

## Surface Spherical Harmonics and Intensity and Strain Pole Figures of Cubic Textured Materials

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### Abstract

The equations for diffraction strain pole figures measured on textured cubic materials exhibit an  $hkl$  dependence. This is expressed by an  $hkl$ -permutation-invariant 'surface' spherical harmonic. Four types of new harmonics are defined. These harmonics differ in essence from those for the  $hkl$ -dependent expression obtained for diffraction intensity pole figures. In the latter case associated Legendre polynomials arise whereas in the former (Jacobi type) generalizations of these polynomials occur. Equations exhibiting diffraction intensity and diffraction strain expressions are given for all cubic point groups. Structure factors arise in the expressions. The treatment is given for both anomalous and normal scattering modes. Surface spherical harmonics do not satisfy Laplace's equation. This only occurs upon conversion into 'solid spherical harmonics'. Then the harmonics associated with intensity pole figures satisfy Laplace's equation. The 'diffraction strain harmonics' do not, however. Orthonormalization is also different from the case of conventional  $hkl$ -permutation-invariant surface spherical harmonics. Stereographic projections are given for a few examples of harmonics.

### 1. Introduction

Spherical harmonics occur in (among others) quantum mechanics, electromagnetism, geodesics and crystal physics. The present treatment deals with spherical surface harmonics in conjunction with crystal physics and, more specifically, residual stress analysis of textured cubic materials.

The new harmonics occur in the  $hkl$ -dependent part of the expression for diffraction line-shift strain pole figures obtained on textured materials for the general  $\{hkl\}$  reflection case. Diffraction strain may be defined as the averaged strain of the crystals engaged in the diffraction experiment. It depends on  $hkl$ , the loading of the specimen and the orientation of the scattering vector  $\mathbf{Q}$  with respect to the specimen's reference axes [definition of  $\mathbf{Q}$  according to Sayers (1984)]. It is determined from the shift of the diffraction line's profile with respect to *some* initial

condition. Crystals exhibit different strains depending on their orientation, when subject to an even uniform stress system. If the specimen exhibits crystallographic texture some orientations may occur frequently, others may be absent. Consequently the texture enters in the diffraction strain equations (Brakman, 1983; Sayers, 1984). Since the diffraction strain depends on the orientation of  $\mathbf{Q}$  it can be represented in terms of a pole figure. Like the conventional diffraction intensity pole figure, it can be depicted in a stereographic projection with respect to specimen axes. By contrast with the intensity pole figure the strain pole figure depends on the stress state (residual or externally applied) of the volume of material irradiated. If the texture is homogeneous over the specimen, the intensity pole figure does not depend on the size of the volume irradiated provided grain statistics are adequately retained. The intensity pole figure is therefore an *intensive* quantity. Although stress and strain are also intensive quantities, the strain pole figure is an *extensive* quantity.

For instance, the case of neutron diffraction (Sayers, 1984; Pintschovius & Macherlauch, 1982) could yield a totally different strain pole figure from that obtained using X-rays. In the case of X-ray diffraction the strain pole figure represents a very local average of the specimen strains due to the strong attenuation of the radiation. In the neutron diffraction experiment the specimen as a whole may be irradiated. For each specimen the conditions of mechanical equilibrium must be satisfied. Then the strains due to *macro*stresses (assumed to be constant over distances large compared with the grain size) compensate. This does not occur in the X-ray experiment.

The intensity pole figure presents data in a (normalized) compact form allowing visual interpretation and presentation. The texture type can be determined from one or more pole figures. The strain pole figure may serve a similar purpose. If the texture is known the approximate stress state can be determined from the strain pole figure. Practical examples are given by Hoffmann, Neff, Scholtes & Macherlauch (1984) and Hauk, Vaessen & Weber (1985). The orientation distribution function (o.d.f.) of the crystals of a textured specimen is usually expanded in a series

(Bunge, 1982, p. 47). The expansion coefficients can be calculated from intensity pole figures. However, *these* pole figures only allow even-order (Bunge, 1982, pp. 100–107) expansion coefficients. The strain pole figures also exhibit odd-order expansion coefficients (Brakman, 1985). Consequently they provide a means of determining the complete o.d.f.

For point group 432 the expression for the diffraction strain pole figure (according to the Reuss model of elasticity), for the  $hkl$  reflection contribution only, is given in Brakman (1986).<sup>\*</sup> It reads (only stress-tensor elements  $\sigma_{11}$  and  $\sigma_{22}$  are taken non-zero; the expression can easily be extended to the full stress tensor):

$$\begin{aligned}
 \langle \varepsilon'_{zz}(\psi, \alpha) \rangle = & s_1(hkl)(\sigma_{11} + \sigma_{22}) + \frac{1}{2}s_2(hkl)\sigma_\alpha \sin^2 \psi \\
 & + [-s_0/2P_{hkl}(\psi, \alpha)] \\
 & \times [(\sigma_{11} - \sigma_{22}) \cos 2\alpha - \sigma_\alpha \sin^2 \psi] \\
 & \times \sum_{\substack{j=4 \\ \mu=1}} [F_1(hkl, j, \mu)B_j^\mu(\psi, \alpha) \\
 & + F_2(hkl, j, \mu)C_j^\mu(\psi, \alpha)] \\
 & + [s_0/2P_{hkl}(\psi, \alpha)] \\
 & \times (\sigma_{11} - \sigma_{22}) \sin 2\alpha \cos \psi \\
 & \times \sum_{\substack{j=4 \\ \mu=1}} [-F_1(hkl, j, \mu)C_j^\mu(\psi, \alpha) \\
 & + F_2(hkl, j, \mu)B_j^\mu(\psi, \alpha)] \\
 & + [s_0/2P_{hkl}(\psi, \alpha)] \\
 & \times (\sigma_{11} - \sigma_{22}) \sin 2\alpha \sin \psi \\
 & \times \sum_{\substack{j=4 \\ \mu=1}} [F_3(hkl, j, \mu)E_j^\mu(\psi, \alpha) \\
 & - F_4(hkl, j, \mu)D_j^\mu(\psi, \alpha)] \\
 & + [s_0/2P_{hkl}(\psi, \alpha)]\sigma_\alpha \sin 2\psi \\
 & \times \sum_{\substack{j=4 \\ \mu=1}} [F_3(hkl, j, \mu)D_j^\mu(\psi, \alpha) \\
 & + F_4(hkl, j, \mu)E_j^\mu(\psi, \alpha)], \quad (1)
 \end{aligned}$$

where  $s_1(hkl)$  and  $\frac{1}{2}s_2(hkl)$  are the so-called quasi-isotropic diffraction elastic constants (Dölle, 1979).  $P_{hkl}$  is defined in equation (15c). Apart from its corresponding structure factor *and* a proportionality factor, it is equal to the *single*  $hkl$  reflection's scattered intensity.

<sup>\*</sup> For the complete mathematical treatment of the  $\{hkl\}$  diffraction experiment it is necessary to take at least the  $\overline{hkl}$  reflection contribution also into account. This is discussed in more detail in § 4. In order to avoid confusion, the commonly used angle  $\Phi$  is replaced in this paper by the symbol  $\alpha$  in the expression for the diffraction strain,  $\langle \varepsilon'_{zz}(\psi, \alpha) \rangle$  [cf. equation (1)]. The angles  $\psi$  and  $\alpha$  determine the orientation of  $Q$  with respect to specimen axes (cf. Fig. 1).

Functions  $B_j^\mu(\psi, \alpha) \dots E_j^\mu(\psi, \alpha)$  are defined in Brakman (1986). They depend on the texture and need not be discussed here.  $s_0$  stands for single cubic crystal elastic anisotropy:

$$s_0 = s_{1111} - s_{1122} - 2s_{1212} \quad (1a)$$

$$\sigma_\alpha = \sigma_{11} \cos^2 \alpha + \sigma_{22} \sin^2 \alpha. \quad (1b)$$

The stresses  $\sigma_{11}$  and  $\sigma_{22}$  are taken to operate parallel to the specimen's axes of symmetry.\* They are assumed to be macrostresses. It is the purpose of this paper to deal with the harmonics denoted by the symbols  $F_1, F_2, F_3$  and  $F_4$  in (1). They depend on  $hkl, j$  and  $\mu$  similar to the  $K_j^\mu$  harmonics defined in (6) and (18) to (20). An example of a diffraction strain pole figure of a cold-rolled steel specimen *calculated* for an assumed stress state of  $\eta = \sigma_{22}/\sigma_{11} = 10$  is given in Fig. 1.

In the calculation use of the  $\{211\}$  b.c.c. reflection has been assumed and expressions involving functions  $F_2$  and  $F_4$  were omitted.

## 2. Definitions

Spherical harmonics are usually divided into three classes:

(i) Zonal spherical harmonics: the 'normal' Legendre polynomials  $P_j(\cos \Phi)$  and tesseral spherical harmonics: the 'associated' Legendre polynomials  $P_j^m(\cos \Phi)$ .

\* Equation (1) represents the Reuss model of elasticity. For the Voigt model an  $\{hkl\}$ -independent diffraction strain pole figure results. The Kröner model is not (yet) feasible for textured materials. Expression (1) averaged with the Voigt model prediction allows calculation of the stress state of the irradiated volume according to the well known 'sin<sup>2</sup>  $\psi$  method' (Dölle, 1979; Hauk, 1984; Hauk & Macherlauch, 1984; James & Cohen, 1980).

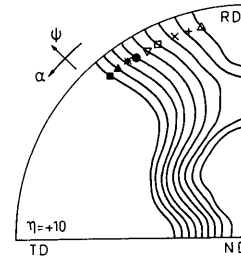


Fig. 1. Calculated  $\{211\}$  diffraction strain for a textured cold-rolled steel sheet sample. Stress state assumed:  $\eta = \sigma_{22}/\sigma_{11} = 10$ . All other stresses taken equal to zero.  $\sigma_{11}$  operates parallel to RD (rolling direction). Diffraction strain  $\langle \varepsilon'_{zz} \rangle$  divided by  $s_0 \sigma_{11}$ . Isostrain values:  $\Delta$   $-1.60$ ,  $+$   $-1.20$ ,  $\times$   $-0.80$ ,  $\square$   $-0.40$ ,  $\nabla$   $0.00$ ,  $\bullet$   $+0.40$ ,  $*$   $+0.80$ ,  $\blacktriangle$   $+1.20$ ,  $\blacksquare$   $+1.60$ . Angle  $\alpha$  measured in the plane of RD and TD (transverse direction). Angle  $\psi$  is the deviation angle from ND (normal direction). The orientation of the scattering vector with respect to the specimen axes is indicated by  $\psi, \alpha$ .

