Acta Cryst. (1988). A**44**, 6–7

Normalization Factors for Spherical Harmonic Density Functions

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(*Received* 30 *April* 1987; *accepted* 7 *July* 1987)

Abstract

Normalization factors N_{lm} for spherical harmonic density functions c_{lmp} defined by $\int N_{lm}|c_{lmp}|\,d\tau = 2-\delta_{l0}$ have been derived for $l \le 7$, from both analytical and numerical integration methods.

Introduction

The increasing accuracy of X-ray diffraction data has led to more widespread use of spherical atom scattering formalisms. In the multipole formalism the atomic density is described by a series of real spherical harmonic functions y_{lmp} with $p = +$ or $-$, multiplied by a normalized radial function $R(r)$ (Dawson, 1967; Stewart, 1976; Hansen & Coppens, 1978; Kurki-Suonio, 1968; Price & Maslen, 1978). The functions y_{lmp} are defined as

$$y_{lmp} = N_{lmp}P_l^m(\cos\theta)\begin{matrix}\cos m\varphi & \text{for } p=+ \\ \sin m\varphi & \text{for } p=-\end{matrix} \quad (1)$$

where $P_l^m(\cos\theta)$ are the associated Legendre polynomials (Arfken, 1970). A number of authors (Price & Maslen, 1978; Hansen & Coppens, 1978) have normalized the angular functions y_{lmp} such that

$$\begin{aligned}\int|y_{lmp}(\theta,\varphi)|\,d\tau &= 1 \quad \text{if } l=0\\ &= 2 \quad \text{if } l>0.\end{aligned} \quad (2)$$

We shall adopt the convention that density functions normalized according to (2) are labelled d_{lmp} (Coppens, 1988).

For a monopole function ($l = 0$) the normalization implies that a population parameter $P_{00} = 1$ corresponds to a population of one electron, while for the higher-order poles $P_{lm} = 1$ implies that one electron has been shifted from the negative to the positive lobes of the function. Normalization factors N_{lm} for $l \le 4$ have been published (Hansen & Coppens, 1978). Though truncation of the expansion at $l = 4$ is often warranted, this is not always the case. In particular, in highly symmetric environments the lowest symmetry-allowed multipole with $m \ne 0$ may be of higher order than four in l. An example is an atom at a site with sixfold symmetry, for which the first allowed multipole with non-zero m is y_{66+}. We report here on the values of N_{lm} as defined by (1) and (2) for $l \le 7$.

Method of calculation

From (1) and (2) N_{lm} is given by

$$N_{lm} = 1/I_{lm}J_m \quad \text{or} \quad N_{lm} = 2/I_{lm}J_m \quad (3)$$

with

$$I_{lm} = \int_{-1}^{+1}|P_l^m(z)|\,dz$$

and

$$J_m = \int_0^{2\pi}|\cos m\varphi|\,d\varphi$$

or

$$J_m = \int_0^{2\pi}|\sin m\varphi|\,d\varphi,$$

which gives

$$J_m = 4 \text{ for } m > 0; \quad J_m = 2\pi \text{ for } m = 0.$$

Evaluation of I_{lm} requires calculation of the roots of the equation $P_l^m(z) = 0$ and integration between boundaries equal to the root values. This is most easily done when the associated Legendre functions are expressed as a series in powers of z:

$$P_l^m(z) = (1/l!2^l)(1-z^2)^{m/2}p_{lm}(z), \quad (4)$$

where

$$p_{lm}(z) = \sum_{k=k_i}^{l} A_k z^{2k-l-m}$$

with

$$A_k = \frac{(-1)^{k-l}l!}{k!(l-k)!}\frac{(2k)!}{(2k-l-m)!}$$

$$k_i = \begin{matrix}(l+m)/2 & \text{if } l+m \text{ odd}\\ (l+m+1)/2 & \text{if } l+m \text{ even.}\end{matrix}$$

Thus $p_{lm}(z)$ is a polynomial of degree $l-m$. Since the exponent increases by two between successive terms either even or odd powers occur.

The integration (2) requires evaluation of the roots of p_{lm}. As it is not possible to find analytically the roots of a polynomial of degree larger than 4, the

