An Empirical Correction for Absorption Anisotropy

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Abstract

A least-squares procedure is described for modeling an empirical transmission surface as sampled by multiple symmetry-equivalent and/or azimuth rotation-equivalent intensity measurements. The fitting functions are sums of real spherical harmonic functions of even order, $y_{lm}(-\mathbf{u}_0) + y_{lm}(\mathbf{u}_1)$, $2 \le l = 2n \le 8$. The arguments of the functions are the components of unit direction vectors, $-\mathbf{u}_0$ for the reverse incident beam and $\mathbf{u}_1$ for the scattered beam, referred to crystal-fixed Cartesian axes. The procedure has been checked by calculations against standard absorption test data.

Introduction

Absorption-corrected Bragg intensities are given by

$$I_{\text{corr}} = I_{\text{meas}}/A, \qquad (1)$$

where

$$A = V^{-1} \int_V \exp[-\mu(t_0 + t_1)]\mathrm{d}^3 t, \qquad (2)$$

and t_0 and t_1 are, respectively, the incident-beam path length to and the scattered-beam path length from each crystal-volume element $\mathrm{d}^3 t$.

For a crystal of well defined size and shape, the transmission factors A can be evaluated by Gaussian numerical integration over a grid of incident and scattered beam paths through the crystal for each reflection (Busing & Levy, 1957; Wuensch & Prewitt, 1965; Coppens, Leiserowitz & Rabinovich, 1965; Coppens, 1970; DeTitta, 1985) or by an analytical method based on subdivision of the crystal into Howells polyhedra (Howells, 1950; Wells, 1960; de Meulenaer & Tompa, 1965; Alcock, 1970).

For a crystal of ill defined size and shape, in the absence of information needed to calculate the beam path lengths, we express the transmission factor as

$$A = A_{\text{sph}}/A_{\text{aniso}}, \qquad (3)$$

where $A_{\text{sph}} = A_{\text{sph}}(\mu,R,2\theta)$ is the transmission factor for an 'equivalent' spherical crystal and A_{aniso} is a correction factor, *i.e.* a reciprocal transmission factor, for absorption anisotropy. All the scattering-angle dependence of the overall transmission factor

is contained in A_{sph}, which increases very nearly linearly with $(\sin \theta)^2$ [Bond (1967), as illustrated by Dunitz (1979)]. The quantity A_{aniso} is intended to correct only for anisotropy of the crystal dimensions.

An equivalent spherical crystal radius can be estimated from an average of crystal diameters measured with the aid of a microscope or as the radius of a sphere or the semidimension of a cube of volume equal to that estimated for the crystal. If the crystal is tablet-, plate-, or blade-shaped, an equivalent spherical radius can be estimated from the minimum crystal thickness and the minimum anisotropy correction factor,

$$\exp(-\mu t_{\text{min}}) \simeq A_{\text{sph}}/A_{\text{aniso, min}},$$

by interpolation on $\ln A_{\text{sph}} = \ln[A_{\text{aniso, min}} \times \exp(-\mu t_{\text{min}})]$ to find R from $A_{\text{sph}}(\mu,R,2\theta)$ tables (Bond, 1967; Dwiggins, 1975).

We formulate the anisotropy correction as

$$A_{\text{aniso}} = 1 + \sum_{l=1}^{l_{\text{max}}} \sum_{m=-l}^{l} a_{lm} f_{lm}, \qquad (4)$$

where

$$f_{lm} = [y_{lm}(-\mathbf{u}_0) + y_{lm}(\mathbf{u}_1)]/2 \qquad (5)$$

and the y_{lm} are real spherical harmonic functions whose arguments $-\mathbf{u}_0$ and $\mathbf{u}_1$ are unit direction vectors referred to crystal-fixed orthonormal axes for the reverse incident beam and scattered beam, respectively. The beam-direction vectors are calculated from the crystal orientation and diffraction geometry for each intensity measurement and the empirical coefficients a_{lm} are obtained by a least-squares analysis of the differences among intensity measurements that are equivalent by symmetry and/or azimuthal rotation. The key requirement is a sufficient multiplicity of equivalent measurements so that the transmission paths through the crystal are sampled thoroughly.