(ii) Surface spherical harmonics:

$$K_j^m(\Phi, \beta) = P_j^m(\cos \Phi) \exp(im\beta) \quad (2)$$

(angles  $\Phi$  and  $\beta$  according to Fig. 2). These functions are often used in normalized form:

$$K_j^m(\Phi, \beta) = (2\pi)^{-1/2} P_j^m(\cos \Phi) \exp(im\beta). \quad (2a)$$

If we use a definition of the associated Legendre polynomials such that (Bunge, 1982, p. 356)

$$\int_0^\pi P_j^m(\cos \Phi) P_j^m(\cos \Phi) \sin \Phi d\Phi = \delta_{jj'}, \quad (3)$$

it follows that\*

$$\oint K_j^m(\Phi, \beta) K_j^{m'}(\Phi, \beta)^* \sin \Phi d\Phi d\beta = \delta_{jj'} \delta_{mm'}. \quad (4)$$

(In what follows the spherical surface element  $\sin \Phi d\Phi d\beta$  will be denoted by  $dA$ .) These functions occur (apart from a factor) as eigenfunctions of the squared angular momentum operator of a single particle in quantum mechanics (Landau & Lifshitz, 1974, p. 54) and also as eigenfunctions of the squared differential operator for infinitesimal rotations (Gel'fand, Minlos & Shapiro, 1963, p. 41).

(iii) Solid spherical harmonics:

$$u = r^j K_j^m(\Phi, \beta) \quad \text{or} \quad u = r^{-j-1} K_j^m(\Phi, \beta). \quad (5)$$

These functions are particular solutions of Laplace's equation  $\Delta u = 0$  (case of  $m = 0$  included) and  $r$  is a radius vector.

The  $K_j^m$  satisfy mathematical constraints:

- (i) They are continuous in the poles of the sphere, *i.e.* exhibit the same value for all  $\beta$  if  $\Phi = 0$  or  $\Phi = \pi$ .
- (ii) They exhibit the same value if  $\beta$  is replaced by  $\beta + 2\pi$ ; 'cyclic behaviour'.
- (iii) They satisfy the orthonormalization condition of (4).
- (iv) Upon multiplication by  $r^j$  or  $r^{-j-1}$  they satisfy Laplace's equation. However, for the physics dis-

cussed in this paper Laplace's equation does not need to be satisfied.

For the application to crystal physics one often needs harmonics satisfying the symmetry of the point group of the crystal under consideration (Betts, Bhatia & Wyman, 1956; Von der Lage & Bethe, 1947; Meyer, 1954; Fox & Krohn, 1977; Kara & Kurki-Suonio, 1981).

### 2.1. Symmetrized harmonics

The symmetry elements of a crystal point group can be seen as transformations of crystal reference axes. They operate on *orientations*. For such orientations, the parent and product crystal orientations are physically indistinguishable. In texture analysis, the symbol  $g$  is commonly used for orientations and transformations of orientations.  $g$  can be written as a  $3 \times 3$  matrix.

There are three types of crystal point-group symmetry elements:

- (i) Pure rotations of which the determinant of  $g$  is equal to +1, 'proper rotations'.
- (ii) Mirror-like operations without an explicit centre of inversion.
- (iii) Centres of inversion.

In the two latter cases  $\det(g)$  is equal to  $-1$ . The harmonics treated in this paper are all defined with respect to crystal reference axes.

For the construction of harmonics satisfying point-group symmetry one can proceed in two different ways:

(a) Construct linear combinations of harmonics  $K_j^m$  and impose the symmetry elements upon them. This method can be applied for the case of cubic harmonics associated with diffraction *intensity* pole figures:

$$K_j^\mu(\Phi, \beta) = \sum_{m=-j}^j A_j^{\mu m} K_j^m(\Phi, \beta). \quad (6)$$

The coefficients can be chosen real (Bunge, 1982, p. 267) and they can be made to satisfy the symmetry requirements of the point group. Equation (6) represents a linear combination of the normal harmonics  $K_j^m$  and  $\mu$  enumerates the number of possible linearly independent constructions  $K_j^\mu$ .

It is required that

$$\oint K_j^\mu(\Phi, \beta) K_j^{\mu'}(\Phi, \beta)^* dA = \delta_{jj'} \delta_{\mu\mu'}. \quad (7)$$

and

$$\sum_{m=-j}^j A_j^{\mu m} A_j^{\mu' m} = \delta_{\mu\mu'}. \quad (8)$$

The total number of possible linearly independent constructions according to (6), *i.e.*  $\mu_{\max}$ , has been given by Bunge (1982, pp. 62 and 365) for even  $j$  and

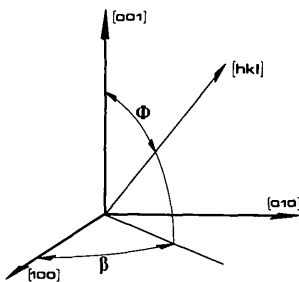


Fig. 2. Definition of angles  $\Phi$  and  $\beta$  with respect to crystallographic axes. The expressions relating  $\Phi$  and  $\beta$  to  $[hkl]$  are given in equations (1-14) and (1-15).

\* The asterisk in mathematical expressions denotes the complex conjugate.

odd  $j$ , respectively.  $\mu_{\max}$  depends on the point group. For non-cubic point groups the coefficients  $A_j^{m\mu}$  take a very simple form (Bunge, 1982, p. 383). For the cubic point groups the full series expansion [cf. (6)] is needed owing to the threefold axis. The coefficients\* cannot be written in a simple form; they have been tabulated by Bunge (1982, p. 501) for point group 432.

(b) For both intensity *and* strain pole figures the  $hkl$ -dependent harmonics arise as a consequence of the physics of the problem. For both types of pole figure the value of the function in the direction of  $\mathbf{Q}$  is obtained as an average over the crystal orientations engaged in the diffraction. For textured materials, the orientation distribution function (o.d.f.)† of the crystals then occurs in the equations for the averaging procedure.

The o.d.f. is a function of *orientations*. It is defined for orientations obtained *via* rotations only. The rotational symmetry elements of the crystal's point group can be imposed upon the o.d.f. with the cubic symmetry coefficients  $A_j^{m\mu}$  treated in § 3. See also Bunge (1982, p. 384).

In the eventual equations for both types of pole figure, harmonics arise satisfying the rotational symmetry elements of the point group considered. However, in some cases the harmonics satisfy additional symmetry elements not imposed on the o.d.f. This may be due to:

(i) The physical nature of the diffraction experiment. Expressions for the contributions from crystal orientations exhibiting  $[hkl]$  parallel to  $\mathbf{Q}$  and  $[\bar{h}\bar{k}\bar{l}]$  parallel to  $\mathbf{Q}$  have to be added. Harmonics satisfying only point group 432 are then cancelled whereas harmonics satisfying  $m3m$  are retained. The same holds for point group 23 with respect to  $m3$ .

(ii) The value of both intensity and strain in the direction of the scattering vector  $\mathbf{Q}$  is an average over basically the two orientation types mentioned above. The averaging covers  $2\pi$  rad rotation about  $\mathbf{Q}$  [angle  $\varphi'_2$  in Brakman (1985)]. The o.d.f. is a function of three independent (Euler) rotation angles for fixed orientation of  $\mathbf{Q}$ . The three angles determine the crystal's orientation with respect to the laboratory frame. The harmonics can be seen as a result of integration of the o.d.f. with respect to one (Euler) angle, *i.e.*  $\varphi'_2$ . For the diffraction strain the same holds

but the o.d.f. *times* the single-crystal strain\* is averaged with respect to  $\varphi'_2$ . As a consequence of the averaging with respect to this angle, the behaviour with respect to symmetry of both harmonic types may be different from that of the o.d.f.

In this paper method (b) is used since the harmonics associated with the strain pole figures cannot be derived using method (a).

## 2.2. Symmetry

A clear distinction must be made between the symmetry exhibited by the o.d.f. and by the harmonics. The *symmetry of the o.d.f.* may be defined as follows. Consider a crystal symmetry element leaving the value of the o.d.f. unchanged in the crystal's product orientation with respect to the parent orientation. These orientations are physically equivalent. Then the o.d.f. may be said to exhibit symmetry with respect to the crystal symmetry element. The symmetry of the o.d.f. is defined with respect to *orientations*. The *symmetry the harmonics satisfy* may be defined as follows. The harmonic is some continuous function of two independent angles defined with respect to crystal reference axes. The harmonic is a function of *directions*. If a crystal symmetry element is applied to crystal reference axes, the behaviour of the harmonic's function may be left unchanged. That is, the (curved) surface representing the function (plotted, for instance, with respect to a reference sphere) coincides exactly in the product and parent orientations of the crystal reference axes. However, this only occurs for the pure rotational symmetry elements (cf. Figs. 3 to 8). Therefore it is better to define the symmetry of the harmonics with respect to *directions*. The symmetry definition then reads: apply a crystal symmetry element to the crystal reference axes. The same vector fixed in space exhibits different elements in the parent and product orientations. The harmonic's function value may be left unchanged in the *direction* of the vector in the parent and product orientation. Then the harmonic exhibits symmetry with respect to the crystal symmetry element.

\* In Bunge's formalism the cubic symmetry coefficients  $A_j^{m\mu}$  and also the harmonics  $K_j^\mu$  are usually written with two dots  $\dot{A}_j^{m\mu}$  etc. In this paper only cubic point groups are treated and since no confusion is to be expected the quantities are written *without* dots.

† The o.d.f. may be defined as follows: consider the volume fraction  $dV/V$  of the crystals (of one metallurgical phase) with orientations in the interval between  $g$  and  $g + dg$ . The proportionality factor is the o.d.f.,  $f(g)$ ;  $dV/V = f(g) dg$ . Evidently it is normalized such that the integral over all orientations yields unity.

\* If single-crystal strain is the consequence of *some* stress state, it follows (since the single-crystal compliances and stiffnesses are the same for all cubic point groups) that the single-crystal strain in the direction of  $\mathbf{Q}$  is invariant for all crystal symmetry operations according to point group  $m3m$ . The symmetry of the o.d.f. is lower (432 at most). Hence the symmetry of the product of o.d.f. and single-crystal strain is determined by the o.d.f.'s symmetry. One would expect that upon integration with respect to the rotation angle about  $\mathbf{Q}$  the same symmetry behaviour of harmonics  $F_1, \dots, F_4$  would result as was found for the  $K_j^\mu$  case. This does not occur, however. The reason is that the intensity only depends on the zero-order variation of the o.d.f. with respect to the rotation angle. The diffraction strain depends on the first- and second-order variations of the o.d.f. *and* the single-crystal strain with respect to this angle (Brakman, 1985).

