because of the short wavelength. This fact at present
limits the amount of detail available in the TDS
profiles, and direct extraction of the elastic constants
is not possible. The vertical resolution can be
improved by the incorporation of horizontal Soller
slits, such that the observed TDS profiles would fall
off more quickly with \( q \) in both the transverse and
longitudinal directions. If the counting-rate reduction
were not too severe, this improvement would provide
greater detail within the limits of the one-phonon
model.

In summary, the analysis of the resolution function
for Mössbauer \( \gamma \)-ray scattering presented here has
led to a good understanding of the strengths and
limitations of this experimental probe. This better
understanding is crucial to the interpretation of TDS
measurements and of direct measurements of inelastic
and quasi-elastic scattering which are planned for the
QUEGS instrument.

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function.

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On the Fast Rotation Function

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Abstract

An analysis of the mathematical structure of the rotation
function is presented. The effect of truncation of
the expansions used in the fast rotation function is
discussed and an alternative procedure of calculation
which drastically reduces the errors is proposed. A
method of sampling on spherical surfaces is
developed. The rotation function can thus be obtained
from the values it takes at the sampling points. The
method can also be used to compute expansions in
spherical harmonics of Patterson functions restricted
to arbitrary domains. Topological properties of the
rotation group are used to obtain distortion-free plots
of the different sections of the rotation function.

Introduction

In Crowther's formulation of the fast rotation function,
emphasis is placed on the expansion of the
Patterson functions in terms of the spherical har-
monics and the spherical Bessel functions (Crowther,
1972). These expansions lead to slowly convergent
series and the relative errors of some contributions
can exceed 100% for reflections corresponding to
certain resolutions, under the conditions of application of the current version of the program. It will be shown that these errors can be drastically reduced if the expansion in radial functions is replaced by a numerical integration rule.

Patterson functions restricted to non-spherical domains will be discussed. In order to take full advantage of the properties of the rotation group these Patterson functions have to be expanded in spherical harmonics, but the coefficients of the expansions have now to be evaluated by numerical integration over the spherical surface. A method of sampling for functions defined on the spherical surface will thus be proposed.

An extension of this method will allow us to recover the rotation function from the values it takes on a set of sampling points.

Finally it will be shown that some topological properties of the rotation group lead, in a unique way, to the metric introduced by Burdina (1971) and Lattman (1972) in order to produce distortion-free plots of the rotation function.

The rotation function

The rotation function is defined as the integral over a spherical region 12 of the product of a given Patterson function \( \mathbb{P}(\mathbf{r}) \) with the rotated version of either itself or another Patterson function \( \mathbb{P}(\mathbf{r}) \) (Rossman & Blow, 1962).

For any rotation \( \Phi \), the rotated version of \( \mathbb{P}(\mathbf{r}) \) is the function \( T(\Phi)\mathbb{P}(\mathbf{r}) \), defined as

\[
[T(\Phi)\mathbb{P}(\mathbf{r})](\mathbf{r}) = \mathbb{P}(\mathbf{r})[R^{-1}(\Phi)\mathbf{r}],
\]

(1)

\( R(\Phi) \) being the operator associated with \( \Phi \) in a three-dimensional representation of the rotation group.

With this definition the rotation function \( \mathbb{R} \) is a functional of \( \mathbb{P}(\mathbf{r}) \) and \( \mathbb{P}(\mathbf{r}) \),

\[
\mathbb{R}(\Phi; \mathbb{P}, \mathbb{P}) = 1/v \int \mathbb{P}(\mathbf{r})[T(\Phi)\mathbb{P}(\mathbf{r})](\mathbf{r}) \, d^3 \mathbf{r},
\]

(2)

which depends on \( \Phi \) as a parameter (\( v \) is the volume of the spherical region \( \Omega \)). It can also be considered as a particular matrix element of the operator \( T(\Phi) \),

\[
\mathbb{R}(\Phi; \mathbb{P}, \mathbb{P}) = \langle \mathbb{P}|T(\Phi)\mathbb{P} \rangle.
\]

(3)

It is then natural to expand the functions \( \mathbb{P}(\mathbf{r}) (t = 1, 2) \) in terms of the spherical harmonics \( Y_{lm} \), since they are the standard basis of the irreducible representations of the rotation group (Crowther, 1972). In this basis the matrix representing the operator \( T(\Phi) \) is block diagonal, the blocks being irreducible ones.

