DIFFaX *v 1.807*

by

M. M. J. Treacy, M. W. Deem & J. M. Newsam

29th August 2000

EMT, $N = \infty$

EMT, $N = 20$

manual written by
Michael M. J. Treacy

DIFFaX v1.807

29th AUGUST, 2000

Contents

List of Figures

iii

1 Introduction.

1.1 What is DIFFaX?

DIFFaX is a fortran computer program that calculates diffraction intensities from crystals that contain coherent planar defects, such as twins and stacking faults. DIFFaX exploits the recurring patterns found in randomized stacking sequences to compute the average interference wavefunction scattered from each layer type occurring in a faulted crystal [1]. It is then a simple step to calculate the incoherent intensity contribution at hkl. Although the physics behind the diffracted intensity algorithm used by DIFFaX is basically the same as that used by other methods, most notably the Hendricks & Teller [2], the difference equation [3], and the Cowley methods [4], the recursion algorithm is significantly simpler to understand. This simplicity prompted the writing of this general purpose program, which can accommodate any type of coherent planar defect in any crystal system. A brief description of the DIFFaX algorithm is presented in section 8 on page 45. The details of the algorithm are described in reference [1], a copy of which is included in section B.7 on page 93 of this manual.

1.2 About this manual.

The DIFFaX fortran code was written with portability in mind. The source code `DIFFaX.f` is pretty much ansi77-compliant and should compile and run, with few changes, on most modern computers. To run, DIFFaX assumes that a standard tty-type of input/output window is available, such as that found on unix machines. When compiling `DIFFaX.f` on a Macintosh or a Windows-based PC, systems which use a graphical user interface, the compiler must supply a tty-type window that is dedicated to DIFFaX.

The primary goal of this manual is to help explain to the reader how to construct a DIFFaX data file, and how to run the DIFFaX program on this file. Although we have tried to make DIFFaX as simple to run, and as flexible as possible, the preparation of a data file that accurately describes a particular faulted structure can be a considerable challenge. It is the hope that this manual provides enough examples and explanations to simplify the data file building process.

A pre-compiled version of DIFFaX that runs on Macintosh computers, MacDIFFaX, is supplied on the Macintosh diskette that came with this manual (MacDIFFaX is also available on request from the author). MacDIFFaX behaves almost identically to the unix version of DIFFaX. The Macintosh-specific differences between MacDIFFaX and DIFFaX are explained in section 3.8 on page 36.

2 Creating structure data files.

DIFFaX structure data files are text files that contain information about the type of radiation to simulate, the structure of the layers, and details of the interlayer connectivities. Although the DIFFaX data file format is straightforward, in the author's experience the creation of error-free files is surprisingly difficult. This difficulty arises because of the unconventional way in which DIFFaX needs the defect structure to be described. Conventionally, crystals are thought of in terms of unit cells, which in turn can be broken down further into asymmetric sub-units of atoms and a set of space group symmetry operators. To use DIFFaX we need to think of crystals in terms of sheets of atoms, or layers, which can interconnect via stacking operations that occur with a certain probability. Frequently, the plane of the layers will not coincide conveniently with any of the unit cell faces of the parent crystal. This requires a transformation of atom coordinates to a new cell system. For example, to describe 111 twins in face centered cubic materials, the layers (111 planes, and their mirrored counterparts) must be transformed to a hexagonal setting where the new c-axis is parallel to the old cubic [111] direction. The new hexagonal a- and b- axes now lie in the plane of the old cubic 111 layers, and define a two-dimensional hexagonal mesh that is common to all layers in the crystal. The atom coordinates and stacking vectors will need to be defined in terms of the hexagonal setting. Great care needs to be taken when creating the data set. It is wise to compare diffraction spectra for the unfaulted end-members, with those generated from a standard diffraction calculation program to ensure that you are starting from a valid data set. To help the user to avoid wasting time on flawed data files, a conscious design decision was made to make DIFFaX unforgiving when it discovers errors or inconsistencies in the data file. Usually, when a potential problem is found, DIFFaX will quit, forcing the user to repair the data file. This "zero-tolerance" strategy has saved the present author from numerous embarrassing blunders.

2.1 Data line lengths.

The maximum data line length is 200 characters. Anything typed beyond the 200th character may be lost, and may generate error messages. The accepted line length has been increased from the obsolete "punched card" value of 80 characters that applied in DIFFaX versions 1.7xx. 200 characters should be sufficiently wide to handle most reasonable data input lines.

2.2 Case sensitivity.

Data can be written in either upper- or lower-case. DIFFaX is insensitive to the case of the text. However, DIFFaX does assume that it is reading ASCII-defined characters. Some of the text manipulating subroutines will not work if the computer system is not ASCII-based.

2.3   Comments in the data file.

Comments can be embedded in the data files. Anything written between curly braces, *i.e.* {this is a comment}, will be ignored. Comments can be nested, and there can be more than one comment per line. All comments must begin and end on the same line; DIFFaX will assume that the data is faulty if it cannot match pairs of braces on a line. Total line lengths, including comments and braces, must not exceed 200 characters.

2.4   Backus-Naur Description of the datafile.

In Backus-Naur form, the data file format is as follows

```
{user input data file -- in Backus-Naur form}
{comments are in curly braces, blank lines are allowed}
{The pipe symbol | implies that one of a series of arguments must be chosen}
{arguments in parentheses () are optional}

{Header for radiation data} INSTRUMENTAL
{radiation type} X-RAY | NEUTRON | ELECTRON
{wavelength in Å} λ

{Instrumental broadening type} NONE
                             | GAUSSIAN half-width | u v w (TRIM)
                             | LORENTZIAN half-width | u v w (TRIM)
                             | PSEUDO-VOIGT u v w σ (TRIM)

{Header for structure data} STRUCTURAL
{cell dimensions, in Å and °} a b c γ
{diffraction point group symmetry} -1  | 2/M(1) | 2/M(2) | MMM
                                   | -3  | -3M    | 4/M    | 4/MMM
                                   | 6/M | 6/MMM  | AXIAL
                                   | UNKNOWN (% tolerance)
{total number of layer types} n
{in-plane layer widths in Å (optional)} ('width along a' 'width along b')
                                      | ('average diameter')
                                      | (INFINITE)

{New layer definition} LAYER 1
{Layer structure symmetry} NONE | CENTROSYMMETRIC
{Atom data record}
name # x_rel y_rel z_rel 'isotropic Debye-Waller factor B_iso' occupancy
.
.
.

{New layer definition} LAYER number | LAYER = a previous layer number
{If this is not equivalent to a prior layer, then enter ...}
{Layer structure symmetry} NONE | CENTROSYMMETRIC
{Atom data record}
name # x_rel y_rel z_rel 'isotropic Debye-Waller factor B_iso' occupancy
.
.
.

{New layer definition} LAYER number | LAYER = previous layer number
{If this is not equivalent to a prior layer, then enter ...}
```

{Layer structure symmetry} NONE | CENTROSYMMETRIC
{Atom data record}
name # x_rel y_rel z_rel 'isotropic Debye-Waller factor B_{iso}' occupancy
.
.
.

name # x_rel y_rel z_rel 'isotropic Debye-Waller factor B_{iso}' occupancy

{Header for stacking description} STACKING
{How to treat layer sequencing} EXPLICIT | RECURSIVE
{If EXPLICIT}
{either, an explicit sequence} lay# lay# lay# lay# lay# lay# lay# lay#
lay# lay# lay# lay# lay# lay# lay# lay# lay# lay# lay# lay# lay#
.
.
.
{or, generate sequence} RANDOM 'number of layers in crystal'
{If RECURSIVE}
{number of layers in crystal} 'number of layers' | INFINITE

{Header for stacking parameters} TRANSITIONS
{stacking probability, stacking vector (optional anisotropic stacking
uncertainty factor)}

α_{11} R_{11}^x R_{11}^y R_{11}^z (C_{11}^{11} C_{11}^{22} C_{11}^{33} C_{11}^{12} C_{11}^{13} C_{11}^{23})
α_{12} R_{12}^x R_{12}^y R_{12}^z (C_{12}^{11} C_{12}^{22} C_{12}^{33} C_{12}^{12} C_{12}^{13} C_{12}^{23})
$\vdots$
α_{1n} R_{1n}^x R_{1n}^y R_{1n}^z (C_{1n}^{11} C_{1n}^{22} C_{1n}^{33} C_{1n}^{12} C_{1n}^{13} C_{1n}^{23})

α_{21} R_{21}^x R_{21}^y R_{21}^z (C_{21}^{11} C_{21}^{22} C_{21}^{33} C_{21}^{12} C_{21}^{13} C_{21}^{23})
α_{22} R_{22}^x R_{22}^y R_{22}^z (C_{22}^{11} C_{22}^{22} C_{22}^{33} C_{22}^{12} C_{22}^{13} C_{22}^{23})
$\vdots$
α_{2n} R_{2n}^x R_{2n}^y R_{2n}^z (C_{2n}^{11} C_{2n}^{22} C_{2n}^{33} C_{2n}^{12} C_{2n}^{13} C_{2n}^{23})
$\vdots$
α_{n1} R_{n1}^x R_{n1}^y R_{n1}^z (C_{n1}^{11} C_{n1}^{22} C_{n1}^{33} C_{n1}^{12} C_{n1}^{13} C_{n1}^{23})
α_{n2} R_{n2}^x R_{n2}^y R_{n2}^z (C_{n2}^{11} C_{n2}^{22} C_{n2}^{33} C_{n2}^{12} C_{n2}^{13} C_{n2}^{23})
$\vdots$
α_{nn} R_{nn}^x R_{nn}^y R_{nn}^z (C_{nn}^{11} C_{nn}^{22} C_{nn}^{33} C_{nn}^{12} C_{nn}^{13} C_{nn}^{23})

2.5 An example data file

A data file describing random twin faults in diamond, {including comments in braces} is listed below. See appendix A.1 for a more complete description.

```
{data file for cubic diamond, with random intergrowths of hexagonal lonsdaleite}
{probability of cubic stacking = 70%}

INSTRUMENTAL                               {Header for instrumental section}
X-RAY                                      {Simulate X-ray diffraction}
1.5418                                     {X-ray wavelength}
PSEUDO-VOIGT 0.89 -0.32 0.08 0.6 trim      {Instrumental broadening}

STRUCTURAL                                 {Header for structural section}
2.52 2.52 2.06 120.0                       {unit cell coordinates}
6/MMM                                      {hexagonal, c axis = cubic [111]}
2                                          {2 layer types:  111 sheet, plus its mirror}
infinite                                   {infinite layer width}

LAYER 1                                    {cubic (111) layer, centrosymmetric}
CENTROSYMMETRIC
 C 1 -0.333333 -0.166667 -0.125 1.0 1.0
{C 1  0.333333  0.166667  0.125 1.0 1.0 -the 2nd centrosymmetric atom}
{                                  -does not need to be declared}

LAYER 2                                    {mirror image of layer 1}
CENTROSYMMETRIC
 C 1  0.333333  0.166667 -0.125 1.0 1.0
{C 1 -0.333333 -0.166667  0.125 1.0 1.0 -the 2nd centrosymmetric atom}
{                                  -does not need to be declared}

STACKING                                   {Header for stacking description}
recursive                                  {Statistical ensemble}
infinite                                   {Infinite number of layers}

TRANSITIONS                                {Header for transitions}
{Transitions from layer 1}
0.7  0.666667  0.333333  1.0               {layer 1 to layer 1, 70% chance}
0.3  0.0       0.0       1.0               {layer 1 to layer 2, 30% chance}

{Transitions from layer 2}
0.3  0.0       0.0       1.0               {layer 2 to layer 1, 30% chance}
0.7 -0.666667 -0.333333  1.0               {layer 2 to layer 2, 70% chance}
```

The same file, expressed more tersely, could read

```
INSTRUMENTAL
X-RAY
1.5418
PSEUDO-VOIGT .89 -.32 .08 .6 trim
STRUCTURAL
2.52 2.52 2.06 120
6/MMM
2
LAYER 1
CENTROSYMMETRIC
C 1 -1/3 -1/6 -1/8 1 1
LAYER 2
CENTROSYMMETRIC
C 1 1/3 1/6 -1/8 1 1
STACKING
recursive
infinite
TRANSITIONS
.7 2/3 1/3 1
.3 0 0 1
.3 0 0 1
.7 -2/3 -1/3 1
```

Note that the data line relating to the layer widths can be omitted (this exception ensures backwards compatibility with earlier DIFFaX versions). All other data lines must appear in the correct sequence.

In DIFFaX v1.804 and later, atom coordinates and stacking vectors can be expressed as fractions. For irrational fractions such as 1/3, this ensures maximum machine precision.

We now describe the data file format in more detail.

2.6   INSTRUMENTAL section.

2.6.1   INSTRUMENTAL header.

Line 1: The first non-comment, non-blank line must contain the keyword INSTRUMENTAL (either upper or lower case). No abbreviations are accepted. The data that follows describes the type of radiation and the instrumental conditions.

2.6.2   The radiation type.

Line 2: The choice is X-RAY, NEUTRON or ELECTRON, in either upper- or lower-case. No abbreviations are accepted.

2.6.3   The radiation wavelength.

Line 3: The numerical value of the radiation wavelength in Ångstroms.

2.6.4 The instrumental broadening.

Line 4: The type of instrumental broadening to simulate in powder pattern calculations. Although instrumental broadening applies only to powder pattern calculations, this line must always be supplied. There are four types supported by DIFFaX namely `NONE`, `GAUSSIAN`, `LORENTZIAN` and `PSEUDO-VOIGT`. We now describe these options in more detail.

2.6.5 Instrumental broadening type, `NONE`.

The raw powder pattern data only is saved to file, with no instrumental broadening applied. The resolution of the spectrum is determined solely by the sampling step size $\Delta\theta$ (see pages 34–36).

2.6.6 `GAUSSIAN` instrumental broadening.

The data is "convolved" with a Gaussian function, whose width may vary across the spectrum. A full-width half-maximum (NOT the standard deviation), in degrees, needs to be specified for this option. The line

```
GAUSSIAN 0.1
```

will specify a 0.1° constant full-width half-maximum Gaussian profile. The data line

```
Gaussian 0.0
```

is equivalent to specifying `NONE`. The full-width half-maximum must not be negative. The data line

```
Gaussian -0.1
```

will generate an error message. Alternatively, a variable full-width half-maximum $\Gamma(\theta)$ can be specified, having the form

$$\Gamma(\theta) = \sqrt{u\tan^2(\theta/2) + v\tan(\theta/2) + w} \tag{1}$$

u, v and w describe how the the full-width half-maximum (FWHM) varies as a function of the scattering angle θ. The intensity profile I_G of the peak centered at θ_1 is described by the equation

$$I_G(\theta_1; \theta_1 - \theta_2) = \sqrt{\frac{4\ln(2)}{\pi\Gamma(\theta_1)^2}} \ \exp\left(\frac{-4\ln(2)(\theta_1 - \theta_2)^2}{\Gamma(\theta_1)^2}\right) \tag{2}$$

where $\theta_1 - \theta_2$ represents the angular distance from the peak center.

The standard deviation Δ of the peak intensity distribution is

$$\Delta(\theta_1) = \frac{\Gamma(\theta_1)}{\sqrt{8\ln(2)}} \tag{3}$$

The data line

```
Gaussian 0.1 -0.036 0.009
```

specifies a Gaussian broadening profile with $u = 0.1$, $v = -0.036$ and $w = 0.009$. Note that the variable FWHM option is computationally expensive.

2.6.7 `LORENTZIAN` instrumental broadening.

The data is "convolved" with a Lorentzian function, whose width may vary across the spectrum. As for the `GAUSSIAN` option, a full-width half-maximum, in degrees, needs to be specified. The full-width half-maximum must be positive. The entry

```
Lorentzian 0.1
```

will specify a 0.1 degree constant full-width half-maximum, Lorentzian instrumental broadening profile. The line

```
Lorentzian 0.0
```

is equivalent to specifying 'NONE'. In the same way as for the GAUSSIAN option, a variable full-width half-maximum can be specified, with associated u, v and w parameters. The full-width half-maximum is given by equation 1. For a Lorentzian profile, the broadening is described by the equation

$$I_{\mathrm{L}}(\theta_1; \theta_1 - \theta_2) = \frac{2\Gamma(\theta_1)}{\pi \left[\Gamma(\theta_1)^2 + 4(\theta_1 - \theta_2)^2\right]} \tag{4}$$

Thus, the data line

```
Lorentzian 0.1 -0.036 0.009
```

specifies a Lorentzian broadening profile with $u = 0.1$ $v = -0.036$ and $w = 0.009$. Note that the variable FWHM option is computationally expensive.

2.6.8 `PSEUDO-VOIGT` instrumental broadening.

The data is broadened according to the pseudo-Voigt function, which is a weighted sum of Gaussian and Lorentzian functions. The broadening can vary with scattering angle. The pseudo-Voigt function requires 4 parameters, u, v, w and σ.

u, v and w control the full-width half-maximum according to equation 1. σ controls the relative amount of Gaussian and Lorentzian components, and must have a value between 0 and 1. $\sigma = 0$ is equivalent to pure Gaussian everywhere, and $\sigma = 1$ is equivalent to pure Lorentzian everywhere. The pseudo-Voigt function is described by the equation

$$I_{\mathrm{PV}}(\theta_1; \theta_1 - \theta_2) = \frac{2\sigma\Gamma(\theta_1)}{\pi\left[\Gamma(\theta_1)^2 + 4(\theta_1 - \theta_2)^2\right]} + (1-\sigma)\sqrt{\frac{4\ln(2)}{\pi\Gamma(\theta_1)^2}} \, \exp\left(\frac{-4\ln(2)(\theta_1 - \theta_2)^2}{\Gamma(\theta_1)^2}\right) \quad (5)$$

The pseudo-Voigt function takes considerably longer to compute than do the Gaussian and Lorentzian options that specify a constant full-width half-maximum. The reason is that the full-width half-maximum changes as a function of scattering angle, and fresh Gaussian and Lorentzian profiles must be computed for each point in the spectrum. When the full-width half-maximum is constant, the broadening profile is computed once, and then rapidly applied to every point. Whenever possible, DIFFaX will attempt to speed up the computation if certain redundancies are detected. For example, the data line

        PSEUDO-VOIGT 0.1 -0.036 0.009 0.6

specifies pseudo-Voigt broadening with $u = 0.1$, $v = -0.036$ $w = 0.009$ and $\sigma = 0.6$. However, the data line

        PSEUDO-VOIGT 0.0 0.0 0.0 0.6

is equivalent to specifying `NONE`, and DIFFaX will not apply any instrumental broadening. The entry

        PSEUDO-VOIGT 0.0 0.0 0.16 1.0

will be interpreted by DIFFaX as if

        LORENTZIAN 0.4

had been specified, since in this instance the FWHM is given by $\Gamma = \sqrt{w}$ (see equation 1), which is independent of scattering angle. Similarly, the data line

        PSEUDO-VOIGT 0.0 0.0 0.16 0.0

is read by DIFFaX as

$$\text{GAUSSIAN 0.4.}$$

It should be noted, however, that the `GAUSSIAN` and `LORENTZIAN` options, when supplied with non-zero u, v and w, call the pseudo-Voigt function with $\sigma = 0$ and 1, respectively, and are therefore computationally expensive functions.

For most powder pattern simulations, a `GAUSSIAN` or `LORENTZIAN` broadening profile with a constant full-width half maximum should be adequate, and are significantly faster computationally than the `PSEUDO-VOIGT` broadening profile.

2.6.9 `TRIM` keyword.

If one of `GAUSSIAN`, `LORENTZIAN`, or `PSEUDO-VOIGT` are specified, there is a further optional keyword that can be supplied, `TRIM`. `TRIM` tells DIFFaX to ignore intensity information close to the origin when simulating the instrumental broadening for a powder diffraction pattern. The peak at the origin is usually many orders of magnitude more intense than any other peak in the spectrum, and the background that is generated by the tails of this peak, when broadened, can easily swamp peaks close to the origin. When `TRIM` is specified, the unbroadened spectrum that is written to file retains the data at the origin, whereas the broadened data that is written to file suppresses the peak at the origin. It is generally desirable to specify `TRIM` whenever the powder pattern angular range includes the origin. `TRIM` has no effect if the powder pattern angular range requested by the user does not include the origin.

2.6.10 Examples of the `INSTRUMENTAL` data section.

Examples of a typical `INSTRUMENTAL` data section, with optional comments, might look like

```
INSTRUMENTAL
{Radiation type = } X-RAY
{wavelength = } 1.5418
{Instrumental broadening} PSEUDO-VOIGT 0.1 -0.036 0.009 0.6 trim
```

or

```
INSTRUMENTAL
electron      {Radiation type}
0.037         {wavelength}
none          {Instrumental broadening}
```

or simply

```
instrumental
x-ray
```

```
0.7
gaussian 0.1 trim
```

2.7 STRUCTURAL section.

The next part of the data section relates to the cell mesh that defines the symmetry of the crystal layers.

2.7.1 STRUCTURAL header.

Line 5: This section must begin with the keyword STRUCTURAL (either upper or lower case).

2.7.2 Cell dimensions.

Line 6: The cell dimensions, **a**, **b**, **c** and γ. All layers must share the same in-plane mesh dimensions, **a**, **b** and γ. The value of **c**, perpendicular to the **a**-**b** plane (and therefore parallel to the stacking direction), is needed to provide a reference scale along the stacking direction with which to define the z-component of the stacking vectors. **c** does not have to correspond to any special periodic dimension along the stacking direction for any of the layers. The data line

$$17.437 \ 17.437 \ 14.238 \ 120.0$$

defines a cell with **a** = **b** = 17.437 Å, **c** = 14.238 Å, and γ = 120°. Note that the cell axes must be defined so that **c** is parallel to the fault direction, that is, perpendicular to **a** and **b**. This will not always coincide with the **c**-axis of the original crystal system that the layer was clipped from. In the present version of DIFFaX, it is up to the user to transform the cell axial system to that expected by DIFFaX.

2.7.3 Laue symmetry.

Line 7: The expected Laue symmetry of the diffraction data. DIFFaX uses the Laue group symmetry to speed up powder diffraction calculations by reducing the volume of reciprocal space that needs to be included in the intensity integration. It is usually best to specify UNKNOWN, and to let DIFFaX work out the symmetry. There are ten possible diffraction point groups in the presence of streaking along one axis. These ten are a subset of the 12 Laue point groups. The Laue groups are, in turn, that subset of the 32 crystallographic point groups which possess a center of symmetry so that the diffracted intensities obey Friedel's law. The two Laue groups, 3M and M3M, are excluded since the non-orthogonal 3-fold axes will be violated by any streaking that may occur along **c***. The user can specify one of

the ten "streaky" point groups, or, if the symmetry is not known, the keyword `UNKNOWN`, whereby DIFFaX will establish the symmetry by randomly sampling reciprocal space. If `UNKNOWN` is entered, the user can specify a % tolerance on intensity deviations for this search. The default tolerance in DIFFaX is 1% (*i.e.* if the symmetry turns out to be `4/MMM`, the maximum deviation in intensity between the hkl, $\overline{k}hl$, $\overline{hk}l$ and $k\overline{h}l$ reflections did not exceed the average intensity of the four reflections by more than one part in a hundred). A twelfth option, `AXIAL`, is provided, which constrains DIFFaX to integrate along the $00l$ axis only. This is useful for turbostratically disordered systems, where lateral coherence between layers is lost. The twelve options are

```
-1
 2/M(1) {diad along streaks}
 2/M(2) {diad perpendicular to streaks}
 MMM
 -3
 -3M
 4/M
 4/MMM
 6/M
 6/MMM
 AXIAL
 UNKNOWN (tolerance)
```

If the user suggests one of the ten point groups, DIFFaX will always check the user's choice, and will override it if the choice is inconsistent with the unit cell dimensions (*i.e.* if **a** is not equal to **b**, then `-3, -3M, 4/M, 4/MMM, 6/M, 6/MMM` are not possible). DIFFaX will provide a measure of the 'goodness of fit', which is the maximum deviation from the symmetry encountered during a random sampling of reciprocal space. In the author's experience, this point group symmetry parameter is useful for debugging new data files, particularly if the cell axes used by DIFFaX are not the same as those of the crystal system from which the layers were clipped.

As a rule, it is easiest and safest to specify `UNKNOWN`, and to let DIFFaX do the work. However, there are rare occasions when DIFFaX finds a higher symmetry than is actually the case. This is caused by the limited random sampling of reciprocal space (the default is 25 tests of symmetry-related hkl values per symmetry element). Erroneous high symmetries can be avoided by using one of three strategies. (1) Specifying a more stringent tolerance, *i.e.* `UNKNOWN 0.01` will specify a tolerance on the comparisons of 0.01% (one part in 10,000). Note that DIFFaX imposes a lower limit on the tolerance of 0.01%. (2) If the user already knows the symmetry, then simply declare it outright. (3) If the cautious user wishes to override the symmetry evaluation altogether, then specify '`-1`', the lowest possible symmetry consistent with Friedel's law. This option may take significantly longer to compute, since all

hkl ($l \geq 0$) are visited.