This definition allows all possible symmetry elements, those of the 'second kind'\* (changing a right-handed frame of reference into a left-handed one), included. Examples are given in Table 7 and Figs. 3 to 8.

### 2.3. The o.d.f. vs crystal point-group symmetry elements of the second kind

Crystals belonging to one metallurgical phase may exhibit right-handed or left-handed behaviour (Bunge, 1982, p. 100). They may occur in different fractions and exhibit different o.d.f.'s. Consequently, a distinction has to be made between the left-handed† and right-handed o.d.f.'s (Bunge, 1982, p. 103) indicated by  $f^L(g)$  and  $f^R(g)$ . If the crystal's point group only exhibits pure rotations, these two o.d.f.'s are not related. They both enter in the eventual scattered intensity expression (Bunge, 1982). Consequently, the fractions of left- and right-handed crystals then have to be known.

If the point group exhibits a centre of inversion the crystals can be seen as left- and right-handed at the same time (Bunge, 1982, p. 105). Then  $f^L$  and  $f^R$  are identical.

For the point groups exhibiting mirror-like symmetry elements but *not* an explicit centre of inversion (for instance,  $\bar{4}3m$ ) a similar condition follows. Then  $f^L$  and  $f^R$  are one-to-one correlated.‡ From this it follows that for all three possible crystal point-group symmetry cases treatment of the pure rotation elements is sufficient. For the rotational point groups, the treatment of  $f^L$  is self-evident (Bunge, 1982). Only  $f^R$  is used in what follows.

### 3. The cubic symmetry coefficients $A_j^{m\mu}$

The purpose of these coefficients is to render the value of the o.d.f. invariant with respect to (rotational) point-group symmetry elements (Bunge, 1982, pp. 363–369). In the expressions for diffraction intensity and diffraction strain pole figures the same cubic symmetry coefficients  $A_j^{m\mu}$  occur. In this section they are studied for the lowest possible cubic symmetry, *i.e.* point group 23. The coefficients  $A_j^{m\mu}$  for the higher-symmetry point groups are shown to be a subset of these.

\* Terminology due to Bunge, Esling & Muller (1981). The determinant of the matrices of these elements is equal to  $-1$ .

† The left-handed o.d.f. is also a function of rotations only.

‡ The o.d.f. series-expansion coefficients of the left- and right-handed o.d.f.'s can be expressed in each other, accordingly: see equation (79) of Bunge, Esling & Muller (1981). Note that for this point group one physical crystal is right- and left-handed at the same time [Bunge, 1982, equation (27)], like the centrosymmetric crystal case.

#### 3.1. Point group 23

With an expression given by Esling, Bechler-Ferry & Bunge (1981) it can be shown that

$$\mu_{\max}(j) = \frac{1}{12} \left[ (2j+1) + 8 \frac{\sin(2j+1)\pi/3}{\sin\pi/3} + 3(-1)^j \right] \quad (9)$$

( $\mu_{\max}$  determines the number of linearly independent  $A_j^{m\mu}$  solution vectors for a given  $j$ ), from which it follows that

$$\mu_{\max} = 0 \quad \text{for } j = 1, 2, 5, \quad (10a)$$

$$\mu_{\max} = 1 \quad \text{for } j = 0, 3, 4, \quad (10b)$$

and

$$\mu_{\max}(j+6) = \mu_{\max}(j) + 1. \quad (11)$$

It can be shown that the symmetry elements of this (pure rotation) point group yield (*via* the o.d.f.) the following 'selection rules':

(i) Twofold axis in [001]:

$$m = \text{multiple of } 2. \quad (12)$$

(ii) Twofold axis in [100]:

$$A_j^{-m\mu} = (-1)^j A_j^{m\mu}. \quad (13)$$

The twofold axis in [010] does not lead to anything new.

(iii) Threefold axis in [111]:

$$A_j^{s\mu} = \sum_{m=-j(2)}^j (-1)^{m/2} A_j^{m\mu} P_j^{ms}(\cos\pi/2) \quad (14a)$$

for a rotation of  $2\pi/3$  about [111];

$$A_j^{s\mu} = (-1)^{s/2} \sum_{m=-j(2)}^j A_j^{m\mu} P_j^{ms}(\cos\pi/2) \quad (14b)$$

for a rotation of  $4\pi/3$  about [111] (in both cases  $s = \text{multiple of } 2$ ).

It is suggested by (14a) and (14b) that two series of possible solutions exist for the coefficients  $A_j^{m\mu}$ . However, it can be shown for both even and odd  $j$  that (14a) and (14b) lead to the same solution.

Two kinds of solutions for both equations are alternately obtained [using (14a) or (14b) and (8)]:

(i) A solution vector [consisting of all  $A_j^{m\mu}$  with  $m = -j(2)j$  for a given value of  $\mu$ ] where the  $A_j^{m\mu}$  for both even and odd  $j$  are only significantly different from zero if  $m = \text{multiple of } 4$ . Then (14a) and (14b) are identical. On inspection (Bunge, 1982, p. 501) it is seen that the  $A_j^{m\mu}$  coefficients for point group 432 are obtained. This is logical since the higher symmetry (432) also satisfies the lower (23).

(ii) A solution vector where the  $A_j^{m\mu}$  are only significantly different from zero for even  $m$  not a multiple of 4 for both even and odd  $j$ . Then, (14a) and (14b) are also identical. These  $A_j^{m\mu}$  are the 'pure' point-group 23 cubic symmetry coefficients. Computer outputs exhibiting these coefficients for  $j=4(2)52$  and  $j=3(2)51$  can be obtained from the author.

Upon analysis, the solutions are alternately obtained, the index  $\mu$  enumerating them in systematic order. For instance,  $\mu=1$  yields a '432' solution,  $\mu=2$  stands for a 'pure 23' solution *etc.* This choice is purely arbitrary and could lead to confusion among different laboratories.

### 3.2. The other cubic point groups

The coefficients  $A_j^{m\mu}$  for point group 432 are contained within the solutions obtained for the 23 case as indicated in § 3.1. The  $A_j^{m\mu}$  coefficients for point groups  $m3$  and  $\bar{4}3m$  are identical to those of 23. The  $A_j^{m\mu}$  coefficients for point group  $m3m$  are identical to those of 432.

However, a difficulty now arises. The symmetry elements of the second kind [ $\det(g) = -1$ ] of these point groups *could* be taken as imposing additional restrictions on the  $A_j^{m\mu}$ . For instance:

(i) It could be taken from the  $K_j^\mu$  harmonics that the  $A_j^{m\mu}$  should be equal to zero for odd  $j$  for point groups  $m3$  and  $m3m$  using their centre of inversion. The same holds with respect to their  $\{h00\}$  mirror planes.

(ii) The  $\{hh0\}$  mirror planes occurring in point groups  $m3m$  and  $\bar{4}3m$  could lead to the condition that  $A_j^{-m\mu} = (-1)^{m/2} A_j^{m\mu}$  for the  $K_j^\mu$  harmonics (Bunge, 1982, p. 385) in addition to (13).

However, these restrictions do *not* lead to fulfilment of the same symmetry elements in harmonics  $F_1, \dots, F_4$ .

If one starts from the harmonics  $F_1, \dots, F_4$  the imposed symmetry is not satisfied in the  $K_j^\mu$  harmonics. From a principal point of view there is no preference for one type of harmonic. Therefore,

(i) The cubic symmetry coefficients should be defined using the *rotational* symmetry elements of the crystal point group considered. The symmetry elements should be taken as transformations of *orientations*.

(ii) The coefficients generate the symmetry of the o.d.f. with respect to (rotational) crystal point-group symmetry elements (Bunge, 1982, pp. 48 and 366). They should be derived *via* expressions obtained from the o.d.f. (Bunge, 1982, pp. 363–369). They should not be derived from the harmonics  $K_j^\mu$  or  $F_1, \dots, F_4$ .

(iii) Fulfilment of symmetry elements of the second kind in the harmonics  $K_j^\mu$  and  $F_1, \dots, F_4$  is obtained by imposing selection rules on  $j$  and  $m$  as indicated in Table 7. No additional restrictions on the  $A_j^{m\mu}$  are necessary.

### 4. The polycrystalline diffraction experiment performed on textured materials

According to § 2.3 only point groups 23 and 432 are necessary for the mathematical treatment of the diffraction experiment for *all* cubic point groups. This holds whether crystals belong to  $m3$  or  $\bar{4}3m$  (as compared to 23) or  $m3m$  (as compared to 432).

Only the right-handed o.d.f. is taken into account. Upon diffraction, all crystal orientations exhibiting an  $\langle hkl \rangle$  vector parallel to the scattering vector  $\mathbf{Q}$  contribute. The number of physically equivalent crystal orientations contributing is determined by the number of symmetry elements of the point group considered.

Use of the rotation elements of the point group leads to an expression for the diffracted intensity and the diffraction strain *via* the o.d.f. For the mathematical treatment of the  $\{hkl\}$  diffraction experiment only the expressions for the  $hkl$  and  $\bar{h}\bar{k}\bar{l}$  reflections have to be considered for the 432 case\* (Brakman, 1985). Multiplied by their appropriate weight factors,† their sum represents physical reality.

Point group 23 does not exhibit fourfold rotation symmetry elements. Consequently, the treatment of, say, the  $k\bar{h}l$  and  $\bar{k}h\bar{l}$  reflections in addition to those mentioned is necessary for this point group. The structure factors for the  $hkl$  and  $k\bar{h}l$  reflections are different for point group 23. For the case of normal scattering the structure factors of  $hkl$  and  $\bar{h}\bar{k}\bar{l}$  reflections are equal (all point groups, Friedel's law). The same holds for the  $k\bar{h}l$  with respect to the  $\bar{k}h\bar{l}$  reflection. In the case of anomalous scattering the structure factors are unequal and Friedel's law does not hold.

For all cubic point groups it holds that 48 permutations of  $h$ ,  $k$  and  $l$  leading to 48  $\{hkl\}$  lattice planes diffract at the *same*  $2\theta$ , equal structure factors or not.

The relationships between the structure factors for the various cubic crystal classes in conjunction with both anomalous and normal scattering are summarized in Table 1.

