, 183–191.

Thomas, L. H. (1963). *Using a Computer to Solve Problems in Physics, Applications of Digital Computers*. Boston: Ginn & Co.

Tolimieri, R., An, M. & Lu, C. (1989). *Algorithms for Discrete Fourier Transform and Convolution*. New York: Springer.

Winograd, S. (1978). *Math. Comput.* **32**, 175–199.

Acta Cryst. (1992). A**48**, 418-423

Exact Conditional Joint Probability Distribution of a Three-Phase Invariant in Space Group *P*2. I. Derivation of the Fourier Coefficients

By Uri Shmueli

School of Chemistry, Tel Aviv University, 69978 *Tel Aviv, Israel*

and George H. Weiss

Physical Sciences Laboratory, Division of Computer Sciences and Technology, National Institutes of Health, Bethesda, Maryland 20892, *USA*

(*Received* 20 *May* 1991; *accepted* 30 *September* 1991)

Abstract

We extend our study of the conditional probability density function (c.p.d.f.) of the three-phase invariant for the space group $P1$ [Shmueli, Rabinovich & Weiss (1989). *Acta Cryst.* A**45**, 361-367] to the monoclinic space group $P2$. A detailed derivation of the characteristic function (and hence Fourier coefficients) of the latter c.p.d.f. is presented in this paper, as well as some simplifications of the resulting expressions.

Introduction

The first exact study of the conditional probability density function (p.d.f.) of a three-phase invariant was presented recently (Shmueli, Rabinovich & Weiss, 1989*a*) in terms of a Fourier representation of the relevant hexavariate p.d.f. The resulting series was then adapted to computer evaluation by suitably partitioning the sums and taking their symmetry into account (Shmueli, Rabinovich & Weiss, 1989*b*). Although the formalism appeared to be extremely complicated it was seen that by properly exploiting the symmetry inherent in the Fourier summations it is possible to reduce the computing efforts sufficiently that conventional mainframes and workstations are able to cope with the relevant computations. The study cited earlier contains a derivation of the general form of the conditional p.d.f. as well as its evaluation for the space group $P1$ - the simplest and, so far, the only example for which noncentrosymmetric direct-methods formalisms have been extensively developed. Our earlier study shows that the general form of the conditional p.d.f. is given by a summation of the form

$$\sum_{\mathbf{u}} C_{\mathbf{u}} Z_{\mathbf{u}} \qquad (1)$$

where $\mathbf{u}$ is a vector of the (six) summation indices and the $C_{\mathbf{u}}$ are coefficients depending on the composition and symmetry of the crystal. The function $Z_{\mathbf{u}}$ depends on the magnitudes of the normalized structure factors and is the same for all the symmetries and compositions. While the conditional p.d.f. for the three-phase invariant in $P1$, in either its approximate (Cochran, 1955) or exact (Shmueli *et al.*, 1989*a*) form, may lead to satisfactory practical algorithms, we believe that it is also desirable to examine the effect of symmetry on this important statistic. To do this, we need only calculate $C_{\mathbf{u}}$ in (1). As pointed out, *e.g.* by Shmueli & Weiss (1985), these Fourier coefficients are just the values of the characteristic function $C(\omega_1, \omega_2, \ldots, \omega_k, \ldots)$ of the p.d.f. at the

0108-7673/92/040418-06$06.00

points $\omega_k = \pi\alpha u_k$, where α is the reciprocal of the maximum value of the magnitude of the normalized structure factor and u_k is a summation index of a Fourier series. Thus all we need to do is to derive the characteristic function for the space-group symmetry of interest.

The next section presents a derivation of the general form of the characteristic function of the condition p.d.f. for the space group $P2$ and the Appendixes deal with some background material and the solution of an integral needed in the text.