2.7.4 Number of layer types.

Line 8: Enter here the number of layer types that the faulted structure contains. Layers which are structurally identical, but exhibit different stacking behavior, count as distinct layers.

2.7.5 Layer characteristic widths.

Line 9: This line is optional, and was introduced with DIFFaX v1.801. Enter here the characteristic width, in Ångstroms, of the crystal along the cell **a** and **b** directions. These parameters are used to estimate a Debye-Scherrer type of peak broadening due to a finite layer width. There are several legal formats for this line;

```
'width along a' 'width along b'
'layer diameter'
'INFINITE'
```

In order to maximize compatibility with versions of DIFFaX preceding version 1.80, this line may be omitted altogether. In this instance an `INFINITE` layer diameter is the default.

Note that the peak-broadening contribution due to a finite number of layers stacking along **c** is handled differently by DIFFaX (see the `STACKING` description in section 2.9).

2.7.6 Examples of the `STRUCTURAL` data section.

Typical data entries for the first part of the `STRUCTURAL` section might look like

```
STRUCTURAL
{a, b, c, gamma} 17.437 17.437 14.238 120.0
{Expected diffraction point symmetry} 6/mmm
{Number of layers = } 4
{layer width (optional)} infinite
```

and

```
STRUCTURAL
{a, b, c, gamma} 4.437 3.431 1.2 87.1
{Point symmetry unknown, tolerance = 1%} unknown 1.0
{Number of layers = } 2
{layer widths along a and b} 200 600
```

2.8 Layer structure data.

The data that follows describes the structure of the layers themselves.

2.8.1 Layer header

Line 10: Header for the first layer. It must read `LAYER 1`. This can be in upper or lower case. Abbreviations are not accepted.

2.8.2 Layer symmetry

Line 11: Symmetry of the layer structure. Only two options are accepted, `NONE` and `CENTROSYMMETRIC`. If `CENTROSYMMETRIC`, only the asymmetric half of the atom coordinates need be entered, *taking care to halve the occupancy of those atoms that are shared by the two asymmetric halves.* If centrosymmetric, all layer scattering factors will be treated as having real values in DIFFaX with the complex component ignored. Therefore, specifying centrosymmetric can provide a useful gain in computation speed, particularly for complicated structures containing many atoms per layer. Other symmetries, such as mirror planes etc. are not supported by DIFFaX because they do not provide any opportunities for significant computational speedups.

2.8.3 Layer atom data

Line 12: First record of the layer atomic structure. For each atom, the data must be entered on a single line in the form

`name id# x_rel y_rel z_rel B_iso Occ`

where
`name` = atom name (4 character wide field),
`id#` = numeric specifier, (*i.e.* 1 for the first atom, 2 for 2nd etc...)
`x_rel` = x coordinate relative to a
`y_rel` = y coordinate relative to b
`z_rel` = z coordinate relative to c
`B_iso` = isotropic Debye-Waller factor (Å^2)
`Occ` = occupancy factor (must be between 0 and 1)

Legal atom names are exactly 4 characters wide, including any spaces. A full list of the names accepted by DIFFaX is

```
'H   ' 'H.  ' 'D   ' 'H 1-' 'He  ' 'Li  ' 'Li1+' 'Be  ' 'Be2+' 'B   '
'C   ' 'C.  ' 'N   ' 'O   ' 'O 1-' 'O 2-' 'F   ' 'F 1-' 'Ne  ' 'Na  '
'Na1+' 'Mg  ' 'Mg2+' 'Al  ' 'Al3+' 'Si  ' 'Si. ' 'Si4+' 'P   ' 'S   '
'Cl  ' 'Cl1-' 'Ar  ' 'K   ' 'K 1+' 'Ca  ' 'Ca2+' 'Sc  ' 'Sc3+' 'Ti  '
'Ti2+' 'Ti3+' 'Ti4+' 'V   ' 'V 2+' 'V 3+' 'V 5+' 'Cr  ' 'Cr2+' 'Cr3+'
```

```
'Mn  ' 'Mn2+' 'Mn3+' 'Mn4+' 'Fe  ' 'Fe2+' 'Fe3+' 'Co  ' 'Co2+' 'Co3+'
'Ni  ' 'Ni2+' 'Ni3+' 'Cu  ' 'Cu1+' 'Cu2+' 'Zn  ' 'Zn2+' 'Ga  ' 'Ga3+'
'Ge  ' 'Ge4+' 'As  ' 'Se  ' 'Br  ' 'Br1-' 'Kr  ' 'Rb  ' 'Rb1+' 'Sr  '
'Sr2+' 'Y   ' 'Y 3+' 'Zr  ' 'Zr4+' 'Nb  ' 'Nb3+' 'Nb5+' 'Mo  ' 'Mo3+'
'Mo5+' 'Mo6+' 'Tc  ' 'Ru  ' 'Ru3+' 'Ru4+' 'Rh  ' 'Rh3+' 'Rh4+' 'Pd  '
'Pd2+' 'Pd4+' 'Ag  ' 'Ag1+' 'Ag2+' 'Cd  ' 'Cd2+' 'In  ' 'In3+' 'Sn  '
'Sn2+' 'Sn4+' 'Sb  ' 'Sb2+' 'Sb5+' 'Te  ' 'I   ' 'I 1-' 'Xe  ' 'Cs  '
'Cs1+' 'Ba  ' 'Ba2+' 'La  ' 'La3+' 'Ce  ' 'Ce3+' 'Ce4+' 'Pr  ' 'Pr3+'
'Pr4+' 'Nd  ' 'Nd3+' 'Pm  ' 'Pm3+' 'Sm  ' 'Sm3+' 'Eu  ' 'Eu2+' 'Eu3+'
'Gd  ' 'Gd3+' 'Tb  ' 'Tb3+' 'Dy  ' 'Dy3+' 'Ho  ' 'Ho3+' 'Er  ' 'Er3+'
'Tm  ' 'Tm3+' 'Yb  ' 'Yb2+' 'Yb3+' 'Lu  ' 'Lu3+' 'Hf  ' 'Hf4+' 'Ta  '
'Ta5+' 'W   ' 'W 6+' 'Re  ' 'Os  ' 'Os4+' 'Ir  ' 'Ir3+' 'Ir4+' 'Pt  '
'Pt2+' 'Pt4+' 'Au  ' 'Au1+' 'Au3+' 'Hg  ' 'Hg1+' 'Hg2+' 'Tl  ' 'Tl1+'
'Tl3+' 'Pb  ' 'Pb2+' 'Pb4+' 'Bi  ' 'Bi3+' 'Bi5+' 'Po  ' 'At  ' 'Rn  '
'Fr  ' 'Ra  ' 'Ra2+' 'Ac  ' 'Ac3+' 'Th  ' 'Th4+' 'Pa  ' 'U   ' 'U 3+'
'U 4+' 'U 6+' 'Np  ' 'Np3+' 'Np4+' 'Np6+' 'Pu  ' 'Pu3+' 'Pu4+' 'Pu6+'
```

In DIFFaXversion 1.80, atom names are case insensitive. Thus 'SI ' and 'Si ' are equivalent.

An example data line for an atom might be

$$C \qquad 1 \ -0.333333 \ -0.166667 \ -0.125 \ 1.0 \ 1.0$$

This specifies that the first atom is neutral carbon, 'C␣␣␣', ('C' followed by 3 spaces), at cell position (-0.333333 -0.166667 -0.125) relative to $\mathbf{a}$, $\mathbf{b}$ and $\mathbf{c}$, with Debye-Waller B_{iso} factor of 1.0Å^2, and occupancy of 1. Although DIFFaX ignores leading spaces in data lines, care must be taken to ensure that the first non-blank character and the three succeeding characters, including blanks, are part of the atom name. The data line

$$C \ 1 \ -.333333 \ -.166667 \ -.125 \ 1.0 \ 1.0$$

would imply that the atom type is 'C 1 ' (*i.e.* 'C␣1␣'). This would generate an error, since DIFFaX would become confused about the subsequent arguments on the line. The data line

$$C \ 111 \ -.333333 \ -.166667 \ -.125 \ 1.0 \ 1.0$$

would imply that the first atom was of type 'C␣11', with an id number of 1. The subsequent parameters would be read correctly, but DIFFaX would be unable to find atomic scattering data for 'C␣11' in the file `data.sfc` and would display an error message and stop running.

The atom id number is simply a label to help you distinguish between identical atom names. DIFFaX uses these labels when it generates error messages, to identify which atom's data is suspect. The numbers do not need to be consecutive, and the same number can be used more than once, although this entails the risk of creating confusion if error messages are generated.

It is usual to enter the atomic relative coordinates so that they are within the range 0 to 1. However, DIFFaX will accept numbers beyond these bounds. Internally, DIFFaXtruncates coordinates to the 0–1 range.

Coordinates can also be entered as fractions, thus

<pre>
C 1 -1/3 -1/6 -1/8 1.0 1.0
</pre>

is acceptable.

The isotropic B_{iso} factors are in Å^2 units, and are equivalent to $8\pi^2 U_{iso}$, where U_{iso} is the mean square atomic vibration amplitude. The B_{iso} factors must not be negative.

The atom occupancy must be between 0 and 1. DIFFaX carries out a rudimentary check for other atoms that may be on (or near) the same position, including those atoms in potentially neighboring layers, which cumulatively might imply a site occupancy greater than 1.

Lines 13 to 13 + no_atoms(layer 1): Data for the remaining atoms in layer 1, entered as for line 12.

2.8.4 Second Layer.

Line 14 + no_atoms(layer 1): Header for the second layer. If it is structurally distinct from layer 1, it must read LAYER 2. If it is structurally identical to layer 1, then the header can be written LAYER 2 = 1. In this instance, no symmetry or atomic coordinate data is needed and the next line can continue with data for layer 3, or, if there are no more layers, the next section.

Line 15 + no_atoms(layer 1): If structurally distinct from layer 1, then enter symmetry of the layer structure, as for line 11.

Lines 16 + no_atoms(layer 1) to 16 + no_atoms(layer 1) + no_atoms(layer 2): Layer atomic structure data, as for line 12.

Continue adding data for the number of layers specified in line 8.

2.8.5 Examples of the layer structure data section.

Layer structure data describing clustered faulting in diamond might look like

<pre>
LAYER 1
None
C   1   -0.333333   -0.166667   -0.125 1.0 1.0
C   2   0.333333   0.166667   0.125 1.0 1.0

LAYER 2
None
C   1   0.333333   0.166667   -0.125 1.0 1.0
</pre>

```
C       2    -0.333333    -0.166667      0.125  1.0  1.0

LAYER 3
None
C       1    -0.333333    -0.166667     -0.125  1.0  1.0
C       2     0.333333     0.166667      0.125  1.0  1.0

LAYER 4
None
C       1     0.333333     0.166667     -0.125  1.0  1.0
C       2    -0.333333    -0.166667      0.125  1.0  1.0
```

The same data could be entered as

```
LAYER 1
CENTROSYMMETRIC
C       1    -1/3     -1/6     -1/8  1.0  1.0

LAYER 2
CENTROSYMMETRIC
C       1     1/3      1/6     -1/8  1.0  1.0

LAYER 3 = 1

LAYER 4 = 2
```

and would run significantly faster, because DIFFaX will now take advantage of the center of symmetry of the layers, and only two layer structure factors need to be computed. Here, we have exploited the fact that layers 1 and 3, and layers 2 and 4, are structurally identical, and therefore have the same layer scattering factors. With the second data set, DIFFaX knows that it needs to compute only the real components for 2 layer structure factors at each reciprocal lattice point hkl. With the first data set, DIFFaX will compute four complex scattering factors. For structures with many atoms per layer, such as zeolites, a significant speedup can be obtained if centers of symmetry and layer equivalences can be spelled out in the data file. However, in the author's experience, errors can easily creep into such simplifications. For example, the hypothetical centrosymmetric layer

```
LAYER 1
None
O    1    -0.2    -0.2    -0.125  1.0  1.0
Si   2     0.0     0.0     0.0    1.0  1.0
O    3     0.2     0.2     0.125  1.0  1.0
```

can be optimized to

```
LAYER 1
centrosymmetric
O    1    -0.2    -0.2    -0.125  1.0  1.0
Si   2     0.0     0.0     0.0    1.0  0.5
```

Note that the occupancy of the `Si` ' atom at the origin has been halved, since it is shared by the two centrosymmetric halves of the layer cell.

17

2.9 STACKING section.

2.9.1 `STACKING` header.

Line N = 13+2×n + no_atoms(layer 1) + no_atoms(layer 2) + ... + no_atoms(layer n): Header for the stacking definition data. This line must be the keyword `STACKING`. It can be written in upper or lower case. No abbreviation is accepted.

2.9.2 Layer sequencing.

Line N + 1: Describe the layer sequencing in the crystal. There are two options, `EXPLICIT` and `RECURSIVE`. `EXPLICIT` indicates that diffraction intensities are to be calculated for a crystal containing a unique, fixed sequence of layers, which will be described by the user in the lines that follow. `RECURSIVE` indicates that diffraction intensities are to be calculated for a statistical ensemble of crystallites, each with a distinct stacking sequence, but weighted by the probability that such a sequence will occur.

2.9.3 `EXPLICIT` layer sequencing.

Line N + 2: Define the layer sequence. If `EXPLICIT` has been specified in the previous line, this can be entered as an explicit sequence of layers, such as

```
1 1 2 1 2 2 2 3 2 3 2 3 2 1 1 1 3 3 3 3 3 2 1 1 2 3 2 1 2 3 2 2
2 3 3 2 3 2 1 1 1 3 3 3 3 3 2 1
```

The list can occupy more than one line, but the data should be confined to 200 characters per line, including spaces, otherwise some of the layer data will be skipped, or misread. The maximum number of explicit layers that can be defined has been set at 5000. The explicit list must not contain transitions which have zero probability (as defined in the `TRANSITIONS` section, next), as DIFFaX treats this as an error and will stop running. Alternatively, DIFFaX can be asked to generate a sequence of M layers automatically ($M \leq 5000$), by specifying

```
RANDOM M
```

The probability that a specific layer to layer transition will occur in the sequence will be weighted by the transition probabilities listed in the `TRANSITIONS` section, described next. If you are curious about the sequence that the computer generated, this will be listed in the `DUMP` file, which can be requested at run-time (see sections 3.5 and B.3).

If the layer sequencing is specified as `RECURSIVE`, then this line contains the number of layers that the crystal is to contain. Any number larger than $M > 1022$, will be treated as

infinite [1]. Alternatively, an infinite number can be specified by the keyword `INFINITE`.

2.9.4 Examples of the `STACKING` data section.

Typical `STACKING` data sections might look like

```
{consider every possible permutation of a large number of layers}
STACKING
recursive
infinite
```

or

```
{include every possible permutation of 250 layers}
STACKING
recursive
250
```

or

```
{generate a single random sequence of 250 layers}
STACKING
explicit
random 250
```

or

```
{140 layers explicitly sequenced as follows...}
STACKING
explicit
1 1 2 4 2 2 2 3 2 3 2 3 2 1 1 1 3 3 3 3 3 2 1 1 2 2 3 3 2 3
2 3 2 1 2 3 2 2 2 3 3 2 3 2 4 1 1 3 3 3 3 3 2 1 3 3 2 3 2 1
1 1 2 1 2 4 1 2 1 2 3 3 3 3 3 4 1 2 2 1 2 1 2 3 3 2 2 2 3 3
2 3 3 4 2 4 2 1 1 1 1 1 1 1 1 1 1 1 1 2 1 2 2 3 3 2 3 2 1 1 1
1 1 1 3 2 1 3 1 3 4 4 4 2 3 2 1 1 1 3 3
```

2.10   TRANSITIONS section.

2.10.1   `TRANSITIONS` header.

Line N + 3: Header for the stacking transition data. This line must be the keyword `TRANSITIONS`, and is case-insensitive.

---

[1]There is, in fact, a good reason for this strange number, 1022. For the `EXPLICIT` option, DIFFaX must compute the $(M + 1)$th power of a complex matrix $\mathbf{T}$, *i.e.* $\mathbf{T}^{M+1}$. This can be computationally expensive. In the interests of efficiency, this matrix power can be computed quickly by first representing the value of $M + 1$ in binary format, and then computing and storing the values of $\mathbf{T}$, $\mathbf{T}^2$, $\mathbf{T}^4$, $\mathbf{T}^8$ etc... by successive squaring. $\mathbf{T}^{M+1}$ can then be computed by multiplying the appropriate powers of $\mathbf{T}$, the relevant powers being given by the binary flags. In DIFFaX the limit has been set to 10-bit representation, thus the largest value that M+1 can have is $2^{10} - 1 = 1023$.

2.10.2   Layer 1 to layer 1 transition data.

Line N + 4: Stacking data for layer 1 to layer 1 transitions. The data record is usually in the form

$$\texttt{alpha\_ij Rx\_ij Ry\_ij Rz\_ij}$$

where

$\texttt{alpha\_ij}$ = stacking transition probability (between 0 and 1),
$\texttt{Rx\_ij}$ = x component of stacking vector (relative to **a**),
$\texttt{Ry\_ij}$ = y component of stacking vector (relative to **b**),
$\texttt{Rz\_ij}$ = z component of stacking vector (relative to **c**)

a, **b**, and **c** are the cell constants defined on line 6 of the data file (see section 2.7.2 on page 11).

An optional set of anisotropic *Debye-Waller*-type factors, C_{11}, C_{22}, C_{33}, C_{12}, C_{13}, C_{23}, can be specified in parentheses after the first 4 numbers, *i.e.*

$$\texttt{alpha\_ij Rx\_ij Ry\_ij Rz\_ij (C11 C22 C33 C12 C13 C23)}$$

For the want of a better term, and for the author's amusement, these C_{ij} coefficients shall be referred to as the *Fats-Waller* factors in this manual. Note that the ordering of the C_{ij} terms has been modified from that used in versions of DIFFaX prior to version 1.80. Previously, the ordering of the cross-terms had been "cyclic", *i.e.* `C11 C22 C33 C12 C23 C31`. However, it turns out that by convention, *Debye-Waller* anisotropic temperature factor cross-terms are always listed in "anti-cyclic" order in the literature, that is, in the order B_{11}, B_{22}, B_{33}, B_{12}, B_{13}, B_{23} [1]. To comply with this convention, DIFFaX now reads the *Fats-Waller* factors "anti-cyclically".

As for the atomic *Debye-Waller* factors, the *Fats-Waller* temperature factors must be in units of Å^2. The *Fats-Waller* factors are equivalent to specifying an ellipsoidal error spread for the stacking vectors R_{ij}. These are useful for modeling systems where there is some coherence between nearest neighbor layers, but no long-range coherence, as in the case of liquid crystals or pillared clays. If these factors are not specified, this is equivalent to setting them all to zero.

If the stacking probability $\texttt{alpha\_ij}$ is zero, everything on the line after it is ignored and set to zero.

---

[1] Note that C_{31} and C_{13} are equivalent.

2.10.3 Subsequent transition data.

Lines (N + 4) to (N + n*n): Stacking data for the layer i to layer j transitions. The data is expected in the order

```
layer 1 to layer 1
layer 1 to layer 2
.
.
.
layer 1 to layer n

layer 2 to layer 1
layer 2 to layer 2
.
.
.
layer 2 to layer n
.
.
.
layer n to layer 1
layer n to layer 2
.
.
.
layer n to layer n
```

The sum of the probabilities starting from each layer must all sum to 1.0, *i.e.* $\sum_j \alpha_{ij} = 1.0$ for each i, otherwise DIFFaX will assume an error has been made, and will quit.

2.10.4 Examples of the stacking `TRANSITIONS` data section.

Typical stacking `TRANSITIONS` data section might look like

```
TRANSITIONS                     {Header for transitions}
{Transitions from layer 1}
0.7   0.66667   0.333333  1.0    {layer 1 to layer 1, 70% chance}
0.3   0.0       0.0       1.0    {layer 1 to layer 2, 30% chance}

{Transitions from layer 2}
0.3   0.0       0.0       1.0    {layer 2 to layer 1, 30% chance}
0.7  -0.66667  -0.333333  1.0    {layer 2 to layer 2, 70% chance}
```

Stacking vectors can be entered as fractions, thus

```
TRANSITIONS                        {Header for transitions}
{Transitions from layer 1}
0.7   2/3        1/3       1.0    {layer 1 to layer 1, 70% chance}
0.3   0.0        0.0       1.0    {layer 1 to layer 2, 30% chance}


{Transitions from layer 2}
0.3   0.0        0.0       1.0    {layer 2 to layer 1, 30% chance}
0.7  -2/3       -1/3       1.0    {layer 2 to layer 2, 70% chance}
```

Data with *Fats-Waller* factors might look like

```
TRANSITIONS              {Header for transitions}
{Transitions from layer 1} {add a spread in R_z}
```

```
0.1   0.5   0.5   1.0     (0.0 0.0 1.0 0.0 0.0 0.0)
0.9   0.0   0.0   1.0     (0.0 0.0 1.0 0.0 0.0 0.0)


{Transitions from layer 2}
0.9   0.0   0.0   1.0     (0.0 0.0 1.0 0.0 0.0 0.0)
0.1   0.5   0.5   1.0     (0.0 0.0 1.0 0.0 0.0 0.0)
```

or, more compactly

```
0.1 0.5 0.5 1.0 (0.0 0.0 1.0 0.0 0.0 0.0)
0.9 0.0 0.0 1.0 (0.0 0.0 1.0 0.0 0.0 0.0)
0.9 0.0 0.0 1.0 (0.0 0.0 1.0 0.0 0.0 0.0)
0.1 0.5 0.5 1.0 (0.0 0.0 1.0 0.0 0.0 0.0)
```

2.11   Dependence of diffraction symmetry on the stacking probabilities.

It is perfectly legitimate to set a layer transition probability to be equal to 1.0, and all the other transitions from that layer to be 0.0. This will define a perfect unfaulted structure, and DIFFaX is capable of handling this limit. However, at this limit, the probability that the other layers will actually exist in the crystal may become zero or undefined. This sudden disappearance of layers from the ensemble may modify the apparent Laue symmetry of the diffraction data relative to the faulted intermediates. This can be confusing if a series of diffraction data for transition probabilities ranging from 0.0 to 1.0 are calculated, because the end-members may appear to have different symmetry to the intermediate members. This is most apparent in the selected area diffraction patterns.

The fraction of layers of type i that actually occur in a crystal, g_i, is given by the set of simultaneous equations

$$g_i = \sum_{j=1}^{N} g_j \alpha_{ji} \qquad \sum_{i=1}^{N} g_i = 1 \qquad (6)$$

Therefore, in a two-layer crystal system where the transition probabilities are defined to be

$$\alpha_{11} = 0.9999, \quad \alpha_{12} = 0.0001$$
$$\alpha_{21} = 0.0001, \quad \alpha_{22} = 0.9999$$

the frequency of occurrence of layers 1 and 2 in the crystal are $g_1 = g_2 = 0.5$ (*i.e.* 50% of the crystal is layer 1, 50% is layer 2). The symmetry of the diffraction pattern will reflect the fact that, statistically, both layer types and their associated stacking sequences, are equally represented in the typical crystal. However, if the transition probabilities had been set up as

$$\alpha_{11} = 1.0, \quad \alpha_{12} = 0.0$$
$$\alpha_{21} = 0.0, \quad \alpha_{22} = 1.0,$$

which at first glance would appear to be essentially indistinguishable from the previous example, we find that there is no unique solution to the simultaneous equations 6. DIFFaX resolves the ambiguity in this instance by counting the number of α_{ii} terms, N, that equal 1.0, and assigning $g_i = 1/N$ when $\alpha_{ii} = 1.0$, and $g_j = 0.0$ when $\alpha_{jj} \neq 1.0$. This tactic, however, is a compromise solution. There is no *a priori* justification for this $1/N$ assumption, since, in principle, the α_{ii} can all approach the value 1.0 at different asymptotic rates, *i.e.*

$$\alpha_{11} = 1 - \epsilon, \quad \alpha_{12} = \epsilon$$
$$\alpha_{21} = 2\epsilon, \quad \alpha_{22} = 1 - 2\epsilon,$$

as $\epsilon \to 0$. This would then give $g_1 = 2/3, g_2 = 1/3$, and our compromise $1/N$ solution would be inappropriate for this case.

