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INTEGRATION OF SINGLE CRYSTAL NEUTRON DATA USING A P.S.D: A CASE OF LARGE, WEAK AND OFTEN OVERLAPPING REFLECTIONS

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Abstract. We have extended our data treatment series of programmes, used routinely for several years now for off-line integration of low-resolution neutron diffraction data, to more general conditions of use. The basic principle has not changed: it consists of the a priori calculation of ellipsoidal masks for every reflection. The resolution function algorithm is rapid, as most parameters can be calculated beforehand. The data treatment is on-line, the reflections appearing on the detector at any rotation angle are predicted progressively. Accurate background correction in the case of non-uniform and non-stationary background has been developed using linear two-dimensional spline function interpolation and moving average techniques.

I - INTRODUCTION:

The work presented hereafter is an extension and a modification of a system of data collection already presented in 1982 /1/, and used extensively since then for data collection in neutron diffraction experiments with a 2-dimensional Position Sensitive Detector. The experiments were carried out on the small angle scattering spectrometer D17 at the Institut Laue - Langevin and concerned protein crystallography at low resolution (i.e. for Bragg d spacings > 15 Å). The corresponding diffraction angle range is 2θ < 45 deg., using neutrons of about 10 Å wavelength.

A new instrument for low resolution crystallography, DB21, has recently been constructed at the ILL in cooperation with the EMBL, Grenoble Outstation, and a new system of data collection has been
developed for it. The work is not yet completed, as some parts have still to be tested and integrated into the system.

Both instruments have a flat square $2\times2$-dimensional PSD, $64\times64$ cm$^2$ on D17 with a $5\times5$ mm$^2$ resolution, $20\times20$ cm$^2$ on DB21 with a $1.5\times1.5$ mm$^2$ resolution. In both cases the size of a spectrum is $128\times128$ pixels. A typical size for a diffraction reflection observed on these detectors is $7\times7$ pixels.

II - METHOD OF INTEGRATION:

At low resolution it is possible to use a rather broad beam, with a large wavelength spread (monochromatisation by velocity selector on D17) or a large divergence (intercalated pyrolytic graphite monochromator on DB21). As a consequence (see Fig.1):

1) the omega range of diffraction (in normal beam geometry) for a given reflection is large (>2.0 deg) and variable from one reflection to another.
2) the angle of diffraction of a reflection (i.e. its position on the detector) changes markedly during the scan.

In addition, the reflections are often close to each other and even overlap, because of the large unit cells of the crystals and because the reflections remain excited over a large rotation angle range.

Given this rather complex situation, we have chosen the following method of integration:
- the intensity of a reflection in a given spectrum is integrated by adding the neutron counts inside an appropriate mask whose size and position are calculated a priori from the parameters of the beam and of the crystal.
- the background is estimated from the data outside the masks.

III - A PRIORI CALCULATION OF POSITION AND SIZE OF MASKS ON THE PSD.

A - CALCULATION OF THE DIFFRACTED INTENSITY DISTRIBUTION.

The following assumptions are made:

1) point diffraction geometry, i.e. sample smaller than the PSD spatial resolution.
2) the incident beam intensity distribution is a 3-dimensional Gaussian function with respect to the incident wave vector components:

$$i(k) = \exp\left(-\sum_{i=1}^{3} \left(\frac{k - k_i}{\sigma_i}\right)^2 / \sigma_i^2\right)$$  (1)

$k$ = any incident wave vector, $k_i$ = the average incident wave vector, $\sigma_i$ = the 3 principal orthonormal axes ($i = 1,2,3$) of the distribution $\sigma_i$ = the widths of the distribution along these axes ($i = 1,2,3$). In the version of this calculation for D17 /1/, the intensity distribution is assumed symmetrical about the mean beam direction, an hypothesis hardly compatible with a beam monochromated by Bragg diffraction from a single crystal monochromator.