Derivation of the characteristic function

We recall that the three-phase structure invariant is usually understood as the sum

$$\Phi = \varphi(\mathbf{h}) + \varphi(\mathbf{k}) + \varphi(-\mathbf{h}-\mathbf{k}), \tag{2}$$

where $\varphi(\mathbf{h})$ is the phase of the normalized structure factor $E(\mathbf{h})$. On the assumption that all the atoms are located in general positions and that non-crystallographic symmetry is absent,

$$E(\mathbf{h}) = \sum_{j=1}^{N/g} n_j \sum_{s=1}^{g} \exp[2i\mathbf{h}^T(\mathbf{P}_s\mathbf{r}_j + \mathbf{t}_s)] \equiv A(\mathbf{h}) + iB(\mathbf{h}). \tag{3}$$

In this equation, g is the number of equivalent general positions in the unit cell, N is the number of atoms in the cell, n_j is the normalized scattering factor of the jth atom, $\mathbf{h}$ is the diffraction vector, $\mathbf{P}$ and $\mathbf{t}$ are the rotation and translation parts of the sth space-group operation respectively and r_j is the position vector of the jth atom in the reference asymmetric unit. We shall also abbreviate all the quantities depending on $\mathbf{h}$, $\mathbf{k}$ and $-\mathbf{h}-\mathbf{k}$ by appending to them the subscripts 1, 2 and 3, *e.g.* $A(\mathbf{h})$, $A(\mathbf{k})$ and $A(-\mathbf{h}-\mathbf{k})$ will be written as A_1, A_2 and A_3. The general expression for the characteristic function of the three-phase invariant is given by

$$C(\omega_1, \ldots, \omega_6) = \left\langle \exp\left[i \sum_{k=1}^{3} (\omega_{2k-1}A_k + \omega_{2k}B_k) \right] \right\rangle \tag{4}$$

(Shmueli *et al.*, 1989*a*, *b*) and the components of the normalized structure factor for the space group $P2$ can be written as

$$A_k = 2\sum_{j=1}^{N/2} n_j \cos\theta_{jk} \cos\psi_{jk}, \tag{5}$$

$$B_k = 2\sum_{j=1}^{N/2} n_j \cos\theta_{jk} \sin\psi_{jk}, \tag{6}$$

where $\theta_{jk} = 2\pi(h_k x_j + l_k z_j)$ and $\psi_{jk} = 2\pi k_k y_j$ and h, k, l and x, y, z are components of $\mathbf{h}$ and $\mathbf{r}$ respectively (*International Tables for X-ray Crystallography*, 1965).

The constraints

$$\sum_{k=1}^{g} \theta_{jk} = 0 \quad \text{and} \quad \sum_{k=1}^{g} \psi_{jk} = 0$$

follow from the invariance condition. If we substitute (5) and (6) into (4) and make use of the assumption of independent atomic contributions, we can rewrite (4) as

$$C(\omega_1, \ldots, \omega_6) = \prod_{j=1}^{N/2} C_j(\omega_1, \ldots, \omega_6) \tag{7}$$

where

$$C_j(\omega_1, \ldots, \omega_6) = \left\langle \exp\left[2in_j \sum_{k=1}^{3} \cos\theta_{jk}(\omega_{2k-1}\cos\psi_{jk} + \omega_{2k}\sin\psi_{jk}) \right] \right\rangle_{\theta,\psi} \tag{8}$$

$$= \left\langle \exp\left[i \sum_{k=1}^{3} \Lambda_{jk} \cos\theta_{jk} \cos(\psi_{jk} - \Delta_k) \right] \right\rangle_{\theta,\psi} \tag{9}$$

where $\Lambda_{jk} = 2n_j(\omega_{2k-1}^2 + \omega_{2k}^2)^{1/2}$ and $\Delta_k = \tan^{-1}(\omega_{2k}/\omega_{2k-1})$. We first perform the average over ψ by making use of the relationship

$$\exp[i(\Lambda_{jk}\cos\theta_{jk})\cos(\psi_{jk} - \Delta_k)] = \sum_{q=-\infty}^{\infty} i^q J_q(\Lambda_{jk}\cos\theta_{jk}) \exp[iq(\psi_{jk} - \Delta_k)] \tag{10}$$