In order to avoid such discontinuous (and sometimes surprising) changes of diffraction symmetry at these limits, it is prudent not to exceed about 0.9999 for transition probability values when computing patterns for a range of transition probabilities. Thus, instead of the series of values $\alpha_{11} = 0.0, 0.1, 0.2 \ldots 0.9, 1.0$, use $\alpha_{11} = 0.0001, 0.1, 0.2 \ldots 0.9, 0.9999$. The transition probability sum, $\sum_j \alpha_{ij}$, must still come to 1.0. When computing powder patterns, errors caused by an unexpected symmetry change can be circumvented by always specifying the symmetry as `UNKNOWN` in the data file (see section 2.7.3). DIFFaX will then catch any changes in symmetry, and modify the integration range in reciprocal space accordingly. However, this will be of no help when computing selected area diffraction patterns, where the change in symmetry is visually obvious from the distribution of diffraction spots. In this case, non-zero probabilities should be used.

2.12   A more complex example. Fault clustering.

Fault clustering can be simulated by increasing the number of layer types. To describe clustering in diamond, we need at least two versions of layer type 1 and at least two versions of layer type 2, in order to differentiate between stacking histories. Thus, a layer type 1, when preceded by a layer type 1, might connect to subsequent layers in a different manner to a layer type 1 that is preceded by a layer type 2. This memory, or "Reichweite", can be simulated by differentiating between such layers, even though they may be structurally identical. A data file describing clustered twin faults in diamond might look like (see appendix A.2 for more details).

```
{data file for clustered faults in diamond}
{Probability of a fault in diamond cubic stacking = 10%}
{Probability of a fault in lonsdaleite hexagonal stacking = 20%}

{There are four layers in total, two of which are}
{structurally distinct.  They are related by a mirror}
```

23

```
{in the a-b plane, and are labeled layers A and A'.}
{Layer 1 is an A layer which is preceded by an A layer}
{Layer 2 is an A' layer which is preceded by an A layer}
{Layer 3 is an A layer which is preceded by an A' layer}
{Layer 4 is an A' layer which is preceded by an A' layer}
{1 - 1 is a cubic stacking transition}
{1 - 2 is a stacking fault in a cubic sequence}
{1 - 3 cannot occur}
{1 - 4 cannot occur}
{2 - 1 cannot occur}
{2 - 2 cannot occur}
{2 - 3 is a hexagonal stacking transition}
{2 - 4 is a fault in a hexagonal sequence}
{3 - 1 is a fault in a hexagonal sequence}
{3 - 2 is a hexagonal stacking transition}
{3 - 3 cannot occur}
{3 - 4 cannot occur}
{4 - 1 cannot occur}
{4 - 2 cannot occur}
{4 - 3 is a stacking fault in a cubic sequence}
{4 - 4 is a cubic stacking transition}

INSTRUMENTAL
X-RAY
1.5418
PSEUDO-VOIGT .89 -.32 .08 .6 TRIM

STRUCTURAL
2.52 2.52 2.06 120.0
unknown 1 {We expect -3m.  Let DIFFaX decide to within a tolerance of 1%.}
4


LAYER 1
CENTROSYMMETRIC
C       1      -1/3      -1/6      -1/8      1.0      1.0


LAYER 2
CENTROSYMMETRIC
C       1       1/3       1/6      -1/8      1.0      1.0

LAYER 3 = 1 {layer 3 is structurally identical to layer 1}

LAYER 4 = 2 {layer 4 is structurally identical to layer 2}

STACKING
recursive
infinite


TRANSITIONS
{1}
0.90      2/3       1/3       1.0
0.10      0.0       0.0       1.0
0.00      0.0       0.0       0.0 {1-3 Transition cannot occur}
0.00      0.0       0.0       0.0 {1-4 Transition cannot occur}


{2}
0.00      0.0       0.0       0.0 {2-1 Transition cannot occur}
0.00      0.0       0.0       0.0 {2-2 Transition cannot occur}
0.80      0.0       0.0       1.0
```

```
0.20    - 2/3    - 1/3     1.0

{3}
0.20     2/3      1/3      1.0
0.80     0.0      0.0      1.0
0.00     0.0      0.0      0.0 {3-3 Transition cannot occur}
0.00     0.0      0.0      0.0 {3-4 Transition cannot occur}

{4}
0.00     0.0      0.0      0.0 {4-1 Transition cannot occur}
0.00     0.0      0.0      0.0 {4-2 Transition cannot occur}
0.10     0.0      0.0      1.0
0.90    - 2/3    - 1/3     1.0
```

3 RUNNING DIFFaX.

Over the course of its development, DIFFaX has been tested on several different computer systems and with many different fortran compilers. DIFFaX has been written so that its operation is, as far as is possible, independent of the computer's operating system. There are two ways to run DIFFaX. (i) It can be run interactively, whereby the user is expected to respond to certain prompts via the keyboard, or (ii) it can be run using a control file, which is a text file containing pre-composed answers to DIFFaX's prompts (see section 5 on page 40 for a fuller description of control files). In order to run correctly, DIFFaX expects to find the standard scattering factor data file `data.sfc`, and (if used) the control file `control.dif`, in the same directory as the executable DIFFaX program. If the user wishes to run DIFFaX interactively, the control file `control.dif` must be renamed or removed, otherwise DIFFaX will read it by default. There are currently two Macintosh versions, MacDIFFaX_881 and MacDIFFaX_PPC, which run on 68k-based CPUs and PowerPC-based CPUs respectively. The MacDIFFaX programs do not need the data file `data.sfc`, as this data is built into MacDIFFaX as a resource.

3.1 Invoking DIFFaX.

Once the data file has been written, DIFFaX is ready to run. On the VAX, Cray or Macintosh, the DIFFaX program can usually be invoked by one of these commands

VAX VMS: `run DIFFaX`
unix: `DIFFaX`
Macintosh: "double-click" the application icon "MacDIFFaX_881" (or "MacDIFFaX_PPC").

3.2 DIFFaX start-up screen.

If all is well, the user will be greeted by a start-up message resembling

```
****************************************************
****************************************************
*                                                  *
*  DDDD     II   FFFFFF   FFFFFF           X   X   *
*  D   D    II   F        F          aaaa   X X    *
*  D    D   II   FFFF     FFFF     a    a    X     *
*  D   D    II   F        F        a   aa   X X    *
*  DDDD     II   F        F         aaa a   X   X  *
*                                                  *
****************************************************
***************** DIFFaX v1.801 ******************
****************************************************
************** 14th JULY, 1995 *****************
****************************************************
*                                                  *
*    A computer program for calculating            *
*    Diffraction Intensity From Faulted Crystals    *
*                                                  *
*    Authors: Michael M. J. Treacy                 *
*             Michael W. Deem                      *
*                                                  *
****************************************************

 Enter name of structure file.
```

3.3 Data file name.

The user should respond by typing in the name of the file containing the structure data, and typing a carriage return. If all is well, then, after a few moments, the following lines will appear

```
 Enter name of structure file.
dia.dat
 Looking for structure data file 'dia.dat'
 Opening structure file 'dia.dat'
 Looking for scattering factor data file 'data.sfc'
 Opening scattering factor data file 'data.sfc'
 Enter 1 for DUMP
```

Note, the user's responses are left-indented.

3.4 Data file name errors.

It is common to get errors at this early stage. If the user mistypes the filename (*i.e.* `dia.dot` instead of `dia.dat`), or the file was saved in a directory other than the one that contains DIFFaX, then the user will get an error message that, depending on the operating system, may look like

```
 Enter name of structure file.
dia.dot
```

26

```
 Looking for structure data file 'dia.dot'
 Problems opening structure data file 'dia.dot'
 IOSTAT =     2
 DIFFaX was terminated abnormally.
```

If the standard scattering factor data file, `data.sfc` is stored in a directory other than the one that contains DIFFaX, then the user will get an error message that, depending on the operating system, may look like

```
 Enter name of structure file.
dia.dat
 Looking for structure data file 'dia.dat'
 Opening structure file 'dia.dat'
 Looking for scattering factor data file 'data.sfc'
 Problems opening scattering factor file 'data.sfc'
 IOSTAT =     2
 DIFFaX was terminated abnormally.
```

Make sure that the full path to the data file is correctly specified (*i.e.* under unix a file path such as `../diamond/dia.dat` could be specified), and that the standard scattering factor data file `data.sfc` is in the same directory as DIFFaX (unless the user is using the bare bones Macintosh version MacDIFFaX).

3.5 Generating a dump file.

If there were no problems opening the data files, the next step is to decide whether or not to generate a dump of the data file, as DIFFaX interpreted it. If this is the first time that this file is being run, it is a good idea to create a dump file, just to make sure there are no surprises. Responding with a '1' will generate a dump. Anything else will not generate a dump. DIFFaX then reads the data file and `data.sfc`. If there were no errors encountered, the screen will appear as follows.

```
 Enter 1 for DUMP
1
 Reading 'dia.dat'
 'dia.dat' read in.
 Reading scattering factor datafile 'data.sfc'. . .
 Scattering factor data read in.
 Enter 1 for full atomic position dump
```

Whether or not a dump was requested, DIFFaX carries out a set of checks on the data file, to ensure that the values entered are reasonable, or physically possible. At this stage, if the user opted for a dump, a choice is presented whether or not to list all the atomic coordinate data with a full listing of their scattering factor parameters, in the dump file. Responding with a '1' will give a full listing. Anything else will give a shortened listing.

```
 Enter 1 for full atomic position dump
1
 Writing data dump to file 'dia.dmp'. . .
 Enter 1 for a dump of the symmetry evaluations
```

Note that the dump file has been named `dia.dmp`. See section 6 for a description of file naming conventions.

3.6 Generating a dump of symmetry evaluations.

If there were no problems generating the data dump file, the user will now be asked whether or not to create a dump of DIFFaX's evaluation of the Laue group symmetry of the diffraction intensities in reciprocal space. This facility is useful mainly for tracking down some of the more mysterious errors in the layer atomic data, when the diffraction symmetry is different to that expected. Responding with a '1' will give a listing of the symmetry evaluations. Anything else will not generate a symmetry dump.

```
 Enter 1 for a dump of the symmetry evaluations
1
 Checking for conflicts in atom positions . . .
 Evaluating point group symmetry of diffraction data. . .
 Writing symmetry data to file 'dia.sym'. . .
 The diffraction data fits the point group symmetry '6/MMM'
   with a tolerance of one part in  12376
 Enter function number:
 0 POINT, 1 STREAK, 2 INTEGRATE, 3 POWDER PATTERN, 4 SADP
```

Note that the symmetry evaluation data file is called `dia.sym`. See section 6 for a description of file naming conventions.

3.7 The main menu.

If there were no problems generating the symmetry evaluations file, the user is now provided with five options for the type of diffraction intensity information required.

3.7.1 Intensity at a reciprocal lattice point, hkl.

Menu choice '0' allows the user to calculate the intensity at a reciprocal lattice point hkl, and will also give information about the scattering angle in degrees, the structure factors of each layer, and the ensemble average interference wavefunctions when each of the various layers is at the origin of the crystal. This menu allows the user to repeat the operation for as many points as desired. The user may return to the main menu when done. An example of a typical run is listed below

```
 Enter function number:
 0 POINT, 1 STREAK, 2 INTEGRATE, 3 POWDER PATTERN, 4 SADP
0

 CALCULATING INTENSITY AT A POINT. . .
 Enter h, k, l
1 0 0
 2theta =   41.371      degrees
 d =    2.1824
```

```
 1/d =  0.45821

 Layer scattering factors
 f( 1) = ( -3.022460    , 0.0000000   )
 f( 2) = ( -3.022460    , 0.0000000   )

 Average scattered wavefunctions
 psi( 1) = ( -2.375548    , 0.8722551   )
 psi( 2) = ( -2.375548    ,-0.8722551   )

 Intensity at   1  0 0.00000    =   4.083575
 Another point? (y/n)
y

 CALCULATING INTENSITY AT A POINT. . .
 Enter h, k, l
3 0 0
    3  0 0.00000     exceeds 180 degree scattering angle!
 Re-enter. . .
 Enter h, k, l
2 0 0
 2theta =   89.898     degrees
 d =   1.0912
 1/d =  0.91643

 Layer scattering factors
 f( 1) = ( -1.438112    , 0.0000000   )
 f( 2) = ( -1.438112    , 0.0000000   )

 Average scattered wavefunctions
 psi( 1) = ( -1.130309    ,-0.4150323   )
 psi( 2) = ( -1.130309    , 0.4150323   )

 Intensity at   2  0 0.00000    =  0.5914296
 Another point? (y/n)
n

 Enter 1 to return to function menu.
1
 Enter function number:
 0 POINT, 1 STREAK, 2 INTEGRATE, 3 POWDER PATTERN, 4 SADP
```

3.7.2 Intensity trace along a reciprocal lattice streak, from hkl_0 to hkl_1.

Menu choice '1' will allow the user to calculate the intensity profile as a function of l for given values of h and k. The results are saved to file. The user will be prompted to enter values for h and k, the beginning, l_0, and ending, l_1, values of l, and the stepsize Δl along l. Naturally, the values should be such that $l_1 > l_0$, and $\Delta l < l_1 - l_0$. The user will also be asked whether or not to use adaptive quadrature to perform the integration. If the interval $l_1 - l_0$ is large, adaptive quadrature will be more accurate, although about 3 times slower than non-adaptive. Depending on the computer, and the number of steps needed to span the interval (equal to $(l_1 - l_0)/\Delta l$), this calculation may take a few minutes. DIFFaX will print out progress reports, announcing what value of l has been reached.

```
 Enter function number:
 0 POINT, 1 STREAK, 2 INTEGRATE, 3 POWDER PATTERN, 4 SADP
1
```

```
 CALCULATING INTENSITY ALONG A STREAK. . .
 Enter 1 for adaptive quadrature
1
 Enter h, k, l0, l1, delta l
1 0 0 1 .01
 Writing streak data to file 'dia.str'. . .
 Reached l =     0.04000
 Reached l =     0.09000
 Reached l =     0.14000
 Reached l =     0.19000
 Reached l =     0.24000
 Reached l =     0.29000
 Reached l =     0.34000
 Reached l =     0.39000
 Reached l =     0.44000
 Reached l =     0.49000
 Reached l =     0.54000
 Reached l =     0.59000
 Reached l =     0.64000
 Reached l =     0.69000
 Reached l =     0.74000
 Reached l =     0.79000
 Reached l =     0.84000
 Reached l =     0.89000
 Reached l =     0.94000
 Reached l =     0.99000
 Streak data file, 'dia.str' written to disk.
 Enter 1 to return to function menu.
1
 Enter function number:
 0 POINT, 1 STREAK, 2 INTEGRATE, 3 POWDER PATTERN, 4 SADP
```

Note that the streak data file is called `dia.str`. See section 6 for a description of file naming conventions. The file `dia.str` is written out in tab-delimited text format. Each line contains the following information

'l-value' tab 'intensity' carriage return

3.7.3   Integrating intensity along a streak, within an interval hkl_0 to hkl_1.

Menu choice '2' will allow the user to calculate the integrated intensity between two l values for given values of h and k. The user will be prompted to enter values for h and k, and the beginning and ending values of l. The user will also be asked whether or not to use adaptive quadrature to perform the integration. If the interval $l_1 - l_0$ is large, adaptive quadrature will be more accurate, although about 3 times slower than non-adaptive. Typical output will look like the following.

```
 Enter function number:
 0 POINT, 1 STREAK, 2 INTEGRATE, 3 POWDER PATTERN, 4 SADP
2

 INTEGRATING INTENSITY IN AN INTERVAL ALONG l. . .
 Enter 1 for adaptive quadrature.
1
```

```
 Enter h, k, l0, l1
0 0 .95 1.05
 Integrated intensity =   12.354973
 Enter 1 to integrate another interval.
1

 INTEGRATING INTENSITY IN AN INTERVAL ALONG l. . .
 Enter 1 for adaptive quadrature.
0
 Enter h, k, l0, l1
0 0 .95 1.05
 Integrated intensity =   12.354973
 Enter 1 to integrate another interval.
1

 INTEGRATING INTENSITY IN AN INTERVAL ALONG l. . .
 Enter 1 for adaptive quadrature.
1
 Enter h, k, l0, l1
0 0 .999 1.001
 Integrated intensity =   11.134940
 Enter 1 to integrate another interval.
0
 Enter 1 to return to function menu.
1
 Enter function number:
 0 POINT, 1 STREAK, 2 INTEGRATE, 3 POWDER PATTERN, 4 SADP
```

If the interval $l_1 - l_0$ is small, as in the above example, adaptive and non-adaptive quadrature usually give similar results. However, full adaptive integration takes significantly more time. In the above example, the reciprocal lattice point 001 is a sharp spot, thus intensity varies rapidly in the vicinity of this spot. Although the adaptive algorithm senses the rapid variation and adjusts its integration stepsize accordingly, the result is not necessarily more accurate: the non-adaptive algorithm is robust enough for this example. However, if $l_1 - l_0$ is too large, the answers may diverge. The cautious user should explore the interaction between the stepsize $l_1 - l_0$ and the need for full adaptive integration.

3.7.4   Selected area diffraction pattern.

Menu choice '4' will allow selected area diffraction patterns to be calculated. This option is most useful for calculating kinematical electron diffraction patterns.

```
Enter function number:
0 POINT, 1 STREAK, 2 INTEGRATE, 3 POWDER PATTERN, 4 SADP
4

CALCULATING SELECTED AREA DIFFRACTION PATTERN. . .
Enter 1 for adaptive quadrature.
1
Choose a plane in reciprocal space to view.
      the l-axis is included by default.
1: k = 0.   2: h = 0.   3: h = k.   4: h = -k.
2
Enter maximum value of l.
2
Choose the bit-depth of the image.
```

```
    8: - 8-bits   16: - 16-bits
16
File will be saved in unsigned 16-bit format.
Choose intensity scaling
    0: - Logarithmic   1: - Linear
1
Enter a brightness (must be +ve)
  1 - scales intensities to the range 0 - 32767
 10 - scales intensities to the range 0 - 327670,
      but values above 65535 will be saturated
5
h =   0 k =   0              'dia.dat'
l =   -0.00781 intensity =  0.00000
l =    0.08594 intensity =  0.13091E-01
l =    0.17969 intensity =  0.32829E-02
l =    0.27344 intensity =  0.14872E-02
l =    0.36719 intensity =  0.87177E-03
l =    0.46094 intensity =  0.60308E-03
l =    0.55469 intensity =  0.48016E-03
l =    0.64844 intensity =  0.44465E-03
l =    0.74219 intensity =  0.50383E-03
l =    0.83594 intensity =  0.80688E-03
l =    0.92969 intensity =  0.33175E-02
l =    1.02344 intensity =  0.94814E-02
l =    1.11719 intensity =  0.39026E-03
l =    1.21094 intensity =  0.92740E-04
l =    1.30469 intensity =  0.34490E-04
l =    1.39844 intensity =  0.16146E-04
l =    1.49219 intensity =  0.88008E-05
l =    1.58594 intensity =  0.53895E-05
l =    1.67969 intensity =  0.36399E-05
l =    1.77344 intensity =  0.26848E-05
l =    1.86719 intensity =  0.21521E-05
l =    1.96094 intensity =  0.18708E-05
h =   0 k =   1              'dia.dat'
l =   -0.00781 intensity =  0.63827E-01
l =    0.08594 intensity =  0.73861E-01
l =    0.17969 intensity =  0.11971
l =    0.27344 intensity =  0.29790
l =    0.36719 intensity =  0.35620
l =    0.46094 intensity =  0.92463E-01
l =    0.55469 intensity =  0.27839E-01
l =    0.64844 intensity =  0.61255E-03
l =    0.74219 intensity =  0.65866E-02
l =    0.83594 intensity =  0.85806E-02
l =    0.92969 intensity =  0.96500E-02
l =    1.02344 intensity =  0.12521E-01
l =    1.11719 intensity =  0.20760E-01
l =    1.21094 intensity =  0.50273E-01
l =    1.30469 intensity =  0.18049
l =    1.39844 intensity =  0.15803
l =    1.49219 intensity =  0.82232E-01
l =    1.58594 intensity =  0.86818E-01
l =    1.67969 intensity =  0.52458E-01
l =    1.77344 intensity =  0.68915E-02
l =    1.86719 intensity =  0.99358E-03
l =    1.96094 intensity =  0.69756E-04
h =   0 k =   2              'dia.dat'
l =   -0.00781 intensity =  0.92443E-02
l =    0.08594 intensity =  0.10797E-01
l =    0.17969 intensity =  0.17989E-01
l =    0.27344 intensity =  0.46839E-01
l =    0.36719 intensity =  0.59557E-01
l =    0.46094 intensity =  0.16694E-01
```

```
l =    0.55469 intensity =  0.55018E-02
l =    0.64844 intensity =  0.13381E-03
l =    0.74219 intensity =  0.16145E-02
l =    0.83594 intensity =  0.23738E-02
l =    0.92969 intensity =  0.30345E-02
l =    1.02344 intensity =  0.44995E-02
l =    1.11719 intensity =  0.85570E-02
l =    1.21094 intensity =  0.23813E-01
l =    1.30469 intensity =  0.98215E-01
l =    1.39844 intensity =  0.98521E-01
l =    1.49219 intensity =  0.58444E-01
l =    1.58594 intensity =  0.69768E-01
l =    1.67969 intensity =  0.47128E-01
l =    1.77344 intensity =  0.68275E-02
l =    1.86719 intensity =  0.10684E-02
l =    1.96094 intensity =  0.00000
Writing SADP data to file 'dia.sadp7'. . .
 256 x  256 pixels: 16 bits deep.
Enter 1 to return to function menu.

1
 Enter function number:
 0 POINT, 1 STREAK, 2 INTEGRATE, 3 POWDER PATTERN, 4 SADP
```


Figure 1: Selected area electron diffraction pattern simulation for diamond/lonsdaleite, showing the $0kl$ spots for a diamond-lonsdaleite stacking fault probability of 0.3. (See data file in section A.1).

The user is first asked to choose a plane in reciprocal space to view. Choosing '1' views the plane containing $h0l$ spots; '2' the plane containing $0kl$ spots; '3' the plane containing hhl spots and '4' the plane containing $h\overline{h}l$ spots. Then the user is asked to enter the maximum value of l to go out to. The maximum values of h and k will be automatically defined by this l value, since the calculated diffraction pattern will be square. The user specifies whether an

8 bit or a 16 bit pixel depth is required. The user is then asked whether or not to save the intensity data on a linear or logarithmic scale, by entering '0' or '1' respectively. In linear mode, an intensity scaling factor can be specified. A factor of 1 means that the intensity range (excluding the zero order beam) will be compressed so as to fit within the range 0 to 255 (8 bit binary) or 0 to 32767 (signed 16 bit binary)[1]. A factor of 10 means that the intensity range will be fitted within the range 0 to 2550 (or 0 to 327670), but all values above 255 (32767) will be saturated, and will be set equal to 255 (32767). Since the calculation may take a little while, DIFFaX prints out a progress report, listing the current value of hkl it has reached. Note that the selected area diffraction pattern data file is saved as `dia.sadp`. See section 6 for a description of file naming conventions. The file `dia.sadp` is saved as an unformatted binary file. Each pixel is saved either as an 8 bit number between 0 and 255 or a 16 bit number between 0 and 32767. The image size is 256×256. To display the file the user needs a graphics utility program that can open such binary files. Figure 1 shows the selected area diffraction pattern, `dia.sadp`, as printed from the Macintosh program $Image$[2].

3.7.5 Powder pattern.

Menu choice '3' will allow the user to calculate the powder pattern within a range of 2θ values. The user will be prompted for the beginning and ending 2θ values, and the 2θ stepsize. Then the user will be asked whether or not to use full adaptive quadrature on the broad peaks. When the fault probabilities are high (*i.e.* close to 50%) it is usually safe to ignore the full adaptive quadrature option. However, there is always the risk of errors near any sharper intensity features that may not have been anticipated. Next the user is asked whether or not to use full adaptive quadrature over all l values on the hk rows known to contain sharp peaks. DIFFaX attempts to establish which values of hk are rows of sharp spots. These will always be integrated full-adaptively. If the reply is '1' to this question, DIFFaX will employ full-adaptive integration over all l values, even though most of the intensity is contained near the sharp spots, with relatively nothing in between. Avoiding full adaptive quadrature speeds up the calculation a little. However, there is the risk of integration errors, which may result in some sharp peaks being missed altogether. It is safest always to select full-adaptive quadrature for both these options. Where computation speed is an important criterion and a large series of calculations are to be carried out, it would be prudent to compare the

---

[1] DIFFaX stores the 16-bit data as two sequential bytes. The Fortran code will have to be modified if reverse byte ordering is required.

