Acta Cryst. (1998). A**54**, 186–190

Absorption and Weighted Path Lengths in Cylinders and Spheres

GUNNAR THORKILDSEN* AND HELGE B. LARSEN

Department of Mathematics and Natural Science, Stavanger College, Ullandhaug, 4004 Stavanger, Norway. E-mail: gunnar.thorkildsen@tn.his.no

(*Received 30 May 1997; accepted 22 September 1997*)

Abstract

By using the boundary-value Green-function technique for the Takagi equations, it is possible to calculate the normal absorption factor and weighted path length in cylinders and spheres as double integrals in angular coordinates of well defined functions. The results are easy to implement for numerical computations. Exact analytical expressions for both crystal geometries are found in the limits of the Bragg angle $\theta_{oh} \to 0°$ and $\theta_{oh} \to 90°$.

1. Introduction

In a previous paper (Thorkildsen & Larsen, 1998), hereafter denoted TL, we have shown how to obtain analytical expressions for the primary extinction factor in cylindrical and spherical perfect crystals. The method, which is based on the boundary-value Green-function technique, is also applicable for calculating normal absorption factors in finite sized crystals. This concept involves entrance and exit *surface* integrations and is thus a novel approach for obtaining the absorption factors.

In the literature (Rouse *et al.*, 1970; Dwiggens, 1972, 1975*a*,*b*), it has been shown how the normal absorption factors in cylinders and spheres can be calculated by performing a numerical *volume* integration over the crystal in question. Similar techniques have also been applied by several authors to calculate values for the weighted path length for spherical crystals (Weber, 1969; Flack & Vincent, 1978; Rigoult & Guidi-Morosini, 1980). More recently, Clark & Reid (1995) used a method based on generation of Howells polyhedra (Howells, 1950; de Meulenaer & Tompa, 1965) to calculate path lengths in a 100-sided 'polycylinder' and a 74-sided 'polysphere'. An overview of absorption-factor calculations is given in ch. 6.3 of *International Tables for Crystallography*, Volume C (Maslen, 1995).

All function definitions, in *Mathematica* code, are available from the authors on request.

2. Absorption

2.1. *General expressions*

The generalized extinction factor, y, the attenuation factor for the integrated power from a perfect crystal due to the combined effect of multiple scattering and absorption, is defined in TL. The expression for a cylindrical crystal is†

$$\begin{aligned} y = (1/\pi \sin 2\theta_{oh}) \sum_{r} \sum_{m=m'(r)} \int_0^1 \mathrm{d}z \\ \times \int_{M(m)} \mathrm{d}\psi_M [-\sin(\psi_M + \theta_{oh})] \int_{S(M)} \mathrm{d}\psi_S \sin(\psi_S - \theta_{oh}) \\ \times |G_h(\psi_S, \psi_M | m; r)|^2 \\ \times \exp[-\mu R(\sin\psi_S - \sin\psi_M)/\cos\theta_{oh}]. \end{aligned} \quad (1)$$

This expansion can be used to calculate the normal absorption factor, A, by applying the zeroth-order term for the boundary-value Green functions, $G_h^{(0)}(\psi_S, \psi_M | m'; r)$, in (1). For the regions which then contribute,

$$G_h^{(0)}(\psi_S, \psi_M | m'; r) = 1. \quad (2)$$

Thus, we have for the absorption factor

$$\begin{aligned} A = (1/\pi \sin 2\theta_{oh}) \sum_{r} \sum_{m=m'(r)} \int_0^1 \mathrm{d}z \\ \times \int_{M(m)} \mathrm{d}\psi_M [-\sin(\psi_M + \theta_{oh})] \int_{S(m)} \mathrm{d}\psi_S \sin(\psi_S - \theta_{oh}) \\ \times \exp[-\mu R(\sin\psi_S - \sin\psi_M)/\cos\theta_{oh}]. \end{aligned} \quad (3)$$

The regions that have a non-vanishing zeroth-order term are I_L, II_L and I_B. I_L contributes when $\theta_{oh} \le 45°$ only, *cf.* Figs. 1 and 2. From the figures, we find that the integrations can be organized in three terms as given in Table 1. It is however possible to simplify the calculations by introducing new coordinates x' and y' as shown in Fig. 3. By performing the double integration in x' and y', the number of integrations is reduced from three to one. The range of integrations in the new coordinates is the same for both $0 \le \theta_{oh} \le \pi/4$ and $\pi/4 \le \theta_{oh} \le \pi/2$. Thus, one setup applies in the whole θ_{oh} range $(0, \pi/2)$. With $v \stackrel{\text{def}}{=} 2^{1/2}/2$, we have the transformations:

$$\begin{aligned} x' &= v(\psi_M - \psi_S) \\ y' &= v[\psi_M + \psi_S - 2(\pi - \theta_{oh})] \end{aligned}$$