[*cf.* Gradshteyn & Ryzhik, 1980, equation 8.511(4)]. If we substitute the right-hand side of (10) in (9) and replace ψ_{j3} by $-\psi_{j1} - \psi_{j2}$, we obtain

$$C_j(\omega_1, \ldots, \omega_6) = \left\langle \sum_{q=-\infty}^{\infty} \sum_{r=-\infty}^{\infty} \sum_{s=-\infty}^{\infty} i^{q+r+s} T_{qrs} I_{qrs} \right\rangle_{\theta}, \tag{11}$$

where

$$T_{qrs} = J_q(G_{j1}) J_r(G_{j2}) J_s(G_{j3}) \times \exp[-i(q\Delta_1 + r\Delta_2 + s\Delta_3)], \tag{12}$$

with $G_{jk} = \Lambda_{jk}\cos\theta_{jk}$ and

$$I_{qrs} = (1/4\pi^2) \int_{-\pi}^{\pi}\int_{-\pi}^{\pi} \exp[i(q-s)\psi_1] \times \exp[i(r-s)\psi_2]\, d\psi_1\, d\psi_2 = \delta_{qs}\delta_{rs}. \tag{13}$$

where δ is the Kronecker delta. Equation (11), taking (12) and (13) into account, can therefore be rewritten as

$$C_j(\omega_1, \ldots, \omega_6) = \sum_{q=-\infty}^{\infty} i^{3q} \exp(-iq\Delta) \times \left\langle \prod_{k=1}^{3} J_q(G_{jk}) \right\rangle_{\theta} \tag{14}$$

where $\Delta = \Delta_1 + \Delta_2 + \Delta_3$. It is readily shown, following the analysis of Shmueli *et al.* (1989*a*, *b*), that for Bessel functions of low orders and small values of ω (14) leads to the Cochran approximation, which is based on the central limit theorem. A rigorous averaging over θ can be performed by replacing θ_{j3} in (14) with $-\theta_{j1} - \theta_{j2}$ and making use of one of the forms of the Neumann summation theorem for Bessel functions (Watson, 1922; Gradshteyn & Ryzhik, 1980). However, we can exploit the symmetry inherent in the problem by observing that the three θs are linked by the requirement that their sum must be zero. This allows us to introduce a Fourier representation of the periodic Dirac delta function as

$$\delta_{2\pi}(\theta_1+\theta_2+\theta_3) = (1/2\pi) \sum_{s=-\infty}^{\infty} \exp[is(\theta_1+\theta_2+\theta_3)] \tag{15}$$

(*e.g.* Bremermann, 1985) and, consequently, re-express the average on the right-hand side of (14) as

$$\begin{aligned}
&\left\langle \prod_{k=1}^{3} J_q(\Lambda_{jk}\cos\theta_{jk}) \right\rangle \\
&= (1/4\pi^2) \int_{-\pi}^{\pi}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi} \delta_{2\pi}(\theta_1+\theta_2+\theta_3) \\
&\quad \times \prod_{k=1}^{3} J_q(\Lambda_{jk}\cos\theta_k)\,\mathrm{d}\theta_1\,\mathrm{d}\theta_2\,\mathrm{d}\theta_3 \\
&= \sum_{s=-\infty}^{\infty} \prod_{k=1}^{3} \left[(1/2\pi) \int_{-\pi}^{\pi} J_q(\Lambda_{jk}\cos\theta_k) \right. \\
&\quad \left. \times \exp(is\theta_k)\,\mathrm{d}\theta_k \right] \qquad (16) \\
&= \sum_{s=-\infty}^{\infty} \prod_{k=1}^{3} \left[(1/\pi) \int_{0}^{\pi} J_q(\Lambda_{jk}\cos\theta_k)\cos(s\theta_k)\,\mathrm{d}\theta_k \right]. \qquad (17)
\end{aligned}$$

