Spatial-Distortion Corrections, for Laue Diffraction Patterns Recorded on Image Plates, Modelled using Polynomial Functions

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Abstract

Successful integration of the spot intensities on a single-crystal X-ray diffraction image depends on the ability to predict the spot positions accurately throughout the image. In some image-plate systems, there are spatial distortions, resulting from imperfections in the scanning system, for which corrections need to be made before a satisfactory match between observed and predicted spot positions can be achieved. A procedure to correct for this spatial distortion as part of the refinement procedure used in processing Laue diffraction data is described. The spatial distortion is modelled using polynomial functions. For the test and other images considered, with a pixel size of 88 μm, the root-mean-square (r.m.s.) deviations between the observed and predicted spot positions were reduced from up to 1.4 pixels to 0.4 pixels or less for the 400 or so spots used in the refinements and good predictions throughout the images were obtained.

1. Introduction

When reflection intensities from an X-ray diffraction pattern of a single crystal recorded on an image plate are integrated, it is necessary to predict the spot positions on the image plate accurately with a r.m.s. discrepancy of no more than a pixel and ideally of half a pixel or less; the prediction should be uniformly good over the whole range of the pattern to be integrated. The accurate prediction of spot positions is crucial to the determination of good spot profiles during the integration procedure and in their application, particularly in determining the intensities of the weaker spots (Rossmann, 1979). The principal parameters required for this prediction are the unit-cell parameters, the angles defining the crystal orientation and the crystal-to-detector distance. An initial determination of the crystal orientation is required. In the Daresbury Laboratory Laue Software Suite (Helliwell et al., 1989; Campbell, 1993), the method used involves identifying some nodal spots and measuring their positions. This method requires prior knowledge of the cell parameters and has sufficient tolerance to cope with some spatial distortion in the measured image. After determination of the approximate orientation of a crystal, this orientation is refined by a minimization procedure to get the best fit between the predicted and observed spot positions on the measured image for 200 or more spots. Refinement of the crystal orientation is always carried out but unit-cell-parameter refinement (actually axial-ratio refinement as one cell length must be fixed) is optional. However, in this software suite, as in MOSFLM for measuring intensities of monochromatic oscillation images (Winocott, Dockrell & Brick, 1980; Leslie, 1992), it is also necessary to introduce and refine a number of other parameters that account for the distortions of the actual experimental geometry from the idealized geometry, for example, to allow for the fact that the detector may not be exactly normal to the X-ray beam. For radially scanned images from the MARResearch™ (Hamburg, Germany) image-plate system, two further refineable parameters, roff and toff, had to be introduced into both MOSFLM (Leslie, 1992) and the Laue Software Suite. When the Molecular Dynamics™ (Sunnyvale, California, USA) off-line image-plate system and scanner were introduced at Daresbury Laboratory for the recording of Laue diffraction patterns, it was soon apparent that some other parameters were required to correct for the spatial distortion in the image resulting from the scanning mechanism; without these, satisfactory spot positions, to enable good intensity integrations, could not be obtained. With this type of scanner, Svensson, Hammersley, Thomson, Gonzalez & Usry (1993) and Hammersley, Svensson & Thomson (1994) found systematic errors in the scanning system that result in spatial-distortion errors of up to several pixels in magnitude, although Né, Gazeau, Lambard, Lesieur, Zemb & Gabriel (1993) reported no significant distortion. Such spatial distortion is often found in other types of area detector (Stanton, Philips, Li & Kalata, 1992).