† Notations and symbols are defined in TL.

with an inverse:

$$\psi_M = v(x' + y') + \pi - \theta_{oh}$$
$$\psi_S = v(y' - x') + \pi - \theta_{oh}.$$

The Jacobian of this transformation is 1. The setup is further simplified by introducing $x = vx'$ and $y = vy'$, which finally leads to

$$\sin(\psi_M + \theta_{oh}) = -\sin(x + y)$$
$$\sin(\psi_S - \theta_{oh}) = \sin(x - y + 2\theta_{oh})$$
$$\sin\psi_S - \sin\psi_M = 2\sin x\cos(y - \theta_{oh})$$
$$\mathrm{d}\psi_S\,\mathrm{d}\psi_M = 2\,\mathrm{d}x\,\mathrm{d}y.$$

The transition to a sphere involves a z integration, which can be performed as follows:†

$$\begin{aligned}\int_0^1 \mathrm{d}z\,\tfrac{3}{2}(1 - z^2)\exp[-a(1 - z^2)^{1/2}] &= \tfrac{3}{2}\int_0^{\pi/2} \mathrm{d}\varphi\,\sin^3\varphi\exp(-a\sin\varphi)\\ &= \tfrac{3}{2}\sum_{n=0}^{\infty}[(-1)^n/n!]a^n\int_0^{\pi/2}\mathrm{d}\varphi\,\sin^{3+n}\varphi\\ &= {}_1F_2[2, (\tfrac{1}{2}, \tfrac{5}{2}), a^2/4] - (9\pi/4a)I_2(a) - (3\pi/4)I_3(a),\end{aligned} \quad (4)$$

where

$$\begin{aligned}a &= \mu R(\sin\psi_S - \sin\psi_M)/\cos\theta_{oh}\\ &= 2\mu R[\sin x\,\cos(y - \theta_{oh})]/\cos\theta_{oh},\end{aligned}$$

${}_pF_q$ is a generalized hypergeometric function and I_n is a modified Bessel function of order n (Abramowitz & Stegun, 1965).

† The function $f(z) = \frac{3}{2}(1 - z^2)$ is the shape function defined in TL.

The formula for the absorption factor is then in general written

$$A(\mu R, \theta_{oh}) = (2/\pi\sin 2\theta_{oh}) \times \int_0^{2\theta_{oh}}\mathrm{d}y\int_0^{\pi-2\theta_{oh}}\mathrm{d}x\,h(\mu R, \theta_{oh}, x, y), \quad (5)$$

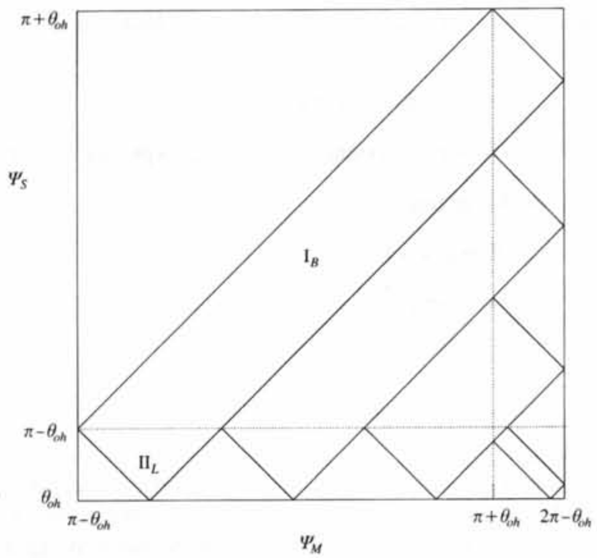

Fig. 2. Contributing fields to zeroth order. An example for $\theta_{oh} \geq 45°$ where the region I_L does not exist on the exit surface. Here $s = 6$ or $75 \leq \theta_{oh} \leq 77.14°$.