Equations (4) and (5) neglect scattering and approximate A_{aniso} as the average of corrections for 'straight-through' beams in the directions $-\mathbf{u}_0$ and $\mathbf{u}_1$. Allowance for scattering is made by the factor A_{sph} in (3). These approximations have been validated in earlier work on empirical absorption correc-

tions (North, Phillips & Mathews, 1968; Flack, 1974; Walker & Stuart, 1983; Katayama, 1986).

Although the A_{aniso} are intended to correct only for absorption anisotropy due to a nonspherical crystal shape, they will of course be affected by any and all differences among equivalent reflection intensities, whatever their source. If, for example, it is suspected that the crystal exhibits anisotropic extinction, then the strongest reflections should be excluded from the least-squares fit of the a_{lm}.

Since we are concerned with an anisotropy correction to a spherical transmision surface, an expansion in multipolar spherical harmonic functions is a quite natural choice. The orthogonality of the y_{lm} makes fitting the coefficients a_{lm} easier than fitting coefficients a_{ijkl} and b_{ijkl} for a Fourier expansion in the diffractometer setting angles,

$$A = \sum_{-i_{\max}}^{i_{\max}} \sum_{-j_{\max}}^{j_{\max}} \sum_{-k_{\max}}^{k_{\max}} \sum_{-l_{\max}}^{l_{\max}} a_{ijkl} \cos{(i2\theta + j\omega + k\chi + l\varphi)}$$

$$+ b_{ijkl} \sin{(i2\theta + j\omega + k\chi + l\varphi)},$$

for which the determination of appropriate expansion orders $i_{\max}$, $j_{\max}$, $k_{\max}$ and $l_{\max}$ is not .trivial (Flack, 1977). Of course, either a spherical harmonic or a Fourier expansion can, in principle, follow the absorption anisotropy more faithfully than can azimuthal correction curves $A = A(\varphi|\chi = 90°)$ or $A = A(\psi|\chi \simeq 90°)$ (North, Phillips & Mathews, 1968; Kopfmann & Huber, 1968; Huber & Kopfmann, 1969).

If a crystal of any shape is uniformly irradiated, its anisotropic transmission surface must be centrosymmetric, since reversal of the beam direction, or 180° rotation of the crystal about its centroid, gives the same absorption paths. Absorption anisotropy should, therefore, be modelled with only even functions, y_{lm} with $l = 2n$. Odd functions, with $l = 2n + 1$, can be introduced to attempt to deal with problems of nonuniform irradiation due to an inhomogeneous quasiparallel beam incident from a crystal monochromator or to a specimen crystal that is over-sized or mis-centered.

Least-squares fitting of the expansion coefficients

The empirical coefficients a_{lm} are obtained by a linear least-squares fit minimizing a total residual

$$\chi^2 = \chi_I^2 + w_A \chi_A^2, \tag{6}$$

in which the first term is an intensity-fit residual for $A_{\text{aniso}} = A_{hi}$ and the second term is an anisotropy-restraint residual to restrain the fitted A_{hi} toward an average value $\langle A_{hi} \rangle = 1$. The subscript h indexes the unique reflections and i indexes the $n = n_h$ equivalent measurements of a reflection h.

The intensity-fit residual is defined as

$$\chi_I^2 = \sum_h \sum_{i=1}^{n} w_{hi} \ (I_{hi}A_{hi} - \langle I_{hi}A_{hi} \rangle_h)^2,$$

$$\chi_I^2 = \sum_h \sum_{i=1}^{n} w_{hi} \left[I_{hi}A_{hi} \right.$$

$$\left. - \left(\sum_{j=1}^{n} w_{hj}I_{hj}A_{hj} \right) \middle/ \left(\sum_{j=1}^{n} w_{hj} \right) \right]^2, \tag{7}$$

where $w_{hi} = 1/\sigma^2(I_{hi})$ and the $I_{hi} = |F(h)|_i^2$ are multiple symmetry-equivalent and/or azimuth rotation-equivalent intensity measurements reduced to squared structure-factor magnitudes.