[2] $Image$ is an image-processing program for the Macintosh (requires 8-bit video), originally written by Wayne Rasband, National Institutes of Health, Bethesda, MD. The $Image$ program, including Macintosh THINK pascal source code and documentation, is in the public domain and may be freely copied and distributed. $Image$ can be obtained via anonymous ftp from `zippy.nimh.nih.gov[128.231.98.32]`. Enter "anonymous" as the user name, and your email address as the password. The `/pub/nih-image` directory contains the latest version of $Image$.

results using the adaptive and non-adaptive options, and explore how they interact with the 2θ stepsize. Making the stepsize too small will also slow up the calculation. For most calculations, it is a matter of trial and error to establish the best combination of parameters. Because this is a time consuming calculation, DIFFaX will periodically print out a progress report, announcing the values of h and k reached. When finished, DIFFaX will inspect the intensities calculated, particularly those near the origin, where the intensities can swamp the values at other angles. These will be suppressed if the TRIM option was included in the instrumental broadening input of the structure data file. If the data file has requested it, an instrumental broadening function will be applied to the trimmed data. The 2θ values, the raw data, and the broadened data are then written to a '.spc' file. If running interactively, DIFFaX will then end. Example screen output is listed below.

```
 Enter function number:
 0 POINT, 1 STREAK, 2 INTEGRATE, 3 POWDER PATTERN, 4 SADP
3

 CALCULATING POWDER DIFFRACTION PATTERN. . .
 Enter angular range:
   2theta min, 2theta max, 2theta increment.
0 170 0.05
 Enter 1 for adaptive quadrature on broad peaks.
1
 Enter 1 for adaptive quadrature over all l values
 on rows with "sharp" spots
1
 Integrating along l at    0   0      'dia.dat'
 Full adaptive integration
 Integrating along l at    1   0      'dia.dat'
 Full adaptive integration
 Integrating along l at    1   1      'dia.dat'
 Full adaptive integration
 Integrating along l at    2   0      'dia.dat'
 Full adaptive integration
 Integrating along l at    2   1      'dia.dat'
 Full adaptive integration
 Adding pseudo-Voigt instrumental broadening . . .
 Writing spectrum data to file 'dia.spc'
 Spectrum written.

 DIFFaX ended normally.
```

The file dia.spc is written out in tab-delimited text format. Each line contains the following information

'2θ degrees' tab 'unbroadened intensity' tab 'broadened intensity' carriage return

The broadened intensity is not written if the data file specified NONE in the line for instrumental broadening type.

```
*******************************************************
***********
*
*   DDDD
*   D   D
*   D    D
*   D    D
*   DDDD
*
***********
***********
***********
***********
***********
*
*    A comp
*    Diffra
*
*    Authors: Michael M. J. Treacy                *
*             Michael W. Deem                     *
*                                                 *
*******************************************************
```

Enter name of structure file.

Figure 2: MacDIFFaX "Open data file..." dialog box.

3.8   Running MacDIFFaX on the Macintosh.

DIFFaX is usually distributed by the author on a double-sided 1.4MB-formatted Macintosh diskette. There are two Macintosh-compatible precompiled versions of DIFFaX supplied on this diskette, named MacDIFFaX_881 and MacDIFFaX_PPC. These were compiled using the Language Systems MPW fortran compilers for Motorola 68k and PowerPC processors respectively. MacDIFFaX_881 will run on a Macintosh II style machine, with a 68020, 68030 or 68040 CPU, equipped with a 68881, 68882 or 68883 math co-processor. MacDIFFaX_881 will not run on the PowerMac machines – MacDIFFaX_PPC is needed for the PowerMac. To maintain compatibility with the mainframe version, MacDIFFaX_881 and MacDIFFaX_PPC are bare-bones applications, and thus, regretfully, do not take much advantage of the Macintosh interface. They are System 7 compatible, and can run in the background. There are a few minor changes to the interface described previously. In the discussion below, "MacDIFFaX" refers generically to either "MacDIFFaX_881" or "MacDIFFaX_PPC".

- The file `data.sfc` is built into MacDIFFaX as a resource, and therefore is not needed.

- If the user is running interactively, the data file is selected using the Macintosh "Open File" dialog box (see Fig. 2). Output files will be saved to the same directory as the input data files.

- When running with the control file `control.dif`, the behavior of MacDIFFaX is identical to that described above for the unix version. However, if data files are not stored in the same directory as MacDIFFaX, then the full path names should be specified in the control file, *i.e.* `Hard Disk:Diamond folder:dia.dat`, instead of just `dia.dat`.

- The default value of the "detune" parameter in MacDIFFaX is 0.001. Ordinarily, this parameter need never be changed (see discussion in Section 8.3 on page 49). To change the detune parameter, you need to use a Macintosh resource editing tool, such as Resedit[TM] or Resorcerer[TM], to modify a string resource in MacDIFFaX (see below).

3.8.1   Modifying the "detune" parameter in MacDIFFaX.

Launch Resedit and open the MacDIFFaX application. The resources used by MacDIFFaX should appear in a window as shown below.

Figure 3: MacDIFFaX resources as viewed by Resedit[TM].

Open the `STR#` resources by double-clicking the `STR#` icon. The window shown below should appear, containing the `STR#` resources.

Double click on the line `1050        9    ''Detune Value''` , and a dialog box resembling the one below will appear.

The editable string `1.0e-3` is the default detune value. This can be changed to the desired value, (*i.e.* to `1.0e-4`, or `0.0001`, which is the same thing) by typing the new value into the edit field. The changes should be saved, and then quit Resedit. USER BEWARE: a detune value of 0, or a value too close to zero, may produce unpredictable results. 0.001 has been found to work best for most powder pattern simulations.

Figure 4: MacDIFFaX string resources.

4 Speeding up DIFFaX computations.

Despite the fact that the DIFFaX code is replete with tricks that attempt to speed up the computation, DIFFaX runs can take a considerable time to finish, particularly for powder patterns, streaks and selected area diffraction patterns. The most computationally expensive part of the DIFFaX code is the intensity integration. For a powder pattern computation DIFFaX cannot get away with merely sampling the intensity at integral multiples of h, k and l, as can be done for perfectly periodic crystals. DIFFaX needs to integrate along the reciprocal lattice vector l in variable steps of Δl such that $\Delta\theta = \theta(h, k, l + \Delta l) - \theta(h, k, l)$ is constant. Depending on the value of $\Delta\theta$ chosen by the user, Δl can be quite large relative to the width of a typical sharp peak, particularly when l is small or zero. To detect reliably such sharp peaks, DIFFaX employs an adaptive 16-point Gauss-Legendre integration algorithm. In Gauss-Legendre integration, the intensity in the interval Δl is sampled at 16 points. These points are not uniformly spaced, but are carefully chosen so as to sample the intensity optimally. The 16 intensities are added, after being multiplied by equally carefully chosen weights. If the intensity is varying smoothly, the integration can be remarkably accurate. In adaptive integration, the region Δl is further subdivided into two regions, each of length $\Delta l/2$, and the 16-point integration is re-applied to each region, and the results added. If the first 16-point result, and the subsequent 32-point result give identical answers within some error limit, DIFFaX moves on to the next region. If the answers do not agree, usually because the intensity varies rapidly within the region Δl, the regions are repeatedly subdivided and re-integrated until a self-consistent answer is obtained. Adaptive integration,

Figure 5: MacDIFFaX string resource for modifying the "detune" parameter.

therefore, takes at least three times as long to perform as non-adaptive integration (at least 48 intensity samplings versus 16 intensity samplings).

The user can control whether or not DIFFaX uses adaptive integration. The user is prompted twice

```
 Enter 1 for adaptive quadrature on broad peaks.
```

and

```
Enter 1 for adaptive quadrature over all l values
 on rows with ''sharp'' spots
```

Entering a "0" in response to these prompts disables adaptive integration.

DIFFaX attempts to make intelligent guesses as to when it is encountering a row of sharp spots, so generally it is safe to answer "0" to the latter prompt. DIFFaX will always perform adaptive integration on the sharp peak itself, but will ignore the vast expanses of low intensity that lie between them. However, there is a danger to switching off adaptive integration on broad peaks. In those instances when fault probabilities are low, peaks that are normally susceptible to broadening, can in fact be quite sharp. It generally is safe to switch off adaptive integration for broad peaks, but the user should be aware that there are risks at the extreme values of stacking probability.

DIFFaX can store spectra with up to 20,000 points. The number of points, N, is given by $N = (\theta_{\max} - \theta_{min})/\Delta\theta$. The time-conscious user should refrain from overzealously specifying unnecessarily small values for $\Delta\theta$. Clearly $\Delta\theta$ should be somewhat smaller than the instrumental peak broadening half-width that is specified in the data file. However, there is usually little to be gained by making $\Delta\theta$ significantly less than about one-tenth the instrumental peak broadening half-width. In fact, a ratio of about one-third or one-fourth will usually be adequate for all but the most demanding graphics. Smaller values of $\Delta\theta$ will increase the number of regions to be integrated and will proportionately slow down the computation.

Another rate-limiting factor is the number of layers, n. The matrix inversion takes $O(n^3)$ to compute. Further, the computation time is linear in the number of atoms in a layer.

If a layer is centrosymmetric, specifying CENTROSYMMETRIC in the data file, and listing only the asymmetric half of the atoms (and being careful to halve the occupancies of those atoms that are shared by the two asymmetric halves) will speed up the computation considerably. This is not just because there are fewer atoms, but also because the imaginary components of the wave functions can be ignored. If all layers are centrosymmetric, this trick can more than halve the computation time. DIFFaX does not accept other symmetry descriptions for layer unit cells, such as rotational or mirror symmetries, because there is no significant speed benefit.

When two layers are structurally identical, but merely stack differently, making use of the layer equivalence description (see section 2.8.4 on page 16),

LAYER 2 = 1

can save a little time.

Finally, be warned that the PSEUDO-VOIGT peak-broadening option is computationally expensive (section 2.6.8, page 9). Unless it is vitally necessary, use the fixed-width GAUSSIAN (section 2.6.6, page 7) or LORENTZIAN (section 2.6.7, page 8) options, as these are substantially faster.

5   DIFFaX control files.

In normal use, DIFFaX will be used to run batches of data files, usually with differing stacking probabilities. After the run, the powder diffraction simulations are compared with data for the best fit. A typical run might require 11 files for stacking probabilities ranging from 0.001 to 0.999 inclusive (see the discussion in section 2.11 on page 22 for an explanation of why I deliberately avoided specifying the range 0.0 to 1.0). Such a computation can take DIFFaX a considerable time, which makes the interactive mode an unsuitable way to execute such a run.

DIFFaX provides a way to run batch jobs by storing pre-composed answers to the DIFFaX prompts in a control file. The control file default name is "`control.dif`", and should be stored in the same directory as the DIFFaX executable code. A simple way to create a control file is to try a dummy run of DIFFaX, and to record the responses to the prompts. Terminate the DIFFaX run, and create the `control.dif` file with the responses listed one per line. For example, a control file for a typical powder pattern run would look like

```
dia001.dat          {data file name}
0                   {no dump file}
0                   {no symmetry file}
3                   {powder pattern}
0 100 .05           {theta_min, theta_max, delta_theta}
1                   {do adaptive quadrature on broad peaks}
1                   {do adaptive quadrature on all l for rows of sharp spots}
dia100.dat
0
0
3
0 100 .05
1
1
dia200.dat
0
0
3
0 100 .05
1
1
dia300.dat
0
0
3
0 100 .05
1
1
dia400.dat
0
0
3
0 100 .05
1
1
dia500.dat
0
0
3
0 100 .05
1
1
dia600.dat
0
0
3
0 100 .05
1
1
dia700.dat
0
0
3
0 100 .05
```

```
1
1
dia800.dat
0
0
3
0 100 .05
1
1
dia900.dat
0
0
3
0 100 .05
1
1
dia999.dat
0
0
3
0 100 .05
1
1
end                 {no more files, end normally}
```

Note that comments {in curly braces} can be embedded in the control file. At the end of each run on a data file, DIFFaX reads the next line for a new data file name. If it finds a legal file name, it will re-run using the new data. A line containing the keyword "end", tells DIFFaX that the control file is ended. It is not fatal to leave off the keyword end, however, erroneous, and possibly confusing, error messages will be generated.

The control file can also be used to compute the POINT, STREAK, INTEGRATE and SADP options. Care should be taken to compose responses in the correct order, usually by trying a dummy run of DIFFaX and recording the responses.

An example control file, with output, is given in appendix B on page 77.

6   File naming conventions used in DIFFaX.

DIFFaX tries to name output files so that it is reasonably clear from the name what the file contains, and with which data set it is associated. If the structure data file was called name, or name.dat then the output files will be named as follows;

name.dmp data dump file

name.sym data symmetry evaluation file

name.spc powder diffraction spectrum

name.str intensity profile along a streak

name.sadp selected area diffraction pattern

Note that DIFFaX reads the filename from the left up to the first period ('.') (or space
(' ') on the Macintosh), and uses that as the root filename. Thus the powder diffraction
pattern file for the files `dia.dat` or `dia`, would be named equally `dia.spc`. If a file of
that name already exists, then DIFFaX will enumerate the filenames sequentially. Thus, if
the file `dia.spc` already exists, then it will name the next file `dia.spc1`. If, for example,
files `dia.spc`, `dia.spc1` to `dia.spc13`, already exist, then the next file will be `dia.spc14`.
However, if the file `dia.spc6` in the previous example had been deleted, then the new file
will take the available name, `dia.spc6`, despite the fact that the previous file had been
named `dia.spc13`, and therefore will be out of sequence. Similar considerations apply to
the `name.dmp`, `name.sym`, `name.str` and `name.sadp` files.

7 Compiling DIFFaX.

DIFFaX source code is contained in the three text files `DIFFaX.f`, `DIFFaX.par` and
`DIFFaX.inc`. `DIFFaX.par` and `DIFFaX.inc` are 'include' files and normally the compiler
expects them to be in the same directory as `DIFFaX.f` when it is compiled. DIFFaX is writ-
ten, as far as is practical, in fortran 77, using commonly found extensions, such as `real*8`,
rather than `DOUBLE PRECISION`. For legibility the source code is written using lower- and
upper-case characters. Useful FORTRAN extensions such as `DO WHILE. . .`, `CYCLE` etc.
were scrupulously avoided. Instead, the equivalent (but ugly) `IF . . . GOTO` construc-
tions were used. DIFFaX was developed mainly on Macintosh systems. However, DIFFaX
v1.7xx has been compiled and run successfully on various computer systems and FORTRAN
compilers. DIFFaX v1.801 has been tested on Macintosh and Silicon Graphics systems.
Small modifications need to be made to the source code in order to get DIFFaX to compile
properly on the Cray cft77 compiler. This is mainly due to cft77's stricter adherence to
the ANSI FORTRAN77 standard, and to the fact that the Cray is a 64-bit machine, which
introduces ambiguities as to the meaning of "double precision" (see section 7.3 for further
details concerning running on the Cray).

Built-in constants used by DIFFaX, such as fixed array sizes etc. (except for the "detune"
parameter discussed in sections 3.8.1 and 8.3) are assigned in the file `DIFFaX.par`. The
meaning of each of the constants is explained in more detail in the `DIFFaX.par` file.

7.1 Binary files.

A problem can arise when writing the binary file `name.sadp`, which is supposed to be an
unformatted $256 \times 256 \times 8$ bit file, exactly 65,536 bytes big. Some compilers pre-pend a
record header to each 256 byte row of data, increasing the physical length of each row of

data. There are two ways around this problem (*i*) Find out the actual row length (usually 260 bytes) and open the image file as a 260 x 256 byte image (and ignore the first 4 pixels in each row), or (*ii*) modify the binary file open statement in the source code. In VAX fortran the binary file open statement should have the specifier `recordtype = 'stream'` added, *i.e.* the specific statement in `DIFFaX.f` should read

```
if(sa.ne.op) open(unit = sa, file = outfile, status = 'new',
|        form = 'unformatted', err = 990, iostat = io_err,
|        recordtype = 'stream')
```

The exact fix, however, will depend on the compiler.

7.2   Precision.

DIFFaX uses double precision (`real*8`) floating point variables. On most machines it will run faster using single precision. DIFFaX has been tested most extensively using double precision variables. There is a risk of numeric overflow in the vicinity of sharp peaks if single precision (`real*4`) is used on some complicated structures.

7.3   Double Precision and the Cray.

On the Cray, however, single precision should be specified, using the '`-dp`' cft77 compiler option. Cray double precision is equivalent to `real*16`. This slows up the Cray unnecessarily and uses up a lot of memory. Consequently, if `DIFFaX.f` is compiled in unmodified form, the cft77 compiler will flag assignment statements such as `REAL*8`, `COMPLEX*16` and `INTEGER*4`, and will generate a screed of warning messages. All variable declarations of the form `REAL*4`, `REAL*8`, `INTEGER*1`, `INTEGER*2`, `INTEGER*4`, `LOGICAL*1` etc. should be replaced with the generic `REAL, INTEGER, LOGICAL` etc. The Cray does not recognize the non-standard functions `DCMPLX, DIMAG and DBLE`. These should be replaced everywhere by the single precision `CMPLX, IMAG and REAL` intrinsic functions.

7.4   The "saveall" option.

With some compilers, DIFFaX may crash with bus errors. The author has not traced down the cause of every problem explicitly. For one compiler it had something to do with ambiguities in the ansi77 standard about the `save` statement. There is, of course, the possibility that there is an elusive bug in the DIFFaX code – perhaps an uninitialized variable is used prematurely. The DIFFaX code has been scoured relentlessly for bugs of this kind. However, there remains the chance that there are still insidious bugs lurking there. If you find bugs, please let the author know about them. He will be delighted to fix them! Meanwhile, declaring all variables to be static invariably 'fixes' the run-time problems. With some

compilers, all variables can be forced to be treated as static at compile time by specifying `-saveall` as a compiler option (sometimes this is specified by '-s').

If your compiler does not provide a `-saveall` (or equivalent compiler option), then you can uncomment the `save` statement at the top of the `DIFFaX.inc` file. `DIFFaX.inc` is included in almost every subroutine in `DIFFaX.f`. Further, you should uncomment the `save` statement at the top of those subroutines that do not include `DIFFaX.inc`. These subroutines can be found by searching the file `DIFFaX.f` for the word "`save`".

However, use of the `-saveall` command may significantly slow down program execution, so it may pay to determine whether or not `-saveall` is needed by first compiling without it, and making a trial run of the DIFFaX executable. Run-time messages about bus errors are usually a hint that the `-saveall` statement is needed.

8   How DIFFaX works.

8.1   Diffraction Described Recursively.

DIFFaX takes advantage of the self-similarity of stacking sequences, present in periodic objects constructed by non-deterministic layer stacking, to rapidly compute diffraction intensity. A simple model of the type of recursion found in crystals is provided by a tall brick wall that contains only one type of brick, where the bricks have been cemented in place with flawless regularity. Neglecting the finite height of the wall, the wall has a recursive property, namely that a brick wall is equivalent to a row of bricks plus another (displaced) brick wall (see Figure 6). Crystals can be viewed similarly in terms of sheets of unit cells. This recursive property is reflected in many physical properties of crystals, such as the scattered wavefunction when such crystals are irradiated. Thus, the scattered wave function from a crystal centered on any layer, is equivalent to the scattering contribution from that layer plus the scattered wavefunction from the displaced crystal centered at the next layer. More formally,

$$\Psi(\mathbf{u}) = F(\mathbf{u}) + \exp(-2\pi i \mathbf{u} \cdot \mathbf{R}) \ \Psi(\mathbf{u}) \tag{7}$$

Figure 6: Illustration of the recursive property of periodic objects.

Here, $\Psi(\mathbf{u})$ is the scattered wavefunction at reciprocal lattice vector $\mathbf{u}$, $F(\mathbf{u})$ the scattering contribution from the layer at the origin, and $\mathbf{R}$ is the shift between the two origins. This equation embodies Laue's diffraction equation, and leads immediately to a solution for the scattered wavefunction from the whole crystal.

$$\Psi(\mathbf{u}) = \frac{F(\mathbf{u})}{1 - \exp(-2\pi i \mathbf{u} \cdot \mathbf{R})} \tag{8}$$

This equation tells us that there are sharp diffracted peaks at values of $\mathbf{u} = (h, k, l)$ that satisfy $\mathbf{u} \cdot \mathbf{R} = hR_x + kR_y + lR_z = 2n\pi$, where n is an integer, and $\mathbf{R} = (R_x, R_y, R_z)$ is in relative cell coordinates.

This recursive property can be extended to crystals containing defects. Figure 7 shows our brick wall, now containing two types of brick arranged in homogeneous layers. In the crystalline analog, recursion relations between the wavefunctions for crystals centered on the two types of layers can be defined. The stacking of sheets is now determined probablistically, by the introduction of the transition probabilities α_{ij}.

$$\Psi_i(\mathbf{u}) = F_i(\mathbf{u}) + \sum_{j=1,2} \alpha_{ij} \exp(-2\pi i \mathbf{u} \cdot \mathbf{R}_{ij}) \, \Psi_j(\mathbf{u}) \tag{9}$$

The simultaneous equations are readily solved to yield the wavefunctions $\Psi(\mathbf{u})$. Since we are computing the diffraction from a statistical ensemble of crystallites, the intensity is given by the incoherent sum

$$\frac{I(\mathbf{u})}{N} = \sum_{i=1,2} g_i \left(F^*(\mathbf{u})\Psi(\mathbf{u}) + F(\mathbf{u})\Psi^*(\mathbf{u}) - |F_i(\mathbf{u})|^2 \right) \tag{10}$$

where the layer existence probability factors g_i are given by equation 6. Interestingly, the recursive property holds even when the crystal contains only a finite number of layers (see the reprint in section B.7 for more details).

8.2   Finite layer widths.

Debye-Scherrer type peak broadening due to finite layer widths is emulated phenomenologically in DIFFaX v1.801. The proper full calculation would require integration over non-integer values of h and k, which would be forbiddingly time-consuming. Here we present an outline of the methodology used in DIFFaX.

A fair representation of shape-broadening can be introduced by assuming that, from a statistical ensemble of crystallites, the average half-width of the intensity profile in the vicinity of reciprocal lattice point $h0l$ for an average crystal width W_a along $\mathbf{a}$ is given by

$$\Gamma = \frac{1}{2W_a} \tag{11}$$

Figure 7: Illustration of recursion in randomly faulted periodic objects.

The intensity profile is assumed to approximate a Lorentzian in shape, with half-width Γ. The Lorentzian is intended to approximate the more formally accurate 'sinc' function form factor,

$$\left(\frac{\sin(\pi(h'-h)W_a)}{\pi(h'-h)W_a}\right)^2 \tag{12}$$

after having averaged out the oscillations due to the presence of a spread in the W_a values in the powder sample. (Here h' is a continuous variable whose value is in the vicinity of the integer value h).

All crystal shapes are assumed to be rectangular (or cylindrical, depending on whether two widths, or a diameter, were specified), so the more complicated form factors due to special crystal morphology are ignored. When the widths W_a and W_b are not equal, it is assumed that the contours of equal intensity about a peak are elliptical. This intensity profile in the h–k plane is intercepted by the Ewald sphere at an angle that is determined by the l-value of the reflection (see Figure 8). Thus, the half-width of the peak at scattering vector

hkl due to shape broadening can be shown to be (for perpendicular **a** and **b** axes)

$$\Gamma = \frac{1}{2Q_{hkl}} \left[\left(\frac{Q_{h00}}{W_a} \right)^2 + \left(\frac{Q_{0k0}}{W_b} \right)^2 \right] \tag{13}$$

where Q_{hkl} is the magnitude of the reciprocal lattice scattering vector at *hkl*, and is given by

$$Q_{hkl}^2 = a_0 h^2 + b_0 k^2 + c_0 l^2 + d_0 hk \tag{14}$$

with the constants

$$
\begin{aligned}
a_0 &= \frac{1}{a^2 \sin^2(\gamma)} \\
b_0 &= \frac{1}{b^2 \sin^2(\gamma)} \\
c_0 &= \frac{1}{c^2} \\
d_0 &= \frac{-2\cos(\gamma)}{ab\sin^2(\gamma)}
\end{aligned}
\tag{15}
$$

Depending on the relative values of W_a and W_b, and the value of l, different peaks will be broadened differently. For non-perpendicular axes, equation 13 is modified slightly to reflect the fact that the W_a and W_b measurements are not mutually perpendicular. Consequently, the shape-broadened half-width of the peak at scattering angle θ in the powder pattern is given by

$$\Gamma_\theta = \frac{\lambda}{2Q_{hkl}} \left[a_0 \left(\frac{h + k\sin(\gamma)}{W_a \cos(\gamma)} \right)^2 + b_0 \left(\frac{k}{W_b} \right)^2 \right] \tag{16}$$

provided $h \neq k = 0$.

The broadening of $00l$ peaks is handled a little differently. When both h and k are zero, the Ewald sphere is tangential to the plane of the shape broadening profile of the $00l$ peak. This produces an asymmetrical peak with a sharp onset and a long tail, whose half-width depends in a complicated way on the relative values of W_a and W_b. If W_a and W_b are close in value, the tail drops off the slowest.