Methods of calculating a spatial-distortion correction based on analysis of an image that has been exposed using a mask with a rectangular grid of holes have been described by Stanton, Philips, Li & Kalata (1992), Svensson, Hammersley, Thomson, Gonzalez & Usry (1993) and Hammersley, Svensson & Thomson (1994). An alternative approach, described here, is the deriva-
MODELLING OF SPATIAL DISTORTION CORRECTIONS

Table 1. Summary of standard parameters refined

<table>
<thead>
<tr>
<th>Type of parameter</th>
<th>Parameter names</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mis-setting angles</td>
<td>( \varphi_x, \varphi_y, \varphi_z )</td>
</tr>
<tr>
<td>Variable cell parameters</td>
<td>( a, b, c \text{ etc. (fix one)} )</td>
</tr>
<tr>
<td>Crystal-to-detector distance</td>
<td>( c_{to,f} )</td>
</tr>
<tr>
<td>Pattern offset from measured centre</td>
<td>( x_c, y_c )</td>
</tr>
<tr>
<td>Raster: ( x:y ) ratio</td>
<td>( y_{scale} )</td>
</tr>
<tr>
<td>Distortion parameters</td>
<td>tilt, twist</td>
</tr>
</tbody>
</table>

tion of a spatial-distortion correction function using the diffraction pattern itself. If successful, it has the advantage that it can be used even if no suitable calibration image is available. In particular, it has been devised as part of the processing procedure for Laue diffraction patterns. The parameters used in the refinements described below are summarized in Table 1.

The tilt and twist distortion corrections are applied as follows:

\[
x_{\text{corr}} = k_{\text{factor}} \times x_p,
\]

\[
y_{\text{corr}} = k_{\text{factor}} \times y_p,
\]

where

\[
k_{\text{factor}} = 1.0 + \left( x_p \times \text{tilt}/c_{\text{to,f}} \right) + \left( y_p \times \text{twist}/c_{\text{to,f}} \right)
\]

and where \( x_p \) and \( y_p \) are the predicted spot coordinates with reference to an origin at the pattern centre.

2. Method

Because plates such as those used in the Molecular Dynamics™ image-plate system are scanned across the plate in one direction and the scanning carriage is moved at right angles to this during the scanning process, independent distortion corrections in the two directions can be expected and may be modelled in a general way using, for example, polynomial functions. The determination of such distortion corrections was incorporated as an option in a development version of the program LAUEGEN from the Daresbury Laboratory Laue Software Suite.

After corrections for the camera constants and the tilt and twist parameters have been applied, the predicted positional coordinates on the detector \( x \) and \( y \) are further corrected using a polynomial function:

\[
x = x + a_1 x + a_2 x^2 + a_3 x^3 + \ldots
\]

\[
y = y + b_1 y + b_2 y^2 + b_3 y^3 + \ldots
\]

The order of the polynomial fitted (up to a maximum of eight) may be selected by the user.

From the results obtained using this correction, it appeared that a cross term between \( x \) and \( y \) might give a further improvement in the matching. As a result, an additional correction is applied just prior to the polynomial correction as follows:

\[
x = x + c_1 y. \tag{3}
\]

3. Experimental test

The method was tested on an image of a ruthenium-based organometallic compound (RUP) (space group \( P2_12_12_1 \); \( a = 9.37, \ b = 13.69 \) and \( c = 23.04 \) Å) recorded on station 9.7 of the Daresbury Synchrotron Radiation Source using the Molecular Dynamics™ image-plate and scanning system. The crystal was supplied by Dr C. D. Reynolds (Liverpool John Moores University). A determination of the structure of this compound using diffractometer data has been reported by Blake, Crook, Mawby, Reid & Reynolds (1992).

The orientation of the crystal was determined and refined using a version of LAUEGEN. As the Laue image did not cover the complete \( 253 \times 200 \) mm image plate, the processing of the data was restricted to an area of about \( 199 \times 182 \) mm.

With the standard distortion refinement of tilt and twist, the best refinement gave a r.m.s. deviation between the observed and predicted spot positions of 0.127 mm for 408 reflections. The raster size for the scanner was 88 μm so this represents a deviation of about 1.4 pixels. This is unacceptably high for successful integration.