#### 4.1. Diffraction intensity pole figures

It can be shown that the  $\{hkl\}$  intensity pole figure exhibits the following expression for the case of

\* This corresponds to two physically different orientation types contributing at the same time: (i) Crystal orientations exhibiting  $[hkl]$  parallel to  $\mathbf{Q}$ . All orientations obtained from this one using a (point-group allowed) rotational symmetry element are physically equivalent and hence not treated. (ii) Crystal orientations exhibiting  $[\bar{h}\bar{k}\bar{l}]$  parallel to  $\mathbf{Q}$ . For the general  $hkl$  case the latter orientation cannot be obtained from the former by a rotation (Brakman, 1985) nor does it imply an inversion of the crystal's reference axes. Generally, the o.d.f. differs in these two orientation types. Consequently, the  $hkl$  and  $\bar{h}\bar{k}\bar{l}$  intensities are different, too.

† I.e.  $S_{HKL}^2 / (S_{HKL}^2 + S_{\bar{H}\bar{K}\bar{L}}^2)$  and  $S_{\bar{H}\bar{K}\bar{L}}^2 / (S_{HKL}^2 + S_{\bar{H}\bar{K}\bar{L}}^2)$  where  $S_{HKL}$  is the geometric structure factor. See also Bunge & Esling (1981).

Table 1. Relationships between squares of geometric structure factors for cubic point groups in conjunction with anomalous scattering and normal scattering (note the use of Laue indices)

Cubic point group	Anomalous scattering (a.s.)	Normal scattering (n.s.)
23	Four structure factors all different from each other; no relationships	$S_{\overline{H}KL}^2 = S_{HKL}^2$ $S_{\overline{K}HL}^2 = S_{KHL}^2$
$m\overline{3}$	—	As above
$\overline{4}3m$	$S_{\overline{K}HL}^2 = S_{HKL}^2$ $S_{\overline{K}\overline{H}L}^2 = S_{\overline{H}KL}^2$	As left-hand column + $S_{\overline{H}KL}^2 = S_{HKL}^2$ $S_{\overline{K}HL}^2 = S_{KHL}^2$
432	$S_{\overline{K}\overline{H}L}^2 = S_{HKL}^2$ $S_{\overline{K}HL}^2 = S_{\overline{H}KL}^2$	As left-hand column + $S_{\overline{H}KL}^2 = S_{HKL}^2$ $S_{\overline{K}HL}^2 = S_{KHL}^2$
$m\overline{3}m$	—	As above

crystals belonging to point group 23 in conjunction with anomalous scattering:

$$\begin{aligned} \tilde{P}_{hkl}(\psi, \alpha) &= \sum_{j\mu\nu} C_j^{\mu\nu} \sum_n \dot{A}_j^{\nu\mu} (-1)^{n/2} \exp(in\alpha) P_j^{0n}(\cos\psi) \\ &\times \sum_{m=-j}^j (-1)^{m/2} A_j^{m\mu} \exp(-im\beta) P_j^{m0}(\cos\Phi) \\ &\times [12S_{HKL}^2 + 12(-1)^{m/2} S_{\overline{K}\overline{H}L}^2 \\ &+ 12(-1)^j S_{\overline{H}KL}^2 + 12(-1)^{j+m/2} S_{\overline{K}HL}^2] \\ &\times (12S_{HKL}^2 + 12S_{\overline{K}\overline{H}L}^2 + 12S_{\overline{H}KL}^2 + 12S_{\overline{K}HL}^2)^{-1}, \end{aligned} \quad (15a)$$

where  $\psi, \alpha$  is the direction of the scattering vector  $\mathbf{Q}$  with respect to specimen reference axes. The  $C_j^{\mu\nu}$  are the well known o.d.f. series-expansion coefficients.  $S_{HKL}^2$  stands for  $S_{HKL}$  times its complex conjugate.

A clear distinction is made between the quantities  $P_{hkl}$  and  $\tilde{P}_{hkl}$ .  $\tilde{P}_{hkl}$  stands for the (structure-factor) weighted  $\{hkl\}$  scattered intensity constituted by the reflections according to the physically different permutations of  $h, k$  and  $l$ . Apart from a proportionality factor, it is equal to the measured scattered intensity. The denominator in (15a) serves as a normalization with respect to the structure factors. As a consequence, the integral

$$\oint \tilde{P}_{hkl}(\psi, \alpha) \sin\psi \, d\psi \, d\alpha \equiv 4\pi \quad (15b)$$

as required.

The quantity  $P_{hkl}$  is defined via the o.d.f.:

$$P_{hkl}(\psi, \alpha) = (2\pi)^{-1} \int_0^{2\pi} \text{o.d.f.}(\psi, \alpha, [hkl], \varphi'_2) \, d\varphi'_2. \quad (15c)$$

The equation is written for the single  $hkl$  reflection. It may occur that the structure factor of the reflection is equal to zero whereas the o.d.f. is non-zero. There-

fore, it should be written

$$I_{HKL} = AS_{HKL}^2 P_{hkl} \quad (15d)$$

where  $HKL$  are Laue indices.  $I_{HKL}$  is the integrated (with respect to  $2\theta$ ) intensity of the  $HKL$  reflection and  $A$  is a proportionality factor.  $P_{hkl}$  is written using Miller indices since it does not depend on the order of the reflection. The relationship between  $\tilde{P}_{hkl}$  and  $P_{hkl}$  is given in Table 2.

The harmonics of interest are given by the  $m$  sum in (15a).

Although the equations given may look complicated, essentially only one type of  $hkl$ -dependent harmonic emerges:

$$\begin{aligned} K_j^\mu(\Phi, \beta) &= \sum_{m=-j(2)}^j (-1)^{m/2} A_j^{m\mu} P_j^m(\cos\Phi) \exp(-im\beta). \end{aligned} \quad (16)$$

$P_j^{m0}(x)$  in (15a) can be reduced to  $P_j^m(x)$  using

$$P_j^{m0}(x) = (-1)^{m/2} [2/(2j+1)]^{1/2} P_j^m(x). \quad (17)$$

The factor  $(-1)^{m/2}$  in (15a) can then be omitted ( $m \equiv$  multiple of 2 in all cases). Evidently, on some occasions [(15a)] an additional factor  $(-1)^{m/2}$  is introduced. It seems that an additional type of harmonic arises, different from (16) in that the factor  $(-1)^{m/2}$  is absent:

$$K_j^\mu(\Phi, \beta) = \sum_{m=-j(2)}^j A_j^{m\mu} P_j^m(\cos\Phi) \exp(-im\beta). \quad (18)$$

However, for the 432 and  $m\overline{3}m$  case ( $m$  only equal to a multiple of 4) (16) and (18) are identical.

Exactly the same holds for the 23,  $m\overline{3}$  and  $\overline{4}3m$  cases if the '432 solution' for the  $A_j^{m\mu}$  coefficients is used. And it follows from the 'pure 23 solution' (§ 3.1) for these coefficients that (16) and (18) then differ only by a minus sign.

Consequently, (16) is taken as the definition of the harmonics associated with intensity pole figures. Note the similarity of (16) to (6). It also follows that in all cases (16) reduces to

$$K_j^\mu(\Phi, \beta) = \sum_{m=-j(2)}^j (-1)^{m/2} A_j^{m\mu} P_j^m(\cos\Phi) \cos m\beta \quad (19)$$

for even  $j$  and

$$K_j^\mu(\Phi, \beta) = \sum_{m=-j(2)}^j (-1)^{m/2} A_j^{m\mu} P_j^m(\cos\Phi) \sin m\beta \quad (20)$$

for odd  $j$  [a factor  $(-1)^{1/2}$  has been omitted in (20)].

In Table 3 the other cubic point groups in conjunction with both anomalous and normal scattering are dealt with. The intensity expression is similar to (15a). The quotient involving the structure factors, however,

Table 2.  $\{hkl\}$  intensity expression  $\tilde{P}_{hkl}$  [cf. equation (15a)] for combinations of cubic point groups and scattering modes

For point group 23, four physically different  $hkl$  reflections occur (same  $2\theta$ ). The quantities  $P_{k\bar{h}l}$  etc. are defined in equation (15c). Weighted with their structure-factor squares, they constitute  $\tilde{P}_{hkl}$ . The denominator  $D$  normalizes the expression with respect to structure factors. Apart from a proportionality factor,  $\tilde{P}_{hkl}$  stands for the measured intensity. It depends on  $\psi$ ,  $\alpha$  (i.e. the orientation of the scattering vector with respect to specimen axes). It is equal to the  $\{hkl\}$  intensity pole figure. The expressions for the pure-rotation point groups do not exhibit the left-handed crystal contributions. Their treatment is not essentially different (Bunge, 1982, p. 102)

Point group and scattering case	Constitution of $\{hkl\}$ scattered intensity expression	$D$
23 and a.s.	$[S_{HKL}^2 P_{hkl} + S_{\bar{H}RL}^2 P_{\bar{h}kl} + S_{K\bar{H}L}^2 P_{k\bar{h}l} + S_{RHL}^2 P_{rhl}] D^{-1}$	$S_{HKL}^2 + S_{\bar{H}RL}^2 + S_{K\bar{H}L}^2 + S_{RHL}^2$
23 and n.s. or $m\bar{3}$	$[S_{HKL}^2 (P_{hkl} + P_{\bar{h}kl}) + S_{K\bar{H}L}^2 (P_{k\bar{h}l} + P_{rhl})] D^{-1}$	$2[S_{HKL}^2 + S_{K\bar{H}L}^2]$
$\bar{4}3m$ and a.s.	$[S_{HKL}^2 (P_{hkl} + P_{\bar{h}kl}) + S_{\bar{H}RL}^2 (P_{\bar{h}kl} + P_{k\bar{h}l})] D^{-1}$	$2[S_{HKL}^2 + S_{\bar{H}RL}^2]$
$\bar{4}3m$ and n.s.	$[P_{hkl} + P_{\bar{h}kl} + P_{k\bar{h}l} + P_{rhl}] D^{-1}$	4
432 and a.s.	$[S_{HKL}^2 P_{hkl} + S_{\bar{H}RL}^2 P_{\bar{h}kl}] D^{-1}$	$[S_{HKL}^2 + S_{\bar{H}RL}^2]$
432 and n.s. or $m\bar{3}m$	$[P_{hkl} + P_{\bar{h}kl}] D^{-1}$	2

Table 3. Elaboration of scattered intensity expression  $\tilde{P}_{hkl}$ 

The quotient in equation (15a) should be replaced by the second-column expression. The symmetry satisfied by the  $hkl$ -dependent part of the intensity expression is displayed in the fourth column. On some occasions this expression satisfies symmetries other than those introduced *via* the point group (cf. § 2.2). An entry  $m\bar{3}m$  in the fourth column signifies that all cubic point groups are satisfied. The data of the third column lead to the so-called ghosts (Esling, 1981; Bunge, 1982, p. 116). Left-handed crystal contributions not shown.