Besides, as \( T(\Phi) \) is already diagonal in the radial components of \( \mathbb{P}(\mathbf{r}) \), it is not in principle necessary to expand them in any particular set of radial functions.

The Patterson functions are represented by truncated Fourier series

\[
\mathbb{P}(\mathbf{r}) = \sum_{H \in \mathcal{H}(t)} |F_H(t)|^2 \exp(2\pi i \mathbf{H} \cdot \mathbf{r}),
\]

(4)

\( \mathcal{H}(t) \) being the set of the experimentally available reciprocal vectors of crystal \( t \). Their expansions in spherical harmonics are of the form

\[
\sum_{l=0}^{\infty} \sum_{m=-l}^{l} c_{lm}(r) Y_{lm}(\hat{\mathbf{r}}),
\]

(5)

where \( r = |\mathbf{r}| \) and \( \hat{\mathbf{r}} = \mathbf{r}/r \).

The radial functions \( c_{lm}(r) \) are determined by the formula

\[
c_{lm}(r) = \int_{\sigma_r} \mathbb{P}(\mathbf{r}) Y_{lm}^*(\hat{\mathbf{r}}) \, d\hat{\mathbf{r}},
\]

(6)

where \( \sigma_r \) is the spherical surface of radius \( r \).

Even if the \( \mathbb{P}(\mathbf{r}) \) are defined for all points in a three-dimensional space, we may be interested in the comparison of the Patterson functions restricted to particular regions \( \Omega(t) \). These regions are only used to obtain the radial functions \( c_{lm}(r) \), and need not be spherical ones.

For an arbitrary domain \( \Omega(t) \), the integral (6) extends over the intersection of the spherical surface \( \sigma_r \), with \( \Omega(t) \), and has to be evaluated by numerical methods to be discussed later. In this case the expansion (5) represents in fact a new function, say \( \hat{\mathbb{P}}(t) \), which coincides with \( \mathbb{P}(\mathbf{r}) \) in \( \Omega(t) \) and is zero outside it.

In order to take full advantage of the properties of the rotation group, it is however necessary that \( \Omega \), which enters the definition of the rotation function, is a spherical domain.

If one recalls the transformation properties of the spherical harmonics (Landau & Lifschitz, 1972)

\[
T(\Phi) Y_{lm} = \sum_{m'} D_{lm,m'}(\Phi) Y_{lm'},
\]

(7)

the rotated Patterson function is easily obtained:

\[
[T(\Phi)\mathbb{P}(\mathbf{r})](\mathbf{r}) = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} c_{lm}(r) Y_{lm}(\hat{\mathbf{r}}) D_{lm,m}^*(\Phi).
\]

(8)

\( D^*(\Phi) \) is the matrix associated with the rotation \( \Phi \) in the \( l \)th canonical irreducible representation of the rotation group.

The rotation function is then (Crowther, 1972)

\[
\langle \mathbb{P}|T(\Phi)\mathbb{P} \rangle = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} D_{lm,m}^*(\Phi) c_{lm}(P(1), P(2)),
\]

(9)

with

\[
c_{lm}(P(1), P(2)) = (1/v) \int_{\sigma_r} c_{lm}^*(r) c_{lm}(r) r^2 \, d\mathbf{r}
\]

(10)
Table 1. Maximum values of the arrays $T^l$ [equation (14)]

<table>
<thead>
<tr>
<th>$l$</th>
<th>$h$</th>
<th>$T^l(h, h) \times 10^{-4}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>4.034</td>
<td>248.64</td>
</tr>
<tr>
<td>4</td>
<td>6.606</td>
<td>92.07</td>
</tr>
<tr>
<td>6</td>
<td>9.008</td>
<td>47.57</td>
</tr>
<tr>
<td>8</td>
<td>11.329</td>
<td>28.86</td>
</tr>
<tr>
<td>10</td>
<td>13.601</td>
<td>19.28</td>
</tr>
<tr>
<td>12</td>
<td>15.839</td>
<td>13.73</td>
</tr>
<tr>
<td>14</td>
<td>18.053</td>
<td>10.24</td>
</tr>
<tr>
<td>16</td>
<td>20.248</td>
<td>7.91</td>
</tr>
<tr>
<td>18</td>
<td>22.427</td>
<td>6.28</td>
</tr>
<tr>
<td>20</td>
<td>24.594</td>
<td>5.10</td>
</tr>
<tr>
<td>22</td>
<td>26.751</td>
<td>4.21</td>
</tr>
<tr>
<td>24</td>
<td>28.898</td>
<td>3.53</td>
</tr>
<tr>
<td>26</td>
<td>31.038</td>
<td>3.00</td>
</tr>
</tbody>
</table>

($a$ is the radius of the region $\Omega$). We see that it can be interpreted as an expansion in terms of the complete orthogonal set of functions $D^l_{mm'}$ (Wigner, 1959).