With a distortion correction of tilt and twist and a fifth-order polynomial in both the \( x \) and \( y \) directions, a r.m.s. deviation of 0.065 mm for 408 reflections was obtained. This is still higher than ideal but is significantly less than one pixel, which is about the upper usable limit.

Inclusion of the additional cross term between \( x \) and \( y \) gave a further significant improvement. This gave a r.m.s. deviation of 0.035 mm for 408 reflections (about 0.4 pixels). This is now within the target range for a good integration of the spot intensities to be obtained.

The deviations between the observed and predicted positions for the different cases are shown in Fig. 1. The final case (Fig. 1c) gives a very good fit across the whole pattern. The forms of the \( x \) and \( y \) polynomial distortion corrections are shown in Fig. 2. The exact values of the corrections derived should be treated with some caution as there tend to be correlations between the derived polynomial functions and the other parameters refined (especially the other distortion parameters). Also, the derived corrections should only be used within ranges of \( x \) and \( y \) defined by the spot positions used in the refinement. However, the primary objective of satisfactory spot-position prediction prior to integration has been achieved.

4. Comparison with a grid-based correction

A further analysis of the method was carried out with use of the procedure and software of Hammersley, Svensson
& Thomson (1994), which uses distortion-corrected images derived from the analysis of a calibration image exposed using a mask with a rectangular grid of holes. Such calibration images were recorded at Daresbury some months later than the Laue data described above and thus there is a limit to the validity of comparisons. It should also be borne in mind that the method was developed so that images could be processed for which no such calibration was available and, if this was done successfully, to obviate the need for such a procedure. The test RUP image was corrected using a calibration image and the crystal-orientation, cell and camera constants were refined together with the tilt and twist parameters, i.e. all the refinable parameters except for the new spatial-distortion corrections that form the subject of this paper. This refinement proceeded reasonably successfully to give a r.m.s. deviation of 0.050 mm for 443 reflections. An inspection of a difference plot indicated that there were still some minor residual systematic errors, probably indicating some change in the spatial-distortion correction required since the Laue images were recorded. The parameters from this refinement were then used as a starting point for determining the spatial-distortion correction from the uncorrected Laue image. The only parameters that were now refined were the polynomial and spatial-distortion cross terms and the offsets $x_c$ and $y_c$ from the pattern centre. This gave a r.m.s. deviation of 0.042 mm for 443 reflections. This same refinement was repeated with a higher-order (eighth-order) polynomial and more reflections, giving a r.m.s. deviation of 0.044 mm for 923 reflections. The spatial-distortion corrections from the two methods based on this latter refinement are compared in Fig. 3. Nowhere is the difference greater than 0.2 mm and mostly it is much less. Across the laser scan direction, where the distortion is large, the agreement is generally good. In the carriage travel direction, the size of the corrections is much smaller. Also, it has been shown by Hammersley, Svensson & Thomson (1994) that the corrections required in this direction are much less reproducible and can vary from day to day. Indeed, the varying nature of such a correction gives added value to a method such as the one described in this paper where no external calibration is used. There was also a problem in

![Image](a)

![Image](b)

![Image](c)

Fig. 1. (a) Plot of the RUP Laue diffraction pattern showing the difference vectors between the observed and predicted spot positions after refinement using the standard distortion correction. Different parts of the image show different systematic errors. (b) Plot of the RUP Laue diffraction pattern showing the difference vectors between the observed and predicted spot positions after refinement using the polynomial-based distortion correction. There is clearly significant improvement over the standard correction but closer inspection still shows residual errors of a systematic nature. (c) Plot of the RUP Laue diffraction pattern showing the difference vectors between the observed and predicted spot positions after refinement using the polynomial-based distortion correction with the addition of an $xy$ cross term. There is again significant improvement over the previous case with no obvious systematic errors remaining. The difference vectors in all the plots are multiplied by a factor of 25. Only the 408 spot positions used in the refinement are indicated. The vertical direction in the plots corresponds to the laser scan direction.
determining the registration between the images in the carriage travel direction and there is perhaps evidence of this in the plots. The spatial distortion in the laser scan direction results mainly from the geometry of the scanning mechanism though it does vary between 88 and 176 µm scans and the distortion in the carriage travel direction results from the mechanism in which a wire is wound around a spindle (A. Hammersley, private communication). This latter correction has a periodic component and, in another grid-based image-distortion correction procedure developed by R. Pilz (Daresbury Laboratory, unpublished results), this has been modelled using a sine-wave-based correction. Such an approach is more detector specific and has not been followed by us.