3) the crystal mosaicity is assumed to be isotropic and uniform in reciprocal space with a 2-dimensional Gaussian angular distribution.
An approximate analytical solution of the problem can be found in the form of a Gaussian distribution of the intensity in the diffracted beam, in the case where the angular width of mosaicity is smaller than the beam divergence. It reads:

\[ I(X,Y,\omega) = \exp \left[ -\frac{(\omega-\omega_0)^2}{\sigma_\omega^2} - \frac{(\theta-\theta_m)^2}{\sigma_\theta^2} - \frac{(\phi-\phi_m)^2}{\sigma_\phi^2} \right] \] (2)

\( \omega \) = rotation angle
\( \omega_0 \) = rotation angle corresponding to maximum intensity
\( \theta, \phi \) = diffraction angles (spherical coordinates)
\( X(\theta, \phi) \) = corresponding reflection coordinates on detector
\( Y(\theta, \phi) \)
\( \theta_m, \phi_m \) = diffraction angles of maximum intensity at a given \( \omega \) setting.

These parameters and the corresponding variances and covariance appearing in relation (2) are functions of the crystal orientation, the reciprocal lattice vector \( /H,K,L/ \) under consideration, the primary beam intensity distribution parameters, the crystal mosaicity and the diffractometer geometry. Their rather lengthy expressions will not be given here.

**B - SIZE OF THE MASKS.**

To calculate the size of a mask using the expression of \( I(X,Y,\omega) \) it is necessary to define a cut-off limit to the Gaussian function in relation (2):

i) Definition of the cut-off \( c \): one considers a pixel of coordinates \( (X,Y) \) as belonging to the mask \( M(c) \) of the reflection under consideration, for a given angle setting of the crystal, if the value of \( I(X,Y,\omega) \) corresponding to it satisfies the inequality:

\[ I(X,Y,\omega) > c \] (3)

This is analogous to the definition of a statistical test. \( M(c) \) represents the domain of validity of the hypothesis: "pixel \( (X,Y) \) belongs to the reflection", and \( I(X,Y,\omega) \), if properly normalized, represents the corresponding probability distribution function.

The threshold of the test is given by:

\[ S = 1 - \frac{\int_{M(c)} I(X,Y,\omega) \sin \theta \, d\theta \, d\phi \, d\omega}{\int_{\infty} I(X,Y,\omega) \sin \theta \, d\theta \, d\phi \, d\omega} \] (4)

\( s \) is the relative cut-off error due to integration in the limited mask \( M(c) \).

ii) To give a value to \( s \), we use the following criterion: The absolute cut-off error on the integrated intensity \( I_c \), corrected for background, has to be small compared to the standard deviation of \( I_c \), i.e.:

\[ s \cdot I_c \ll \sqrt{I_c + n \cdot B} \] (5)

where: \( B \) = average background per pixel
\( n \) = number of pixels used to integrate the reflection.
In (5), it is assumed that the error on $B$ can be neglected, and that the uncorrected integrated intensity $(I_0 + n \cdot B)$ is a Poisson random variable. In practice we use the relation:

$$s \cdot I_0 = \sqrt{n \cdot B} \quad (6)$$

This leads to the definition of a mask whose size varies as a function of the real intensity of the reflection considered: the size of the masks varies both as a function of $\omega - \omega_0$, and from one reflection to another.

The value of $n$ can be calculated a priori from the value of the experiment parameters and the value of $c$. To make use of (5), we need to know the scale factor between the real intensity of a given reflection in a given rotation setting and its theoretical counterpart (2). This factor is calculated by integrating in $X$ and $Y$, with background correction, the diffracted intensity in a single spectrum measured at $\omega$ near to $\omega_0$ for the reflection under consideration, and dividing the result by the corresponding integrated value of $I(X,Y,\omega)$.

The calculation of masks must be modified in two cases:

- for strong reflections, the masks obtained are enlarged by adding a ring of one pixel width. This has to be done to take into account the scattering of neutrons of the diffracted beam by the material of the entrance window of the detector.
- for very weak reflections, the relation (6) is not applicable ($s$ would be $> 1$). The cut-off $c$ is then given a value such that the number of pixels included in the mask at $\omega = \omega_0$ (i.e. at maximum intensity) is equal to 2.

IV - DETERMINATION OF CRYSTAL MOSAICITY.