The atomic characteristic function for this space group therefore becomes

$$C_j(\omega_1,\ldots,\omega_6) = \sum_{q=-\infty}^{\infty} i^{3q} \exp(-iq\Delta)\mathbf{K}_{q,j}, \tag{18}$$

where the summation $\mathbf{K}_{q,j}$, restricted to non-negative values of s, is given by

$$\begin{aligned}
\mathbf{K}_{q,j} &= \prod_{k=1}^{3} \left[(1/\pi) \int_{0}^{\pi} J_q(\Lambda_{jk}\cos\theta_k)\,\mathrm{d}\theta_k \right] \\
&\quad + 2 \sum_{s=1}^{\infty} \prod_{k=1}^{3} \left[(1/\pi) \int_{0}^{\pi} J_q(\Lambda_{jk}\cos\theta_k) \right. \\
&\quad \left. \times \cos(s\theta_k)\,\mathrm{d}\theta_k \right]. \qquad (19)
\end{aligned}$$

Note for further reference that $\mathbf{K}_{-q,j} = (-1)^q \mathbf{K}_{q,j}$ and that the change of the sign of s in the integral in (19) does not affect its value.

The formal expression for the required Fourier coefficient is now obtained by replacing ω_q in (7), (18) and (19) with $\pi\alpha q$, where α is the reciprocal of the maximum value of $|E|$. For example, Λ_{jk} in (19) then becomes $2\pi\alpha n_j (u_{2k-1}^2 + u_{2k}^2)^{1/2}$. We recall in Appendix *A* the general structure of the Fourier representation of the conditional p.d.f. of the three-phase invariant and show that the conditional p.d.f.s for the space groups $P1$ and $P2$ differ only in the functional form of the *real* composition- and symmetry-dependent factor $\mathbf{K}_{q,j}$.

The real and imaginary parts of the atomic characteristic function can be obtained as follows. Equation (18) can be decomposed as

$$C_j = R_j + iI_j, \tag{20}$$

where

$$\begin{aligned}
R_j &= \tfrac{1}{2}(C_j + C_j^*) \\
&= \tfrac{1}{2} \sum_{q=-\infty}^{\infty} [i^{-q}\exp(-iq\Delta) + i^q \exp(iq\Delta)]\mathbf{K}_{q,j} \\
&= \mathbf{K}_{0,j} + \sum_{q=1}^{\infty} \cos\{q[(\pi/2)+\Delta]\}\mathbf{K}_{q,j}[1+(-1)^q] \qquad (21) \\
&= \mathbf{K}_{0,j} + 2\sum_{q=1}^{\infty} (-1)^q \cos(2q\Delta)\mathbf{K}_{2q,j}, \qquad (22)
\end{aligned}$$

since the sum of a $(q, -q)$ pair in (21) must vanish if q is odd. We further have

$$\begin{aligned}
I_j &= (1/2i)(C_j - C_j^*) \\
&= (1/2i) \sum_{q=-\infty}^{\infty} [i^{-q}\exp(-iq\Delta) - i^q \exp(iq\Delta)]\mathbf{K}_{q,j} \\
&= -\sum_{p=1}^{\infty} \sin\{q[(\pi/2)+\Delta]\}\mathbf{K}_{q,j}[1-(-1)^q] \qquad (23) \\
&= 2\sum_{q=1}^{\infty} (-1)^q \cos[(2q-1)\Delta]\mathbf{K}_{2q-1,j} \qquad (24)
\end{aligned}$$

since the sum of a $(q, -q)$ pair in (23) must vanish if q is even.