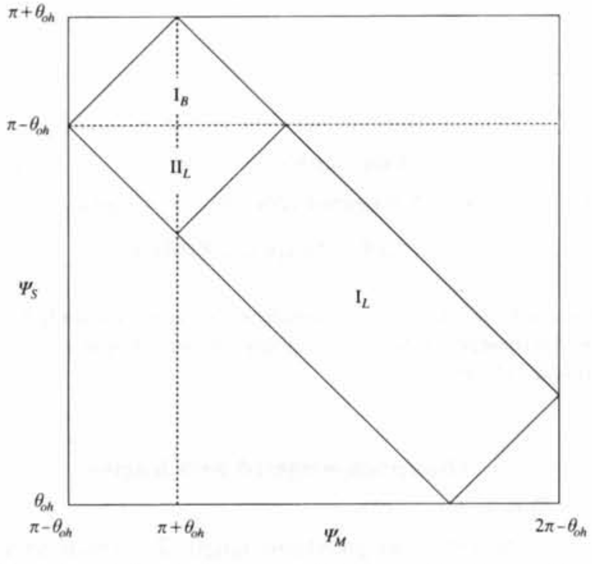

Fig. 1. Contributing fields to zeroth order. $s = 1$ or $0 \leq \theta_{oh} \leq 45°$.

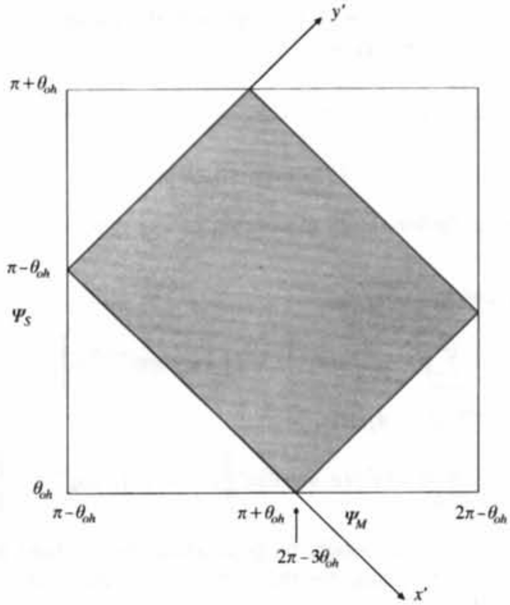

Fig. 3. Defining new coordinates x' and y' to simplify the integrations. The shaded area shows the actual region of integration.

Table 1. *Surface integrations, limits in ψ_M and ψ_S for calculating the absorption factor*

θ_{oh}	ψ_M	ψ_S
$(0, \pi/4)$	$(\pi - \theta_{oh}, \pi + \theta_{oh})$	$(-\psi_M + 2\pi - 2\theta_{oh}, \psi_M)$
	$(\pi + \theta_{oh}, 2\pi - 3\theta_{oh})$	$(-\psi_M + 2\pi - 2\theta_{oh}, -\psi_M + 2\pi + 2\theta_{oh})$
	$(2\pi - 3\theta_{oh}, 2\pi - \theta_{oh})$	$(\psi_M - 2\pi + 4\theta_{oh}, -\psi_M + 2\pi + 2\theta_{oh})$
$(\pi/4, \pi/2)$	$(\pi - \theta_{oh}, 2\pi - 3\theta_{oh})$	$(-\psi_M + 2\pi - 2\theta_{oh}, \psi_M)$
	$(2\pi - 3\theta_{oh}, \pi + \theta_{oh})$	$(\psi_M - 2\pi + 4\theta_{oh}, \psi_M)$
	$(\pi + \theta_{oh}, 2\pi - \theta_{oh})$	$(\psi_M - 2\pi + 4\theta_{oh}, -\psi_M + 2\pi + 2\theta_{oh})$

where the function h in the case of a cylinder is defined by

$$h = h_c(\mu R, \theta_{oh}, x, y) \stackrel{\text{def}}{=} \sin(x+y)\sin(x-y+2\theta_{oh})\exp(-a); \tag{6}$$

and for the sphere:

$$h = h_s(\mu R, \theta_{oh}, x, y) \stackrel{\text{def}}{=} \sin(x+y)\sin(x-y+2\theta_{oh})\{{}_1F_2[2, (\tfrac{1}{2}, \tfrac{5}{2}), a^2/4] - (9\pi/4a)I_2(a) - (3\pi/4)I_3(a)\}. \tag{7}$$

2.2. *Absorption in the limits $\theta_{oh}\to 0$ and $\theta_{oh}\to\pi/2$*

When $\theta_{oh} \to 0$, we have $\psi_M + \psi_S = 2\pi$. A close examination of the three terms in the surface integrals in Table 1 reveals that it is only the second term that contributes to the absorption factor in this limit. We find†

$$A_c(\theta_{oh} = 0) = (2/\pi)\int_0^\pi d\psi_M \sin^2\psi_M \exp(-2\mu R \sin\psi_M). \tag{8}$$