If the $\Omega^{(t)}$ are spherical regions, the radial functions are readily obtained when the expansion (Landau & Lifschitz, 1972)

$$\exp(ipr) = 4\pi \sum_{l=0}^{\infty} \sum_{m=-l}^{l} i^l j_l(pr) Y^*_{lm}(\hat{\mathbf{r}}) Y_{lm}(\hat{\mathbf{r}})$$

($j_l$ is the spherical Bessel function of order $l$) is substituted into (4), to give

$$c^{(t)}_{lm}(r) = 4\pi r^l \sum_{H \in \mathcal{R}(t)} |F^{(t)}_H|^2 j_l(2\pi Hr) Y^*_{lm}(\hat{H})$$

A closed expression for the coefficients $C^{l'}_{mm'}$ can now be obtained. With the dimensionless quantities $h = 2\pi Ha$ and $k = 2\pi Ka$, the result is

$$C^{l'}_{mm'}(P^{(1)}, P^{(2)}) = 12\pi \sum_{H \in \mathcal{R}(1)} \sum_{K \in \mathcal{R}(2)} |F^{(1)}_H|^2 Y^*_{lm}(\hat{H}) \times T^l(h, k) |F^{(2)}_K|^2 Y^*_{lm}(\hat{K}),$$

where (Watson, 1958)

$$T^l(h, k) = \int_0^h (h x) j_l(k x) x^2 dx$$

$$= \begin{cases} [h j_{l-1}(h) j_l(k) - k j_{l-1}(k) j_l(h)]/((k^2 - h^2)), \\ 2j^2_l(h) - j_{l-1}(h) j_{l+1}(h), \quad \text{if } h \neq k. \end{cases}$$

The two-dimensional array $T^l$ takes its maximum value on the diagonal $h = k$. This value is listed as a function of $l$ in Table 1. The contour levels of the arrays $T^l$, normalized to their maximum values, are shown in Fig. 1.

On practical grounds it is, however, important that the summations on $H$ and $K$ be performed independently of each other, as was the case in the original equations [(10)--(12)]. This property can be restored in several ways.

One possibility is to expand the $c^{(t)}_{lm}$ in terms of functions $f_n$ ($n = 1, \ldots, N$) which are orthogonal in the interval $(0, a)$, with measure $r^2 \, dr$. Furthermore, we have the possibility of choosing different expansion functions for different $l$’s because they refer to distinct irreducible representations of the rotation group. For example, Crowther (1972) chooses Fourier-Bessel expansions based on the $j_l$’s. The $C^{l'}_{mm'}$ will be given by an expression of the form

$$C^{l'}_{mm'}(P^{(1)}, P^{(2)}) \equiv \sum_{n=1}^N a^{(1)}_{lm} a^{(2)}_{lm}.$$  

Another possibility is the evaluation of the integral in (10) by means of an approximation of the form

$$C^{l'}_{mm'}(P^{(1)}, P^{(2)}) \approx \sum_{n=1}^M c^{(1)}_{lm}(r_n) c^{(2)}_{lm}(r_n) w_n,$$

where $w_n$ is the weight associated with the sampling point $r_n$.

In both cases we are ultimately calculating $T^l$ in an approximate way. The confrontation with the exact result [(14)] will show the errors implied in any one of the approximations.

Despite the errors in the $C^{l'}_{mm'}$, which can sometimes be very important, and the subsequent errors in the rotation function [(9)], the results still have a full physical sense. The reason is that the effect of the approximation [(15) or (16)] on the rotation function is strictly equivalent to that of some particular projection operator which projects the Patterson functions on a subspace spanned by functions labeled
by the numbers \((l, m, n)\). The important point is that the projection is correctly calculated.

Qualitatively the situation is the same as if we computed an \(R\) factor using less reflections than available.

This suggests that a selection of the \(l\)'s entering into (9) can be made on physical grounds. For example, the term \(l=0\) may be omitted not only because it simply adds a constant contribution to the rotation function, but also because it represents the contribution of the origin peaks of \(P^{(1)}\) and \(P^{(2)}\). In fact, according to (6), \(c^{(1)}(r)\) is the spherical average of \(P^{(1)}\) at distance \(r\), so that the origin peak, which is normally spherical in shape, will be correctly described by this radial function.