The anisotropy-restraint residual is defined as

$$w_A \chi_A^2 = w_A \sum_h \sum_{i=1}^{n} (A_{hi} - 1)^2,$$

$$w_A \chi_A^2 = w_A \sum_h \sum_{i=1}^{n} \left(\sum_{l=1}^{l_{\max}} \sum_{m=-l}^{l} a_{lm} f_{lm,hi} \right)^2. \tag{8}$$

A constant weighing factor w_A multiplies χ_A^2 to scale the restraint residual relative to the fit residual and the value of w_A governs how tightly the restraint toward $\langle A_{hi} \rangle = 1$ applies. Depending upon the multiplicity of equivalent I_{hi} data and, in particular, upon how well the data sample the absorption paths through the crystal, using $w_A = 0$ might allow unreasonable extreme excursions of the fitted transmission surface in regions not sampled by multiple equivalent data.

To choose an appropriate value for w_A, we note that, if the differences among the equivalent I_{hi} are due mainly to the differences among the A_{hi}, we expect the normalized mean-square deviation of the A_{hi} from unity to be

$$\frac{\chi_A^2}{\sum_h \sum_i A_{hi}^2} \simeq \frac{\sum_h \sum_i w_{hi}(I_{hi} - \langle I_{hi} \rangle_h)^2}{\sum_h \sum_i w_{hi}I_{hi}^2} = R_w^2. \tag{9}$$

If $\langle A_{hi} \rangle \simeq 1$, the denominator on the left is

$$\sum_h \sum_i A_{hi}^2 \simeq N,$$

where $N = \sum_h n_h$ is the total number of measurements used in the fitting. Thus, we expect

$$\chi_A^2 \simeq N R_w^2$$

and, in order to have a standardized mean-square deviation

$$w_A \chi_A^2/N \simeq 1,$$

we should choose

$$w_A = 1/R_w^2. \tag{10}$$

This has the sensible property that the larger the differences among the equivalent I_{hi}, the more slack the restraint toward $\langle A_{hi} \rangle \simeq 1$.

Least-squares normal equations

For minimization of the intensity-fit residual, a change of variable (Katayama, 1986) simplifies the development and gives least-squares normal equations that are explicitly linear in the desired coefficients a_{lm}. The intensity-fit residual, (7), becomes

$$\chi_I^2 = \sum_h^n \sum_{i=1}^n w_{hi} \left(\sum_{j=1}^n J_{hj} A_{hj} \right)^2 \qquad (11)$$

with the change of variable

$$\begin{cases} J_{hj} = -\left(w_{hj} \Big/ \sum_{k=1}^n w_{hk} \right) I_{hj} & \text{if } j \neq i, \\[3mm] J_{hj} = \left[1 - \left(w_{hj} \Big/ \sum_{k=1}^n w_{hk} \right) \right] I_{hj} & \text{if } j = i. \end{cases} \qquad (12)$$

Then, rewriting (4) with a single dummy index for the expansion orders,

$$A_{hi} = 1 + \sum_{l=1}^{l_{max}} \sum_{m=-l}^l a_{lm} f_{lm,hi} = 1 + \sum_{k=1}^{k_{max}} a_k f_{k,hi},$$

we get

$$\chi_I^2 = \sum_h \sum_{i=1}^n w_{hi} \left[\sum_{j=1}^n J_{hj} \left(1 + \sum_{k=1}^{k_{max}} a_k f_{k,hj} \right) \right]^2$$

$$\chi_I^2 = \sum_h \sum_i w_{hi} \left(\sum_j J_{hj} + \sum_k a_k \sum_j J_{hj} f_{k,hj} \right)^2$$

and the condition for a minimum,

$$\partial \chi_I^2 / \partial a_l = 2 \sum_h \sum_i w_{hi} \left(\sum_j J_{hj} + \sum_k a_k \sum_j J_{hj} f_{k,hj} \right)$$

$$\times \left(\sum_j J_{hj} f_{l,hj} \right)$$

$$= 0,$$

gives the normal equations for the intensity fit,

$$\sum_k a_k \sum_h \sum_j w_{hi} \left(\sum_j J_{hj} f_{k,hj} \right) \left(\sum_j J_{hj} f_{l,hj} \right)$$