As shown in Appendix *B*, the integral in (19) can be represented in terms of Bessel functions,

$$\begin{aligned}
I &= (1/\pi) \int_{0}^{\pi} J_q(\Lambda_{jk}\cos\theta_k)\cos(s\theta_k)\,\mathrm{d}\theta_k \\
&= \begin{cases} J_{(q+s)/2}\left(\dfrac{\Lambda_{jk}}{2}\right) J_{(q-s)/2}\left(\dfrac{\Lambda_{jk}}{2}\right), & q+s=2n \\ 0, & q+s=2n+1 \end{cases} \qquad (25)
\end{aligned}$$

where n is any integer. It follows that only Bessel functions of integer order are required for this calculation. The symmetry of this integral with respect to the interchange of the q and s indices is also discussed

in Appendix *B*. If we define

$$J_{(q+s)/2}(\Lambda_{jk}/2)J_{(q-s)/2}(\Lambda_{jk}/2) \equiv T^k_{qs} \tag{26}$$

the relation

$$T^k_{qs} = \begin{cases} T^k_{sq} & \text{for } q-s=4n \\ -T^k_{sq} & \text{for } q-s=4n-2 \end{cases} \tag{27}$$

obtained from that discussion may also be useful in attempts at reducing the extent of the computing effort needed. If we abbreviate the product appearing in (17) by

$$\prod_{k=1}^{3}\left[(1/\pi)\int_0^{\pi} J_q(\Lambda_{jk}\cos\theta_k)\cos(s\theta_k)\,d\theta_k\right] = \prod_{k=1}^{3} T^k_{qs} \equiv W_{q,s}, \tag{28}$$

the forms of $\mathbf{K}_{q,j}$ required by (22) and (24) can be obtained explicitly. Since q and s must have the same parity, the composition- and symmetry-dependent functions for the real part R_j are

$$\mathbf{K}_{0,j} = \sum_{s=-\infty}^{\infty} W_{0,s} = W_{0,0} + 2\sum_{s=1}^{\infty} W_{0,2s} \tag{29}$$

$$= \prod_{k=1}^{3} J_0^2(\Lambda_{jk}/2) + 2\sum_{s=1}^{\infty}\prod_{k=1}^{3} J_s(\Lambda_{jk}/2)J_{-s}(\Lambda_{jk}/2) \tag{30}$$

$$= \prod_{k=1}^{3} J_0^2(\Lambda_{jk}/2) + 2\sum_{s=1}^{\infty}(-1)^s\prod_{k=1}^{3} J_s^2(\Lambda_{jk}/2) \tag{31}$$

and

$$\mathbf{K}_{2q,j} = \sum_{s=-\infty}^{\infty} W_{2q,s}$$

$$= W_{2q,0} + 2\sum_{s=1}^{\infty} W_{2q,2s} \tag{32}$$

$$= W_{2q,0} + 2W_{2q,2q} + 2\sum_{s<q}(W_{2q,2s} + W_{2s,2q}) \tag{33}$$

$$= \prod_{k=1}^{3} J_q^2(\Lambda_{jk}/2) + 2\prod_{k=1}^{3} J_{2q}(\Lambda_{jk}/2)J_0(\Lambda_{jk}/2) + 2\sum_{s<q} Q_R, \tag{34}$$

where

$$Q_R = 2\prod_{k=1}^{3} J_{q+s}\left(\frac{\Lambda_{jk}}{2}\right)J_{q-s}\left(\frac{\Lambda_{jk}}{2}\right), \quad q-s=2n \tag{35}$$

and

$$Q_R = 0, \qquad q-s=2n+1.$$

and that entering the imaginary part I_j is

$$\mathbf{K}_{2q-1,j} = \sum_{s=-\infty}^{\infty} W_{2q-1,2s-1}$$

$$= 2W_{2q-1,2q-1} + 2\sum_{s<q}(W_{2q-1,2s-1} + W_{2s-1,2q-1}) \tag{36}$$

$$= 2\prod_{k=1}^{3} J_{2q-1}(\Lambda_{jk}/2)J_0(\Lambda_{jk}/2) + 2\sum_{s<q} Q_I, \tag{37}$$

where

$$Q_I = 2\prod_{k=1}^{3} J_{q+s-1}\left(\frac{\Lambda_{jk}}{2}\right)J_{q-s}\left(\frac{\Lambda_{jk}}{2}\right), \quad q-s=2n \tag{38}$$

and

$$Q_I = 0, \qquad q-s=2n+1.$$

The computation is then running over one eighth of the (q, s) index plane, where only terms with $q+s=2n$ need be considered, *i.e.* only about one sixteenth of the possible combinations of the (q, s) indices has to be used.