Assume now that a certain Patterson function is roughly ellipsoidal in shape. Then, if one recalls that the \(Y_{lm}\) are intimately related to the homogeneous polynomials of degree \(l\) in \(x, y, z\) (Smirnov, 1972), the terms with \(l=2\) will dominate the expansion of that Patterson function in spherical harmonics. If we are looking for a finer structure, such terms should be omitted from (9).

Inspection of Fig. 1 shows that the omission of small \(l\)'s is somewhat equivalent to omitting low-resolution data in the computation of the Patterson functions, except that now high angular resolution is obtained without omitting any data.

Approximate evaluation of \(T^f\)

(a) Fourier–Bessel expansion

We shall discuss exclusively the case (essentially the formulation of Crowther) in which \(j_l(hx)\) is approximated by the truncated expansion

\[
j_l(hx) \approx \sum_{n=1}^{N} b_{ln}(h) j_l(\lambda_{ln}x),
\]

(17)

The \(\lambda_{ln}\) are such that \(j_l(\lambda_{ln})=0\), i.e. they are the zeros of \(j_l\). The coefficients \(b_{ln}\) are (Watson, 1958)

\[
b_{ln}(h) = 2 \int_{0}^{1} j_l(hx) j_l(kx)x^2 \, dx / j_l(\lambda_{ln})^2
\]

\[= 2j_l(h) \lambda_{ln} / \left[j_{l-1}(\lambda_{ln})(h^2 - \lambda_{ln}^2]\right], \quad h \neq \lambda_{ln},
\]

(18)

and, of course, \(b_{ln}(\lambda_{ln}) = 1\). The array \(T^f\) is now approximated by

\[
T^f(h, k) \approx T^f_N(h, k)
\]

\[= \sum_{n=1}^{N} b_{ln}(h) b_{ln}(k) j_{l-1}(\lambda_{ln}) / 2
\]

\[= 2j_l(h) j_l(k) \sum_{n=1}^{N} \lambda_{ln}^2 / \left[(h^2 - \lambda_{ln}^2)(k^2 - \lambda_{ln}^2]\right].
\]

(19)

This expansion has the advantage of giving the exact result whenever \(h\) or \(k\) coincides with one of the zeros of \(j_l\), provided it was included in the summation.

When \(N\) tends to infinity this series converges because the large zeros of the Bessel functions behave as \(\lambda_{ln} = (n + l/2)\pi\). For \(h\) and \(k \ll \lambda_{ln}\) the error is of the order of

\[
T^f(h, k) - T^f_N(h, k) \approx 2j_l(h) j_l(k)(\pi \lambda_{ln})^{-1}.
\]

(20)

For particular values of \(h\) and \(k\) we have exact summation rules which can be used to assess the error introduced by the truncation. In particular, if \(h\) and \(k\) are equal to two different zeros of \(j_{l-1}\), say \(h = \lambda_{l-1,p}\) and \(k = \lambda_{l-1,q}\), then \(T^f\) vanishes because of (14) and

\[
\sum_{n=1}^{\infty} \lambda_{ln}^2 / \left[(\lambda_{l-1,p}^2 - \lambda_{ln}^2)(\lambda_{l-1,q}^2 - \lambda_{ln}^2]\right] = 0.
\]

(21)

Similarly, if \(h\) and \(k\) are both equal to a root of \(j_{l-1}\) or \(j_{l+1}\), say \(h = k = \lambda_{l+1,p}\), then

\[
\sum_{n=1}^{\infty} \lambda_{ln}^2 / \left[(\lambda_{l+1,p}^2 - \lambda_{ln}^2)^2\right] = 1.
\]

(22)

In practical applications the value \(N\) at which the summations are truncated depends on \(l\) and on the maximum value \(h_{max}\) taken by \(h\) or \(k\). For example, the terms included in the fast rotation function are such that \(\lambda_{ln} \ll h_{max}\) (Crowther, 1972; Dodson, 1985). The relative errors \(|1 - T^f_N / T^f|\) introduced in this case are shown in Fig. 2. Similar calculations, but including

Fig. 2. Percentage errors of \(T^f(h, k)\) when evaluated under the conditions of the fast rotation function (Crowther, 1972), for \(h_{max} = 20\) and \(l = 2, \ldots, 12\). The contour levels are 20 and 100%.
4 and 20 supplementary roots for each \( l \), are shown in Figs. 3 and 4 respectively.