Tables 2 and 3 take on a simpler form for the more symmetric  $\{hk0\}$  and  $\{hhl\}$  reflections

Point group and scattering case	Expression instead of the quotient in equation (15a)	Values of $j$ and $m$ blotted out from intensity expression	Symmetry satisfied by $hkl$ -dependent part of intensity expression (cf. Table 7)
23 and n.s. or $m\bar{3}$	$\frac{1}{2}[1 + (-1)^j][S_{HKL}^2 + (-1)^{m/2} S_{K\bar{H}L}^2] / (S_{HKL}^2 + S_{K\bar{H}L}^2)$	$j = \text{odd}$	23 and $m\bar{3}$
$\bar{4}3m$ and a.s.	$\frac{1}{2}[1 + (-1)^{j+m/2}][S_{HKL}^2 + (-1)^j S_{\bar{H}RL}^2] / (S_{HKL}^2 + S_{\bar{H}RL}^2)$	1. $j = \text{even}$ $m = \pm 2, \pm 6, \dots$ 2. $j = \text{odd}$ $m = 0, \pm 4, \dots$	1. $j = \text{even}$ $m = 0, \pm 4, \dots$ } $m\bar{3}m$ 2. $j = \text{odd}$ $m = \pm 2, \pm 6, \dots$ } 23 and $\bar{4}3m$
$\bar{4}3m$ and n.s.	$\frac{1}{4}[1 + (-1)^{j+m/2}][1 + (-1)^j]$	1. $j = \text{odd}$ 2. $j = \text{even}$ $m = \pm 2, \pm 6, \dots$	$j = \text{even}$ $m = 0, \pm 4, \dots$ } $m\bar{3}m$
432 and a.s.	$\frac{1}{2}[1 + (-1)^{m/2}][S_{HKL}^2 + (-1)^j S_{\bar{H}RL}^2] / (S_{HKL}^2 + S_{\bar{H}RL}^2)$	$m = \pm 2, \pm 6, \dots$	1. $j = \text{even}$ $m = 0, \pm 4, \dots$ } $m\bar{3}m$ 2. $j = \text{odd}$ $m = 0, \pm 4, \dots$ } 432
432 and n.s. or $m\bar{3}m$	$\frac{1}{4}[1 + (-1)^{m/2}][1 + (-1)^j]$	1. $j = \text{odd}$ 2. $m = \pm 2, \pm 6, \dots$	$j = \text{even}$ $m = 0, \pm 4, \dots$ } $m\bar{3}m$

should be replaced by the corresponding second-column entry of Table 3. Except for the 23+ anomalous scattering case a certain amount of harmonics does *not* arise in the final intensity expression (third column of Table 3). This represents the so-called 'ghost' phenomenon well known in texture analysis.

#### 4.2. Diffraction strain pole figures

Instead of the harmonics  $K_j^\mu$  of § 4.1 the harmonics  $F_1, \dots, F_4$  [cf. equation (1)] arise as the  $hkl$ -dependent part in the theoretical expression for diffraction strain pole figures (Brakman, 1986). They can be seen as surface spherical harmonics, but their properties differ from those of the  $K_j^\mu$ . These harmonics can only be arrived at *via* the o.d.f. Hence, they cannot

be constructed using method (a) of § 2.1. In Appendix I\* they are defined as a function of  $hkl$ ,  $j$  and  $\mu$ . However, use of (I-14) and (I-15) allows them to be expressed generally in  $\Phi$  and  $\beta$ . The definitions

$$A(\Phi, \beta) = \frac{1}{8}[2 \sin^2 \Phi \sin^2 2\beta - (2 - \frac{1}{2} \sin^2 2\beta) \sin^2 2\Phi] \quad (21)$$

and

$$B(\Phi, \beta) = \frac{1}{4} \sin 4\beta \cos \Phi \sin^2 \Phi \quad (22)$$

\* Appendices I-III, which give further details of complex mathematical expressions, have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 43428 (7 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.



lead to

$$F_1(\Phi, \beta, j, \mu) = A(\Phi, \beta) \sum_{m=-j(2)}^j (-1)^{m/2} A_j^{m\mu} \\ \times P_j^{m2}(\cos \Phi) \cos m\beta \\ - B(\Phi, \beta) \sum_{m=-j(2)}^j (-1)^{m/2} A_j^{m\mu} \\ \times P_j^{m2}(\cos \Phi) \sin m\beta \quad (23)$$

$$F_2(\Phi, \beta, j, \mu) = A(\Phi, \beta) \sum_{m=-j(2)}^j (-1)^{m/2} A_j^{m\mu} \\ \times P_j^{m2}(\cos \Phi) \sin m\beta \\ + B(\Phi, \beta) \sum_{m=-j(2)}^j (-1)^{m/2} A_j^{m\mu} \\ \times P_j^{m2}(\cos \Phi) \cos m\beta. \quad (24)$$

The definitions

$$C(\Phi, \beta) = \frac{1}{4}(\sin 4\Phi + \sin 2\Phi \sin^2 \Phi \sin^2 2\beta) \quad (25)$$

and

$$D(\Phi, \beta) = \frac{1}{4} \sin^3 \Phi \sin 4\beta \quad (26)$$

lead to

$$F_3(\Phi, \beta, j, \mu) = C(\Phi, \beta) \sum_{m=-j(2)}^j (-1)^{m/2} A_j^{m\mu} \\ \times P_j^{m1}(\cos \Phi) \cos m\beta \\ - D(\Phi, \beta) \sum_{m=-j(2)}^j (-1)^{m/2} A_j^{m\mu} \\ \times P_j^{m1}(\cos \Phi) \sin m\beta, \quad (27)$$

$$F_4(\Phi, \beta, j, \mu) = C(\Phi, \beta) \sum_{m=-j(2)}^j (-1)^{m/2} A_j^{m\mu} \\ \times P_j^{m1}(\cos \Phi) \sin m\beta \\ + D(\Phi, \beta) \sum_{m=-j(2)}^j (-1)^{m/2} A_j^{m\mu} \\ \times P_j^{m1}(\cos \Phi) \cos m\beta. \quad (28)$$

The expressions for  $A, \dots, D$  arise as a consequence of cubic-crystal compliance or stiffness-tensor transformations (Brakman, 1983, Appendix II). Functions  $F_1$  and  $F_2$  are purely real whereas  $F_3$  and  $F_4$  are purely imaginary. These functions behave as (symmetrized) harmonics.\* The symmetries these harmonics satisfy are summarized in Table 7.

\* The definition of functions  $P_j^{mn}$  is given by Gel'fand, Minlos & Shapiro (1963, p. 85) and by Bunge (1982, p. 351). The general differential equation these functions obey reads (Gel'fand, Minlos & Shapiro, 1963, p. 82):

$$d^2 P_j^{mn}(\cos \Phi) / d\Phi^2 + \cot(\Phi) dP_j^{mn}(\cos \Phi) / d\Phi \\ + [j(j+1) - (n^2 - 2mn \cos \Phi + m^2) / \sin^2 \Phi] P_j^{mn}(\cos \Phi) = 0.$$

For  $n=0$  the 'normal' Legendre differential equation is obtained.

From the definition

$$g(hkl) = \int_0^{2\pi} [\text{o.d.f.}(\psi, \alpha, [hkl], \varphi'_2) \\ \times \text{compliance}([hkl], \varphi'_2) \\ \times \text{stress}(\varphi'_2)] d\varphi'_2, \quad (29a)$$

it follows that the expression for the  $hkl$  reflection diffraction strain [equation (1)] is the consequence of

$$\langle \varepsilon'_{zz}(\psi, \alpha) \rangle = g(hkl) / \int_0^{2\pi} \text{o.d.f.}(\psi, \alpha, [hkl], \varphi'_2) d\varphi'_2. \quad (29b)$$

The strain is determined in the direction of the  $Z$  axis of a laboratory frame. This  $Z$  axis is parallel to  $\mathbf{Q}$  and consequently parallel to the diffracting lattice plane normal  $[hkl]$ . The integration only includes those crystals with  $[hkl]$  parallel to  $\mathbf{Q}$ . The denominator is a normalization factor, and is defined in (15c). In the numerator, the o.d.f. expression depends on  $\psi$  and  $\alpha$  and the orientations of the crystallites with  $[hkl]$  parallel to  $\mathbf{Q}$ . The numerator comprises a summation over products of compliance and stress-tensor elements. For the Reuss model, the stresses are taken out of the integration.

Equation (29) is written for the  $hkl$  reflection only. However, the  $\bar{h}\bar{k}\bar{l}$  reflection also contributes at the same time. A similar expression to (29) can then be written.

The question arises of how the 'measured' diffraction strain (*i.e.* the composition of the weighted contributions of the various reflections) expression is constituted. The  $2\theta$  shifts of the centroids of the  $hkl$  and  $\bar{h}\bar{k}\bar{l}$  line profiles can be shown (*via* Bragg's law) to lead to an expression:

$$\langle \varepsilon'_{zz} \rangle = [S_{HKL}^2 g(hkl) + S_{\bar{H}\bar{K}\bar{L}}^2 g(\bar{h}\bar{k}\bar{l})] \\ \times [2\pi(S_{HKL}^2 + S_{\bar{H}\bar{K}\bar{L}}^2) \bar{P}_{hkl}]^{-1} \quad (30)$$

[the factor  $\pi$  is cancelled upon integration of (29a)]. The procedure can be extended to all combinations of crystal point groups and scattering modes. The result is shown in Table 4, which exhibits a close analogy to Table 2.

It can be seen from (1) that four different expressions occur involving the stresses  $\sigma_{11}$  and  $\sigma_{22}$  in conjunction with the functions  $F_1, \dots, F_4$ . They are a part of the  $g(hkl)$  function.

All four of them are displayed in Tables 5 and 6 together with the coefficients they exhibit in the evaluated Table 4\* expressions. The texture-free terms  $s_1(hkl)[\sigma_{11} + \sigma_{22}]$  and  $\frac{1}{2}s_2(hkl)\sigma_\alpha \sin^2 \psi$  are not written. They are independent of the permutation of  $h, k$  and  $l$  and these two terms remain unchanged in the eventual equation. The complete  $\{hkl\}$  diffraction strain equation is obtained as follows:

(i) Start with the  $s_1$  and  $\frac{1}{2}s_2$  expressions.