The width of the Gaussian distribution of the mosaicity is determined by a least-squares adjustment of the theoretical and observed variations of the values of $X_m$ and $Y_m$, the coordinates of the intensity maximum of a given reflection in the spectra where it is active, and of its intensity integrated in these spectra, as a function of the rotation angle. This determination has to be made on a small number of reflections before the start of the full data reduction.

V - FLOW DIAGRAM OF ON-LINE DATA TREATMENT.

The data treatment has to be made in two stages: Since a correct background calculation can only be made on a spectrum where all masks have been calculated, and because the size of the masks depends on the intensity of the reflections, it is necessary to make a first evaluation of the intensity of the predicted reflections (this is made, for every reflection, on the spectrum where the average Bragg conditions are satisfied). They are treated effectively later on, with a constant lag NL sufficiently large to be sure that all reflections active at N-NL have already had their maximum at N.
Figure 1: Example of variations, as a function of rotation angle $\omega$, of: $I$, the integrated intensity per spectrum; $B$, the average background per pixel in a box around the reflection; and $X_m$ and $Y_m$, the coordinates of the position of the intensity maximum, in the case of reflection $/111/$ of a Nucleosome Core Particle Crystal in 100% $\text{D}_2\text{O}$. 
<table>
<thead>
<tr>
<th>Spectrum number</th>
<th>Measurement in progress</th>
</tr>
</thead>
<tbody>
<tr>
<td>N+1</td>
<td>First evaluation of reflection parameters on reflections which are at their maximum on this spectrum:</td>
</tr>
<tr>
<td>N</td>
<td>1) predict the values of HKL of these reflections.</td>
</tr>
<tr>
<td></td>
<td>2) calculate a first estimate of their intensity and determine their integration cutoff c.</td>
</tr>
<tr>
<td></td>
<td>3) calculate their omega limits ((\omega)<em>{\text{min}}) and ((\omega)</em>{\text{max}}).</td>
</tr>
<tr>
<td></td>
<td>4) store HKL, ((\omega)<em>{\text{min}}), ((\omega)</em>{\text{max}}), c and other parameters in an indexed scratch file with ((\omega)_{\text{min}}) as key (VAX-specific).</td>
</tr>
<tr>
<td>N-NL</td>
<td>Full integration of the reflections, spectrum by spectrum: reflections that are being integrated are kept in a temporary array which is updated at each step.</td>
</tr>
<tr>
<td></td>
<td>1) recall from the scratch file the HKL's which are beginning to be active on this spectrum (search with key ((\omega)_{\text{min}})) and put them into the temporary array.</td>
</tr>
<tr>
<td></td>
<td>2) calculate the masks for all reflections occurring on this spectrum.</td>
</tr>
<tr>
<td></td>
<td>3) calculate background.</td>
</tr>
<tr>
<td></td>
<td>4) integrate intensities within masks and treat overlaps.</td>
</tr>
<tr>
<td></td>
<td>5) for any reflections finished ((\omega)_{\text{max}}) reached:</td>
</tr>
<tr>
<td></td>
<td>- output the total integrated intensity, corrected for background and detector efficiency,</td>
</tr>
<tr>
<td></td>
<td>- calculate the standard deviation and Lorentz correction</td>
</tr>
<tr>
<td></td>
<td>- erase from the temporary array.</td>
</tr>
</tbody>
</table>

VI - ON-LINE (STEP BY STEP) H,K,L PREDICTION.

The method used to predict the reflections HKL whose intensity maximum (average Bragg condition) occurs in the omega range:

\[ \omega_1 = \omega - \frac{\delta \omega}{2}, \quad \omega_2 = \omega + \frac{\delta \omega}{2} \]  \hspace{1cm} (7)

where \(\delta \omega\) is the omega step, is the following:

1) Calculation of \(H_1, H_2, K_1, K_2, L_1, L_2\) defining the set \(E_1\) of HKL's:

\[ H_1 < H < H_2, \quad K_1 < K < K_2, \quad L_1 < L < L_2 \]

where \(H_1, H_2, K_1, K_2, L_1, L_2\) are the narrowest limits to be used for generating the HKL's to be selected. These limits are calculated as the extrema of \(H, K,\) and \(L\) with the constraints:

a) Bragg condition for that omega setting,

b) diffracted beam inside a cone surrounding the PSD, using a Lagrange multiplier method proposed by Taupin /2/.