(b) Numerical integration

Here again we have the possibility of choosing a special integration formula for each \( l \) and \( h_{\text{max}} \). Since the purpose of this presentation is not to find the best quadrature rule but rather to show its usefulness, we chose a unique \( M \)-point Legendre-Gauss integration formula

\[
T^l(h, k) = T^l_M(h, k) = \sum_{n=1}^{M} j_n(hx_n)j_l(kx_n)x_n^2 w_n. \tag{23}
\]

The sampling points \( x_n \) are the \( M \) positive roots of the Legendre polynomial of degree \( 2M \). These values as well as their associated weights \( w_n \) have been tabulated by several authors (e.g. Abramowitz & Stegun, 1964). For a given accuracy, the number \( M \) of sampling points is certainly related to the number of oscillations of the integrand, which in turn depends on the order \( l \) of the Bessel function and \( h_{\text{max}} \). The relative errors of \( T^l \) for a 12-point integration formula are shown in Fig. 5. They clearly show the improvement in accuracy that can be expected when using numerical integration techniques.

Sampling and numerical integration on spherical surfaces

We have seen that the coefficients \( C^l_{mn} \) can be calculated from the values of the radial functions \( c^{(l)}_{im} \) taken at selected radial points. When the domain
of integration is the whole sphere, these values are
given by analytical expressions \([12]\), but for
arbitrary regions \(I_2(\cdot)\) and selected radial points \(r_\nu\),
the quantities \(c_\nu(r_\nu)\) have in general to be evaluat6d
by numerical methods of integration.

In general terms, our problem is to find points \(\hat{\varphi}\)
on the surface of the unit sphere \(\sigma\), and corresponding
weights \(W_s\), such that
\[
\int f(\hat{\varphi}) Y_{lm}(\hat{\varphi}) \, d\sigma = \sum_{s=1}^{S} f(\hat{\varphi}_s) Y_{lm}(\hat{\varphi}_s) \, W_s \quad (24)
\]
is exact for a sufficiently large class of functions.

The answer to this problem will depend on the
prior knowledge we may have about the function \(f\).
We shall assume that \(f\) can be faithfully represented
by a limited expansion in spherical harmonics,
\[
f(\hat{\varphi}) = \sum_{l=0}^{L} \sum_{m=-l}^{l} A_{lm} Y_{lm}(\hat{\varphi}). \quad (25)
\]
Functions of this type (i.e. with \(A_{lm} = 0\) for \(l > L\)) will
be called belonging to class \(\mathcal{C}_L\).

Since the \(Y_{lm}\) are orthogonal on the spherical surface
and the product of two functions of classes \(\mathcal{C}_L\) and \(\mathcal{C}_L'\) is a function of class \(\mathcal{C}_{L+L'}\), the coefficients
\(A_{lm}(l \leq L)\) will be given by (24) if the approximate
integration formula gives the correct result whenever
the integrand is a \(Y_{lm}\) of order at most \(2L\), that is
\[
\int \sum_{s=1}^{S} Y_{lm}(\hat{\varphi}_s) \, W_s = (4\pi)^{1/2} \delta_{l0}, \quad l \leq 2L.
\]
(26)

One solution to this problem can be obtained by
slightly modifying a result of Peirce (1957), concerning
the integration of polynomials over the spherical
shell.

The result is the following: (26) is satisfied if the
points \(\hat{\varphi}_s = (\theta_\nu, \varphi_\nu)\) are taken at all the intersections of the
cones \(\theta_\nu\) \((i = 1, \ldots, L + 1)\), with the half planes \(\varphi_\nu\)
\((k = 1, \ldots, 2L + 1)\). The corresponding weights \(W_s\) are of
the product form \(W_s = A_\nu B_\nu\), where \(A_\nu\) are the weights
of the \((L + 1)\)-point Legendre-Gauss quadrature
formula and the \(B_\nu\) are equal to \(2\pi/2L + 1\).

In fact, \(Y_{lm}\) depends on \(\varphi\) through the exponential
term \(\exp\left(i m\varphi\right)\), so that its integral is exactly calculated
if the function is sampled according to Shannon's theorem of sampling. What survives this first
integration is a polynomial of degree \(l\) in \(\cos \theta\), whose
integral is correctly given by a Gaussian quadrature
formula.