Table 4. Diffraction strain pole figure expression  $\langle \epsilon'_{zz} \rangle$  in terms of structure factors, the function  $g(hkl)$  defined in equation (29a) and the quantity  $\tilde{P}_{hkl}$  defined in equation (15a) and Table 2

No elasticity model has yet been used since  $g(hkl)$  is written for the general case. Function  $g(hkl)$  is basically the numerator of the RHS of equation (1).  $\langle \epsilon'_{zz} \rangle$  represents the  $\{hkl\}$  measured diffraction strain. In equation (1) only the  $hkl$  reflection is treated. Even in the most symmetric case [bottom row of this table and equation (30)] the additional treatment of the  $\bar{h}\bar{k}\bar{l}$  reflection is required. For point groups not exhibiting fourfold axes, four reflections according to four physically different permutations of  $h, k$  and  $l$  constitute the  $\langle \epsilon'_{zz} \rangle$  expression. Through  $g(hkl)$  and  $\tilde{P}_{hkl}$ , the diffraction strain depends on  $\psi$  and  $\alpha$ .  $\langle \epsilon'_{zz} \rangle$  can be seen as a *diffraction strain pole figure*. Left-handed crystal contributions not shown.

Point group and scattering case	Constitution of $\{hkl\}$ diffraction strain $\langle \epsilon'_{zz} \rangle$ [ $g(hkl)$ according to equation (29a)]	$D[\tilde{P}_{hkl}$ according to equation (15a) and Table 3]
23 and a.s.	$[S^2_{HKL}g(hkl) + S^2_{\bar{H}\bar{K}\bar{L}}g(\bar{h}\bar{k}\bar{l}) + S^2_{K\bar{H}L}g(k\bar{h}l) + S^2_{\bar{K}HL}g(\bar{k}h\bar{l})]D^{-1}$	$[S^2_{HKL} + S^2_{\bar{H}\bar{K}\bar{L}} + S^2_{K\bar{H}L} + S^2_{\bar{K}HL}]2\pi\tilde{P}_{hkl}$
23 and n.s. or $m_3$	$\{S^2_{HKL}[g(hkl) + g(\bar{h}\bar{k}\bar{l})] + S^2_{K\bar{H}L}[g(k\bar{h}l) + g(\bar{k}h\bar{l})]\}D^{-1}$	$2[S^2_{HKL} + S^2_{K\bar{H}L}]2\pi\tilde{P}_{hkl}$
$\bar{4}3m$ and a.s.	$\{S^2_{HKL}[g(hkl) + g(\bar{k}h\bar{l})] + S^2_{\bar{H}\bar{K}\bar{L}}[g(\bar{h}\bar{k}\bar{l}) + g(k\bar{h}l)]\}D^{-1}$	$2[S^2_{HKL} + S^2_{\bar{H}\bar{K}\bar{L}}]2\pi\tilde{P}_{hkl}$
$\bar{4}3m$ and n.s.	$[g(hkl) + g(\bar{h}\bar{k}\bar{l}) + g(k\bar{h}l) + g(\bar{k}h\bar{l})]D^{-1}$	$8\pi\tilde{P}_{hkl}$
432 and a.s.	$[S^2_{HKL}g(hkl) + S^2_{\bar{H}\bar{K}\bar{L}}g(\bar{h}\bar{k}\bar{l})]D^{-1}$	$[S^2_{HKL} + S^2_{\bar{H}\bar{K}\bar{L}}]2\pi\tilde{P}_{hkl}$
432 and n.s. or $m3m$	$[g(hkl) + g(\bar{h}\bar{k}\bar{l})]D^{-1}$ [case treated in Brakman (1985)]	$4\pi\tilde{P}_{hkl}$

Table 5. Elaboration of  $\{hkl\}$  diffraction strain expression (cf. Table 4) for point groups 23,  $m3$ , 432 and  $m3m$

Functions  $B_j^\mu(\psi, \alpha), \dots, E_j^\mu(\psi, \alpha)$  defined in Brakman (1986). Functions  $F_1, \dots, F_4$  (depending on  $hkl, j$  and  $\mu$ ) defined in equations (21) to (28). For point groups 23,  $m3$  and  $\bar{4}3m$  the  $\langle \epsilon'_{zz} \rangle$  expression is evaluated by multiplying alternately the second or third column by the stress expressions in the left-hand column. The resulting four expressions are multiplied by  $s_0(2\tilde{P}_{hkl})^{-1}$  and added. The texture-free contribution  $s_1(hkl)[\sigma_{11} + \sigma_{22}] + \frac{1}{2}s_2(hkl)\sigma_\alpha \sin^2 \psi$  is not shown since it does not depend on the point group or the scattering mode. Only stresses  $\sigma_{11}$  and  $\sigma_{22}$  are taken into account. Coefficients  $c_1, \dots, c_5$  depend on the structure factors. They are defined in equations (31a) to (31e). Note the vanishing of the non-centrosymmetric functions  $F_2$  and  $F_4$  upon changing from the anomalous to the normal scattering mode. The symmetries to which the functions  $F_1, \dots, F_4$  are subject are given in Table 7. In Tables 2 to 5 the cases of 23 + n.s. and 432 + n.s. have been taken equivalent to  $m3$  and  $m3m$ , respectively. However, although normal scattering renders  $S^2_{HKL}$  and  $S^2_{\bar{H}\bar{K}\bar{L}}$  equal, an essential difference remains. For  $m3$  and  $m3m$  the left-handed o.d.f. does not have to be treated. For 23 and 432 the left-handed o.d.f. may be essentially different from the right-handed one. This fact is *not* changed in normal scattering.

	Coefficients of left-hand column in $\langle \epsilon'_{zz} \rangle$ expression '23 solution' for $A_j^{m\mu}$ coefficients	Coefficients of left-hand column in $\langle \epsilon'_{zz} \rangle$ expression '432 solution' for $A_j^{m\mu}$ coefficients		Coefficients of left-hand column in $\langle \epsilon'_{zz} \rangle$ expression only '432 solution' for $A_j^{m\mu}$
23 and a.s.			432 and a.s.	
$-(\sigma_{11} - \sigma_{22}) \cos 2\alpha - \sigma_\alpha \sin^2 \psi$	$\sum_{j,\mu} [c_2 B_j^\mu F_1 + c_1 C_j^\mu F_2]$	$\sum_{j,\mu} [B_j^\mu F_1 + c_3 C_j^\mu F_2]$		$\sum_{j,\mu} [B_j^\mu F_1 + c_3 C_j^\mu F_2]$
$-(\sigma_{11} - \sigma_{22}) \sin 2\alpha \cos \psi$	$\sum_{j,\mu} [c_2 C_j^\mu F_1 - c_1 B_j^\mu F_2]$	$\sum_{j,\mu} [C_j^\mu F_1 - c_3 B_j^\mu F_2]$		$\sum_{j,\mu} [C_j^\mu F_1 - c_3 B_j^\mu F_2]$
$(\sigma_{11} - \sigma_{22}) \sin 2\alpha \sin \psi$	$\sum_{j,\mu} [c_2 E_j^\mu F_3 - c_1 D_j^\mu F_4]$	$\sum_{j,\mu} [E_j^\mu F_3 - c_3 D_j^\mu F_4]$		$\sum_{j,\mu} [E_j^\mu F_3 - c_3 D_j^\mu F_4]$
$\sigma_\alpha \sin 2\psi$	$\sum_{j,\mu} [c_2 D_j^\mu F_3 + c_1 E_j^\mu F_4]$	$\sum_{j,\mu} [D_j^\mu F_3 + c_3 E_j^\mu F_4]$		$\sum_{j,\mu} [D_j^\mu F_3 + c_3 E_j^\mu F_4]$
23 and n.s. or $m3$			432 and n.s. or $m3m$	
$-(\sigma_{11} - \sigma_{22}) \cos 2\alpha - \sigma_\alpha \sin^2 \psi$	$\sum_{j,\mu} c_4 B_j^\mu F_1$	$\sum_{j,\mu} B_j^\mu F_1$		$\sum_{j,\mu} B_j^\mu F_1$
$-(\sigma_{11} - \sigma_{22}) \sin 2\alpha \cos \psi$	$\sum_{j,\mu} c_4 C_j^\mu F_1$	$\sum_{j,\mu} C_j^\mu F_1$		$\sum_{j,\mu} C_j^\mu F_1$
$(\sigma_{11} - \sigma_{22}) \sin 2\alpha \sin \psi$	$\sum_{j,\mu} c_4 E_j^\mu F_3$	$\sum_{j,\mu} E_j^\mu F_3$		$\sum_{j,\mu} E_j^\mu F_3$
$\sigma_\alpha \sin 2\psi$	$\sum_{j,\mu} c_4 D_j^\mu F_3$	$\sum_{j,\mu} D_j^\mu F_3$		$\sum_{j,\mu} D_j^\mu F_3$

(ii) For the 23,  $m3$  and  $\bar{4}3m$  (whatever the scattering mode) cases the summations over  $\mu$  comprise both odd and even  $\mu$  values. As  $\mu$  steadily increases, 'pure 23 solutions' and '432 solutions' for the  $A_j^{m\mu}$  coefficients occur alternately. Then the left-hand expressions of Tables 5 and 6 must be alternately multiplied by the second- or third-column expressions. For the 432 and  $m3m$  (whatever the

scattering mode) cases only the '432 solution' for the  $A_j^{m\mu}$  can exist. Consequently, only one column persists.

(iii) The completed  $j, \mu$  sums are multiplied by  $s_0$  [not shown in Tables 5 and 6 but also a factor of  $g(hkl)$ ].

(iv) The four  $j, \mu$  sums are added and divided by the quantity  $D$  of Table 4.

Table 6. As Table 5 but point group  $\bar{4}3m$  treated

Left-handed crystal contributions not shown in Tables 5 and 6. In both tables the Reuss model of elasticity has been used. Note that in Tables 5 and 6 no 'ghost' expressions arise by contrast to those given in Table 3, third column, for the intensity expression case.