The cross term would suggest that the two scan directions are not quite orthogonal. This may result, in part at least, from the fact that the carriage is in continuous motion during the scanning process. From the calibration image, the distortions at each hole position along the carriage travel direction were averaged for each hole along the laser scan direction. This showed a linear trend across the laser scan direction requiring a correction of -0.422 mm over the calibrated width of 200.9 mm, which would correspond to a cross term of -0.0021. This compares with a range of values of -0.0024 to -0.0035 obtained from the processed Laue images. The higher-order polynomial and additional spots gave a slightly improved fit over that obtained from the fifth-order polynomial and around 400 spots used in other refinements though, for the 443 refinement spots used in the RUP test refinement, the r.m.s. deviation was only very marginally reduced by an increase in the polynomial order.

5. Correlation of refined parameters

Further investigation based on the grid-based comparison indicated that it is the tilt and twist parameters that are most highly correlated to the polynomial-based correction terms in the refinement procedure and it is for this reason that the distortion corrections in Fig. 2 have additional slopes compared with those derived via the protocol using the grid-corrected image. If all parameters are refined for the RUP test image, except for the tilt and twist values derived from the grid-corrected image, then a r.m.s. value of 0.037 mm for 442 reflections was obtained. The polynomial-based distortion corrections in this case are very close to those obtained when the cell, orientation angles and crystal-to-plate distance were not refined, differing by less than 0.08 mm in the laser scan direction and 0.04 in the carriage travel direction.

Although the primary aim of the proposed method is the ability to make accurate predictions of the spot positions, it is also important to know whether it is acceptable to refine the cell parameters as part of the procedure. The cell parameters derived from the image corrected using the calibration grid were compared with those obtained at two stages of the RUP test-image refinement using the original uncorrected image and also with the diffractometer-data-derived cell parameters. The results are shown in Table 2. The cell parameters, when the new spatial-distortion correction has been included in the refinement, agree well with

Table 2. Comparison of RUP cell-parameter values

<table>
<thead>
<tr>
<th>Description</th>
<th>a (Å)</th>
<th>b (Å)</th>
<th>c (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diffractometer data</td>
<td>9.3667 (8)</td>
<td>13.6888 (20)</td>
<td>23.0421 (20)</td>
</tr>
<tr>
<td>Grid-corrected image</td>
<td>9.37 (fixed)</td>
<td>13.686</td>
<td>22.982</td>
</tr>
<tr>
<td>All parameters except new spatial distortion</td>
<td>9.37 (fixed)</td>
<td>13.673</td>
<td>23.135</td>
</tr>
<tr>
<td>All parameters including new spatial distortion</td>
<td>9.37 (fixed)</td>
<td>13.680</td>
<td>23.033</td>
</tr>
</tbody>
</table>

Fig. 3. Comparison of the polynomial-based spatial-distortion terms with those derived from a grid-based calibration image. Curve (a) is the polynomial function in the laser scan direction and curve (b) is the corresponding grid-based correction in that direction averaged along the carriage travel direction. Curve (c) is the polynomial function in the carriage travel direction and curve (d) is the corresponding grid-based correction in that direction averaged along the laser scan direction. (Because the spatial-distortion corrections for the calibrated image are relative to an arbitrary reference position, an offset, equal to its calculated value at this reference position, has been subtracted from each polynomial-derived distortion correction.)
compound are also shown in Table 4 and are discussed below. (Cell dimensions from a series of images of another parameters derived as part of the refinement procedure. The cell parameters derived from other images gives a maximum variation of about 0.5% and this probably gives a more realistic view of the accuracy of any cell parameters derived as part of the refinement procedure. (Cell dimensions from a series of images of another compound are also shown in Table 4 and are discussed below.)