Substituting for \(A_{lm}\) from (24) into (25) and using
the addition theorem for spherical harmonics and the recurrence relationship of the Legendre polynomials
\(P_l\), we obtain
\[
f(\hat{\varphi}) = \sum_{s=1}^{S} f(\hat{\varphi}_s) \sum_{l=0}^{L} \sum_{m=-l}^{l} Y_{lm}(\hat{\varphi}_s) Y_{lm}(\hat{\varphi}) \, W_s
\]
\[
= \sum_{s=1}^{S} f(\hat{\varphi}_s) \sum_{l=0}^{L} \left[(2l+1)/4\pi\right] P_l(\hat{\varphi}_s, \hat{\varphi}) \, W_s
\]
\[
= \sum_{s=1}^{S} f(\hat{\varphi}_s) \left[P_{l+1}(\hat{\varphi}_s, \hat{\varphi}) - P_l(\hat{\varphi}_s, \hat{\varphi})/(\hat{\varphi}_s, \hat{\varphi} - 1)\right]
\]
\[
\times W_s(L + 1)/4\pi. \quad (27)
\]

We see that \(f\) can be recovered from its values
taken on the set \(\{\hat{\varphi}_s, s = 1, \ldots, S\}\). Other values of \(f\)
taken on another set of points may also determine
the function \(f\), but it may not be possible or practical
to reconstruct \(f\) starting from these values. As in
Dodson & Silva (1985), a set of points which determines
\(f\) and allows a reconstruction of \(f\) in terms of
its values on the set will be called a sampling set of
\(f\). Then the abscissas of an approximate integration
formula which gives the exact values for all spherical harmonics of order at most \(2L\) are a sampling set of
functions of class \(\mathcal{C}_L\).

It must be noted that the number of independent complex coefficients for a function of class \(\mathcal{C}_L\) is
\((L + 1)^2\), whereas the number of integration points is
\((2L + 1)(L + 1)\). A sampling set for which the number
of independent coefficients and that of samples of
the function are equal will be called an optimal
sampling set.

If the set \(\{\hat{\varphi}_s, q = 1, \ldots, Q\}\) is an optimal one, the
\(f(\hat{\varphi}_q)\) are all independent values and (27) gives
\[
\sum_{q=1}^{Q} (2l+1) P_l(\hat{\varphi}_q, \hat{\varphi}_q) = 4\pi/ W_{q', q}. \quad (28)
\]

The first result is that the weights for this sampling,
if it exists, are all equal to \(4\pi/(Q + 1)^2\). A second
result is that the angle \(\theta_{q', q}\) between any two different
sampling points should satisfy the equation
\[
\sum_{q=0}^{Q} (2l+1) P_l(\cos(\theta_{q, q}) = 0, \quad q' \neq q. \quad (29)
\]
For a given \(Q\), the roots of (29) give the cosines of
all the possible angles. For \(Q = 0\) there is no root so
that the optimal sampling set is constituted by a single
point. For \(Q = 1\) there is a single root, \(\cos(\theta_{q, q}) = -\frac{1}{2}\),
which gives rise to a tetrahedron. For \(Q = 2\), we have
two possible angles, 73.15 and 133.62°, which do not
give any closed figure inscribed on the sphere.

Hence, any rule of approximate integration [(26)]
must in general involve more than the theoretical
minimum number of points. This assertion does not
mean that there is no optimal sampling set for functions of class \(\mathcal{C}_L\), but rather that a sampling set
obtained from an integration rule is not in general
an optimal one.
The results of this paragraph can be applied to the rotation function \([9]\) which, in all practical applications, is also a limited expansion in terms of the orthogonal functions \(D^l_{mm'}\).

\[
\mathcal{R}(\phi) = \sum_{l=0}^{L} \sum_{m,m'=-l}^{l} D^l_{mm'}(\phi) C^l_{mm'}.
\]  

Since \(D^l_{mm'}\), when written in terms of the Euler angles \((\alpha, \beta, \gamma)\), depends on \(\alpha\) and \(\gamma\) through the expression \(\exp[i(m\alpha + m'\gamma)]\), and since for \(m' = 0\) it is proportional to the spherical harmonic \(Y_{lm}\), a quadrature rule can be obtained:

\[
\begin{align*}
\int_0^{2\pi} \int_0^{\pi} \cos \beta \int_0^{2\pi} d\gamma \mathcal{R}_L(\alpha, \beta, \gamma) D^{*l}_{mm'}(\alpha, \beta, \gamma) \\
= \sum_{u=1}^{U} \mathcal{R}_L(\phi_u) D^{*l}_{mm'}(\phi_u) W_u \\
= C_{mm'} 8\pi^2/(2L+1).
\end{align*}
\]  

(31)

It follows that the set \(\phi_u = (\alpha_i, \beta_j, \gamma_k); \ i, k = 1, \ldots, 2L+1; \ j = 1, \ldots, L+1\) where \(\alpha_i\) and \(\gamma_k\) are regularly spaced in the interval \([0, 2\pi]\) and \(\cos \beta_j\) are the roots of \(P_{l+1}\), is a sampling set of \(\mathcal{R}_L\). The weights \(W_u\) are again of the product form, derived from the Legendre-Gauss and Fourier quadrature rules.