$$= -\sum_h \sum_i w_{hi} \left(\sum_j J_{hj} \right) \left(\sum_j J_{hj} f_{l,hj} \right),$$

$$l = 1, 2, \ldots, k_{max}. \qquad (13)$$

Introducing the restraint toward $\langle A_{hi} \rangle \simeq 1$, and minimizing the total residual, we have

$$\chi^2 = \chi_I^2 + w_A \chi_A^2 = \chi_I^2 + w_A \sum_h \sum_i \left(\sum_{k=1}^{k_{max}} a_k f_{k,hi} \right)^2,$$

$$\partial \chi^2 / \partial a_l = (\partial \chi_I^2 / \partial a_l) + w_A (\partial \chi_A^2 / \partial a_l)$$

$$= \partial \chi_I^2 / \partial a_l + 2 w_A \sum_h \sum_i \left(\sum_k a_k f_{k,hi} \right) f_{l,hi}$$

$$= 0,$$

and the restrained normal equations, still linear in the coefficients a_{lm}, become

$$\sum_k a_k \sum_h \sum_i \left[w_A f_{k,hi} f_{l,hi} + w_{hi} \left(\sum_j J_{hj} f_{k,hj} \right) \left(\sum_j J_{hj} f_{l,hj} \right) \right]$$

$$= -\sum_h \sum_i w_{hi} \left(\sum_j J_{hj} \right) \left(\sum_j J_{hjh} f_{l,hj} \right),$$

$$l = 1, 2, \ldots, k_{max}. \qquad (14)$$

Evaluation of the fitting functions from the diffraction geometry

Cartesian arguments of the spherical harmonic functions are obtained *via* the rotation matrix for Eulerian diffractometer setting angles:

$$\omega, \chi, \varphi \rightarrow \mathbf{R}_\varphi \mathbf{R}_\chi \mathbf{R}_\omega = \mathbf{R} = (\mathbf{XYZ});$$

$$\mathbf{X}, \mathbf{Y}, \theta, \lambda \rightarrow \mathbf{u}_0, \quad \mathbf{u}_1 \rightarrow y_{lm}(-\mathbf{u}_0), \; y_{lm}(\mathbf{u}_1). \qquad (15)$$

We employ diffractometer axes defined (Hamilton, 1974) such that, for each reflection measurement, components of orthonormal vectors $\mathbf{X}$, $\mathbf{Y}$ and $\mathbf{Z}$ referred to crystal-fixed orthonormal axes are given by the columns of the rotation matrix

$$\mathbf{R} = \begin{pmatrix} \cos\varphi & \sin\varphi & 0 \\ -\sin\varphi & \cos\varphi & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos\chi & \sin\chi \\ 0 & -\sin\chi & \cos\chi \end{pmatrix} \begin{pmatrix} \cos\omega & \sin\omega & 0 \\ -\sin\omega & \cos\omega & 0 \\ 0 & 0 & 1 \end{pmatrix},$$

$$\mathbf{R} = \begin{pmatrix} \cos\varphi \cos\omega - \sin\varphi \sin\omega \cos\chi & \cos\varphi \sin\omega + \sin\varphi \cos\omega \cos\chi & \sin\varphi \sin\chi \\ -\sin\varphi \cos\omega - \cos\varphi \sin\omega \cos\chi & -\sin\varphi \sin\omega + \cos\varphi \cos\omega \cos\chi & \cos\varphi \sin\chi \\ \sin\chi \sin\omega & -\sin\chi \cos\omega & \cos\chi \end{pmatrix}, \qquad (16)$$

$$X = R \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, \quad Y = R \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} \quad \text{and} \quad Z = R \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}. \quad (17)$$