Despite the simplifications and approximations used, the addition of layer-width broadening to powder pattern peaks still introduces a significant computational overhead to the powder pattern calculations. In particular, shape-broadening adds an extra degree of freedom to the simulations that can easily overwhelm the user with data. As a rule, layer widths should be specified as `INFINITE` when exploring the effects of stacking fault probabilities. The addition of the shape broadening profiles should be reserved for final refinements of simulations in systems that are known to have small crystallites, and where the simulations for large crystals look promising.

Figure 8: Geometry of the Ewald sphere intercepting a shape-broadened peak at *hkl*.

8.3 The "detune" parameter.

Most users will not need to modify the internal parameters used within the DIFFaX program, and thus these have all been set at compile time and cannot be modified dynamically by the user. These need to be changed within the file `DIFFaX.par`, and `DIFFaX.f` needs to be recompiled. Under special circumstances, however, the "detune" parameter may need to be adjusted by the user. What is this "detune" parameter? Why is it hidden from the user in this way? To understand the role of the detune parameter, we must first examine what the computer is trying to do when it integrates intensities under sharp peaks. Sharp diffraction peaks from perfect crystals arise from a resonance condition between the incoming wave and the diffracting lattice. Ideally, such a resonance produces a peak in intensity that can be described as a delta function, with infinite height and essentially zero width, although the integrated intensity under the peak is finite. True delta functions cannot be properly represented in a computer. In particular, they cannot be detected or integrated properly. To

help DIFFaX detect and integrate these delta functions, DIFFaX resorts to a trick whereby the diffraction resonance is effectively detuned slightly. The Q-factor of the resonance is no longer infinite (but still large), and importantly, the peak develops a detectable width. The result is that the peak can be integrated by the computer. The detuning is brought about by reducing all the stacking probabilities by a small amount, so that the sum of all transition probabilities from any given layer is slightly less than unity. This is a mathematical trick, and the principle behind this trick is explained in more detail in the reprint in section B.7 on page 93. The fraction by which all the stacking probabilities is reduced we refer to as the detune parameter. One way to understand this is to examine equation 7 for the scattered wavefunction from a perfect crystal containing only one layer type

$$\Psi(\mathbf{u}) = \frac{F(\mathbf{u})}{1 - \exp(-2\pi i \mathbf{u} \cdot \mathbf{R})} \tag{17}$$

There is a resonance condition (a sharp peak) when $\exp(-2\pi i \mathbf{u} \cdot \mathbf{R}) = 1$, when the denominator is zero. To make equation 17 more palatable to a computer, we modify the denominator so that

$$\Psi(\mathbf{u}) = \frac{F(\mathbf{u})}{1 - \Omega \exp(-2\pi \mathbf{u} \cdot \mathbf{R})} \tag{18}$$

where Ω is a number slightly less than unity, $i.e.$ $\Omega = 1 - \text{detune}$. The denominator now will never be zero and the real part will always be positive. Ordinarily, Ω should be unity, the probability that a layer is followed by another identical layer. A consequence of detuning the sharp peaks in this way, is that they are broadened slightly. This broadening becomes apparent if one plots spectra on a log-linear scale, and can become a nuisance if background diffuse scattering is being simulated and fitted to a power law, or if satellite peaks close to a sharp peak are being simulated. The default value of detune is 0.001. To sharpen the sharp peaks, the value of detune should be decreased. However, if detune is made too small, there is a risk that DIFFaX will miss the sharp peaks, or will fail to integrate them properly. Setting detune to zero may cause DIFFaX to fail, because of a "divide by zero" condition. Sharp peaks will not be integrated properly. Spectra may even contain negative intensities. Consequently, detune is kept away from casual misuse by not including it as a run time variable.

The impact of "detune" on peak width is illustrated in Fig. 9, where the continuous line is for detune $= 10^{-3}$, and the dotted line is for detune $= 10^{-4}$. The peak width is reduced by a factor of ten when detune is reduced by a factor of ten. Similarly, the peak heights increase by a factor of ten, such that the integrated area is approximately constant. To change detune, locate the subroutine DETUN in the file DIFFaX.f.

```
subroutine DETUN()
include 'DIFFaX.par'
```

Figure 9: Intensity trace along l showing the effect of changing the parameter "detune" on the width of the 004 peak in unfaulted GaAs (see appendix A.5, page 74).

```
        include 'DIFFaX.inc'
*
        integer*4 i, j
        real*8 delta
*
        delta = eps3
*
* A value of delta = 0.001 is found to be optimum.
* If preferred, user can specify 'delta' interactively
* using the following three lines.
C       write(op,400) 'Enter detune parameter'
C       read(cntrl,*) delta
C       if(CFile) write(op,401) delta
*
        do 10 i = 1, n_layers
          do 20 j = 1, n_layers
            detune(j,i) = ONE - abs(delta)
   20     continue
   10 continue
*
        return
C  400 format(1x, a)
C  401 format(1x, g12.5)
        end
```

Either, change the value `delta` (note `eps3` is defined to be 10^{-3}, and `eps4` is defined to be 10^{-4} etc.) to the new value required, or uncomment the five lines of code beginning with

"C" that allow the user to set "detune" interactively. DIFFaX then needs to be recompiled. An explanation how to change detune for the Macintosh version MacDIFFaX is given in section 3.8.1 on page 37.

9   References

References

[1] M. M. J. Treacy, J. M. Newsam & M. W. Deem 1991 "A General Recursion Method for Calculating Diffracted Intensities From Crystals Containing Planar Faults" *Proc. Roy. Soc. Lond.* **A433**, 499–520.

[2] S. Hendricks and E. Teller 1942 "X-ray interference in partially ordered layer lattices" *J. Chem. Phys.* **10**, 147–167.

[3] B. E. Warren 1959 "X-ray diffraction", pp. 275–312. Reading, Massachusetts: Addison-Wesley.

[4] J. M. Cowley 1981 "Diffraction Physics", pp. 388–400. New York and London: North-Holland.

10   Differences between DIFFaX versions 1.807 and 1.7xx

DIFFaX v1.807 behaves pretty much the same as previous versions. These are the important differences;

1. DIFFaX v1.804 now simulates peak broadening in powder patterns due to finite crystal dimensions perpendicular to the stacking direction. This feature requires an additional line in the structure data file specifying the average layer widths parallel to **a** and **b** (see section 2.7.5 on page 13). Note that broadening due to a finite number of layers is handled differently by DIFFaX (see section 2.9, page 18).

2. The six layer uncertainty stacking factors, the *Fats-Waller* factors C_{ij}, have been re-ordered. They must now be entered in the sequence `C11 C22 C33 C12 C13 C23`, as opposed to `C11 C22 C33 C12 C23 C31` (see section 2.10.1, page 19). This was done to maintain consistency with the conventional ordering of the anisotropic Debye-Waller temperature factors.

3. The maximum allowable length of a data line has been extended from 80 to 200 characters.

4. Atom names are now case-insensitive.

5. In DIFFaX v1.805 atom coordinates and stacking vectors can be entered as fractions. That is, "1/3" is now accepted in place of 0.3333.

6. DIFFaX behaves differently when it encounters a stacking probability $\alpha_{ii} = 1.0$. DIFFaX now counts the number of layers N that have $\alpha_{ii} = 1.0$, and assigns a layer existence probability $g_i = 1/N$ to each of these layers. Otherwise $g_i = 0$. See section 2.11 on page 22 for more details. DIFFaX v1.7xx usually returned a "singular matrix error".

7. DIFFaX v1.806 and higher can now generate 16-bit greyscale SAD patterns, as well as 8-bit patterns.

8. There have been some minor bug fixes and code improvements.

A Appendices

A.1 Example data file for random faults in diamond.

Diamond belongs to the cubic space group Fd$\overline{3}$m. It supports twinning on the cubic {111} planes, thus the [111] direction becomes the new DIFFaX **c** axis. The structure is best represented in the hexagonal setting, where $(111)_{\mathrm{cubic}}$ is equivalent to $(001)_{\mathrm{hexagonal}}$, and the new cell axes are along $[1\overline{1}0]_{\mathrm{cubic}}$ and $[10\overline{1}]_{\mathrm{cubic}}$. The (111) planes are centrosymmetric about the origin, and two distinct forms occur in any given crystal, related by a mirror plane (or, equivalently, a 60° rotation). When like layers stack, the sequence has (local) cubic symmetry. When mirror-related planes stack alternately, to form the lonsdaleite structure, the sequence has (local) hexagonal symmetry. See figure 10 for a graphic description.

```
Data file dia.dat

{data file for diamond, with random hexagonal intergrowths}
{probability of cubic stacking = 70%}

INSTRUMENTAL                        {Header for instrumental section}
X-RAY                               {Simulate X-ray diffraction}
1.5418                              {X-ray wavelength}
{gaussian 0.1 trim}                 {Instrumental broadening (much faster)}
PSEUDO-VOIGT 0.1 -0.036 0.009 0.6 TRIM {Instrumental broadening (much slower)}

STRUCTURAL                          {Header for structural section}
2.52 2.52 2.06 120.0                {unit cell coordinates, a, b, c, gamma}
6/MMM                               {hexagonal, c = cubic [111]}
2                                   {111 sheet, plus its mirror}
infinite                            {Layers are very wide in the a-b plane}

LAYER 1
CENTROSYMMETRIC
 C    1  -.333333  -.166667  -.125  1.0  1.0
{C    2   .333333   .166667   .125  1.0  1.0, related to 1 by -1}

LAYER 2
CENTROSYMMETRIC
 C    1   .333333   .166667  -.125  1.0  1.0
{C    2  -.333333  -.166667   .125  1.0  1.0, related to 1 by -1}

STACKING                            {Header for stacking description}
recursive                           {Statistical ensemble}
infinite                            {Infinite number of layers}

TRANSITIONS                         {Header for stacking transition data}

{Transitions from layer 1}
0.7   0.666667   0.333333  1.0     {layer 1 to layer 1}
0.3   0.0        0.0       1.0     {layer 1 to layer 2}

{Transitions from layer 2}
0.3   0.0        0.0       1.0     {layer 2 to layer 1}
0.7  -0.666667  -0.333333  1.0     {layer 2 to layer 2}
```

[1$\bar{1}$0] view of diamond layers

Figure 10: [1$\bar{1}$0] view of diamond/lonsdaleite layers.

A.2   Example data file describing clustered faults in diamond.

Appendix A.1 described diamond/lonsdaleite intergrowths, where the twin faults were randomly dispersed throughout the crystal, without correlation. It is also possible to describe the faulting as if there were a 'memory', or Reichweite, so that the stacking history plays a role in determining the fault probability. For example, a crystal may be stacking in a cubic sequence with a low probability that it will twin. Once twinned, however, the probability of a fault in the new hexagonal sequence may also be low, and the crystal may continue for some while before faulting back to a cubic sequence. Figure 11 illustrates how to describe

such faulting by defining four layer types.

4 layers are required to describe fault clustering in diamond

Structure type A layer

Structure type A' layer

Type 1 layer

Type 2 layer

Type 3 layer

Type 4 layer

A

A

A'

A

A

A'

A'

A'

Cubic stacking

1
1
1
1

R_{11}

4
4
4
4

R_{44}

Hexagonal stacking

3
2
3
2
3

R_{23}

R_{32}

Typical faulted sequence

4
4
4
2
1
1
1
3
2
3
2
1
3
4
4 or 2

cubic

hexagonal

cubic

hexagonal

cubic
hexagonal
cubic

Figure 11: [1$\overline{1}$0] view showing fault clustering in diamond/lonsdaleite.

```
Data file Clustdia.dat

{data file describing clustered faulting in diamond}

{There are four layers in total, two of which are}
{structurally distinct. They are related by a mirror}
{in the a-b plane, and are labeled layers A and A'.}
```

```
{Layer 1 is an A layer which is preceded by an A layer}
{Layer 2 is an A' layer which is preceded by an A layer}
{Layer 3 is an A layer which is preceded by an A' layer}
{Layer 4 is an A' layer which is preceded by an A' layer}
{1 - 1 is a cubic stacking transition}
{1 - 2 is a stacking fault in a cubic sequence}
{1 - 3 cannot occur}
{1 - 4 cannot occur}
{2 - 1 cannot occur}
{2 - 2 cannot occur}
{2 - 3 is a hexagonal stacking transition}
{2 - 4 is a fault in a hexagonal sequence}
{3 - 1 is a fault in a hexagonal sequence}
{3 - 2 is a hexagonal stacking transition}
{3 - 3 cannot occur}
{3 - 4 cannot occur}
{4 - 1 cannot occur}
{4 - 2 cannot occur}
{4 - 3 is a stacking fault in a cubic sequence}
{4 - 4 is a cubic stacking transition}

{In this file. . .}
{the probability of a fault in cubic stacking = 1-2 = 4-3 = 10%}
{                                    thus, 1-1 = 4-4 = 90%}
{the probability of a fault in hexagonal stacking = 2-4 = 3-1 = 20%}
{                                    thus, 2-3 = 3-2 = 80%}

INSTRUMENTAL
X-RAY
1.5418
PSEUDO-VOIGT .89 -.32 .08 .6 trim

STRUCTURAL
2.52 2.52 2.06 120.0
-3M        {Symmetry is not necessarily 6/MMM, as there}
           {may not be equal numbers of 1 layers and 4 layers}
4
infinite

LAYER 1
CENTROSYMMETRIC
C     1   -.333333    -.166667    -.125  1.0  1.0

LAYER 2
CENTROSYMMETRIC
C     1    .333333     .166667    -.125  1.0  1.0

LAYER 3 = 1

LAYER 4 = 2

STACKING
recursive
infinite

TRANSITIONS
{1}
0.90  0.666667  0.333333  1.0
0.10  0.0       0.0       1.0
0.00  0.0       0.0       0.0       {1-3 Transition cannot occur}
0.00  0.0       0.0       0.0       {1-4 Transition cannot occur}

{2}
0.00  0.0       0.0       0.0       {2-1 Transition cannot occur}
```

```
0.00  0.0        0.0       0.0        {2-2 Transition cannot occur}
0.80  0.0        0.0       1.0
0.20 -0.666667 -0.333333  1.0

{3}
0.20  0.666667  0.333333  1.0
0.80  0.0        0.0       1.0
0.00  0.0        0.0       0.0        {3-3 Transition cannot occur}
0.00  0.0        0.0       0.0        {3-4 Transition cannot occur}

{4}
0.00  0.0        0.0       0.0        {4-1 Transition cannot occur}
0.00  0.0        0.0       0.0        {4-2 Transition cannot occur}
0.10  0.0        0.0       1.0
0.90 -0.666667 -0.333333  1.0
```

A.3 Example data file describing faults in Faujasite.

Faujasite is a zeolite which, like diamond, has topological space group symmetry Fd$\overline{3}$m. Unlike, diamond, which has 8 atoms per unit cell, faujasite has 576 framework atoms. However, despite the fact that each layer contains significantly more atoms, and that the unit cell dimensions are larger, the data file will resemble that for diamond. In place of each carbon atom, faujasite has a 78-atom sodalite cage. The morphology of the sodalite cage is that of a truncated cuboctahedron, or tetrakaidekahedron. Faujasite is, strictly speaking, an alumino-silicate, with average framework composition, $Si_xAl_{1-x}O_2$, where typically, $x \sim 0.75$. The aluminum is essentially randomly distributed on the framework, substituting for Si, but with the proviso that no two aluminum atoms share a bridging oxygen (Loewenstein's rule). To balance the framework charge introduced by the trivalent aluminum atoms occupying tetrahedrally coordinated sites, there are extra-framework cations, such as Na^+ and K^+. There is also a considerable amount of water associated with the framework. Most of the cations and water molecules are mobile. In this data file, we assume that $x = 1$, *i.e.* there is no aluminum. This is not a bad approximation since Al and Si have similar x-ray scattering factors. The cations and water molecules are also ignored. Figure 12 below emphasizes the resemblance to the diamond case described in appendix A.1.

```
Data file FAU.dat

{data file for Faujasite/Breck's structure 6}
{data is good for random distribution of faults only}
{A high value of alpha(1,1) (= alpha(2,2)) implies a FAU-rich material}
{A low value of alpha(1,1) (= alpha(2,2)) implies a BSS-rich material}
{This file has alpha(1,1) = 0.99, and thus is FAU-rich,}
{but the crystal contains only three layers}

INSTRUMENTAL
{Radiation type = } X-RAY
{wavelength = } 1.5418
{Instrumental broadening (faster)} {Lorentzian 0.1 trim}
{Instrumental broadening (slower)} PSEUDO-VOIGT 0.1 -0.036 0.009 0.6 TRIM
```

[1$\bar{1}$0] view of faujasite layers

Figure 12: [1$\bar{1}$0] view of faujasite 111 layers.

```
STRUCTURAL
{a, b, c, gamma} 17.437 17.437 14.238 120.0
{Expected diffraction point symmetry} 6/mmm
{Number of layers = } 2

LAYER 1
centrosymmetric
Si4+  1  0.15373 -0.01354 -0.14201 1.0 1.0
Si4+  2  0.33274 -0.01354 -0.14201 1.0 1.0
Si4+  3  0.15373  0.16726 -0.14201 1.0 1.0
Si4+  4  0.51354  0.16726 -0.14201 1.0 1.0
Si4+  5  0.33274  0.34627 -0.14201 1.0 1.0
Si4+  6  0.51354  0.34627 -0.14201 1.0 1.0
Si4+  7  0.09514 -0.13074  0.03379 1.0 1.0
Si4+  8  0.27413 -0.13074  0.03379 1.0 1.0
Si4+  9  0.09514  0.22587  0.03379 1.0 1.0
Si4+ 10  0.63074  0.22587  0.03379 1.0 1.0
Si4+ 11  0.27413  0.40486  0.03379 1.0 1.0
Si4+ 12  0.63074  0.40486  0.03379 1.0 1.0
Si4+ 13  0.03487 -0.07047  0.21460 1.0 1.0
Si4+ 14  0.39468 -0.07047  0.21460 1.0 1.0
Si4+ 15  0.03487  0.10532  0.21460 1.0 1.0
Si4+ 16  0.57047  0.10532  0.21460 1.0 1.0
Si4+ 17  0.39468  0.46513  0.21460 1.0 1.0
Si4+ 18  0.57047  0.46513  0.21460 1.0 1.0
Si4+ 19  0.15421 -0.01080  0.39358 1.0 1.0
Si4+ 20  0.33501 -0.01080  0.39358 1.0 1.0
```

```
Si4+ 21   0.15421   0.16499   0.39358 1.0 1.0
Si4+ 22   0.51080   0.16499   0.39358 1.0 1.0
Si4+ 23   0.33501   0.34579   0.39358 1.0 1.0
Si4+ 24   0.51080   0.34579   0.39358 1.0 1.0
O 1-   1   0.07207  -0.07207  -0.21620 1.0 0.5
O 1-   2   0.35587  -0.07207  -0.21620 1.0 0.5
O 1-   3   0.07207   0.14413  -0.21620 1.0 0.5
O 1-   4   0.57207   0.14413  -0.21620 1.0 0.5
O 1-   5   0.35587   0.42793  -0.21620 1.0 0.5
O 1-   6   0.57207   0.42793  -0.21620 1.0 0.5
O 1-   7   0.23754  -0.02494  -0.18761 1.0 0.5
O 1-   8   0.23754   0.26246  -0.18761 1.0 0.5
O 1-   9   0.52494   0.26246  -0.18761 1.0 0.5
O 1-  10   0.18854   0.09426  -0.14561 1.0 1.0
O 1-  11   0.40574   0.09426  -0.14561 1.0 1.0
O 1-  12   0.40574   0.31146  -0.14561 1.0 1.0
O 1-  13   0.12808  -0.04067  -0.03050 1.0 1.0
O 1-  14   0.33127  -0.04067  -0.03050 1.0 1.0
O 1-  15   0.12808   0.16873  -0.03050 1.0 1.0
O 1-  16   0.54067   0.16873  -0.03050 1.0 1.0
O 1-  17   0.33127   0.37192  -0.03050 1.0 1.0
O 1-  18   0.54067   0.37192  -0.03050 1.0 1.0
O 1-  19   0.00000  -0.21620   0.00000 1.0 0.5
O 1-  20   0.28380  -0.21620   0.00000 1.0 0.5
O 1-  21   0.00000   0.21620   0.00000 1.0 0.5
O 1-  22   0.71620   0.21620   0.00000 1.0 0.5
O 1-  23   0.28380   0.50000   0.00000 1.0 0.5
O 1-  24   0.71620   0.50000   0.00000 1.0 0.5
O 1-  25   0.16248  -0.17507   0.03760 1.0 0.5
O 1-  26   0.16247   0.33752   0.03760 1.0 0.5
O 1-  27   0.67507   0.33753   0.03760 1.0 0.5
O 1-  28   0.09240  -0.09801   0.14278 1.0 1.0
O 1-  29   0.30961  -0.09801   0.14278 1.0 1.0
O 1-  30   0.09240   0.19039   0.14278 1.0 1.0
O 1-  31   0.59801   0.19039   0.14278 1.0 1.0
O 1-  32   0.30961   0.40760   0.14278 1.0 1.0
O 1-  33   0.59801   0.40760   0.14278 1.0 1.0
O 1-  34   0.47086   0.02912   0.17893 1.0 1.0
O 1-  35   0.05826   0.02914   0.17893 1.0 1.0
O 1-  36   0.47088   0.44174   0.17893 1.0 1.0
O 1-  37  -0.07206  -0.14414   0.21619 1.0 0.5
O 1-  38   0.42794  -0.14414   0.21619 1.0 0.5
O 1-  39  -0.07206   0.07206   0.21619 1.0 0.5
O 1-  40   0.64414   0.07206   0.21619 1.0 0.5
O 1-  41   0.42794   0.57206   0.21619 1.0 0.5
O 1-  42   0.64414   0.57206   0.21619 1.0 0.5
O 1-  43   0.06667  -0.07927   0.32500 1.0 0.5
O 1-  44   0.35408  -0.07927   0.32500 1.0 0.5
O 1-  45   0.06667   0.14592   0.32500 1.0 0.5
O 1-  46   0.57927   0.14592   0.32500 1.0 0.5
O 1-  47   0.35408   0.43333   0.32500 1.0 0.5
O 1-  48   0.57927   0.43333   0.32500 1.0 0.5
O 1-  49   0.23720  -0.02561   0.35998 1.0 1.0
O 1-  50   0.52561   0.26279   0.35998 1.0 1.0
O 1-  51   0.23721   0.26280   0.35998 1.0 1.0
O 1-  52   0.40313   0.09686   0.38212 1.0 1.0
O 1-  53   0.19373   0.09687   0.38212 1.0 1.0
O 1-  54   0.40314   0.30627   0.38212 1.0 1.0
O 1-  55   0.11714  -0.04954   0.50000 1.0 0.5
O 1-  56   0.33333  -0.04954   0.50000 1.0 0.5
O 1-  57   0.11714   0.16667   0.50000 1.0 0.5
O 1-  58   0.54954   0.16667   0.50000 1.0 0.5
O 1-  59   0.33333   0.38286   0.50000 1.0 0.5
O 1-  60   0.54954   0.38286   0.50000 1.0 0.5
```

```
LAYER 2
centrosymmetric
Si4+   1   0.15373  -0.01354   0.14201 1.0 1.0
Si4+   2   0.33274  -0.01354   0.14201 1.0 1.0
Si4+   3   0.15373   0.16726   0.14201 1.0 1.0
Si4+   4   0.51354   0.16726   0.14201 1.0 1.0
Si4+   5   0.33274   0.34627   0.14201 1.0 1.0
Si4+   6   0.51354   0.34627   0.14201 1.0 1.0
Si4+   7   0.09514  -0.13074  -0.03379 1.0 1.0
Si4+   8   0.27413  -0.13074  -0.03379 1.0 1.0
Si4+   9   0.09514   0.22587  -0.03379 1.0 1.0
Si4+  10   0.63074   0.22587  -0.03379 1.0 1.0
Si4+  11   0.27413   0.40486  -0.03379 1.0 1.0
Si4+  12   0.63074   0.40486  -0.03379 1.0 1.0
Si4+  13   0.03487  -0.07047  -0.21460 1.0 1.0
Si4+  14   0.39468  -0.07047  -0.21460 1.0 1.0
Si4+  15   0.03487   0.10532  -0.21460 1.0 1.0
Si4+  16   0.57047   0.10532  -0.21460 1.0 1.0
Si4+  17   0.39468   0.46513  -0.21460 1.0 1.0
Si4+  18   0.57047   0.46513  -0.21460 1.0 1.0
Si4+  19   0.15421  -0.01080  -0.39358 1.0 1.0
Si4+  20   0.33501  -0.01080  -0.39358 1.0 1.0
Si4+  21   0.15421   0.16499  -0.39358 1.0 1.0
Si4+  22   0.51080   0.16499  -0.39358 1.0 1.0
Si4+  23   0.33501   0.34579  -0.39358 1.0 1.0
Si4+  24   0.51080   0.34579  -0.39358 1.0 1.0
O 1-   1   0.07207  -0.07207   0.21620 1.0 0.5
O 1-   2   0.35587  -0.07207   0.21620 1.0 0.5
O 1-   3   0.07207   0.14413   0.21620 1.0 0.5
O 1-   4   0.57207   0.14413   0.21620 1.0 0.5
O 1-   5   0.35587   0.42793   0.21620 1.0 0.5
O 1-   6   0.57207   0.42793   0.21620 1.0 0.5
O 1-   7   0.23754  -0.02494   0.18761 1.0 0.5
O 1-   8   0.23754   0.26246   0.18761 1.0 0.5
O 1-   9   0.52494   0.26246   0.18761 1.0 0.5
O 1-  10   0.18854   0.09426   0.14561 1.0 1.0
O 1-  11   0.40574   0.09426   0.14561 1.0 1.0
O 1-  12   0.40574   0.31146   0.14561 1.0 1.0
O 1-  13   0.12808  -0.04067   0.03050 1.0 1.0
O 1-  14   0.33127  -0.04067   0.03050 1.0 1.0
O 1-  15   0.12808   0.16873   0.03050 1.0 1.0
O 1-  16   0.54067   0.16873   0.03050 1.0 1.0
O 1-  17   0.33127   0.37192   0.03050 1.0 1.0
O 1-  18   0.54067   0.37192   0.03050 1.0 1.0
O 1-  19   0.00000  -0.21620  -0.00000 1.0 0.5
O 1-  20   0.28380  -0.21620  -0.00000 1.0 0.5
O 1-  21   0.00000   0.21620  -0.00000 1.0 0.5
O 1-  22   0.71620   0.21620  -0.00000 1.0 0.5
O 1-  23   0.28380   0.50000  -0.00000 1.0 0.5
O 1-  24   0.71620   0.50000  -0.00000 1.0 0.5
O 1-  25   0.16248  -0.17507  -0.03760 1.0 0.5
O 1-  26   0.16247   0.33752  -0.03760 1.0 0.5
O 1-  27   0.67507   0.33753  -0.03760 1.0 0.5
O 1-  28   0.09240  -0.09801  -0.14278 1.0 1.0
O 1-  29   0.30961  -0.09801  -0.14278 1.0 1.0
O 1-  30   0.09240   0.19039  -0.14278 1.0 1.0
O 1-  31   0.59801   0.19039  -0.14278 1.0 1.0
O 1-  32   0.30961   0.40760  -0.14278 1.0 1.0
O 1-  33   0.59801   0.40760  -0.14278 1.0 1.0
O 1-  34   0.47086   0.02912  -0.17893 1.0 1.0
O 1-  35   0.05826   0.02914  -0.17893 1.0 1.0
O 1-  36   0.47088   0.44174  -0.17893 1.0 1.0
O 1-  37  -0.07206  -0.14414  -0.21619 1.0 0.5
```

```
O 1- 38  0.42794 -0.14414 -0.21619 1.0 0.5
O 1- 39 -0.07206  0.07206 -0.21619 1.0 0.5
O 1- 40  0.64414  0.07206 -0.21619 1.0 0.5
O 1- 41  0.42794  0.57206 -0.21619 1.0 0.5
O 1- 42  0.64414  0.57206 -0.21619 1.0 0.5
O 1- 43  0.06667 -0.07927 -0.32500 1.0 0.5
O 1- 44  0.35408 -0.07927 -0.32500 1.0 0.5
O 1- 45  0.06667  0.14592 -0.32500 1.0 0.5
O 1- 46  0.57927  0.14592 -0.32500 1.0 0.5
O 1- 47  0.35408  0.43333 -0.32500 1.0 0.5
O 1- 48  0.57927  0.43333 -0.32500 1.0 0.5
O 1- 49  0.23720 -0.02561 -0.35998 1.0 1.0
O 1- 50  0.52561  0.26279 -0.35998 1.0 1.0
O 1- 51  0.23721  0.26280 -0.35998 1.0 1.0
O 1- 52  0.40313  0.09686 -0.38212 1.0 1.0
O 1- 53  0.19373  0.09687 -0.38212 1.0 1.0
O 1- 54  0.40314  0.30627 -0.38212 1.0 1.0
O 1- 55  0.11714 -0.04954 -0.50000 1.0 0.5
O 1- 56  0.33333 -0.04954 -0.50000 1.0 0.5
O 1- 57  0.11714  0.16667 -0.50000 1.0 0.5
O 1- 58  0.54954  0.16667 -0.50000 1.0 0.5
O 1- 59  0.33333  0.38286 -0.50000 1.0 0.5
O 1- 60  0.54954  0.38286 -0.50000 1.0 0.5

STACKING
recursive
3

TRANSITIONS
{1}
0.99    0.666666667     0.333333333    1.0
0.01    0.0             0.0            1.0

{2}
0.01    0.0             0.0            1.0
0.99   -0.666666667    -0.333333333    1.0
```