**Plotting the rotation function**

Several authors have discussed different ways of plotting the rotation function in order to obtain undistorted maps (Burdina, 1971; Lattman, 1972).

Distortion appears for example when \(\mathcal{R}\) is evaluated and plotted on a grid with equal steps in the Euler angles; if \(\beta = 0\) all rotations with a constant value of \(\alpha + \gamma\) are the same. This situation is somewhat analogous to that encountered when a function of the Cartesian variables \((x, y, z)\) is computed on a regularly spaced grid in the spherical coordinates \((r, \theta, \varphi)\): the value of the function is independent of \(\varphi\) when \(\theta = 0\) or \(\pi\).

This source of distortion can be avoided, to some extent, if we are able to define a metric, in order to give a sense to the notion of nearness of two elements of the group. This can be done because the rotation group is compact, so that a unique positive-definite invariant metric exists (up to a multiplicative constant) (Normand, 1971)

\[
ds^2 = \frac{1}{2} \text{Tr} (dR \, dR^*),
\]  

\[dR\] being the variation of the matrix \(R\) associated with a rotation in a three-dimensional representation of the rotation group.

This metric confers to the space of the parameters of the group the structure of a Riemannian variety, and cannot be reduced to a Cartesian metric.

Expression (32) is an intrinsic one, i.e. independent of the parametrization of the group. When parametrized in terms of the Euler angles, it gives

\[
ds^2 = d\alpha^2 + 2\cos(\beta) \, d\alpha \, dy + d\gamma^2 + d\beta^2,
\]  

(33)

which, when written in the new variables \(\theta_+ = \alpha \pm \gamma\), coincides with the expression given by Lattman [1972, equation (7)],

\[
ds^2 = \cos^2(\beta/2) \, d\theta_+^2 + \sin^2(\beta/2) \, d\theta_-^2 + d\beta^2.
\]  

(34)

This metric shows that the topology of the surfaces of constant \(\beta\) is that of a plane. In fact, the change of variables

\[
u = \sin(\beta/2) \theta_-
\]

(35)

gives the Euclidean metric \(ds^2 = du^2 + dv^2\) all over any section. Distortion-free \(\beta\) sections of the rotation function can thus be obtained.

Surfaces of constant \(\theta_+\) have instead the topology of a spherical surface. Therefore, to obtain distortion-free \(\theta_+\) sections the rotation function should be represented on the surface of a sphere, with \(\beta/2\) and \(2\theta_+\) playing the role of the colatitude and longitude respectively.

If the group is parametrized by \((\chi, \hat{n})\), \(\hat{n}\) being the unit vector along the axis of rotation and \(\chi\) the spin about it, the metric is

\[
ds^2 = d\chi^2 + 2(1 - \cos \chi) \, d\hat{n}^2
\]  

[\text{Lattman, 1972, equation (9)}].

The distance between any two rotations, defined as the integral of \(ds\) along a path joining them, is path dependent. In particular, since the parameters \((\pi, \hat{n})\) and \((-\pi, \hat{n})\) represent the same rotation, any path joining them is closed but it cannot be reduced, by means of a continuous deformation, to one of vanishingly small length. This implies that the rotation group is not simply connected.

The important point is that these topological properties are intrinsic ones, and cannot be removed by any particular choice of parametrization of the group.

**Some numerical results**

A program was written which samples the function \(c_m(r)\) \([12]\) at different points \(r_n\). The quantities \(w_n^{1/2} r_n c_m(r_n)\) can thus be used as an input to the Crowther program, instead of the \(a_{imn}\), and the rotation function can be calculated. For a given \(l\), the function \(j_l(hx) j_l(kx)\) vanishes at most twice in the interval between any two consecutive zeros of \(j_l\). A good estimate of its integral can thus be obtained by sampling it at three points within such an interval. Crowther's method should then be faster than any simple integration method by at most a factor of three.
Actually, this ratio was two for the example here reported.