The incident- and scattered-beam wave vectors s_0 and s_1 are obtained from the diffraction vector $D = s_1 - s_0$ and its normal $T = s_1 + s_0$ in the equatorial plane, as shown in Fig. 1 (Coppens, 1970). The definition of diffractometer axes (Hamilton, 1974) is such that the unit vector Y is parallel to D and the unit vector X is antiparallel to T. Thus,

$$s_1 - s_0 = D = Y[2(\sin \theta)/\lambda]$$

$$s_1 + s_0 = T = -X[2(\cos \theta)/\lambda],$$

$$s_1 = (T + D)/2$$

$$s_0 = (T - D)/2$$

and the corresponding unit direction vectors are given by

$$u = \lambda s,$$

$$u_1 = -X \cos \theta + Y \sin \theta$$

$$u_0 = -X \cos \theta - Y \sin \theta. \quad (18)$$

The components of the unit vectors u are their direction cosines, which are Cartesian arguments for the real spherical harmonic functions $y_{lm}(u)$. Cartesian forms for evaluating the functions have been published for $l \le 7$ (Paturle & Coppens, 1988) and the forms for $l = 8$ are given in the Appendix, which was kindly provided by Dr Antoine Paturle (1990).

Test calculations

Routines for carrying out the calculations outlined above have been incorporated into the program
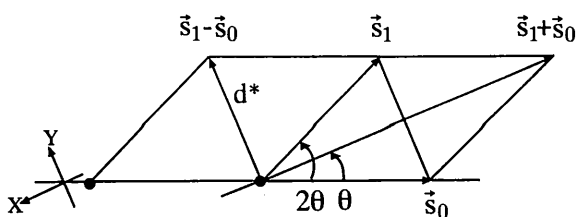
SORTAV for sorting, inter-subset scaling, averaging and analysis of variance for multiple equivalent intensity measurements [Blessing (1989) and earlier references cited therein]. The new routines have been tested against synthetic standard absorption test data for the so-called Alcock irregular crystal (Flack, Vincent & Alcock, 1980). With the program ABSORB (DeTitta, 1985), Eulerian diffractometer setting angles $(2\theta,\omega,\chi,\varphi)$ were calculated for the test crystal for $-3 \le h,k,l \le +3$ and $\psi = 0$, 10, 20, ..., 180° and transmission factors $A(h,k,l,\psi)$ were computed by Gaussian numerical quadrature on a $16 \times 16 \times 16$ grid. This gave a set of 6498 transmission factors for 171 unique reflections in Laue group $\bar{1}$. The transmission factors ranged from 0.121 to 0.344 and these were taken to be intensity data for testing the new routines.

Table 1 and Fig. 2 summarize tests of the multipole expansion limits. The statistics of fit for the transmission surface improved significantly as even multipoles were added through order eight, but, as anticipated, added odd multipoles had no significant effect. The anisotropy-corrected and then averaged unique data from the fit with $l = 1$ through 8 and $N_{par} = 80$ were essentially the same as those from the fit with $l = 2$, 4, 6, 8 and $N_{par} = 44$. Averaging the two unique data sets gave $R_{merge} = [\Sigma(I - \langle I \rangle)^2/\Sigma I^2]^{1/2} = 0.0001$, a further confirmation that the odd multipoles were superfluous because the transmission surface was centrosymmetric.

In the calculations summarized in Table 1, the models were fitted against all the generated data, with weight $w_A = 0$ for the anisotropy restraint residual. Table 2 summarizes a test of the restraint condition with the $l = 2$, 4, 6, 8 model. Test 1, against all

Fig. 1. Diffraction geometry for incident beam s_0 and scattered beam s_1 with wave-vector magnitudes $|s_0| = |s_1| = 1/\lambda$.

$$|s_0| = |s_1| = 1/\lambda$$

$$s_1 - s_0 = D \qquad |D| = 2(\sin\theta)/\lambda$$

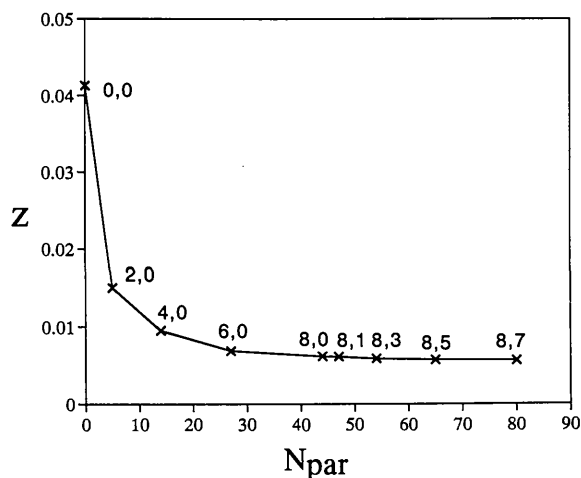$$s_1 + s_0 = T \qquad |T| = 2|s_0| \cos \theta = 2(\cos \theta)/\lambda$$