The foregoing development leads to computable expressions, the programming of which is in progress. The decomposition of the conditional p.d.f. into partial summations follows a similar route to that described elsewhere (Shmueli *et al.*, 1989*a*) and is aided by the analogous structure of the equations (*cf.* Appendix *A*). It is interesting to note that the functional forms of some partial summations indicate that the discrepancy between the exact conditional p.d.f.s for *P*2 and the corresponding ones resulting from the central limit theorem is likely to be smaller than that observed for the space group *P*1. However, conclusive statements can be made only after the computations are completed.

This work was supported in part by research grants from the United States–Israel Binational Science Foundation (BSF) and the Israel Academy of Sciences.

APPENDIX *A*

The purpose of this Appendix is to recall the general form of the conditional p.d.f. for the three-phase invariant and compare the general expressions for the space groups *P*1 (Shmueli *et al.*, 1989*a*) and *P*2 (this paper). The p.d.f. of the three-phase invariant, Φ, as defined by (2) in the text, conditioned on the magnitudes of three normalized structure factors, crystallographic symmetry and atomic composition, is given by

$$p(\Phi|E_1, E_2, E_3, \ldots) = \mathbf{K}\sum_{\mathbf{u}} C_{\mathbf{u}}(\Delta)Z_{\mathbf{u}}(\Phi, \Delta), \tag{A1}$$

where $\mathbf{K}$ is a normalization factor; $\mathbf{u}^T = (u_1\ u_2\ u_3\ u_4\ u_5\ u_6)$ is the vector of the Fourier summation indices for the hexavariate joint p.d.f., from which the conditional p.d.f. is derived; $C_{\mathbf{u}}(\Delta)$ is the Fourier coefficient corresponding to a certain sextet of summation indices and expressible as

$$C_{\mathbf{u}}(\Delta) = \prod_{j=1}^{N/g} C_{\mathbf{u},j}(\Delta) \qquad (A2)$$

where N is the number of atoms in the unit cell and g is 1 or 2 for $P1$ or $P2$ respectively; the atomic contribution to the Fourier coefficient is

$$C_j(\Delta) = \sum_{q=-\infty}^{\infty} i^{-q} \exp(-iq\Delta)\mathbf{K}_{q,j} \qquad (A3)$$

and the integrated phase factor $Z_{\mathbf{u}}(\Phi, \Delta)$ is given by

$$Z_{\mathbf{u}}(\Phi, \Delta) = \sum_{p=-\infty}^{\infty} i^p \exp[ip(\Phi - \Delta)]\mathbf{G}_p. \qquad (A4)$$

It is shown by Shmueli *et al.* (1989*a*) that the quantity $\mathbf{G}_p$ is given by

$$\mathbf{G}_p = \prod_{k=1}^{3} J_p(E_k\Omega_k), \qquad (A5)$$

where

$$\Omega_k = \pi\alpha(u_{2k-1}^2 + u_{2k}^2)^{1/2} \qquad (A6)$$

and is therefore independent of the atomic composition and crystal symmetry. The above-quoted reference along with the text of this paper show that the only difference between the structures of the conditional p.d.f.s for the space groups $P1$ and $P2$ lies in the quantity $\mathbf{K}_{q,j}$, which is given by

$$\mathbf{K}_{q,j} = \prod_{k=1}^{3} J_q(n_j\Omega_k) \qquad (A7)$$

for the space group $P1$ (Shmueli *et al.*, 1989*a*) and by

$$\mathbf{K}_{q,j} = \sum_{s=-\infty}^{\infty} \prod_{k=1}^{3} \left\{ (1/\pi) \int_0^{\pi} J_q[2n_j\Omega_k \cos(\phi_k)] \times \cos(s\phi_k)\, \mathrm{d}\phi_k \right\} \qquad (A8)$$

for the space group $P2$ (see text).