A.4   Data file describing chiral twinning in zeolite beta.

Zeolite beta, like faujasite, has a 3 dimensional pore system with the minimum constricting apertures being 12-rings. However, the (tertiary) building units stack in either a right-handed, 4_1, or left-handed, 4_3, manner. Zeolite beta exhibits near-random faulting probability, with a 40–50% chance that the next layer will continue in the same handedness as the previous two layers. Due to the near-random nature of the stacking, there is no reason to believe that the as-synthesized zeolite exhibits any significant residual chirality. The zeolite beta structure can be described by a minimum of four layers. However, with four layers, the two end-members, represented by the extreme stacking transition probabilities of 0 and 1, will be either the right-handed or the left-handed enantiomer. Only at a stacking probability of 0.5 will the crystal be racemic. In general, the data will describe a material which exhibits a preferred handedness, which contains faults which immediately attempt to repair the error by following quickly with a second fault which brings the sequence back into the preferred handedness. These are generally known as deformation faults. To describe a

structure that (as a statistical ensemble of crystallites at least) is always racemic, we need to define 8 layers; 4 right-handed layers, and 4 left-handed layers. Under these circumstances, once the fault has happened, the sequence continues in this new handedness until another fault is encountered. The fault probability is now the chance that the stacking will reverse handedness. Thus one end-member, 'fault' probability 0, represents the chiral crystal system (*i.e.* an equal mixture of right-handed and left-handed crystallites). The other end-member represents the achiral crystal system, where stacking is a strictly alternating LRLRLR..., which is a c-glide operation. By increasing the number of layers we have introduced the ability to add a 'memory' to the layer description, that they remember the stacking sense of the previous layer (Reichweite = 1). The range of this 'memory' can be increased by increasing the number of layers.

[010] view of a faulted zeolite beta crystallite

Figure 13: [010] view of a faulted zeolite beta framework.