Fig. 2. Goodness of fit versus number of parameters from the tests of the multipole expansion limits summarized in Table 1. The points are labelled with the maximum even and maximum odd multipole orders.

Table 1. *Tests of even and even-plus-odd multipole expansion limits*

The statistics of fit are the standardized root-mean-square deviation of equivalent data from their means,

$$Z = [\sum_h \sum_i w_{hi}(I_{hi}A_{hi} - \langle I_{hi}A_{hi}\rangle_h)^2 / (N_{data} - N_{means} - N_{par})]^{1/2}$$

and the normalized root-mean-square deviation,

$$R = [\sum_h \sum_i w_{hi}(I_{hi}A_{hi} - \langle I_{hi}A_{hi}\rangle_h)^2 / \sum_h \sum_i w_{hi}(I_{hi}A_{hi})^2]^{1/2}.$$

Since the test calculations were made with 'error-free' synthetic data, all $w_{hi} = 1$.

l_{max}					A_{aniso}		
Even	Odd	N_{par}	Z	R	Minimum	Maximum	Mean ± r.m.s. deviation
0	0	0	0.04129	0.1662	1.0	1.0	1.0 ± 0.0
2	0	5	0.01506	0.0630	0.607	1.31	1.01 ± 0.168
4	0	14	0.00950	0.0399	0.658	1.45	1.01 ± 0.187
6	0	27	0.00689	0.0289	0.655	1.56	1.01 ± 0.193
8	0	44	0.00616	0.0258	0.651	1.62	1.01 ± 0.195
8	1	47	0.00613	0.0257	0.650	1.62	1.01 ± 0.195
8	3	54	0.00587	0.0246	0.650	1.62	1.01 ± 0.195
8	5	65	0.00574	0.0241	0.650	1.62	1.01 ± 0.195
8	7	80	0.00564	0.0236	0.651	1.62	1.01 ± 0.196

Table 2. *Test calculations fitting $N_{par} = 44$ coefficients, a_{lm} with $l = 2, 4, 6, 8$*

Tests 1 and 2 are without and test 3 is with an anisotropy restraint. The agreement statistic is

$$R = [\sum_h \sum_i w_{hi}(I_{hi}A_{hi} - \langle I_{hi}A_{hi}\rangle_h)^2 / \sum_h \sum_i w_{hi}(I_{hi}A_{hi})^2]^{1/2},$$

where all $A_{hi} = 1$ before the least-squares fitting and all $w_{hi} = 1$. The minimum and maximum values of $A_{aniso} = A_{hi}$ are followed by their standard deviations (in parentheses) estimated from the least-squares variance–covariance matrix for the fitted a_{lm}. The mean values are followed by the root-mean-square deviations from the mean.

			R		A_{aniso}		
Test	N_{data}	N_{means}	Before	After	Minimum	Maximum	Mean ± r.m.s. deviation
1	6498	171	0.1662	0.0258	0.649 (2)	1.621 (4)	1.01 ± 0.195
2	3204	171	0.1681	0.0235	0.567 (5)	1.430 (11)	0.843 ± 0.197
3	3204	171	0.1681	0.0281	0.667 (3)	1.511 (7)	0.947 ± 0.198