0108-7673/88/010006-02\$03.00

Table 1. *Normalization factors of spherical harmonic density functions*

l	m	p	$c_{lmp}(x, y, z)$*	C†	N_{lm} numerical	N_{lm} analytical
0	0		1	(1)	0·0795776	0·0795774
1	0		z	(1)	0·3183106	0·3183099
	1	+	x	(1)	0·3183103	0·3183099
		−	y			
2	0		$3z^2-1$	(1/2)	0·2067485	0·2067483
	1	+	xz	(3)	0·7500008	0·7500000
		−	yz			
	2	+	x^2-y^2	(3)	0·3750000	0·3750000
		−	$2xy$			
3	0		$5z^3-3z$	(1/2)	0·2448539	0·2448538
	1	+	$(5z^2-1)x$	(3/2)	0·3203333	0·3203331
		−	$(5z^2-1)y$			
	2	+	$z(x^2-y^2)$	(15)	1·0000000	1·0000000
		−	$2xyz$			
	3	+	x^3-3xy^2	(15)	0·4244131	0·4244132
		−	$3x^2y-y^3$			
4	0		$35z^4-30z^2+3$	(1/8)	0·0694175	0·0694175
	1	+	$(7z^3-3z)x$	(5/2)	0·4740028	0·4740025
		−	$(7z^3-3z)y$			
	2	+	$(7z^2-1)(x^2-y^2)$	(15/2)	0·3305913	0·3305913
		−	$2xy(7z^2-1)$			
	3	+	$z(x^3-3xy^2)$	(105)	1·2499999	1·2500000
		−	$z(3x^2y-y^3)$			
	4	+	$x^4-6x^2y^2+y^4$	(105)	0·4687500	0·4687500
		−	$4x^3y-4xy^3$			
5	0		$63z^5-70z^3+15z$	(1/8)	0·0767395	0·0767395
	1	+	$(21z^4-14z^2+1)x$	(15/8)	0·3229812	
		−	$(21z^4-14z^2+1)y$			
	2	+	$(3z^3-z)(x^2-y^2)$	(105/2)	1·6875000	
		−	$2xy(3z^3-z)$			
	3	+	$(9z^2-1)(x^3-3xy^2)$	(105/2)	0·3451455	
		−	$(9z^2-1)(3x^2y-y^3)$			
	4	+	$z(x^4-6x^2y^2+y^4)$	(945)	1·5000000	1·5000000
		−	$z(4x^3y-4xy^3)$			
	5	+	$x^5-10x^3y^2+5xy^4$	(945)	0·5092958	0·5092958
		−	$5x^4y-10x^2y^3+y^5$			
6	0		$231z^6-315z^4+105z^2-5$	(1/16)	0·0417084	
	1	+	$(33z^5-30z^3+5z)x$	(21/8)	0·4172129	0·4172128
		−	$(33z^5-30z^3+5z)y$			
	2	+	$(33z^4-18z^2+1)(x^2-y^2)$	(105/8)	0·3261107	0·3261107
		−	$2xy(33z^4-18z^2+1)$			
	3	+	$(11z^3-3z)(x^3-3xy^2)$	(315/2)	0·6513219	
		−	$(11z^3-3z)(3x^2y-y^3)$			
	4	+	$(11z^2-1)(x^4-6x^2y^2+y^4)$	(945/2)	0·3610405	
		−	$(11z^2-1)(4x^3y-4xy^3)$			
	5	+	$z(x^5-10x^3y^2+5xy^4)$	(10395)	1·7500002	1·7500000
		−	$z(5x^4y-10x^2y^3+y^5)$			
	6	+	$x^6-15x^4y^2+15x^2y^4-y^6$	(10395)	0·5468750	
		−	$6x^5y-20x^3y^3+6xy^5$			
7	0		$429z^7-693z^5+315z^3-35z$	(1/16)	0·0447979	
	1	+	$(429z^6-495z^4+135z^2-5)x$	(7/16)	0·0648780	
		−	$(429z^6-495z^4+135z^2-5)z$			
	2	+	$(143z^5-110z^3+15z)(x^2-y^2)$	(63/8)	0·1573192	
		−	$2xy(143z^5-110z^3+15z)$			
	3	+	$(143z^4-66z^2+3)(x^3-3xy^2)$	(315/8)	0·1109240	
		−	$(143z^4-66z^2+3)(3x^2y-y^3)$			
	4	+	$(13z^3-3z)(x^4-6x^2y^2+y^4)$	(3465/2)	0·7404370	
		−	$(13z^3-3z)(4x^3y-4xy^3)$			
	5	+	$(13z^2-1)(x^5-10x^3y^2+5xy^4)$	(10395/2)	0·3772319	
		−	$(13z^2-1)(5x^4y-10x^2y^3+y^5)$			
	6	+	$z(x^6-15x^4y^2+15x^2y^4-y^6)$	(135135)	2·0000000	
		−	$z(6x^5y-20x^3y^3+6xy^5)$			
	7	+	$x^7-21x^5y^2+35x^3y^4-7xy^6$	(135135)	0·5820523	
		−	$7x^6y-35x^4y^3+21x^2y^5-y^7$			

* $x=\sin\theta\cos\varphi$, $y=\sin\theta\sin\varphi$, $z=\cos\theta$.