Stacking of building units in zeolite beta

LH stacking   RH stacking

Figure 14: Stacking of building units in zeolite beta.

```
Data file beta.dat

{                    DATA FOR ZEOLITE BETA                      }
{           chiral stacking probability  =  0.440              }

{This data file was created by clipping a 16 T-atom 'helicopter'}
{unit from the chiral P4122 structure. Layers 1 to 4 were }
{generated by rotating this unit successively by +90 degrees.}
{The left-hand layers, 5 to 8, were generated by taking layer 1}
{and reflecting it in the vertical glide plane at x = y,}
{essentially by swapping the x and y atom coordinates. This operation,}
{applied to layer 1, generates layer 8. Successive -90 degree}
{rotations of layer 8 generate layers 7, 6 and 5 respectively}
{Sequences of the type 1,2,3,4,1,2,3,4,1,2,3,4,1. . .  are pure}
{right-handed P4122 structures, and 5,8,7,6,5,8,7,6,5,8,. . . are}
{pure left-handed P4322 forms. Sequences such as 1,8,1,8,1. . ,}
{2,5,2,5,2,. . ., 3,6,3,6,3,. . ., and 4,5,4,5,4,. . . are the}
{monoclinic C2/c forms.}
{This description does not allow for any subtle strains in the}
{basic helicopter unit that can arise as a function of stacking}
{probability}

INSTRUMENTAL
X-RAY                                   {radiation type}
1.5418                                  {radiation wavelength}
PSEUDO-VOIGT .89 -.32 .08 .6 trim       {pseudo-Voigt instr. broadening}
```

```
STRUCTURAL
12.423  12.423   26.307   90.000              {cell a, b, c and gamma in Angstroms}
4/mmm                                         {symmetry, averaged over the faults}
8                                             {number of layer types}
infinite

LAYER    1      {basic 'helicopter' unit from bea, Newsam et al, Proc Roy Soc}
NONE           {no special symmetry such as centrosymmetric}
Si4+   1  -0.0040 -0.1310 -0.0690   1.0   1.0
Si4+   2   0.1260 -0.0010  0.0690   1.0   1.0
Si4+   3  -0.0040  0.1220 -0.0690   1.0   1.0
Si4+   4  -0.1260 -0.0010  0.0690   1.0   1.0
Si4+   5  -0.3830 -0.1310 -0.0660   1.0   1.0
Si4+   6   0.1260  0.3790  0.0660   1.0   1.0
Si4+   7  -0.3830  0.1210 -0.0660   1.0   1.0
Si4+   8  -0.1260  0.3790  0.0660   1.0   1.0
Si4+   9   0.3790 -0.1310 -0.0690   1.0   1.0
Si4+  10   0.1270 -0.3840  0.0690   1.0   1.0
Si4+  11   0.3790  0.1220 -0.0690   1.0   1.0
Si4+  12  -0.1260 -0.3840  0.0690   1.0   1.0
Si4+  13  -0.1950 -0.1950  0.0000   1.0   1.0
Si4+  14   0.1900  0.1910  0.0000   1.0   1.0
Si4+  15  -0.1950  0.1910  0.0000   1.0   1.0
Si4+  16   0.1900 -0.1950  0.0000   1.0   1.0
O 1-   1  -0.0030 -0.0040 -0.0600   1.0   1.0
O 1-   2   0.0000 -0.0010  0.0600   1.0   1.0
O 1-   3   0.1510  0.3280  0.1210   1.0   0.5
O 1-   4  -0.0010 -0.1550 -0.1290   1.0   0.5
O 1-   5   0.1510 -0.0030  0.1290   1.0   0.5
O 1-   6  -0.3320 -0.1550 -0.1210   1.0   0.5
O 1-   7   0.0990 -0.1820 -0.0420   1.0   1.0
O 1-   8   0.1780 -0.1030  0.0420   1.0   1.0
O 1-   9  -0.1090 -0.1800 -0.0440   1.0   1.0
O 1-  10   0.1760  0.1050  0.0440   1.0   1.0
O 1-  11  -0.1510  0.3280  0.1210   1.0   0.5
O 1-  12  -0.0010  0.1460 -0.1290   1.0   0.5
O 1-  13  -0.1510 -0.0030  0.1290   1.0   0.5
O 1-  14  -0.3320  0.1460 -0.1210   1.0   0.5
O 1-  15   0.0990  0.1740 -0.0420   1.0   1.0
O 1-  16  -0.1780 -0.1030  0.0420   1.0   1.0
O 1-  17  -0.1100  0.1720 -0.0440   1.0   1.0
O 1-  18  -0.1760  0.1050  0.0450   1.0   1.0
O 1-  19  -0.3850 -0.0050 -0.0570   1.0   1.0
O 1-  20   0.0000  0.3810  0.0570   1.0   1.0
O 1-  21  -0.5000 -0.1770 -0.0640   1.0   0.5
O 1-  22   0.1720  0.5000  0.0640   1.0   0.5
O 1-  23  -0.3130 -0.1860 -0.0230   1.0   1.0
O 1-  24   0.1820  0.3080  0.0230   1.0   1.0
O 1-  25  -0.5000  0.1670 -0.0640   1.0   0.5
O 1-  26  -0.1720  0.5000  0.0640   1.0   0.5
O 1-  27  -0.3130  0.1770 -0.0220   1.0   1.0
O 1-  28  -0.1810  0.3090  0.0230   1.0   1.0
O 1-  29   0.3800 -0.0050 -0.0620   1.0   1.0
O 1-  30   0.0000 -0.3960  0.0620   1.0   1.0
O 1-  31   0.3050 -0.1850 -0.0270   1.0   1.0
O 1-  32   0.1800 -0.3090  0.0270   1.0   1.0
O 1-  33   0.3050  0.1750 -0.0270   1.0   1.0
O 1-  34  -0.1800 -0.3090  0.0270   1.0   1.0
O 1-  35   0.1550 -0.3380  0.1250   1.0   0.5
O 1-  36   0.3330 -0.1590 -0.1250   1.0   0.5
O 1-  37  -0.1540 -0.3380  0.1250   1.0   0.5
O 1-  38   0.3330  0.1500 -0.1250   1.0   0.5
O 1-  39  -0.1720 -0.5000  0.0640   1.0   0.5
```

```
O 1-  40  0.1720 -0.5000  0.0640  1.0  0.5
O 1-  41  0.5000 -0.1770 -0.0640  1.0  0.5
O 1-  42  0.5000  0.1670 -0.0640  1.0  0.5

LAYER    2       {same as layer 1 rotated by +90 degrees about c}
NONE            {no special symmetry such as centrosymmetric}
Si4+  1   0.1310 -0.0040 -0.0690  1.0  1.0
Si4+  2   0.0010  0.1260  0.0690  1.0  1.0
Si4+  3  -0.1220 -0.0040 -0.0690  1.0  1.0
Si4+  4   0.0010 -0.1260  0.0690  1.0  1.0
Si4+  5   0.1310 -0.3830 -0.0660  1.0  1.0
Si4+  6  -0.3790  0.1260  0.0660  1.0  1.0
Si4+  7  -0.1210 -0.3830 -0.0660  1.0  1.0
Si4+  8  -0.3790 -0.1260  0.0660  1.0  1.0
Si4+  9   0.1310  0.3790 -0.0690  1.0  1.0
Si4+ 10   0.3840  0.1270  0.0690  1.0  1.0
Si4+ 11  -0.1220  0.3790 -0.0690  1.0  1.0
Si4+ 12   0.3840 -0.1260  0.0690  1.0  1.0
Si4+ 13   0.1950 -0.1950  0.0000  1.0  1.0
Si4+ 14  -0.1910  0.1900  0.0000  1.0  1.0
Si4+ 15  -0.1910 -0.1950  0.0000  1.0  1.0
Si4+ 16   0.1950  0.1900  0.0000  1.0  1.0
O 1-   1   0.0040 -0.0030 -0.0600  1.0  1.0
O 1-   2   0.0010  0.0000  0.0600  1.0  1.0
O 1-   3  -0.3280  0.1510  0.1210  1.0  0.5
O 1-   4   0.1550 -0.0010 -0.1290  1.0  0.5
O 1-   5   0.0030  0.1510  0.1290  1.0  0.5
O 1-   6   0.1550 -0.3320 -0.1210  1.0  0.5
O 1-   7   0.1820  0.0990 -0.0420  1.0  1.0
O 1-   8   0.1030  0.1780  0.0420  1.0  1.0
O 1-   9   0.1800 -0.1090 -0.0440  1.0  1.0
O 1-  10  -0.1050  0.1760  0.0440  1.0  1.0
O 1-  11  -0.3280 -0.1510  0.1210  1.0  0.5
O 1-  12  -0.1460 -0.0010 -0.1290  1.0  0.5
O 1-  13   0.0030 -0.1510  0.1290  1.0  0.5
O 1-  14  -0.1460 -0.3320 -0.1210  1.0  0.5
O 1-  15  -0.1740  0.0990 -0.0420  1.0  1.0
O 1-  16   0.1030 -0.1780  0.0420  1.0  1.0
O 1-  17  -0.1720 -0.1100 -0.0440  1.0  1.0
O 1-  18  -0.1050 -0.1760  0.0450  1.0  1.0
O 1-  19   0.0050 -0.3850 -0.0570  1.0  1.0
O 1-  20  -0.3810  0.0000  0.0570  1.0  1.0
O 1-  21   0.1770 -0.5000 -0.0640  1.0  0.5
O 1-  22  -0.5000  0.1720  0.0640  1.0  0.5
O 1-  23   0.1860 -0.3130 -0.0230  1.0  1.0
O 1-  24  -0.3080  0.1820  0.0230  1.0  1.0
O 1-  25  -0.1670 -0.5000 -0.0640  1.0  0.5
O 1-  26  -0.5000 -0.1720  0.0640  1.0  0.5
O 1-  27  -0.1770 -0.3130 -0.0220  1.0  1.0
O 1-  28  -0.3090 -0.1810  0.0230  1.0  1.0
O 1-  29   0.0050  0.3800 -0.0620  1.0  1.0
O 1-  30   0.3960  0.0000  0.0620  1.0  1.0
O 1-  31   0.1850  0.3050 -0.0270  1.0  1.0
O 1-  32   0.3090  0.1800  0.0270  1.0  1.0
O 1-  33  -0.1750  0.3050 -0.0270  1.0  1.0
O 1-  34   0.3090 -0.1800  0.0270  1.0  1.0
O 1-  35   0.3380  0.1550  0.1250  1.0  0.5
O 1-  36   0.1590  0.3330 -0.1250  1.0  0.5
O 1-  37   0.3380 -0.1540  0.1250  1.0  0.5
O 1-  38  -0.1500  0.3330 -0.1250  1.0  0.5
O 1-  39   0.5000 -0.1720  0.0640  1.0  0.5
O 1-  40   0.5000  0.1720  0.0640  1.0  0.5
O 1-  41   0.1770  0.5000 -0.0640  1.0  0.5
O 1-  42  -0.1670  0.5000 -0.0640  1.0  0.5
```

```
LAYER    3    {same as layer 2 rotated by +90 degrees about c}
NONE         {no special symmetry such as centrosymmetric}
Si4+  1   0.0040  0.1310 -0.0690  1.0  1.0
Si4+  2  -0.1260  0.0010  0.0690  1.0  1.0
Si4+  3   0.0040 -0.1220 -0.0690  1.0  1.0
Si4+  4   0.1260  0.0010  0.0690  1.0  1.0
Si4+  5   0.3830  0.1310 -0.0660  1.0  1.0
Si4+  6  -0.1260 -0.3790  0.0660  1.0  1.0
Si4+  7   0.3830 -0.1210 -0.0660  1.0  1.0
Si4+  8   0.1260 -0.3790  0.0660  1.0  1.0
Si4+  9  -0.3790  0.1310 -0.0690  1.0  1.0
Si4+ 10  -0.1270  0.3840  0.0690  1.0  1.0
Si4+ 11  -0.3790 -0.1220 -0.0690  1.0  1.0
Si4+ 12   0.1260  0.3840  0.0690  1.0  1.0
Si4+ 13   0.1950  0.1950  0.0000  1.0  1.0
Si4+ 14  -0.1900 -0.1910  0.0000  1.0  1.0
Si4+ 15   0.1950 -0.1910  0.0000  1.0  1.0
Si4+ 16  -0.1900  0.1950  0.0000  1.0  1.0
O 1-   1   0.0030  0.0040 -0.0600  1.0  1.0
O 1-   2   0.0000  0.0010  0.0600  1.0  1.0
O 1-   3  -0.1510 -0.3280  0.1210  1.0  0.5
O 1-   4   0.0010  0.1550 -0.1290  1.0  0.5
O 1-   5  -0.1510  0.0030  0.1290  1.0  0.5
O 1-   6   0.3320  0.1550 -0.1210  1.0  0.5
O 1-   7  -0.0990  0.1820 -0.0420  1.0  1.0
O 1-   8  -0.1780  0.1030  0.0420  1.0  1.0
O 1-   9   0.1090  0.1800 -0.0440  1.0  1.0
O 1-  10  -0.1760 -0.1050  0.0440  1.0  1.0
O 1-  11   0.1510 -0.3280  0.1210  1.0  0.5
O 1-  12   0.0010 -0.1460 -0.1290  1.0  0.5
O 1-  13   0.1510  0.0030  0.1290  1.0  0.5
O 1-  14   0.3320 -0.1460 -0.1210  1.0  0.5
O 1-  15  -0.0990 -0.1740 -0.0420  1.0  1.0
O 1-  16   0.1780  0.1030  0.0420  1.0  1.0
O 1-  17   0.1100 -0.1720 -0.0440  1.0  1.0
O 1-  18   0.1760 -0.1050  0.0450  1.0  1.0
O 1-  19   0.3850  0.0050 -0.0570  1.0  1.0
O 1-  20   0.0000 -0.3810  0.0570  1.0  1.0
O 1-  21   0.5000  0.1770 -0.0640  1.0  0.5
O 1-  22  -0.1720 -0.5000  0.0640  1.0  0.5
O 1-  23   0.3130  0.1860 -0.0230  1.0  1.0
O 1-  24  -0.1820 -0.3080  0.0230  1.0  1.0
O 1-  25   0.5000 -0.1670 -0.0640  1.0  0.5
O 1-  26   0.1720 -0.5000  0.0640  1.0  0.5
O 1-  27   0.3130 -0.1770 -0.0220  1.0  1.0
O 1-  28   0.1810 -0.3090  0.0230  1.0  1.0
O 1-  29  -0.3800  0.0050 -0.0620  1.0  1.0
O 1-  30   0.0000  0.3960  0.0620  1.0  1.0
O 1-  31  -0.3050  0.1850 -0.0270  1.0  1.0
O 1-  32  -0.1800  0.3090  0.0270  1.0  1.0
O 1-  33  -0.3050 -0.1750 -0.0270  1.0  1.0
O 1-  34   0.1800  0.3090  0.0270  1.0  1.0
O 1-  35  -0.1550  0.3380  0.1250  1.0  0.5
O 1-  36  -0.3330  0.1590 -0.1250  1.0  0.5
O 1-  37   0.1540  0.3380  0.1250  1.0  0.5
O 1-  38  -0.3330 -0.1500 -0.1250  1.0  0.5
O 1-  39   0.1720  0.5000  0.0640  1.0  0.5
O 1-  40  -0.1720  0.5000  0.0640  1.0  0.5
O 1-  41  -0.5000  0.1770 -0.0640  1.0  0.5
O 1-  42  -0.5000 -0.1670 -0.0640  1.0  0.5


LAYER    4    {same as layer 3 rotated by +90 degrees about c}
NONE         {no special symmetry such as centrosymmetric}
```

```
Si4+   1  -0.1310   0.0040  -0.0690   1.0   1.0
Si4+   2  -0.0010  -0.1260   0.0690   1.0   1.0
Si4+   3   0.1220   0.0040  -0.0690   1.0   1.0
Si4+   4  -0.0010   0.1260   0.0690   1.0   1.0
Si4+   5  -0.1310   0.3830  -0.0660   1.0   1.0
Si4+   6   0.3790  -0.1260   0.0660   1.0   1.0
Si4+   7   0.1210   0.3830  -0.0660   1.0   1.0
Si4+   8   0.3790   0.1260   0.0660   1.0   1.0
Si4+   9  -0.1310  -0.3790  -0.0690   1.0   1.0
Si4+  10  -0.3840  -0.1270   0.0690   1.0   1.0
Si4+  11   0.1220  -0.3790  -0.0690   1.0   1.0
Si4+  12  -0.3840   0.1260   0.0690   1.0   1.0
Si4+  13  -0.1950   0.1950   0.0000   1.0   1.0
Si4+  14   0.1910  -0.1900   0.0000   1.0   1.0
Si4+  15   0.1910   0.1950   0.0000   1.0   1.0
Si4+  16  -0.1950  -0.1900   0.0000   1.0   1.0
O 1-   1  -0.0040   0.0030  -0.0600   1.0   1.0
O 1-   2  -0.0010   0.0000   0.0600   1.0   1.0
O 1-   3   0.3280  -0.1510   0.1210   1.0   0.5
O 1-   4  -0.1550   0.0010  -0.1290   1.0   0.5
O 1-   5  -0.0030  -0.1510   0.1290   1.0   0.5
O 1-   6  -0.1550   0.3320  -0.1210   1.0   0.5
O 1-   7  -0.1820  -0.0990  -0.0420   1.0   1.0
O 1-   8  -0.1030  -0.1780   0.0420   1.0   1.0
O 1-   9  -0.1800   0.1090  -0.0440   1.0   1.0
O 1-  10   0.1050  -0.1760   0.0440   1.0   1.0
O 1-  11   0.3280   0.1510   0.1210   1.0   0.5
O 1-  12   0.1460   0.0010  -0.1290   1.0   0.5
O 1-  13  -0.0030   0.1510   0.1290   1.0   0.5
O 1-  14   0.1460   0.3320  -0.1210   1.0   0.5
O 1-  15   0.1740  -0.0990  -0.0420   1.0   1.0
O 1-  16  -0.1030   0.1780   0.0420   1.0   1.0
O 1-  17   0.1720   0.1100  -0.0440   1.0   1.0
O 1-  18   0.1050   0.1760   0.0450   1.0   1.0
O 1-  19  -0.0050   0.3850  -0.0570   1.0   1.0
O 1-  20   0.3810   0.0000   0.0570   1.0   1.0
O 1-  21  -0.1770   0.5000  -0.0640   1.0   0.5
O 1-  22   0.5000  -0.1720   0.0640   1.0   0.5
O 1-  23  -0.1860   0.3130  -0.0230   1.0   1.0
O 1-  24   0.3080  -0.1820   0.0230   1.0   1.0
O 1-  25   0.1670   0.5000  -0.0640   1.0   0.5
O 1-  26   0.5000   0.1720   0.0640   1.0   0.5
O 1-  27   0.1770   0.3130  -0.0220   1.0   1.0
O 1-  28   0.3090   0.1810   0.0230   1.0   1.0
O 1-  29  -0.0050  -0.3800  -0.0620   1.0   1.0
O 1-  30  -0.3960   0.0000   0.0620   1.0   1.0
O 1-  31  -0.1850  -0.3050  -0.0270   1.0   1.0
O 1-  32  -0.3090  -0.1800   0.0270   1.0   1.0
O 1-  33   0.1750  -0.3050  -0.0270   1.0   1.0
O 1-  34  -0.3090   0.1800   0.0270   1.0   1.0
O 1-  35  -0.3380  -0.1550   0.1250   1.0   0.5
O 1-  36  -0.1590  -0.3330  -0.1250   1.0   0.5
O 1-  37  -0.3380   0.1540   0.1250   1.0   0.5
O 1-  38   0.1500  -0.3330  -0.1250   1.0   0.5
O 1-  39  -0.5000   0.1720   0.0640   1.0   0.5
O 1-  40  -0.5000  -0.1720   0.0640   1.0   0.5
O 1-  41  -0.1770  -0.5000  -0.0640   1.0   0.5
O 1-  42   0.1670  -0.5000  -0.0640   1.0   0.5


{The following four sheets are the left handed versions of the first four}
{They are related to the first four by a c-glide operation}
LAYER    5      {layer 2 mirrored across a-c and b-c planes}
NONE            {no special symmetry such as centrosymmetric}
Si4+   1 -0.0040   0.1310  -0.0690   1.0   1.0
```

```
Si4+    2   0.1260   0.0010   0.0690   1.0   1.0
Si4+    3  -0.0040  -0.1220  -0.0690   1.0   1.0
Si4+    4  -0.1260   0.0010   0.0690   1.0   1.0
Si4+    5  -0.3830   0.1310  -0.0660   1.0   1.0
Si4+    6   0.1260  -0.3790   0.0660   1.0   1.0
Si4+    7  -0.3830  -0.1210  -0.0660   1.0   1.0
Si4+    8  -0.1260  -0.3790   0.0660   1.0   1.0
Si4+    9   0.3790   0.1310  -0.0690   1.0   1.0
Si4+   10   0.1270   0.3840   0.0690   1.0   1.0
Si4+   11   0.3790  -0.1220  -0.0690   1.0   1.0
Si4+   12  -0.1260   0.3840   0.0690   1.0   1.0
Si4+   13  -0.1950   0.1950   0.0000   1.0   1.0
Si4+   14   0.1900  -0.1910   0.0000   1.0   1.0
Si4+   15  -0.1950  -0.1910   0.0000   1.0   1.0
Si4+   16   0.1900   0.1950   0.0000   1.0   1.0
O 1-    1  -0.0030   0.0040  -0.0600   1.0   1.0
O 1-    2   0.0000   0.0010   0.0600   1.0   1.0
O 1-    3   0.1510  -0.3280   0.1210   1.0   0.5
O 1-    4  -0.0010   0.1550  -0.1290   1.0   0.5
O 1-    5   0.1510   0.0030   0.1290   1.0   0.5
O 1-    6  -0.3320   0.1550  -0.1210   1.0   0.5
O 1-    7   0.0990   0.1820  -0.0420   1.0   1.0
O 1-    8   0.1780   0.1030   0.0420   1.0   1.0
O 1-    9  -0.1090   0.1800  -0.0440   1.0   1.0
O 1-   10   0.1760  -0.1050   0.0440   1.0   1.0
O 1-   11  -0.1510  -0.3280   0.1210   1.0   0.5
O 1-   12  -0.0010  -0.1460  -0.1290   1.0   0.5
O 1-   13  -0.1510   0.0030   0.1290   1.0   0.5
O 1-   14  -0.3320  -0.1460  -0.1210   1.0   0.5
O 1-   15   0.0990  -0.1740  -0.0420   1.0   1.0
O 1-   16  -0.1780   0.1030   0.0420   1.0   1.0
O 1-   17  -0.1100  -0.1720  -0.0440   1.0   1.0
O 1-   18  -0.1760  -0.1050   0.0450   1.0   1.0
O 1-   19  -0.3850   0.0050  -0.0570   1.0   1.0
O 1-   20   0.0000  -0.3810   0.0570   1.0   1.0
O 1-   21  -0.5000   0.1770  -0.0640   1.0   0.5
O 1-   22   0.1720  -0.5000   0.0640   1.0   0.5
O 1-   23  -0.3130   0.1860  -0.0230   1.0   1.0
O 1-   24   0.1820  -0.3080   0.0230   1.0   1.0
O 1-   25  -0.5000  -0.1670  -0.0640   1.0   0.5
O 1-   26  -0.1720  -0.5000   0.0640   1.0   0.5
O 1-   27  -0.3130  -0.1770  -0.0220   1.0   1.0
O 1-   28  -0.1810  -0.3090   0.0230   1.0   1.0
O 1-   29   0.3800   0.0050  -0.0620   1.0   1.0
O 1-   30   0.0000   0.3960   0.0620   1.0   1.0
O 1-   31   0.3050   0.1850  -0.0270   1.0   1.0
O 1-   32   0.1800   0.3090   0.0270   1.0   1.0
O 1-   33   0.3050  -0.1750  -0.0270   1.0   1.0
O 1-   34  -0.1800   0.3090   0.0270   1.0   1.0
O 1-   35   0.1550   0.3380   0.1250   1.0   0.5
O 1-   36   0.3330   0.1590  -0.1250   1.0   0.5
O 1-   37  -0.1540   0.3380   0.1250   1.0   0.5
O 1-   38   0.3330  -0.1500  -0.1250   1.0   0.5
O 1-   39  -0.1720   0.5000   0.0640   1.0   0.5
O 1-   40   0.1720   0.5000   0.0640   1.0   0.5
O 1-   41   0.5000   0.1770  -0.0640   1.0   0.5
O 1-   42   0.5000  -0.1670  -0.0640   1.0   0.5


LAYER    6    {layer 3 mirrored across a-c and b-c planes}
NONE          {no special symmetry such as centrosymmetric}
Si4+    1  -0.1310  -0.0040  -0.0690   1.0   1.0
Si4+    2  -0.0010   0.1260   0.0690   1.0   1.0
Si4+    3   0.1220  -0.0040  -0.0690   1.0   1.0
Si4+    4  -0.0010  -0.1260   0.0690   1.0   1.0
```

```
Si4+    5 -0.1310 -0.3830 -0.0660   1.0   1.0
Si4+    6  0.3790  0.1260  0.0660   1.0   1.0
Si4+    7  0.1210 -0.3830 -0.0660   1.0   1.0
Si4+    8  0.3790 -0.1260  0.0660   1.0   1.0
Si4+    9 -0.1310  0.3790 -0.0690   1.0   1.0
Si4+   10 -0.3840  0.1270  0.0690   1.0   1.0
Si4+   11  0.1220  0.3790 -0.0690   1.0   1.0
Si4+   12 -0.3840 -0.1260  0.0690   1.0   1.0
Si4+   13 -0.1950 -0.1950  0.0000   1.0   1.0
Si4+   14  0.1910  0.1900  0.0000   1.0   1.0
Si4+   15  0.1910 -0.1950  0.0000   1.0   1.0
Si4+   16 -0.1950  0.1900  0.0000   1.0   1.0
O 1-    1 -0.0040 -0.0030 -0.0600   1.0   1.0
O 1-    2 -0.0010  0.0000  0.0600   1.0   1.0
O 1-    3  0.3280  0.1510  0.1210   1.0   0.5
O 1-    4 -0.1550 -0.0010 -0.1290   1.0   0.5
O 1-    5 -0.0030  0.1510  0.1290   1.0   0.5
O 1-    6 -0.1550 -0.3320 -0.1210   1.0   0.5
O 1-    7 -0.1820  0.0990 -0.0420   1.0   1.0
O 1-    8 -0.1030  0.1780  0.0420   1.0   1.0
O 1-    9 -0.1800 -0.1090 -0.0440   1.0   1.0
O 1-   10  0.1050  0.1760  0.0440   1.0   1.0
O 1-   11  0.3280 -0.1510  0.1210   1.0   0.5
O 1-   12  0.1460 -0.0010 -0.1290   1.0   0.5
O 1-   13 -0.0030 -0.1510  0.1290   1.0   0.5
O 1-   14  0.1460 -0.3320 -0.1210   1.0   0.5
O 1-   15  0.1740  0.0990 -0.0420   1.0   1.0
O 1-   16 -0.1030 -0.1780  0.0420   1.0   1.0
O 1-   17  0.1720 -0.1100 -0.0440   1.0   1.0
O 1-   18  0.1050 -0.1760  0.0450   1.0   1.0
O 1-   19 -0.0050 -0.3850 -0.0570   1.0   1.0
O 1-   20  0.3810  0.0000  0.0570   1.0   1.0
O 1-   21 -0.1770 -0.5000 -0.0640   1.0   0.5
O 1-   22  0.5000  0.1720  0.0640   1.0   0.5
O 1-   23 -0.1860 -0.3130 -0.0230   1.0   1.0
O 1-   24  0.3080  0.1820  0.0230   1.0   1.0
O 1-   25  0.1670 -0.5000 -0.0640   1.0   0.5
O 1-   26  0.5000 -0.1720  0.0640   1.0   0.5
O 1-   27  0.1770 -0.3130 -0.0220   1.0   1.0
O 1-   28  0.3090 -0.1810  0.0230   1.0   1.0
O 1-   29 -0.0050  0.3800 -0.0620   1.0   1.0
O 1-   30 -0.3960  0.0000  0.0620   1.0   1.0
O 1-   31 -0.1850  0.3050 -0.0270   1.0   1.0
O 1-   32 -0.3090  0.1800  0.0270   1.0   1.0
O 1-   33  0.1750  0.3050 -0.0270   1.0   1.0
O 1-   34 -0.3090 -0.1800  0.0270   1.0   1.0
O 1-   35 -0.3380  0.1550  0.1250   1.0   0.5
O 1-   36 -0.1590  0.3330 -0.1250   1.0   0.5
O 1-   37 -0.3380 -0.1540  0.1250   1.0   0.5
O 1-   38  0.1500  0.3330 -0.1250   1.0   0.5
O 1-   39 -0.5000 -0.1720  0.0640   1.0   0.5
O 1-   40 -0.5000  0.1720  0.0640   1.0   0.5
O 1-   41 -0.1770  0.5000 -0.0640   1.0   0.5
O 1-   42  0.1670  0.5000 -0.0640   1.0   0.5

LAYER    7    {layer 4 mirrored across a-c and b-c planes}
NONE          {no special symmetry such as centrosymmetric}
Si4+    1  0.0040 -0.1310 -0.0690   1.0   1.0
Si4+    2 -0.1260 -0.0010  0.0690   1.0   1.0
Si4+    3  0.0040  0.1220 -0.0690   1.0   1.0
Si4+    4  0.1260 -0.0010  0.0690   1.0   1.0
Si4+    5  0.3830 -0.1310 -0.0660   1.0   1.0
Si4+    6 -0.1260  0.3790  0.0660   1.0   1.0
Si4+    7  0.3830  0.1210 -0.0660   1.0   1.0
```

```
Si4+   8   0.1260   0.3790   0.0660   1.0   1.0
Si4+   9  -0.3790  -0.1310  -0.0690   1.0   1.0
Si4+  10  -0.1270  -0.3840   0.0690   1.0   1.0
Si4+  11  -0.3790   0.1220  -0.0690   1.0   1.0
Si4+  12   0.1260  -0.3840   0.0690   1.0   1.0
Si4+  13   0.1950  -0.1950   0.0000   1.0   1.0
Si4+  14  -0.1900   0.1910   0.0000   1.0   1.0
Si4+  15   0.1950   0.1910   0.0000   1.0   1.0
Si4+  16  -0.1900  -0.1950   0.0000   1.0   1.0
O 1-   1   0.0030  -0.0040  -0.0600   1.0   1.0
O 1-   2   0.0000  -0.0010   0.0600   1.0   1.0
O 1-   3  -0.1510   0.3280   0.1210   1.0   0.5
O 1-   4   0.0010  -0.1550  -0.1290   1.0   0.5
O 1-   5  -0.1510  -0.0030   0.1290   1.0   0.5
O 1-   6   0.3320  -0.1550  -0.1210   1.0   0.5
O 1-   7  -0.0990  -0.1820  -0.0420   1.0   1.0
O 1-   8  -0.1780  -0.1030   0.0420   1.0   1.0
O 1-   9   0.1090  -0.1800  -0.0440   1.0   1.0
O 1-  10  -0.1760   0.1050   0.0440   1.0   1.0
O 1-  11   0.1510   0.3280   0.1210   1.0   0.5
O 1-  12   0.0010   0.1460  -0.1290   1.0   0.5
O 1-  13   0.1510  -0.0030   0.1290   1.0   0.5
O 1-  14   0.3320   0.1460  -0.1210   1.0   0.5
O 1-  15  -0.0990   0.1740  -0.0420   1.0   1.0
O 1-  16   0.1780  -0.1030   0.0420   1.0   1.0
O 1-  17   0.1100   0.1720  -0.0440   1.0   1.0
O 1-  18   0.1760   0.1050   0.0450   1.0   1.0
O 1-  19   0.3850  -0.0050  -0.0570   1.0   1.0
O 1-  20   0.0000   0.3810   0.0570   1.0   1.0
O 1-  21   0.5000  -0.1770  -0.0640   1.0   0.5
O 1-  22  -0.1720   0.5000   0.0640   1.0   0.5
O 1-  23   0.3130  -0.1860  -0.0230   1.0   1.0
O 1-  24  -0.1820   0.3080   0.0230   1.0   1.0
O 1-  25   0.5000   0.1670  -0.0640   1.0   0.5
O 1-  26   0.1720   0.5000   0.0640   1.0   0.5
O 1-  27   0.3130   0.1770  -0.0220   1.0   1.0
O 1-  28   0.1810   0.3090   0.0230   1.0   1.0
O 1-  29  -0.3800  -0.0050  -0.0620   1.0   1.0
O 1-  30   0.0000  -0.3960   0.0620   1.0   1.0
O 1-  31  -0.3050  -0.1850  -0.0270   1.0   1.0
O 1-  32  -0.1800  -0.3090   0.0270   1.0   1.0
O 1-  33  -0.3050   0.1750  -0.0270   1.0   1.0
O 1-  34   0.1800  -0.3090   0.0270   1.0   1.0
O 1-  35  -0.1550  -0.3380   0.1250   1.0   0.5
O 1-  36  -0.3330  -0.1590  -0.1250   1.0   0.5
O 1-  37   0.1540  -0.3380   0.1250   1.0   0.5
O 1-  38  -0.3330   0.1500  -0.1250   1.0   0.5
O 1-  39   0.1720  -0.5000   0.0640   1.0   0.5
O 1-  40  -0.1720  -0.5000   0.0640   1.0   0.5
O 1-  41  -0.5000  -0.1770  -0.0640   1.0   0.5
O 1-  42  -0.5000   0.1670  -0.0640   1.0   0.5

LAYER   8      {layer 1 mirrored across a-c and b-c planes}
NONE           {no special symmetry such as centrosymmetric}
Si4+   1   0.1310   0.0040  -0.0690   1.0   1.0
Si4+   2   0.0010  -0.1260   0.0690   1.0   1.0
Si4+   3  -0.1220   0.0040  -0.0690   1.0   1.0
Si4+   4   0.0010   0.1260   0.0690   1.0   1.0
Si4+   5   0.1310   0.3830  -0.0660   1.0   1.0
Si4+   6  -0.3790  -0.1260   0.0660   1.0   1.0
Si4+   7  -0.1210   0.3830  -0.0660   1.0   1.0
Si4+   8  -0.3790   0.1260   0.0660   1.0   1.0
Si4+   9   0.1310  -0.3790  -0.0690   1.0   1.0
Si4+  10   0.3840  -0.1270   0.0690   1.0   1.0
```

```
Si4+  11 -0.1220 -0.3790 -0.0690  1.0  1.0
Si4+  12  0.3840  0.1260  0.0690  1.0  1.0
Si4+  13  0.1950  0.1950  0.0000  1.0  1.0
Si4+  14 -0.1910 -0.1900  0.0000  1.0  1.0
Si4+  15 -0.1910  0.1950  0.0000  1.0  1.0
Si4+  16  0.1950 -0.1900  0.0000  1.0  1.0
O 1-   1  0.0040  0.0030 -0.0600  1.0  1.0
O 1-   2  0.0010  0.0000  0.0600  1.0  1.0
O 1-   3 -0.3280 -0.1510  0.1210  1.0  0.5
O 1-   4  0.1550  0.0010 -0.1290  1.0  0.5
O 1-   5  0.0030 -0.1510  0.1290  1.0  0.5
O 1-   6  0.1550  0.3320 -0.1210  1.0  0.5
O 1-   7  0.1820 -0.0990 -0.0420  1.0  1.0
O 1-   8  0.1030 -0.1780  0.0420  1.0  1.0
O 1-   9  0.1800  0.1090 -0.0440  1.0  1.0
O 1-  10 -0.1050 -0.1760  0.0440  1.0  1.0
O 1-  11 -0.3280  0.1510  0.1210  1.0  0.5
O 1-  12 -0.1460  0.0010 -0.1290  1.0  0.5
O 1-  13  0.0030  0.1510  0.1290  1.0  0.5
O 1-  14 -0.1460  0.3320 -0.1210  1.0  0.5
O 1-  15 -0.1740 -0.0990 -0.0420  1.0  1.0
O 1-  16  0.1030  0.1780  0.0420  1.0  1.0
O 1-  17 -0.1720  0.1100 -0.0440  1.0  1.0
O 1-  18 -0.1050  0.1760  0.0450  1.0  1.0
O 1-  19  0.0050  0.3850 -0.0570  1.0  1.0
O 1-  20 -0.3810  0.0000  0.0570  1.0  1.0
O 1-  21  0.1770  0.5000 -0.0640  1.0  0.5
O 1-  22 -0.5000 -0.1720  0.0640  1.0  0.5
O 1-  23  0.1860  0.3130 -0.0230  1.0  1.0
O 1-  24 -0.3080 -0.1820  0.0230  1.0  1.0
O 1-  25 -0.1670  0.5000 -0.0640  1.0  0.5
O 1-  26 -0.5000  0.1720  0.0640  1.0  0.5
O 1-  27 -0.1770  0.3130 -0.0220  1.0  1.0
O 1-  28 -0.3090  0.1810  0.0230  1.0  1.0
O 1-  29  0.0050 -0.3800 -0.0620  1.0  1.0
O 1-  30  0.3960  0.0000  0.0620  1.0  1.0
O 1-  31  0.1850 -0.3050 -0.0270  1.0  1.0
O 1-  32  0.3090 -0.1800  0.0270  1.0  1.0
O 1-  33 -0.1750 -0.3050 -0.0270  1.0  1.0
O 1-  34  0.3090  0.1800  0.0270  1.0  1.0
O 1-  35  0.3380 -0.1550  0.1250  1.0  0.5
O 1-  36  0.1590 -0.3330 -0.1250  1.0  0.5
O 1-  37  0.3380  0.1540  0.1250  1.0  0.5
O 1-  38 -0.1500 -0.3330 -0.1250  1.0  0.5
O 1-  39  0.5000  0.1720  0.0640  1.0  0.5
O 1-  40  0.5000 -0.1720  0.0640  1.0  0.5
O 1-  41  0.1770 -0.5000 -0.0640  1.0  0.5
O 1-  42 -0.1670 -0.5000 -0.0640  1.0  0.5

STACKING
recursive          {use the recursion model}
infinite           {for an infinitely thick crystal, no thickness broadening}

TRANSITIONS

{Right-handed layer 1}
   0.000   0.000   0.000   0.25000  {layer i is never followed by another i}
   0.440   0.000  -0.33333 0.25000  {prob of 0.44 that layer 2 follows layer 1}
   0.000   0.000   0.000   0.25000
   0.000   0.000   0.000   0.25000
   0.000   0.000   0.000   0.25000
   0.000   0.000   0.000   0.25000
   0.000   0.000   0.000   0.25000
   0.560   0.000   0.33333 0.25000  {prob = 0.56 that 1 stacks to LH layer}
```

```
{Right-handed layer 2}
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000
   0.440    0.33333  0.000    0.25000    {RH}
   0.000    0.000    0.000    0.25000
   0.560   -0.33333  0.000    0.25000    {LH}
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000

{Right-handed layer 3}
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000
   0.440    0.000    0.33333  0.25000    {RH}
   0.000    0.000    0.000    0.25000
   0.560    0.000   -0.33333  0.25000    {LH}
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000

{Right-handed layer 4}
   0.440   -0.33333  0.000    0.25000    {RH}
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000
   0.560    0.33333  0.000    0.25000    {LH}
   0.000    0.000    0.000    0.25000

{Left-handed layer 5}
   0.000    0.000    0.000    0.25000
   0.560    0.000   -0.33333  0.25000    {RH}
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000
   0.440    0.000    0.33333  0.25000    {LH}

{Left-handed layer 6}
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000
   0.560    0.33333  0.000    0.25000    {RH}
   0.000    0.000    0.000    0.25000
   0.440   -0.33333  0.000    0.25000    {LH}
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000

{Left-handed layer 7}
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000
   0.560    0.000    0.33333  0.25000    {RH}
   0.000    0.000    0.000    0.25000
   0.440    0.000   -0.33333  0.25000    {LH}
   0.000    0.000    0.000    0.25000
   0.000    0.000    0.000    0.25000

{Left-handed layer 8}
   0.560   -0.33333  0.000    0.25000    {RH}
   0.000    0.000    0.000    0.25000
```

```
0.000    0.000    0.000    0.25000
0.000    0.000    0.000    0.25000
0.000    0.000    0.000    0.25000
0.000    0.000    0.000    0.25000
0.440    0.33333  0.000    0.25000    {LH}
0.000    0.000    0.000    0.25000
```

A.5 Data file describing GaAs/GaAlAs 001 superlattice, `GaAs_GaAlAs.dat.`

The lattice of GaAs has the cubic diamond framework, with the Ga atoms on the (0,0,0) positions and the As on the (1/4,1/4,1/4) positions. GaAlAs also uses the diamond framework, but in this case the Ga and Al share the (0,0,0) sublattice, and the As occupies the (1/4,1/4,1/4) sublattice. Thus, in a supperlattice structure, the As sublattice is continuous. The superlattice can be described by three layer types; type 1 being pure Ga, type 2 pure As, and type 3 being admixed Ga and As. Pure GaAs can be simulated by alternating Ga and As layers. (Pure GaAs could also be described by one layer that is an amalgam of layer types 1 and 2.) In this example we simulate a finite superlattice by defining an explicit sequence of layers, and we list the layer sequence in the data file. Note that GaAlAs has a slightly larger unit cell dimension than GaAs. In the superlattice, however, the in-plane a and b cell dimensions are the same, because of the tetragonal distortion induced by coherency strains. However, the layer separation along c is not the same in the two materials. This difference is accommodated in the data file by defining a common c cell dimension of 5.65 Å, and making the c-component of the stacking vector for the Al-rich layers to be slightly larger. The exact amount is computed from the formula for the strain along z

$$\epsilon_z = \frac{1 + \sigma}{1 - \sigma}\epsilon_0 \tag{19}$$

where the strain $\epsilon_0 = (a_{\text{GaAlAs}} - a_{\text{GaAs}})/(a_{\text{GaAlAs}} + a_{\text{GaAs}})$, with a being the cubic unit cell dimensions for each composition, and σ being the isotropic Poisson ratio for the material. All of the strain is absorbed in the GaAlAs layer because in this instance, we assume that the superlattice is grown on bulk GaAs. Typically, $\sigma = 1/3$, and $\epsilon_0 = 0.0018$, giving $\epsilon_z = 0.0036$. Since for GaAs, $R_z = 0.25$, then for GaAlAs, $R_z = 0.25(1 + 0.0036) = 0.2509$.

```
Data file GaAs_GaAlAs.dat

{data file for an explicit sequence of GaAs/GaAlAs layers}
{with 7 GaAs layers followed by 10 GaAlAs layers}
{In the GaAlAs, Al:Ga ratio is 3:7, with Ga and Al randomly distributed.}
{There are a total of 680 layers}
{The sequence 1 2 1 2 ... represents GaAs, and the sequence 2 3 2 3 ...}
{represents GaAlAs}

INSTRUMENTAL                        {Header for instrumental section}
X-RAY                               {Simulate X-ray diffraction}
1.5418                              {X-ray wavelength}
```

74

Figure 15: [010] views of GaAs and GaAlAs crystals.

```
none                              {No Instrumental braodening}

STRUCTURAL                        {Header for structural section}
5.65 5.65 5.65 90.0               {unit cell coordinates}
Axial                             {We are only interested in the 00l trace}
3                                 {3 layers}
infinite

LAYER 1                                    {(001) plane}
NONE
Ga3+    1  0.0  0.0  0.0  1.0  1.0
Ga3+    2  0.5  0.5  0.0  1.0  1.0          {face centered atom}

LAYER 2                                    {(001) plane}
NONE
As      1  0.0  0.0  0.0  1.0  1.0
As      2  0.5  0.5  0.0  1.0  1.0          {face centered atom}

LAYER 3                                    {(001) plane, Ga and Al randomized}
NONE
Ga3+    1  0.0  0.0  0.0  1.0  0.7
Al3+    1  0.0  0.0  0.0  1.0  0.3
Ga3+    2  0.5  0.5  0.0  1.0  0.7          {face centered Ga3+ atom}
Al3+    2  0.5  0.5  0.0  1.0  0.3          {face centered Al3+atom}

STACKING                          {Explicitly defined superlattice}
explicit
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
```

Figure 16: [010] view of GaAs/GaAlAs superlattice.

```
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
1 2 1 2 1 2 1 2 1 2 1 2 1 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2 3 2
```

```
TRANSITIONS                        {Header for stacking transition data}

{Transitions from layer 1}
0.0    0.25   0.25  0.25    {layer 1 to layer 1}
1.0    0.25   0.25  0.25    {layer 1 to layer 2, diamond glide}
0.0    0.25   0.25  0.25    {layer 1 to layer 3}

{Transitions from layer 2}
```

```
0.5   0.25  -0.25  0.25     {layer 2 to layer 1, diamond glide}
0.0   0.25  -0.25  0.25     {layer 2 to layer 2}
0.5   0.25  -0.25  0.2509  {layer 2 to layer 3, GaAlAs is strained along c-axis}


{Transitions from layer 3}
0.0   0.25   0.25  0.25     {layer 3 to layer 1}
1.0   0.25   0.25  0.2509  {layer 3 to layer 2, GaAlAs is strained along c-axis}
0.0   0.25   0.25  0.25     {layer 3 to layer 3}
```

B   DIFFaX output using a control file.