6. Other examples

The procedure was repeated with the other images from the RUP data set, RUP4 and RUP6. The r.m.s. deviations for the first refinement (all parameters apart from the new spatial-distortion parameters) and for the final refinement (including the new spatial-distortion parameters) are shown in Table 3. A test was carried out to see whether the polynomial corrections derived from the original test image were transferable to the other images of the data set. This was done by transferring the polynomial and cross correction terms to the parameter lists of the other two data sets and allowing the other parameters to be re-refined. The final r.m.s. values were 0.031 mm for 374 reflections (RUP4) and 0.032 mm for 432 reflections (RUP6). If the cell and mis-setting angles were not re-refined, then the r.m.s. values were only slightly higher at 0.033 and 0.035 mm, respectively. The most significant changes were in the other distortion parameters. For RUP4, the tilt and twist values were −29.3 and 6.9 for the independently refined data set and −17.8 and 12.8 for the data set with the transferred polynomial and cross terms from the RUP test image. Similarly, for RUP6, the values changed from −8.4 and −11.1 to −18.0 and −3.6 (tilt and twist units are 0.01°). For both the RUP4 image and the RUP6 image, the refined crystal-to-detector distance decreased by 0.6% and the y_scale parameter increased by 0.7%. It is apparent from this and also from the tests described in the previous section that there tends to be a large correlation between the tilt and twist parameters and the new distortion-correction terms. It may be satisfactory, in many cases, to allow the polynomial distortion correction to model the tilt and twist as well as the spatial distortion arising from the scanning process.

A sequence of images from another set of data, collected on station 9.7 of the Daresbury Synchrotron Radiation Source nine months later, were also processed as a further test of the method. The sample was a crystal of potassium titanyl phosphate (KTP), supplied by Dr P. A. Thomas (University of Warwick), for which a structure determination using diffractometer data has been reported by Thomas, Glazer & Watts (1990). The same refinement protocol as for the RUP data was followed for a set of three images from this data set; the r.m.s. deviations for the first and final refinements are given in Table 3.

Again, as in the case of RUP, it was clear from difference plots that the systematic errors apparent after the first refinement were effectively dealt with by the introduction of the new distortion-correction terms. Two further investigations were carried out with this data set. In the first of these, the polynomial and spatial-distortion cross terms from the RUP test data set were transferred to the list of parameters for the KTP2 data set. The parameters x_c, y_c, c_to_f, y_scale, tilt and twist were re-refined to give a r.m.s. value of 0.041 mm for 449 reflections. The difference plot indicated that this refinement was still quite satisfactory though there was an indication of some minor systematic errors. Although it may be of interest to know whether the distortion correction is transferable, it is not envisaged that this would be used in the normal refinement protocol except perhaps within a data set of several images being processed at the same time.

In the second test, the cell dimensions were looked at more closely and compared with a set of diffractometer-data-derived cell parameters (Thomas, Glazer & Watts, 1990). The refinement was repeated for the KTP2 image, with refinement of all parameters except for the cell parameters. The final r.m.s. value was 0.033 mm for 450 spots, the same as that obtained when the cell parameters were refined. Table 4 shows that, when the cell parameters were refined, those obtained after the new spatial-distortion corrections were included in the refinement were closer to the diffractometer-data-derived values (maximum deviation = 0.13%) than those at the end of the first stage of the refinement (maximum deviation = 0.6%). The maximum variation for a cell