The phase factor $\Delta = \Delta_1 + \Delta_2 + \Delta_3$, with $\Delta_k = \tan^{-1}(u_{2k}/u_{2k-1})$ has played a very important role in the decomposition of the conditional p.d.f. into partial summations (Shmueli *et al.*, 1989*a*, *b*). Appendix *A* of the latter reference describes in detail the procedures of restricting the Fourier summations to positive indices and exploiting the intra-pair and inter-pair symmetries in order to avoid repetitive computations. In fact, most arguments put forward in that Appendix are applicable to the numerical study of the conditional p.d.f. for the three-phase invariant for space group $P2$, which is now in progress.

APPENDIX *B*

We now deal with the solution of the definite integral

$$\int_0^{\pi} J_q(\Lambda_{jk} \cos\theta_k) \cos(s\theta_k)\, \mathrm{d}\theta_k,$$

where q and s are integers, which first appears in (17) in the text. The integral can be written in the form

$$\int_0^{\pi} J_q[2a\cos(x)]\cos(sx)\,\mathrm{d}x = \int_0^{\pi/2} J_q[2a\cos(x)]\cos(sx)\,\mathrm{d}x + \int_{\pi/2}^{\pi} J_q[2a\cos(x)]\cos(sx)\,\mathrm{d}x, \qquad (B1)$$

where

$$\int_0^{\pi/2} J_q(2a\cos x)\cos(sx)\,\mathrm{d}x \equiv K = (\pi/s)J_{(q+s)/2}(a)J_{(q-s)/2}(a) \qquad (B2)$$

is a known definite integral [Gradshteyn & Ryzhik, 1980; equation 6.681(1)]. The second integral on the right-hand side of (*B*1) can be reduced to a tractable form by introducing the change of variable $y = \pi - x$. Hence $x = \pi - y$, $\mathrm{d}x = -\mathrm{d}y$, the integration limits on x, $(\pi/2)$ to π, become $(\pi/2)$ to 0 on y,

$$\cos(sx) = \cos[s(\pi - y)] = \cos(s\pi)\cos(sy) = (-1)^s\cos(sy)$$

$$J_q(2a\cos x) = J_q[2a\cos(\pi - y)] = (-1)^q J_q[2a\cos(y)]$$

and (*B*1) becomes

$$\int_0^{\pi} J_q(2a\cos x)\cos(sx)\,\mathrm{d}x = K - (-1)^{q+s}\int_{\pi/2}^{0} J_q(2a\cos x)\cos(sx)\,\mathrm{d}x = [1+(-1)^{q+s}]K = \begin{cases} \pi J_{(q+s)/2}(a)J_{(q-s)/2}(a) & \text{for } q+s \text{ even} \\ 0 & \text{for } q+s \text{ odd.} \end{cases} \qquad (B3)$$

Of course, the orders of the Bessel functions appearing in (*B*3) must be integers. This product of Bessel functions is symmetric with respect to the interchange of the indices q and s if $q - s = 4n$ and is antisymmetric with respect to such an interchange if $q - s = 4n + 2$. This further reduces the computing time required for the valuation of the double summations in (22) and (24).

References

BREMERMANN, H. (1985). *Distributions, Complex Variables, and Fourier Transforms.* Reading, MA: Addison-Wesley.

COCHRAN, W. (1955). *Acta Cryst.* **8**, 473-478.

GRADSHTEYN, I. S. & RYZHIK, I. M. (1980). *Table of Integrals, Series and Products.* New York: Academic Press.

International Tables for X-ray Crystallography (1965). Vol. I. Birmingham: Kynoch Press.

SHMUELI, U., RABINOVICH, S. & WEISS, G. H. (1989*a*). *Acta Cryst.* **A45**, 361-367.

SHMUELI, U., RABINOVICH, S. & WEISS, G. H. (1989*b*). *Acta Cryst.* **A45**, 367-371.

SHMUELI, U. & WEISS, G. H. (1985). *Acta Cryst.* **A41**, 401-408.