2) Selection in the set \(E_1\) of the subset \(E_2\) verifying the condition (7).

3) Selection from subset \(E_2\) of the subset \(E_3\) of the reflections falling effectively on the detector.

This calculation is very fast for low resolution reflections.

VII - BACKGROUND CALCULATION AND CORRECTION.

METHOD 1: the method used up to now was to calculate an average value of the background around a reflection and to correct the
integrated intensity by subtracting this value multiplied by the number of pixels used in the integration. The average was calculated by sampling the background counts found in the pixels outside the masks in the vicinity of the considered reflection, over all spectra where this reflection is active.

This method is simple and fast, and also precise because a high number of pixels is sampled. Nevertheless it is not safe if the background is not uniform, because there is no control on the background sampling.

Non-uniformity of background is indeed observed frequently, as well as variations of background with the rotation of the crystal. For instance, one observes an apparent increase of background near intense reflections due to scattering of the diffracted beam by the detector, or due to thermal diffuse scattering (see B in Fig. 1).

In the method of Xuong et al. /3/, an average background is calculated for each pixel independently, using a method of autoregressive filtering of the background measured in the same pixel before the appearance of the reflection. This solves the problem of non-uniformity of background rather well but it cannot be used in our case as a given pixel can remain inside masks over a large part of the crystal rotation range.

METHOD 2: this is the method we are using at present. It is a combination of:

a) an interpolation of the background in each spectrum by two-dimensional linear spline functions using the unmasked pixels, and

b) a moving average of the spline parameters over the M most recent spectra.

The method of interpolation with linear splines has been applied by Schwartz /4/. It is based on a distribution of nodes on the detector area, each node being the vertex of a triangle. A linear variation of the background is assumed inside these triangles. The value of background on the nodes is determined by least-squares. The system of linear equations (typical size 100 * 100) is solved in our case by a conjugate gradient method /5/.

The calculation of the moving average is applied to the value of background at the nodes and is made either by assuming stationary conditions or allowing for a linear time evolution over the M spectra, or with some intermediate approximation.

The advantage of this method is that the sampling of the background is less local and can be controlled, permitting the treatment of non-uniform and non-stationary background conditions.

It is, however, rather slow (4 sec/spectrum on a VAX 750) and may involve large arrays.

The background correction is made by adding the interpolated and averaged values of background calculated in the masks, at the same time as it is made for the intensity integration and the total background is subtracted at the end. Tests have shown that the precision is the same as for the first method i.e. it gives the same order of magnitude for the standard deviations of the corrected integrated intensities.
REMARK ON DETECTOR NORMALISATION.

The measured local variations of counting in the case of uniform flux are due mostly to an imbalance between neighbouring pixels (an effect comparable to a non-uniformity of the pixel areas) and not to a local variation in counting efficiency (see for instance /6/). This effect should normally be included in the background calculation but because of the large number of pixels involved in reflection integration, it is presumably averaged out and the relevant correction can be discarded. This is going to be tested.

VIII - DISCUSSION.

The positive features of the method of a priori calculated masks are:
- fast calculation
- a good control of the collected data (i.e. control of the quality of the crystal and of the diffraction conditions)
- an easy treatment of reflections very close to each other or overlapping.

The method also provides an easy way of calculating an approximate value of the integrated intensity from partial data.

Its difficulties and limitations are:
- it requires a good knowledge of the orientation of the crystal
- the reflection prediction and mask calculation have to be exhaustive (no unpredicted reflections)
- the incident beam and the crystal have to be "regular" (clean collimation and no crystal or wavelength splitting).

Our experience seems to show that the Gaussian approximation based on the real parameters of the diffraction experiment is good enough for predicting masks, but it is not accurate enough for a profile fitting. Nevertheless, it may provide a starting point for a profile analysis.

REFERENCES.