the data and with $w_A = 0$, reduced the normalized root-mean-square discrepancy among equivalent data R from 16.6 to 2.6%. In a somewhat more realistic test 2, the test 1 calculation was repeated using only data with $-48 \le \omega + \theta \le +58°$ and $|\omega - \theta| \le 60°$, which would be mechanically accessible on a Siemens (née Nicolet née Syntex) $P3$ diffractometer. This restriction eliminated about half the data and, owing to the 'blind' regions wherein the transmission surface was not sampled by the data, the range and mean of the fitted anisotropy corrections were shifted to smaller values. Nevertheless, the anisotropy-corrected and then averaged unique data set from test 2 agreed with that from test 1 to within a least-squares-fitted scale factor $k_2 = 1.1364$ (5) and $R_{merge} = 0.0076$. In test 3, the test 2 calculation was repeated with the anisotropy-restraint residual given a weight $w_A = 1/R^2 = 1/0.1681^2$. This moved the range and mean of the fitted anisotropy corrections back close to the test 1 values, and the unique data sets from test 3 and test 1 agreed to within a scale factor $k_3 = 1.0180$ (8) and $R_{merge} = 0.0107$.

Application of the new routines to a data set measured for an experimental electron-density study is described elsewhere (Souhassou, Espinosa, Lecomte & Blessing, 1995).

Extensively commented Fortran77 source code and an ASCII file of detailed users' instructions for the *SORTAV* program augmented by the routines for the empirical absorption correction are available on request. Users must bear in mind that reliable anisotropy corrections require a quite substantial multiplicity of symmetry-equivalent and/or azimuthrotation-equivalent measurements, for at least a lower-order subset of data, in order that there be an adequate sampling of absorption paths through the specimen crystal.

APPENDIX

Table 3. *Cartesian forms for the real spherical harmonic functions with l = 8 (Paturle, 1990)*

These extend the published tabulation for $l \leq 7$ (Paturle & Coppens, 1988). The arguments x, y and z are orthonormal components, *i.e.* direction cosines, of a unit vector and the functions

$$y_{l,m} = N_{l,|m|} C_{l,|m|} c_{l,m}(x,y,z)$$

are normalized such that

$$\int_{\Omega} |y_{l,m}| \, d\Omega = 2 - \delta_{0,l}.$$

m	$c_{8,m}$	$C_{8,|m|}$	$N_{8,|m|}$
0	$6435z^8 - 12012z^6 + 6930z^4 - 1260z^2 + 35$	0.0078125	0.0059609
1	$(715z^7 - 1001z^5 + 385z^3 - 35z)x$	0.5625	0.0784858
-1	$(715z^7 - 1001z^5 + 385z^3 - 35z)y$		
2	$(143z^6 - 143z^4 + 33z^2 - 1)(x^2 - y^2)$	19.6875	0.3253786
-2	$(143z^6 - 143z^4 + 33z^2 - 1)(2xy)$		
3	$(39z^5 - 26z^3 + 3z)(x^3 - 3xy^2)$	433.125	0.8780415
-3	$(39z^5 - 26z^3 + 3z)(3x^2y - y^3)$		
4	$(65z^4 - 26z^2 + 1)(x^4 - 6x^2y^2 + y^4)$	1299.375	0.3411683
-4	$(65z^4 - 26z^2 + 1)(4x^3y - 4xy^3)$		
5	$(5z^3 - z)(x^5 - 10x^3y^2 + 5xy^4)$	67567.5	2.4892756
-5	$(5z^3 - z)(5x^4y - 10x^2y^3 + y^5)$		
6	$(15z^2 - 1)(x^6 - 15x^4y^2 + 15x^2y^4 - y^6)$	67567.5	0.3933012
-6	$(15z^2 - 1)(6x^5y - 20x^3y^3 + 6xy^5)$		
7	$z(x^7 - 21x^5y^2 + 35x^3y^4 - 7xy^6)$	2027025	2.2500000
-7	$z(7x^6y - 35x^4y^3 + 21x^2y^5 - y^7)$		
8	$x^8 - 28x^6y^2 + 70x^4y^4 - 28x^2y^6 + y^8$	2027025	0.6152344
-8	$8x^7y - 56x^5y^3 + 56x^3y^5 - 8xy^7$		

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A Matrix-Operator Approach to Reflection High-Energy Electron Diffraction Theory

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Abstract

The amplitudes of beams reflected from a crystal surface by high-energy electrons are expressed in terms of matrix operators based on Bloch waves. The solution is derived in terms of the limiting case of an infinite slab and is therefore applicable to cases involving overlayers of different composition and