In the limit $\theta_{oh} \to \pi/2$, it is still only the second term that contributes. We have:

$$A_c(\theta_{oh} = \pi/2) = (2/\pi)\int_0^\pi d\psi_M \sin^2\psi_M\{(1/4\mu R\sin\psi_M) \times [1 - \exp(-4\mu R\sin\psi_M)]\}. \tag{9}$$

Using a Taylor-series expansion of the exponentials, we get:

$$A_c^{\theta_{oh}=0^\circ}(b = 2\mu R) = \sum_{n=0}^{\infty}[(-1)^n/n!]b^n\left\{(2/\pi)\int_0^\pi dy\ \sin^{(n+2)}y\right\}$$

$$A_c^{\theta_{oh}=90^\circ}(b = 4\mu R) = \sum_{n=0}^{\infty}[(-1)^n/(n+1)!]b^n\left\{(2/\pi)\int_0^\pi dy\ \sin^{(n+2)}y\right\}.$$

These results can easily be generalized to a sphere using the same procedure for the z integration that led to (4).

† A_c is here used for the absorption factor for the cylinder, A_s for the sphere.

We find:

$$A_s^{\theta_{oh}=0^\circ}(b = 2\mu R) = \sum_{n=0}^{\infty}[(-1)^n/n!]b^n\left\{(3/\pi)\left(\int_0^{\pi/2} dx \sin^{(n+3)}x\right) \times \left(\int_0^\pi dy \sin^{(n+2)}y\right)\right\}$$

$$A_s^{\theta_{oh}=90^\circ}(b = 4\mu R) = \sum_{n=0}^{\infty}[(-1)^n/(n+1)!]b^n\left\{(3/\pi)\left(\int_0^{\pi/2} dx \sin^{(n+3)}x\right) \times \left(\int_0^\pi dy \sin^{(n+2)}y\right)\right\}.$$

Using *Mathematica* (Wolfram, 1991), we then obtain closed expressions for the absorption factors in the two limiting cases:

$$A_c^{\theta_{oh}=0^\circ}(\mu R) = 2I_2(2\mu R) + [I_1(2\mu R)/\mu R] - (16\mu R/3\pi) \times {}_1F_2\ [2, (\tfrac{3}{2}, \tfrac{5}{2}), (\mu R)^2] \tag{10}$$

$$A_c^{\theta_{oh}=90^\circ}(\mu R) = [I_1(4\mu R)/2\mu R] - (16\mu R/3\pi) \times {}_1F_2[1, (\tfrac{3}{2}, \tfrac{5}{2}), 4(\mu R)^2] \tag{11}$$

while

$$A_s^{\theta_{oh}=0^\circ}(\mu R) = [3/4(\mu R)^3]\{1 - [1 + 2\mu R + 2(\mu R)^2] \times \exp(-2\mu R)\} \tag{12}$$

$$A_s^{\theta_{oh}=90^\circ}(\mu R) = [3/64\exp(4\mu R)(\mu R)^3]\{1 - \exp(4\mu R) + 4\mu R + 8\exp(4\mu R)(\mu R)^2\}. \tag{13}$$

The last two results are identical to those presented in *International Tables for Crystallography*, Volume C, by Maslen (1995).

3. Absorption-weighted path lengths

3.1. *Results for a cylinder*

The absorption-weighted path length, $\overline{T}_\mu$, which enters into some analysis of extinction, is defined as follows

(Zachariasen, 1968; Becker & Coppens, 1974):

$$\overline{T}_\mu = -(1/A)(\partial A/\partial \mu). \tag{14}$$

Using the definition of h_c, (6), it follows that

$$\frac{\overline{T}_\mu}{R} = \frac{1}{\mu R}\frac{\int_0^{2\theta_{oh}} \mathrm{d}y \int_0^{\pi-2\theta_{oh}} \mathrm{d}x\, \tilde{h}_c(\mu R, \theta_{oh}, x, y)}{\int_0^{2\theta_{oh}} \mathrm{d}y \int_0^{\pi-2\theta_{oh}} \mathrm{d}x\, h_c(\mu R, \theta_{oh}, x, y)} \tag{15}$$

with

$$\tilde{h}_c \stackrel{\text{def}}{=} \sin(x+y)\sin(x-y+2\theta_{oh})\, a\exp(-a). \tag{16}$$