	Coefficients of left-hand column in $\langle \varepsilon'_{zz} \rangle$ '23 solution' for $A_j^{m\mu}$ coefficients	Coefficients of left-hand column in $\langle \varepsilon'_{zz} \rangle$ '432 solution' for $A_j^{m\mu}$ coefficients
$\bar{4}3m$ and a.s.		
$-[(\sigma_{11} - \sigma_{22}) \cos 2\alpha - \sigma_\alpha \sin^2 \psi]$	$\sum_{j,\mu} c_5 C_j^\mu F_2$	$\sum_{j,\mu} B_j^\mu F_1$
$-(\sigma_{11} - \sigma_{22}) \sin 2\alpha \cos \psi$	$-\sum_{j,\mu} c_5 B_j^\mu F_2$	$\sum_{j,\mu} C_j^\mu F_1$
$(\sigma_{11} - \sigma_{22}) \sin 2\alpha \sin \psi$	$-\sum_{j,\mu} c_5 D_j^\mu F_4$	$\sum_{j,\mu} E_j^\mu F_3$
$\sigma_\alpha \sin 2\psi$	$+\sum_{j,\mu} c_5 E_j^\mu F_4$	$\sum_{j,\mu} D_j^\mu F_3$
$\bar{4}3m$ and n.s.		
$-[(\sigma_{11} - \sigma_{22}) \cos 2\alpha - \sigma_\alpha \sin^2 \psi]$	0	$\sum_{j,\mu} B_j^\mu F_1$
$-(\sigma_{11} - \sigma_{22}) \sin 2\alpha \cos \psi$	0	$\sum_{j,\mu} C_j^\mu F_1$
$(\sigma_{11} - \sigma_{22}) \sin 2\alpha \sin \psi$	0	$\sum_{j,\mu} E_j^\mu F_3$
$\sigma_\alpha \sin 2\psi$	0	$\sum_{j,\mu} D_j^\mu F_3$

The factors  $c_1, \dots, c_5$  of Tables 5 and 6 are defined as follows:

$$c_1 = (S_{HKL}^2 - S_{\bar{H}\bar{K}\bar{L}}^2 - S_{K\bar{H}\bar{L}}^2 + S_{\bar{K}H\bar{L}}^2) Z^{-1}, \quad (31a)$$

$$Z = (S_{HKL}^2 + S_{\bar{H}\bar{K}\bar{L}}^2 + S_{K\bar{H}\bar{L}}^2 + S_{\bar{K}H\bar{L}}^2);$$

$$c_2 = (S_{HKL}^2 + S_{\bar{H}\bar{K}\bar{L}}^2 - S_{K\bar{H}\bar{L}}^2 - S_{\bar{K}H\bar{L}}^2) Z^{-1}; \quad (31b)$$

$$c_3 = (S_{HKL}^2 - S_{\bar{H}\bar{K}\bar{L}}^2 + S_{K\bar{H}\bar{L}}^2 - S_{\bar{K}H\bar{L}}^2) Z^{-1}; \quad (31c)$$

$$c_4 = (S_{HKL}^2 - S_{\bar{K}\bar{H}\bar{L}}^2) / (S_{HKL}^2 + S_{\bar{K}\bar{H}\bar{L}}^2); \quad (31d)$$

$$c_5 = (S_{HKL}^2 - S_{\bar{H}\bar{K}\bar{L}}^2) / (S_{HKL}^2 + S_{\bar{H}\bar{K}\bar{L}}^2). \quad (31e)$$

From these definitions it follows that (i)  $c_2$  changes into  $c_4$  if normal scattering is assumed (Table 1); (ii)  $c_1, c_3$  and  $c_5$  are equal to zero for the normal scattering case; (iii) upon changing from 23 to 432 (Table 1)  $c_3$  changes into  $c_5$ .

All these facts are clearly displayed in Tables 5 and 6. It also follows from these tables that upon changing from anomalous to normal scattering the non-centrosymmetric functions ( $F_2$  and  $F_4$ ) always vanish. Note that the '23 solution' and the '432 solution' columns may exhibit the same functions  $F_1, \dots, F_4$  but they satisfy different symmetries (Table 7).<sup>\*</sup> Note also that (23,  $m3$  and  $\bar{4}3m$ ), since the '23 solution' and the '432 solution' for the  $A_j^{m\mu}$  coefficients occur alternately in one expression, the total symmetry (with respect to cubic crystal axes) is determined by the harmonic with the lowest symmetry. For instance,

<sup>\*</sup> It follows then that the  $hkl$ -dependent part of the diffraction strain expression sometimes satisfies a different symmetry from that introduced *via* the crystal's point group. This is analogous to the intensity-expression case (Table 3).

Table 7. Symmetry satisfied by the harmonics  $K_j^\mu$  and  $F_1, \dots, F_4$ 

Definition of symmetry: § 2.2. Definition of  $K_j^\mu$  harmonics: equation (16). Definition of  $F_1, \dots, F_4$  harmonics: equations (21) to (28). For an entry ' $m3m$ ' it is understood that all lower cubic symmetries are also satisfied. An entry ' $m3$ ' or ' $\bar{4}32$ ' implies the same with respect to 23. The 'pure 23' and '432' solutions for the cubic symmetry coefficients  $A_j^{m\mu}$  are dealt with in § 3.

Harmonic	'Pure 23' solution for $A_j^{m\mu}$ $m = \pm 2, \pm 6, \pm 10, \dots$		'432' solution for $A_j^{m\mu}$ $m = 0, \pm 4, \pm 8$ ( $m \neq 0$ for odd $j$ )	
	$j = \text{even}$	$j = \text{odd}$	$j = \text{even}$	$j = \text{odd}$
$K_j^\mu$	$m3$ not $\bar{4}3m$	$23$ and $\bar{4}3m$	$m3m$	$432$ not $\bar{4}3m$
$F_1$ and $F_3$	$m3$ not $\bar{4}3m$	$m3$ not $\bar{4}3m$	$m3m$	$m3m$
$F_2$ and $F_4$	$23$ and $\bar{4}3m$	$23$ and $\bar{4}3m$	$432$ not $\bar{4}3m$	$432$ not $\bar{4}3m$

for  $\bar{4}3m$  + anomalous scattering, the functions  $F_2$  and  $F_4$  satisfy 23 and  $\bar{4}3m$  whereas  $F_1$  and  $F_3$  satisfy  $m3m$ .

Note further that by contrast with the intensity pole figure  $\tilde{P}_{hkl}$  the diffraction strain pole figure is *not* normalized. This could be achieved by dividing  $\langle \varepsilon'_{zz} \rangle$  by  $s_0$  times a stress-tensor invariant (or by  $s_0 \sigma_{11}$ ); *cf.* Fig. 1. However, the normalization then still depends on the stress state.

## 5. Results and discussion

From the definition of symmetry of § 2.2, it can be shown that the harmonics may exhibit symmetry as displayed in Table 7. The definition of the  $K_j^\mu$  harmonics is given by (16) whereas the definition of harmonics  $F_1, \dots, F_4$  is given by (23), (24), (27) and (28). It follows that  $\bar{4}3m$  symmetry is *either* satisfied by a 'pure 23' harmonic *or* by an  $m3m$ -type harmonic. The properties of the  $K_j^\mu$  harmonics are not considered in what follows.

### 5.1. Cyclic behaviour and continuity in the poles of the sphere

It can be taken from (21) to (28) that upon replacement of  $\beta$  by  $\beta + 2\pi$  the same expression results. It is also seen that  $F_1 = F_2 = F_3 = F_4 = 0$  for  $\Phi = 0$  and  $\Phi = \pi$  for all  $\beta$ .

### 5.2. Behaviour with respect to Laplace's equation

If one writes  $u = r^j F_1$  this demonstrates that Laplace's equation  $\Delta u = 0$  is *not* satisfied. The same holds for harmonics  $F_2$  to  $F_4$ .

### 5.3. Orthogonality

5.3.1. Orthogonality of types of functions. It can be shown that

$$\oint F_1(\Phi, \beta, j, \mu) F_2^*(\Phi, \beta, j', \mu') dA = 0, \quad (32)$$

$$\oint F_1(\Phi, \beta, j, \mu) F_4^*(\Phi, \beta, j', \mu') dA = 0, \quad (33)$$

$$\oint F_2(\Phi, \beta, j, \mu) F_3^*(\Phi, \beta, j', \mu') dA = 0, \quad (34)$$

$$\oint F_3(\Phi, \beta, j, \mu) F_4^*(\Phi, \beta, j', \mu') dA = 0. \quad (35)$$

These results hold in all four cases, even if  $j=j'$  and  $\mu=\mu'$ . However,

$$\oint F_1(\Phi, \beta, j, \mu) F_3^*(\Phi, \beta, j', \mu') dA \neq 0, \quad (36)$$

$$\oint F_2(\Phi, \beta, j, \mu) F_4^*(\Phi, \beta, j', \mu') dA \neq 0. \quad (37)$$

The inequalities (36) and (37) both hold, even if  $j \neq j'$  or  $\mu \neq \mu'$ . Obviously, if both functions in the integrand exhibit the same symmetry (*cf.* Table 7) orthogonality is lost.

5.3.2. *Orthogonality within one type of function with respect to  $j$  and  $\mu$ .* It generally follows from Appendices II and III\* that

$$\oint F_1(\Phi, \beta, j, \mu) F_1^*(\Phi, \beta, j', \mu') dA \neq 0 \quad (38)$$

even for  $j \neq j'$  or  $\mu \neq \mu'$ . The same holds true for functions  $F_2$  to  $F_4$ . However, a systematic behaviour can be observed (and predicted): if  $F_1(\Phi, \beta, j, \mu)$  belongs to the 'pure 23' column in Table 7 and  $F_1(\Phi, \beta, j', \mu')$  belongs to the '432' column of Table 7, equation (38) is on all occasions equal to zero. Again, the same holds true for harmonics  $F_2$  to  $F_4$ . Note that the value of the index  $\mu$  (or  $\mu'$ ) determines whether a harmonic belongs to the 'pure 23' column or the '432' column of Table 7.

A certain amount of the conventional orthogonality is retained (Appendices II and III). This is reflected by the appearance in the last terms of (II-1) and (III-1) of a factor  $\delta_{jj'} \delta_{\mu\mu'}$ . (II-1) and (III-1) may be used for the normalization of harmonics  $F_1$  to  $F_4$  but the use in general terms of  $j, j', \mu$  and  $\mu'$  is very cumbersome.