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Table 3. Results of the refinements of other RUP and KTP images

<table>
<thead>
<tr>
<th>Sample name</th>
<th>R.m.s. deviation (mm)</th>
<th>No. of spots in refinement</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>First refinement</td>
<td>Final refinement</td>
</tr>
<tr>
<td>RUP4</td>
<td>0.088</td>
<td>0.032</td>
</tr>
<tr>
<td>RUP6</td>
<td>0.082</td>
<td>0.032</td>
</tr>
<tr>
<td>KTP2</td>
<td>0.084</td>
<td>0.033</td>
</tr>
<tr>
<td>KTP3</td>
<td>0.082</td>
<td>0.036</td>
</tr>
<tr>
<td>KTP4</td>
<td>0.076</td>
<td>0.037</td>
</tr>
<tr>
<td>KTP3</td>
<td>0.081</td>
<td>0.063</td>
</tr>
<tr>
<td>KTP4</td>
<td>0.081</td>
<td>0.035</td>
</tr>
<tr>
<td>KTP5</td>
<td>0.076</td>
<td>0.037</td>
</tr>
</tbody>
</table>

Table 4. Cell parameters derived from KTP refinements

<table>
<thead>
<tr>
<th>Description</th>
<th>a (Å)</th>
<th>b (Å)</th>
<th>c (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diffractometer</td>
<td>12.819 (3)</td>
<td>6.399 (1)</td>
<td>10.584 (2)</td>
</tr>
<tr>
<td>KTP2 first refinement</td>
<td>12.82 (fixed)</td>
<td>6.412</td>
<td>10.572</td>
</tr>
<tr>
<td>KTP3 first refinement</td>
<td>12.82 (fixed)</td>
<td>6.381</td>
<td>10.525</td>
</tr>
<tr>
<td>KTP4 first refinement</td>
<td>12.82 (fixed)</td>
<td>6.375</td>
<td>10.519</td>
</tr>
<tr>
<td>KTP2 final refinement</td>
<td>12.82 (fixed)</td>
<td>6.407</td>
<td>10.589</td>
</tr>
<tr>
<td>KTP3 final refinement</td>
<td>12.82 (fixed)</td>
<td>6.402</td>
<td>10.583</td>
</tr>
<tr>
<td>KTP4 final refinement</td>
<td>12.82 (fixed)</td>
<td>6.400</td>
<td>10.581</td>
</tr>
</tbody>
</table>

Sample First Final No. of spots
KTP3 0.082 0.036 535
KTP2 0.084 0.033 450
KTP4 0.076 0.037 585
parameter within the three fully refined images was 0.11%.

7. Concluding remarks
The results obtained demonstrate that the method described can provide a practical way of coping with the spatial distortion in the scanned Molecular Dynamics™ image-plate images used for recording Laue diffraction patterns and has enabled the successful integration of intensities from such patterns. It has the advantage that it can be used on data images for which the required calibration data from other sources, e.g. data obtained using masks, are not available or if the spatial distortion is not sufficiently stable over a period of time. The same method could probably be used for monochromatic X-ray diffraction images from single crystals. It should also be applicable to other image plates or detectors scanned in orthogonal directions. Because of the correlation of the derived polynomial spatial-distortion corrections with other refined distortion parameters and because the Laue pattern often does not cover the complete plate, it would be unrealistic to try to use this method as a general calibration method.

We are grateful to Dr A. Hammersley (ESRF) for making available his calibration software and for some detailed discussions of his investigations and results using a grid-based calibration method for Molecular Dynamics™ image plates. We thank the staff at Daresbury including Dr J. E. Harries, Dr D. A. A. Myles and Dr C. Nave for the provision of a test grid mask image and for related discussions. We also thank Dr Q. Hao (De Montfort University, Leicester) for help in scanning the Laue images. We acknowledge support from the Science and Engineering Research Council (now EPSRC) and the University of Liverpool.

References