In the limiting cases, we have:

$\theta_{oh} = 0°, b = 2\mu R$:

$$\begin{aligned}\frac{\overline{T}_\mu}{R} &= 2\frac{\sum_{n=0}^{\infty}[(-1)^n/n!]b^n\{\int_0^\pi \mathrm{d}y \sin^{(3+n)} y\}}{\sum_{n=0}^{\infty}[(-1)^n/n!]b^n\{\int_0^\pi \mathrm{d}y \sin^{(2+n)} y\}}\\ &= 2\frac{-9\pi I_2(b) - 3\pi b I_3(b) + 4b\, {}_1F_2[2, (\frac{1}{2}, \frac{5}{2}), b^2/4]}{3\pi I_1(b) + 3\pi b I_2(b) - 4b^2\, {}_1F_2[2, (\frac{3}{2}, \frac{5}{2}), b^2/4]}.\end{aligned} \tag{17}$$

$\theta_{oh} = 90°, b = 4\mu R$:

$$\begin{aligned}\frac{\overline{T}_\mu}{R} &= 4\frac{\sum_{n=0}^{\infty}[(-1)^n/(n+2)n!]b^n\{\int_0^\pi \mathrm{d}y \sin^{(3+n)} y\}}{\sum_{n=0}^{\infty}[(-1)^n/(n+1)!]b^n\{\int_0^\pi \mathrm{d}y \sin^{(2+n)} y\}}\\ &= 4\frac{-3\pi I_2(b) + 2b\, {}_1F_2[1, (\frac{1}{2}, \frac{5}{2}), b^2/4]}{3\pi I_1(b) - 2b^2\, {}_1F_2[1, (\frac{3}{2}, \frac{5}{2}), b^2/4]}.\end{aligned} \tag{18}$$

3.2. *Results for a sphere*

The transition to a sphere will now involve the z integration:

$$\begin{aligned}&\int_0^1 \mathrm{d}z(1-z^2)^{3/2}\exp[-a(1-z^2)^{1/2}]\\ &= \int_0^{\pi/2} \mathrm{d}\varphi \sin^4\varphi \exp(-a\sin\varphi)\\ &= (1/a)\{-(8a^2/15)\,{}_1F_2[3, (\tfrac{3}{2}, \tfrac{7}{2}), a^2/4]\\ &\quad + (3\pi/2a)I_2(a) + 3\pi I_3(a) + (\pi a/2)I_4(a)\}.\end{aligned} \tag{19}$$

We may then formally write the weighted path length as in (15):

$$\frac{\overline{T}_\mu}{R} = \frac{1}{\mu R}\frac{\int_0^{2\theta_{oh}} \mathrm{d}y \int_0^{\pi-2\theta_{oh}} \mathrm{d}x\, \tilde{h}_s(\mu R, \theta_{oh}, x, y)}{\int_0^{2\theta_{oh}} \mathrm{d}y \int_0^{\pi-2\theta_{oh}} \mathrm{d}x\, h_s(\mu R, \theta_{oh}, x, y)} \tag{20}$$

with the function $\tilde{h}_s$ defined by

$$\begin{aligned}\tilde{h}_s &\stackrel{\text{def}}{=} \sin(x+y)\sin(x-y+2\theta_{oh})\\ &\quad\times\{-\tfrac{4}{5}a^2\,{}_1F_2[3, (\tfrac{3}{2}, \tfrac{7}{2}), a^2/4] + (9\pi/4)(1/a)I_2(a)\\ &\quad + (9\pi/2)I_3(a) + (3\pi/4)aI_4(a)\}.\end{aligned} \tag{21}$$

In the limiting cases, we now obtain:

$\theta_{oh} = 0°, b = 2\mu R$:

$$\begin{aligned}\overline{T}_\mu/R &= 2\left\{\sum_{n=0}^{\infty}[(-1)^n/n!]b^n\left[\int_0^{\pi/2} \mathrm{d}y \sin^{(4+n)} y\right]\right.\\ &\quad\times\left.\left[\int_0^{\pi} \mathrm{d}y \sin^{(3+n)} y\right]\right\}\left\{\sum_{n=0}^{\infty}[(-1)^n/n!]b^n\right.\\ &\quad\times\left.\left[\int_0^{\pi/2} \mathrm{d}y \sin^{(3+n)} y\right]\left[\int_0^{\pi} \mathrm{d}y \sin^{(2+n)} y\right]\right\}^{-1}\\ &= \frac{6}{b}\frac{1-(1+b+\frac{1}{2}b^2+\frac{1}{6}b^3)\exp(-b)}{1-(1+b+\frac{1}{2}b^2)\exp(-b)}.\end{aligned} \tag{22}$$