The integrals of equations (II-1) and (III-1) also yield contributions for  $j' \neq j$  and  $\mu' \neq \mu$ . However, it follows from computer calculations (and also from the isodensity contours in Figs. 5 to 8) that the evaluation of the integrals yields a value small compared with unity. This is a consequence of the normalization of the  $A_j^{m\mu}$  coefficients using (8) leading to the orthonormalization of the  $K_j^\mu$  harmonics [equation

(7)]. The harmonics  $F_1, \dots, F_4$  are *not* normalized then. However, this condition cannot be changed since (8) follows from the normalization of the o.d.f. itself (Bunge, 1982, p. 368).

It is expected that it is possible to reduce the expressions (II-1) and (III-1) to a much simpler form (still involving  $j, j', \mu$  and  $\mu'$ ). A solution of this problem has not yet been found.

#### 5.4. Graphical representation of results

In Figs. 3 and 4 two 'normal' symmetrized harmonics  $K_j^\mu$  are depicted. Figs. 5 to 8 display an example of harmonics  $F_1, F_2, F_3$  and  $F_4$  respectively. On all occasions the threefold cubic axes are clearly exhibited.

#### 5.5. 'Desymmetrized' harmonics $F_1$ to $F_4$

These harmonics can be written (in analogy to the  $K_j^m$  harmonics) *without* the cubic symmetrization according to (6). Harmonic  $F_1$  is then written, for instance,

$$F_1(\Phi, \beta, j, m) = [A(\Phi, \beta) \cos m\beta - B(\Phi, \beta) \sin m\beta] \times (-1)^{m/2} P_j^{m2}(\cos \Phi)$$

where  $A(\Phi, \beta)$  and  $B(\Phi, \beta)$  are defined in (21) and (22). The sums over  $m$  and the cubic symmetry coefficients  $A_j^{m\mu}$  vanish from (II-1) and (III-1) but the resulting equations are still complicated. Of course, upon conversion to solid spherical harmonics the desymmetrized harmonics do *not* satisfy Laplace's equation either.

#### 5.6. Extension to more general concepts

The conventional harmonics  $K_j^m$  governing the diffracted-intensity (*i.e.* a scalar quantity) expression exhibit associated Legendre polynomials  $P_j^m(\cos \Phi)$

\* See deposition footnote.

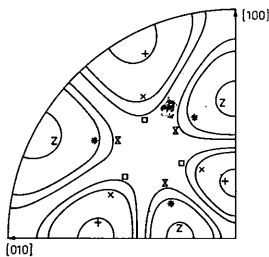


Fig. 3. Stereographic projection of 'normal' symmetrized harmonic  $K_6^2(\Phi, \beta)$  satisfies point group  $m3$  (and  $23$ ), *not*  $43m$  (*cf.* Table 7). Isodensity symbols used:  $\square$ ,  $\otimes$  0.00,  $*$  +0.20,  $\Sigma$  +0.50,  $\times$  -0.20,  $+$  -0.50. Only a quarter is shown since the mirror planes (point group  $m3$ ) perpendicular to the plane of the paper generate the entire pole figure.

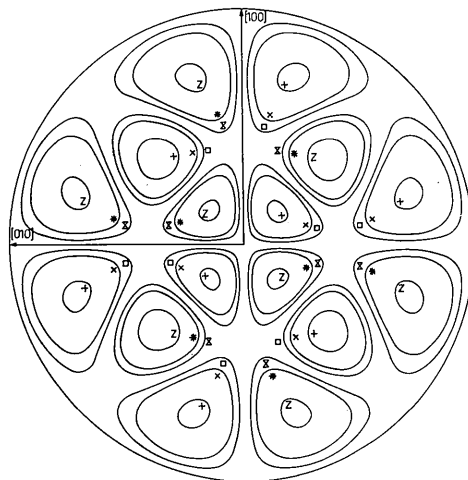


Fig. 4. 'Normal' symmetrized harmonic  $K_1^1(\Phi, \beta)$  satisfies point group  $23$  (and  $43m$ ) *not*  $m3$ . Isodensity symbols: according to Fig. 3. Plane of the paper is *not* a mirror plane. Definition of  $K_j^\mu$  in Figs. 3 and 4 according to equation (16).

or, more generally,  $P_j^{m_0}(\cos \Phi)$ . The new harmonics  $F_1$  to  $F_4$  governing the diffraction-strain (*i.e.* a second-rank tensor) expression exhibit generalized associated Legendre (Jacobi-type) polynomials  $P_j^{m_1}(\cos \Phi)$  and  $P_j^{m_2}(\cos \Phi)$ . It is expected then that harmonics exist exhibiting Jacobi polynomials up to, say,  $P_j^{m_3}(\cos \Phi)$ . They may arise in conjunction with physical properties whose descriptions need higher-rank tensors than required for (diffraction) elastic strain.

### 6. Concluding remarks

Four new types of spherical harmonics have been defined. They can be made to satisfy cubic point-group symmetry. All cubic point groups have been dealt with. The new harmonics originate in the equations for diffraction *strain* pole figures obtained on textured materials. They differ essentially from those occurring in diffraction *intensity* pole figures.

Cubic-symmetry-coefficient equations for point group 23 have been given. The coefficients allow the calculation of both even- and odd-order symmetrized harmonics. The better known cubic symmetry coefficients for point group 432 constitute half of the solutions obtained.

General equations have been given which allow calculation of the normalization factors of the new harmonics. The expressions involve Clebsch-Gordan coefficients *and* the cubic symmetry coefficients leading to complicated expressions.

General diffraction intensity and diffraction strain expressions for all cubic point groups, taking into account structure factors for both anomalous and normal scattering modes, have been given. By contrast with the diffraction *intensity* case, the diffraction *strain* expressions do not exhibit the so-called 'ghosts' often encountered in texture analysis from diffraction intensity pole figures.

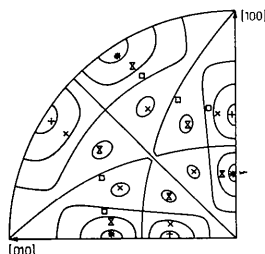


Fig. 5. Symmetrized harmonic  $F_1(\Phi, \beta, 9, 1)$  satisfies point group  $m\bar{3}$  (and 23), not  $\bar{4}3m$  (cf. Table 7). Isodensity symbols used: \* +0.07, X +0.03, Z +0.10, + -0.07, x -0.03,  $\Delta$  -0.10,  $\square$  0.00. Only a quarter is shown since the mirror planes (point group  $m\bar{3}$ ) perpendicular to the plane of the paper generate the entire pole figure. Definition of  $F_1$  according to equation (23).

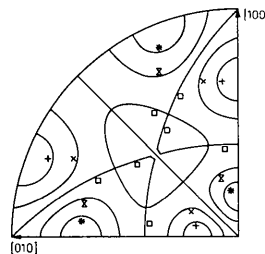


Fig. 7. Symmetrized harmonic  $F_3(\Phi, \beta, 7, 1)$  satisfies point group  $m\bar{3}$  (and 23) not  $\bar{4}3m$ . Isodensity symbols according to Fig. 5. Only a quarter is shown since the mirror planes (point group  $m\bar{3}$ ) perpendicular to the plane of the paper generate the entire pole figure. Definition of  $F_3$  according to equation (27).

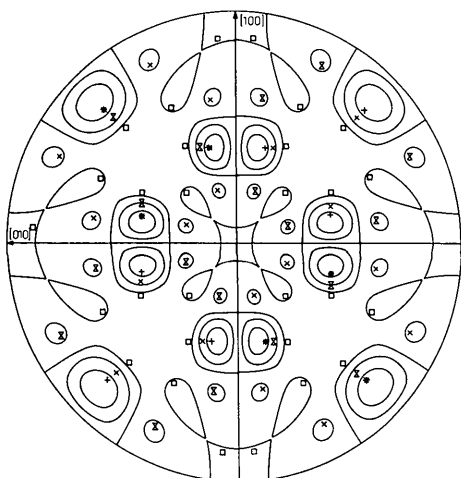


Fig. 6. Symmetrized harmonic  $F_2(\Phi, \beta, 12, 2)$  satisfies point group 23 (and  $\bar{4}3m$ ) not  $m\bar{3}$ . Isodensity symbols according to Fig. 5. Plane of paper is *not* a mirror plane. Definition of  $F_2$  according to equation (24).

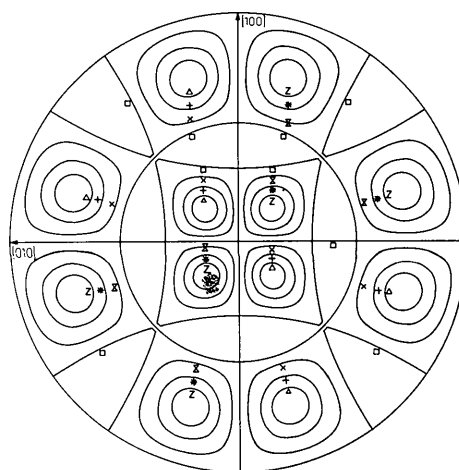


Fig. 8. Symmetrized harmonic  $F_4(\Phi, \beta, 6, 2)$  satisfies point group 23 (and  $\bar{4}3m$ ) not  $m\bar{3}$ . Isodensity symbols according to Fig. 5. Plane of paper is *not* a mirror plane. Definition of  $F_4$  according to equation (28).

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**Comments on From a partial to the complete crystal structure. II. The procedure and its applications, by M. Camalli, C. Giocovazzo & R. Spagna (1985).** By PAUL T. BEURSKENS, *Laboratory for Crystallography, Toernooiveld, 6525 ED Nijmegen, The Netherlands*

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#### Abstract

A recent paper by Camalli, Giocovazzo & Spagna [*Acta Cryst.* (1985). **A41**, 605-613] describes a method which is almost identical to the *DIRDIF* method for the application of direct methods to difference structure factors. The similarities and differences, incorrectly described in that paper, are discussed in the present paper.

#### Introduction

The application of direct methods to the solution of a heavy-atom superstructure problem (Beurskens & Noordik, 1971) led to the concept of the *DIRDIF* method, which

has been extensively developed over fifteen years of practical experience [see Beurskens (1985) and references therein]. It has been intuitively assumed that direct methods are applicable to a hypothetical structure consisting of the complete structure minus the known part of the structure. The program *DIRDIF* can expand a partial structure to the complete structure, if the partial structure comprises as little as ten percent of the total scattering power of the structure. Camalli, Giocovazzo & Spagna (1985) have now described a very similar method, justified in terms of the probabilistic formulae of Giocovazzo (1983). Unfortunately, they make several important errors in the comparison of their method with *DIRDIF*, a comparison which is made more difficult by differences in terminology.