WATSON, G. N. (1922). *A Treatise on the Theory of Bessel Functions.* Cambridge Univ. Press.

Acta Cryst. (1992). A**48**, 423-430

Magnetic *Pendellösung* Effects in Neutron Scattering by Perfect Magnetic Crystals

BY V. V. KVARDAKOV AND V. A. SOMENKOV

I. V. Kurchatov Institute of Atomic Energy, Moscow 123182, *Russia*

(*Received* 18 *December* 1990; *accepted* 6 *December* 1991)

Abstract

Pendellösung effects in magnetic neutron scattering by thin (thickness of the order of the extinction length) crystals of weak ferromagnets $FeBO_3$ and α-Fe_2O_3 were investigated. Dynamical oscillations in the scattering intensity dependences on the crystal's effective thickness, temperature and magnetic field orientation have been found. The influence of irregularities in the magnetic structure related to domains, a magnetic phase transition and magnetoelastic oscillations on the *Pendellösung* oscillation amplitude has been established. The oscillation amplitude and the scattering intensity dependence on the structure-factor magnitude have been used to obtain information on small deformations from perfection of the crystalline and magnetic structures, which are difficult to detect by other methods.

Introduction

In the case of diffraction by a perfect crystal, the wave field is determined by the dynamical interaction between the transmitted and scattered waves (Zachariasen, 1945; Laue, 1960). This results in a number of dynamical effects, one of which is the *Pendellösung* effect, *i.e.* an oscillating dependence of the scattered intensity in Laue geometry on the ratio of the crystal thickness to the extinction length. The dynamical oscillations were studied in detail in X-ray scattering (Kato & Lang, 1959; Utemisov, Somenkova, Somenkov & Shilstein, 1980) and in nuclear neutron scattering (Sippel, Kleinstück & Schulze, 1965; Shull, 1968; Somenkov, Shilstein, Belova & Utemisov, 1978). The theoretical aspects of neutron scattering by perfect magnetically ordered crystals were treated in a number of papers (Stassis & Oberteuffer, 1974; Sivardière, 1975; Gukasov & Ruban, 1975; Schmidt & Deimel, 1976; Belyakov & Bokun, 1975, 1976; Mendiratta & Blume, 1976; Baryshevskii, 1976; Guigay & Schlenker, 1978). However, *Pendellösung* oscillations were observed in the magnetic neutron scattering case only recently (Baruchel, Guigay, Mazure-Espejo, Schlenker & Schweizer, 1982; Kvardakov, Somenkov & Shilstein, 1988; Zelepukhin, Kvardakov, Somenkov & Shilstein, 1989) owing to the small number of perfect magnetically ordered crystals.

Baruchel *et al.* (1982) used the anisotropy of the defect distribution in incompletely perfect yttrium iron garnet crystals. The analyzed reflection had an orientation of the scattering vector such that the effect of defects on the diffraction was minimal. As a result, a peak corresponding to one of the dynamical oscillations was found in the dependence of the mixed (nuclear–magnetic) scattering intensity of polarized neutrons on the wavelength.

Kvardakov, Somenkov & Shilstein (1988) and Zelepukhin, Kvardakov, Somenkov & Shilstein (1989) reported on the *Pendellösung*-effect observation in pure magnetic neutron scattering by using the inclination method. The method was proposed earlier by Somenkov *et al.* (1978) for the case of nuclear neutron scattering and later it was used for *Pendellösung*-effect investigation in X-ray (Utemisov *et al.*, 1980) and synchrotron-radiation (Belova & Kabannik, 1985) scattering, for precise determination of structure factors (Saka & Kato, 1986) and for the study of extinction parameters connected with microdefects (Voronkov, Piskunov, Chukhovskii & Maksimov, 1987).

In works of Kvardakov, Somenkov & Shilstein (1988) and Zelepukhin *et al.* (1989), thin (of the order of the extinction length) crystals of weak ferromagnets iron borate $FeBO_3$ and hematite α-Fe_2O_3 were

0108-7673/92/040423-08$06.00