$\theta_{oh} = 90°, b = 4\mu R$:

$$\begin{aligned}\overline{T}_\mu/R &= 4\left\{\sum_{n=0}^{\infty}[(-1)^n/(n+2)n!]b^n\left[\int_0^{\pi/2} \mathrm{d}y \sin^{(4+n)} y\right]\right.\\ &\quad\times\left.\left[\int_0^{\pi} \mathrm{d}y \sin^{(3+n)} y\right]\right\}\left\{\sum_{n=0}^{\infty}[(-1)^n/(n+1)!]b^n\right.\\ &\quad\times\left.\left[\int_0^{\pi/2} \mathrm{d}y \sin^{(3+n)} y\right]\left[\int_0^{\pi} \mathrm{d}y \sin^{(2+n)} y\right]\right\}^{-1}\\ &= \frac{b}{2}\frac{{}_2F_2[(2,4),(3,5),-b]}{\frac{1}{2}-(1/b^2)[1-(1+b)\exp(-b)]}.\end{aligned} \tag{23}$$

The closed expressions have been obtained using *Maple* (Char *et al.*, 1991*a*,*b*) and are equivalent to those found by Rigoult & Guidi-Morosini (1980).

4. Conclusions

Analytical formulas for the normal absorption factors and weighted path lengths in perfect crystals in the shape of a cylinder and a sphere have been developed based on the concept of a generalized extinction factor.

By applying the formulas for the absorption factors, we have calculated numerical values for the absorption correction A^*. The tabulated values given by Maslen (1995, Tables 6.3.3.2 and 6.3.3.3) are all within ±0.03% of our results. We find larger discrepancies when it comes to weighted path lengths for a sphere, *cf.* Table 6.3.3.4 given by Maslen. For most of the tabulated values, the deviations are within ±0.001. However, for $\mu R = 2.5$, we find deviations within the range (−0.009, 0.018). Our results for the weighted path lengths in a cylinder ($\mu R = 1$) are in perfect agreement with those given in Table 3 of Clark & Reid (1995).

References

Abramowitz, M. & Stegun, I. (1965). *Handbook of Mathematical Functions.* New York: Dover.

Becker, P. & Coppens, P. (1974). *Acta Cryst.* A**30**, 129–147.

Char, B. W., Geddes, K. O., Gonnet, G. H., Leong, B. L., Monagan, M. B. & Watt, S. M. (1991*a*). *Maple V Language Reference Manual.* New York: Springer-Verlag.

Char, B. W., Geddes, K. O., Gonnet, G. H., Leong, B. L., Monagan, M. B. & Watt, S. M. (1991*b*). *Maple V Library Reference Manual.* New York: Springer-Verlag.

Clark, R. C. & Reid, J. S. (1995). *Acta Cryst.* A**51**, 887–897.

Dwiggens, C. W. (1972). *Acta Cryst.* A**28**, 219–220.

Dwiggens, C. W. (1975*a*). *Acta Cryst.* A**31**, 146–148.

Dwiggens, C. W. (1975*b*). *Acta Cryst.* A**31**, 395–396.

Flack, H. D. & Vincent, M. G. (1978). *Acta Cryst.* A**34**, 489–491.

Howells, R. G. (1950). *Acta Cryst.* **3**, 366–369.

Maslen, E. N. (1995). *International Tables for Crystallography,* Vol. C, edited by A. J. C. Wilson, pp. 520–529. Dordrecht: Kluwer Academic Publishers.

Meulenaer, J. de & Tompa, H. (1965). *Acta Cryst.* **19**, 1014–1018.

Rigoult, J. & Guidi-Morosini, C. (1980). *Acta Cryst.* A**36**, 149–151.

Rouse, K. D., Cooper, M. J., York, E. J. & Chakera, A. (1970). *Acta Cryst.* A**26**, 682–691.

Thorkildsen, G. & Larsen, H. B. (1998). *Acta Cryst.* A**54**, 172–185.

Weber, K. (1969). *Acta Cryst.* B**25**, 1174–1178.

Wolfram, S. (1991). *Mathematica – A System for Doing Mathematics by Computer.* New York: Addison-Wesley.

Zachariasen, W. H. (1968). *Acta Cryst.* A**24**, 421–424.