† Common factor such that $Cc_{lmp}=d_{lmp}$, where d_{lmp} is defined by equations (1) and (2).

analytical integration is limited to values of $l\le 9$ (for $l=8$ the expansion contains four even terms; for $l=9$, z can be factored out, which leads to a four-term expansion). In addition, the analytical procedure is extremely cumbersome. An alternative procedure is a numerical evaluation of both the roots and the integrals. Using the commercially available subroutines *ZROOTS* (Laguerre method) and *QROMB* (Romberg integration) (Press, Flannery, Teukolsky & Vetterling, 1986) we have evaluated the integrals I_{lm}. Results of the numerical integration, as well as analytical values for a number of cases are given in Table 1. In the calculation of these values, common integers in the functions d_{lm} have been eliminated (Hansen & Coppens, 1978; Coppens, 1988). The definition of the resulting functions c_{lm} and the common integers are given in the fourth and fifth columns of Table 1.

The comparison with the analytical results is a measure for the numerical accuracy obtained. Since standard deviations in experimental population parameters are considerable [$\sigma(P)/P$ is typically larger than 5%], the accuracy achieved is more than adequate.

Support of this work by the National Science Foundation (CHE8403428) is gratefully acknowledged. Computing was done on a VAX785 made available through the National Science Foundation (CHE8406077).

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Acta Cryst. (1988). A**44**, 8-14

X-ray Topography of Bent Crystals

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(*Received* 18 *November* 1986; *accepted* 20 *May* 1987)

Abstract

The influence of the bending of a crystal on the formation of diffracted images in Bragg section topographs as well as in Laue section and traverse topographs is studied. In the case of Bragg section topography the interferometric fringes due to the interference of waves once and twice internally reflected inside a bent crystal are described. It is established that the maximum positions of diffracted intensity satisfy the law $x_n=[16\pi(2n-1)/5B^2]^{1/3}$, where x_n is the distance from the incidence slit of the X-rays, B is the uniform strain gradient and n is the fringe number. This dependence is found to be in good agreement with experimental data. The Laue section topograph of a bent crystal with a screw dislocation parallel to the diffraction vector is considered. The effects of asymmetry in the *Pendellösung* fringe pattern and of 'splitting' of the direct image with respect to the dislocation line in both the experimental and simulated topographs are accounted for. The variation of the contrast of dislocations with depth inside a bent crystal in Laue traverse topographs is studied by computer simulations using the reciprocity theorem.

1. Introduction

The dynamical scattering of X-rays (DSXR) in distorted crystals with a uniform strain gradient (USG) has been investigated in a number of studies (Kato, 1964; Bonse, 1964; Penning, 1966; Hart, 1966; Blech & Meieran, 1967; Malgrange, 1969; Ando & Kato, 1970; Petrashen', 1973; Chukhovskii, 1974; Petrashen' & Chukhovskii, 1975; Chukhovskii & Petrashen', 1977; Chukhovskii, Gabrielyan & Petrashen', 1978; Kushnir, Suvorov & Mukhin, 1980; Khrupa, Kislovskii & Datzenko, 1980; Petrashen' & Yaroslavskaya, 1981; Balibar, Chukhovskii & Malgrange, 1983; Shulpina, Petrashen', Chukhovskii & Gabrielyan, 1984) and is a starting point for the quantitative theory of X-ray topographic images and the theory of DSXR in distorted crystals in general.

In practice a situation can exist where the USG macroscopic elastic field is superposed on those caused by single defects in a crystal. Their cooperative action was experimentally observed: in X-ray traverse topographs of bent crystals the dislocation contrast is found to be reversed (Blech & Meieran, 1967); in Laue section topographs of a twisted silicon crystal the *Pendellösung* fringe patterns become asymmetrical with respect to a dislocation line (Kushnir, Suvorov & Mukhin, 1980); owing to dislocation loops randomly distributed within a crystal the dependence of the reflecting power on the radius of curvature of the sample is changed (Khrupa, Kislovskii & Datenzko, 1980); and in Bragg diffraction a new kind of USG fringe, curved near local inhomogeneities, has been observed recently (Shulpina, Petrashen', Chukhovskii & Gabrielyan, 1984).

The study of these phenomena is of interest in itself since it permits one to understand better the general features of the formation of X-ray topographic images and it can also demonstrate how to use crystal bending as a new tool for DSXR investigations of crystal-lattice defects.

In the present paper we shall discuss the physical foundations of the formation of X-ray topographic images for bent crystals; these are important from the point of view of applications and further development of DSXR methods. We confine our consideration to the case of the USG $|B|\ll 1$ [the dimensionless USG $B=\frac{1}{4}\partial^2(\mathbf{hu})/\partial s_0\partial s_h$; hereafter the notation is that of Petrashen' & Chukhovskii (1975)], when two wave fields relating to the two branches of the dispersion surface are essential and contribute simultaneously to the formation of images. In § 2 the Green function of the diffracted radiation and its asymptotic expression (the eikonal approximation) are used for the explanation of the *Pendellösung* fringes in Laue section topographs. The eikonal approximation is used to treat the USG fringe formation in Bragg section topographs in § 3.

In the final section some pecularities of the dislocation images in both Laue section and traverse topographs are discussed. The treatment is based on

0108-7673/88/010008-07$03.00