Even if Figs. 1–5 give a clear idea of the errors involved in either method, we present here a test calculation using a model gramicidin A structure (space group $P2_12_12_1$, $a = 24.7$, $b = 32.3$, $c = 32.5\,\text{Å}$). Two identical model monomers consisting of a polypeptide helix with three tryptophans and tilted by arbitrary angles with respect to the $c$ axis constituted the asymmetric unit content. The resulting structure showed a slight overlap of some of the tryptophans belonging to different molecules.

The squared norms of each $l$ component of the expansion of the Patterson function in spherical harmonics,

$$
\sum_{m=-l}^{l} \sum_{n=1}^{N} |a_{l m n}|^2
$$

in Crowther’s method and

$$
\sum_{m=-l}^{l} \sum_{n=1}^{M} |c_{l m}(r_n)|^2 r_n^2 w_n
$$

in the method here presented, should be essentially the same when a great number of $n$ terms are included, so that the differences between them effectively show the relative errors involved in the procedures. With 2 Å data, an overall temperature $B$ factor of 6 Å$^2$ and an outer radius of 6.5 Å, the relative difference in the squared norms was 18, 7, 11, 28, 34, 62 and 36% for $l$ ranging from 2 to 14.

We then computed a cross rotation function using a single monomer to obtain the second Patterson function to be rotated. Both procedures of calculation showed essentially the same features (Figs. 6 and 7) even if the values differed by 6–15% at the local

![Fig. 6. Contour maps of a cross rotation function as produced by the program written by Crowther (1972) and modified by Dodson (1985). GP1 and GP2 denote the true positions. The contour levels are 60 and 80% of the maximum value of the function.](image)

![Fig. 7. Same as Fig. 6, but now the cross rotation function was computed using a 12-point Legendre-Gauss integration formula.](image)

![Fig. 8. Same as Fig. 7, but with the contributions of the terms with $l = 2, 4$ omitted from the calculation.](image)
maxima. This result is not surprising because low l terms contribute a substantial portion of the whole Patterson function (after omitting the $l=0$ term) under the conditions of the test. However, as discussed in the article, these very low- l terms can hinder the true solution from showing up. This is clearly shown in Fig. 8, where the terms $l=2,4$ were omitted from the calculations. It is worth noticing that the two omitted terms contributed 86% of the Patterson squared norm.

References


Measurability of Bijvoet Differences in Triclinic, Monoclinic and Orthorhombic Crystals. II

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Abstract

The probability that the Bijvoet ratio $X$ for the observed reflections of a given crystal is greater than any particular value $X_0$ depends on space-group symmetry, the number of anomalous scatterers per asymmetric unit and the parameters $k$ and $\sigma_2^2$. Numerical values for this probability are obtained as a function of $X_0$ for different values of $k$ and $\sigma_2^2$ for the triclinic, monoclinic and orthorhombic crystals containing $p$ ($=1,2,3$ or 4) anomalous scatterers per asymmetric unit. These results are provided in the form of compact tables; Fortran programs that are useful in computing this probability for any given situation are also provided.

1. Introduction

The success of the anomalous-scattering method of structure determination strongly depends on the measurability of Bijvoet differences. The measurability is defined as the probability of the event \{(X \geq 0.1) \cap (\sigma_{\min} \geq 0.3)\} (Velmurugan & Parthasarathy, 1984; VP, hereafter) where $X$ is the Bijvoet ratio and $\sigma_{\min}$ is the minimum value of the normalized structure-factor magnitudes for the reflection $H$ and the inverse reflection $\bar{H}$. Owing to the importance of the anomalous-scattering method, particularly due to the advent of synchrotron radiation as a source for diffraction studies, it would be useful to know a priori, in the case of a given crystal, the percentage of observed reflections for which the Bijvoet ratio $X$ would be greater than any specific value $X_0$, say. This information can be obtained from the probability value for the event \{(X \geq X_0) \cap (\sigma_{\min} \geq 0.3)\} (= D, say) and we shall denote this probability by $M(X_0,0.3)$. The values of $M(X_0,0.3)$ for the particular case of $X_0 = 0.1$ were obtained in VP for the triclinic, monoclinic and orthorhombic crystals containing one or two heavy atoms per asymmetric unit for values of $k$ (i.e. the ratio of the imaginary to the total real part of the atomic scattering factor of the anomalous scatterer) up to 0.6. Under a pronounced anomalous-scattering effect, $k$ can have larger values for some of the heavy atoms. For example, the values

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