B.1   The Control File

A control file which will run consecutively the 5 data files found in appendices A.1 to A.5 is given below. The file must be called `control.dif`.

```
{Control file script to run the data given in appendices A.1 through A.5}
{Text in curly braces are comments. Matching braces must be on the same line.}
{This file contains the pre-composed responses to DIFFaX's prompts.}
{When DIFFaX finds this file, control.dif, in the same directory, DIFFaX}
{reads this file for its data instead of prompting the user.}
{This allows the user to run long ''batch'' jobs unattended.}

dia.dat               {File describing random faults in diamond}
1                             {We want a data dump}
1                             {We want a full atomic position dump}
1                             {We want a dump of symmetry evaluations}
3                             {We want a powder pattern}
0 170 .05             {0 to 170 degrees, in steps of 0.05 degrees}
1                             {Full adaptive quadrature on broad peaks}
1                             {Full adaptive quadrature over all l on sharp rows}
Clustdia.dat          {File describing clustered faults in diamond}
0                             {We do not want a data dump}
0                             {We do not want a dump of symmetry evaluations}
3                             {We want a powder pattern}
0 170 .05             {0 to 170 degrees, in steps of 0.05 degrees}
1                             {Full adaptive quadrature on broad peaks}
1                             {Full adaptive quadrature over all l on sharp rows}
FAU.dat               {File describing random faults in faujasite}
0                             {We do not want a data dump}
0                             {We do not want a dump of symmetry evaluations}
3                             {We want a powder pattern}
0 25 .01              {0 to 25 degrees, in steps of 0.01 degrees}
1                             {Full adaptive quadrature on broad peaks}
1                             {Full adaptive quadrature over all l on sharp rows}
beta.dat              {File describing random faults, with ''memory'', in zeolite beta}
0                             {We do not want a data dump}
0                             {We do not want a dump of symmetry evaluations}
3                             {We want a powder pattern}
0 25 .01              {0 to 25 degrees, in steps of 0.01 degrees}
1                             {Full adaptive quadrature on broad peaks}
1                             {Full adaptive quadrature over all l on sharp rows}
GaAs_GaAlAs.dat   {File describing an explicitly defined superlattice}
0                             {No data dump. No symmetry dump - file specified 'Axial'}
1                             {We want a line trace, or streak}
1                             {Full adaptive quadrature on broad peaks}
0 0 3.5 4.5 .001      {h, k, l0, l1, delta_l}
0                             {Do not return to the function menu}
end                   {Stop}
```

B.2 Output from the Control File

The output from DIFFaX, using the above control file, is listed below. The values read from the control file are indented.

```
****************************************************
****************************************************
*                                                  *
*  DDDD      II   FFFFFF   FFFFFF        X   X   *
*  D   D    II   F        F           aaaa   X X   *
*  D    D   II   FFFF     FFFF     a    a    X    *
*  D   D    II   F        F        a   aa   X X   *
*  DDDD     II   F        F         aaa a   X   X  *
*                                                  *
****************************************************
***************** DIFFaX v1.801 ******************
****************************************************
**************** 14th JULY, 1995 ****************
****************************************************
*                                                  *
*   A computer program for calculating        *
*   Diffraction Intensity From Faulted Crystals    *
*                                                  *
*   Authors: Michael M. J. Treacy             *
*            Michael W. Deem                   *
*                                                  *
****************************************************

Enter name of structure file.
  dia.dat
Looking for structure data file 'dia.dat'
Opening structure file 'dia.dat'
Enter 1 for DUMP
  1
Reading 'dia.dat'
'dia.dat' read in.
Scattering factor data read in.
Enter 1 for full atomic position dump
  1
Writing data dump to file 'dia.dmp'. . .
Using a ''detune'' value of  0.10000E-02
Enter 1 for a dump of the symmetry evaluations
  1
Checking for conflicts in atom positions . . .
Evaluating point group symmetry of diffraction data. . .
Writing symmetry data to file 'dia.sym'. . .
The diffraction data fits the point group symmetry '6/MMM'
  with a tolerance of one part in  12376
Enter function number:
0 POINT, 1 STREAK, 2 INTEGRATE, 3 POWDER PATTERN, 4 SADP
  3

CALCULATING POWDER DIFFRACTION PATTERN. . .
Enter angular range:
  2theta min, 2theta max, 2theta increment.
 0.0000        170.0         0.5000E-01
Enter 1 for adaptive quadrature on broad peaks.
  1
Enter 1 for adaptive quadrature over all l values
on rows with ''sharp'' spots
  1
Integrating along l at    0   0     'dia.dat'
Full adaptive integration
```

```
Integrating along l at     1   0      'dia.dat'
Full adaptive integration
Integrating along l at     1   1      'dia.dat'
Full adaptive integration
Integrating along l at     2   0      'dia.dat'
Full adaptive integration
Integrating along l at     2   1      'dia.dat'
Full adaptive integration
Adding pseudo-Voigt instrumental broadening
Writing spectrum data to file 'dia.spc'. . .
Spectrum written.

Enter name of structure file.
  Clustdia.dat
Looking for structure data file 'Clustdia.dat'
Opening structure file 'Clustdia.dat'
Enter 1 for DUMP
  0
Reading 'Clustdia.dat'
'Clustdia.dat' read in.
Scattering factor data read in.
Using a ''detune'' value of   0.10000E-02
Enter 1 for a dump of the symmetry evaluations
  0
Checking for conflicts in atom positions . . .
Evaluating point group symmetry of diffraction data. . .
Diffraction point symmetry is 6/MMM
  to within a tolerance of one part in    100
Enter function number:
0 POINT, 1 STREAK, 2 INTEGRATE, 3 POWDER PATTERN, 4 SADP
  3

CALCULATING POWDER DIFFRACTION PATTERN. . .
Enter angular range:
  2theta min, 2theta max, 2theta increment.
 0.0000        170.0         0.5000E-01
Enter 1 for adaptive quadrature on broad peaks.
  1
Enter 1 for adaptive quadrature over all l values
on rows with ''sharp'' spots
  1
Integrating along l at     0   0      'Clustdia.dat'
Full adaptive integration
Integrating along l at     1   0      'Clustdia.dat'
Full adaptive integration
Integrating along l at     1   1      'Clustdia.dat'
Full adaptive integration
Integrating along l at     2   0      'Clustdia.dat'
Full adaptive integration
Integrating along l at     2   1      'Clustdia.dat'
Full adaptive integration
Adding pseudo-Voigt instrumental broadening
Writing spectrum data to file 'Clustdia.spc'. . .
Spectrum written.

Enter name of structure file.
  FAU.dat
Looking for structure data file 'FAU.dat'
Opening structure file 'FAU.dat'
Enter 1 for DUMP
  0
Reading 'FAU.dat'
'FAU.dat' read in.
Scattering factor data read in.
```

```
Using a ''detune'' value of  0.10000E-02
Enter 1 for a dump of the symmetry evaluations
   0
Checking for conflicts in atom positions . . .
Evaluating point group symmetry of diffraction data. . .
Diffraction point symmetry is 6/MMM
   to within a tolerance of one part in    100
Enter function number:
0 POINT, 1 STREAK, 2 INTEGRATE, 3 POWDER PATTERN, 4 SADP
   3

CALCULATING POWDER DIFFRACTION PATTERN. . .
Enter angular range:
   2theta min, 2theta max, 2theta increment.
 0.0000         25.00          0.1000E-01
Enter 1 for adaptive quadrature on broad peaks.
   1
Enter 1 for adaptive quadrature over all l values
on rows with ''sharp'' spots
   1
Integrating along l at    0   0      'FAU.dat'
Full adaptive integration
Integrating along l at    1   0      'FAU.dat'
Full adaptive integration
Integrating along l at    1   1      'FAU.dat'
Full adaptive integration
Integrating along l at    2   0      'FAU.dat'
Full adaptive integration
Integrating along l at    2   1      'FAU.dat'
Full adaptive integration
Integrating along l at    2   2      'FAU.dat'
Full adaptive integration
Integrating along l at    3   0      'FAU.dat'
Full adaptive integration
Integrating along l at    3   1      'FAU.dat'
Full adaptive integration
Integrating along l at    4   0      'FAU.dat'
Full adaptive integration
Adding pseudo-Voigt instrumental broadening
Writing spectrum data to file 'FAU.spc'. . .
Spectrum written.

Enter name of structure file.
   beta.dat
Looking for structure data file 'beta.dat'
Opening structure file 'beta.dat'
Enter 1 for DUMP
   0
Reading 'beta.dat'
'beta.dat' read in.
Scattering factor data read in.
Using a "detune" value of  0.10000E-02
Enter 1 for a dump of the symmetry evaluations
   0
Checking for conflicts in atom positions . . .

WARNING: Atoms from layer  2 extend into layer  1

WARNING: Atoms from layer  8 extend into layer  1

WARNING: Atoms from layer  3 extend into layer  2

WARNING: Atoms from layer  5 extend into layer  2
```

```
WARNING: Atoms from layer  4 extend into layer  3

WARNING: Atoms from layer  6 extend into layer  3

WARNING: Atoms from layer  1 extend into layer  4

WARNING: Atoms from layer  7 extend into layer  4

WARNING: Atoms from layer  2 extend into layer  5

WARNING: Atoms from layer  8 extend into layer  5

WARNING: Atoms from layer  3 extend into layer  6

WARNING: Atoms from layer  5 extend into layer  6

WARNING: Atoms from layer  4 extend into layer  7

WARNING: Atoms from layer  6 extend into layer  7

WARNING: Atoms from layer  1 extend into layer  8

WARNING: Atoms from layer  7 extend into layer  8
Evaluating point group symmetry of diffraction data. . .
Diffraction point symmetry is 4/MMM
   to within a tolerance of one part in     100
Enter function number:
0 POINT, 1 STREAK, 2 INTEGRATE, 3 POWDER PATTERN, 4 SADP
  3

CALCULATING POWDER DIFFRACTION PATTERN. . .
Enter angular range:
  2theta min, 2theta max, 2theta increment.
 0.0000         25.00         0.1000E-01
Enter 1 for adaptive quadrature on broad peaks.
  1
Enter 1 for adaptive quadrature over all l values
on rows with "sharp" spots
  1
Integrating along l at    0   0      'beta.dat'
Full adaptive integration
Integrating along l at    1   0      'beta.dat'
Full adaptive integration
Integrating along l at    1   1      'beta.dat'
Full adaptive integration
Integrating along l at    2   0      'beta.dat'
Full adaptive integration
Integrating along l at    2   1      'beta.dat'
Full adaptive integration
Integrating along l at    2   2      'beta.dat'
Full adaptive integration
Integrating along l at    3   0      'beta.dat'
Full adaptive integration
Integrating along l at    3   1      'beta.dat'
Full adaptive integration
Adding pseudo-Voigt instrumental broadening
Writing spectrum data to file 'beta.spc'. . .
Spectrum written.

Enter name of structure file.
  GaAs_GaAlAs.dat
Looking for structure data file 'GaAs_GaAlAs.dat'
Opening structure file 'GaAs_GaAlAs.dat'
Enter 1 for DUMP
```

```
    0
Reading 'GaAs_GaAlAs.dat'
'GaAs_GaAlAs.dat' read in.
Scattering factor data read in.
Using a ''detune'' value of   0.10000E-02
Checking for conflicts in atom positions . . .
Checking for conflicts in layer stackings . . .
Axial integration only selected.
Enter function number:
0 POINT, 1 STREAK, 2 INTEGRATE, 3 POWDER PATTERN, 4 SADP
   1

CALCULATING INTENSITY ALONG A STREAK. . .
Enter 1 for adaptive quadrature
   1
Enter h, k, l0, l1, delta l
   0   0   3.50000   4.50000   0.00100
Writing streak data to file 'GaAs_GaAlAs.str'. . .
Reached l =      3.54900
Reached l =      3.59900
Reached l =      3.64900
Reached l =      3.69900
Reached l =      3.74900
Reached l =      3.79900
Reached l =      3.84900
Reached l =      3.89900
Reached l =      3.94900
Reached l =      3.99900
Reached l =      4.04900
Reached l =      4.09900
Reached l =      4.14900
Reached l =      4.19900
Reached l =      4.24900
Reached l =      4.29900
Reached l =      4.34900
Reached l =      4.39900
Reached l =      4.44900
Reached l =      4.49900
Streak data file, 'GaAs_GaAlAs.str' written to disk.
Enter 1 to return to function menu.
   0
Enter name of structure file.
end
DIFFaX ended normally.
```

B.3   DIFFaX output for diamond data file `dia.dat` in appendix A.1

Contents of the data dump file, `dia.dmp` for random faults in diamond.

The control file `control.dif` suggested in appendix B.1 requested a dump of the data file. The data file was called `dia.dat`. The data dump is called `dia.dmp`. This file name is assigned automatically by DIFFaX. The file `dia.dmp` should look like;

Contents of file `dia.dmp`

```
                    Number of layers =     2
             Number of unique layers =     2
        Number of different atom types =     1
 cell_a,        cell_b,        cell_c,        cell_gamma
  2.5200         2.5200         2.0600         120.00
 Layers are to be treated as having infinite lateral width.
```

```
radiation type = X-RAY
Wavelength lambda =   1.5418
Diffraction point group symmetry specified = 6/MMM
Instrumental broadening to be simulated = PSEUDO-VOIGT
Full width half-maximum =  0.10000
  Intensity near origin will be ignored


LAYER    1 is equivalent to fundamental LAYER    1
   Existence probability =  0.50000
LAYER    2 is equivalent to fundamental LAYER    2
   Existence probability =  0.50000

fundamental LAYER    1
symmetry = CENTROSYMMETRIC
LAYER composition is:
                   C   :   2.00


ATOM number  1 in layer 1 is 'C   '
   x_rel        y_rel        z_rel         DW          Occ
 -0.33333     -0.16667     -0.12500      1.0000       1.0000
X-ray scattering factor data Ai, Bi
  2.3100       20.844       1.0200       10.207       1.5886
 0.56870       0.86500      51.651       0.21560

fundamental LAYER    2
symmetry = CENTROSYMMETRIC
LAYER composition is:
                   C   :   2.00


ATOM number  1 in layer 2 is 'C   '
   x_rel        y_rel        z_rel         DW          Occ
  0.33333      0.16667     -0.12500      1.0000       1.0000
X-ray scattering factor data Ai, Bi
  2.3100       20.844       1.0200       10.207       1.5886
 0.56870       0.86500      51.651       0.21560


Average crystal composition is:
                   C   : 1.0000


STACKING
Stacking is to be treated RECURSIVELY by DIFFaX.
Number of layers along the fault direction is INFINITE


TRANSITIONS
Layer stacking probabilities and stacking vectors.
  i  j |   alpha_ij       Rx_ij         Ry_ij         Rz_ij
  -----|-------------------------------------------------
       |
  1  1 |  0.70000       0.66667       0.33333       1.0000
  1  2 |  0.30000       0.00000       0.00000       1.0000
       |
  2  1 |  0.30000       0.00000       0.00000       1.0000
  2  2 |  0.70000      -0.66667      -0.33333       1.0000

Anisotropic Layer stacking 'uncertainty' factors.
  i  j |    C11      C22      C33      C12      C13      C23
  -----|-------------------------------------------------------
       |
  1  1 |   0.000    0.000    0.000    0.000    0.000    0.000
```

```
  1  2 |    0.000    0.000    0.000    0.000    0.000    0.000
        |
  2  1 |    0.000    0.000    0.000    0.000    0.000    0.000
  2  2 |    0.000    0.000    0.000    0.000    0.000    0.000
```

B.4   Contents of the symmetry evaluation data file `dia.sym`

Contents of the symmetry evaluation data file

The control file `control.dif` also requested a dump of the symmetry evaluations. The symmetry evaluation file is called `dia.sym`. This file name is assigned automatically by DIFFaX. In abbreviated form, the file `dia.sym` should look like;

Contents of file `dia.sym` (partial listing)

```
SYMMETRY EVALUATIONS FOR DATA IN FILE 'dia.dat'
Number of trials per symmetry element =    25
Threshold intensity =           0.612862E-04

Testing for 2-fold axis

   h     k     l                     Intensity
   0     1    1.2675                   9.535522
   0    -1    1.2675                   9.535522
                         ---------------------
       Average Intensity =             9.535522
     Average variation = +/-           0.000000  (+/- 0.93E-14%)
 Intensity tolerance = +/-             0.095355
 Intensities are consistent with a 2-fold

   h     k     l                     Intensity
   1     0    0.2110                  11.197628
  -1     0    0.2110                  11.197628
                         ---------------------
       Average Intensity =            11.197628
     Average variation = +/-           0.000000  (+/- 0.24E-13%)
 Intensity tolerance = +/-             0.111976
 Intensities are consistent with a 2-fold

.
.
.
.

   h     k     l                     Intensity
   1     1    1.2210                   0.002129
  -1    -1    1.2210                   0.002129
                         ---------------------
       Average Intensity =             0.002129
     Average variation = +/-           0.000000  (+/- 0.00     %)
 Intensity tolerance = +/-             0.000021
 Intensities are consistent with a 2-fold

INTENSITY DISTRIBUTION HAS A 2-FOLD AXIS

Testing for a 3-fold axis

   h     k     l                     Intensity
   1     0    0.3126                  31.816023
  -1     1    0.3126                  31.816217
   0    -1    0.3126                  31.816023
```

```
                      ----------------------
      Average Intensity =                  31.816088
    Average variation = +/-                 0.000086  (+/- 0.27E-03%)
 Intensity tolerance = +/-                  0.318161
 Intensities are consistent with a 3-fold


   1     0     1.7739                       0.763649
  -1     1     1.7739                       0.763618
   0    -1     1.7739                       0.763649
                      ----------------------
      Average Intensity =                   0.763639
    Average variation = +/-                 0.000014  (+/- 0.18E-02%)
 Intensity tolerance = +/-                  0.007636
 Intensities are consistent with a 3-fold


 .
 .
 .


   0     1     1.1768                       3.078191
  -1     0     1.1768                       3.078191
   1    -1     1.1768                       3.078178
                      ----------------------
      Average Intensity =                   3.078186
    Average variation = +/-                 0.000006  (+/- 0.18E-03%)
 Intensity tolerance = +/-                  0.030782
 Intensities are consistent with a 3-fold

 INTENSITY DISTRIBUTION HAS A 3-FOLD AXIS


 Testing for mirror about the h-l plane

   h     k     l                        Intensity
   1     1     0.6100                       0.003514
   2    -1     0.6100                       0.003514
                      ----------------------
      Average Intensity =                   0.003514
    Average variation = +/-                 0.000000  (+/- 0.81E-06%)
 Intensity tolerance = +/-                  0.000035
 Intensities are consistent with an h-l mirror plane


   1     1     0.6782                       0.003939
   2    -1     0.6782                       0.003939
                      ----------------------
      Average Intensity =                   0.003939
    Average variation = +/-                 0.000000  (+/- 0.13E-05%)
 Intensity tolerance = +/-                  0.000039
 Intensities are consistent with an h-l mirror plane


 .
 .
 .


   1     1     0.2655                       0.007861
   2    -1     0.2655                       0.007861
                      ----------------------
      Average Intensity =                   0.007861
    Average variation = +/-                 0.000000  (+/- 0.85E-07%)
 Intensity tolerance = +/-                  0.000079
 Intensities are consistent with an h-l mirror plane


 THERE IS A MIRROR ABOUT THE H-L PLANE
```

```
The diffraction data fits the point group symmetry '6/MMM'
  with a tolerance of one part in  12376
```

B.5   Contents of the spectrum intensity file `dia.spc`

The spectrum data is stored as a text file in `dia.spc`. The spectrum data records are printed in tab-delimited format, *i.e.*

'2θ in degrees' tab 'raw spectrum intensity' tab 'instrumentally broadened intensity'

The spectrum data file, in abbreviated form, should look like;

Contents of file `dia.spc`

```
0.00000E+00    .000000        .000000
0.50000E-01  0.144523E+08     .000000
0.10000E+00  0.174739E+07     .000000
0.15000E+00    446514.        .000000
0.20000E+00    162174.        .000000
0.25000E+00    72398.4        .000000
0.30000E+00    37032.3        .000000
0.35000E+00    20864.0        .000000
0.40000E+00    12634.9        .000000
0.45000E+00    8092.45        .000000
0.50000E+00    5420.24        .000000
0.55000E+00    3765.62        .000000
0.60000E+00    2696.97        .000000
0.65000E+00    1981.96        .000000
0.70000E+00    1488.98        .000000
0.75000E+00    1140.20        .000000
0.80000E+00    887.827        .000000
0.85000E+00    701.580        .000000
0.90000E+00    561.714        .000000
0.95000E+00    455.031        .000000
0.10000E+01    372.518        .000000
.
.
.
.
.
0.42600E+02    4.94228        5.00760
0.42650E+02    5.06303        5.12953
0.42700E+02    5.19179        5.25957
0.42750E+02    5.32847        5.39769
0.42800E+02    5.47297        5.54386
0.42850E+02    5.62516        5.69802
0.42900E+02    5.78480        5.86009
0.42950E+02    5.95161        6.02996
0.43000E+02    6.12520        6.20746
0.43050E+02    6.30506        6.39241
0.43100E+02    6.49056        6.58464
0.43150E+02    6.68092        6.78397
0.43200E+02    6.87521        6.99031
0.43250E+02    7.07237        7.20378
0.43300E+02    7.27120        7.42486
0.43350E+02    7.47046        7.65471
0.43400E+02    7.66894        7.89569
0.43450E+02    7.86566        8.15231
0.43500E+02    8.06030        8.43294
0.43550E+02    8.25393        8.75318
0.43600E+02    8.45062        9.14288
```

```
0.43650E+02    8.66125       9.66243
0.43700E+02    8.91362       10.4459
0.43750E+02    9.28563       11.8648
0.43800E+02    10.0503       15.6333
0.43850E+02    12.7649       28.6585
0.43900E+02    74.8518       54.1308
0.43950E+02    115.226       61.2062
0.44000E+02    13.2889       34.3516
0.44050E+02    9.99240       16.9498
0.44100E+02    9.09912       12.0554
0.44150E+02    8.65015       10.3552
0.44200E+02    8.33094       9.42702
0.44250E+02    8.05508       8.80419
0.44300E+02    7.79252       8.32827
0.44350E+02    7.53186       7.92957
0.44400E+02    7.26908       7.57430
0.44450E+02    7.00346       7.24517
0.44500E+02    6.73576       6.93314
.
.
.
.
.
.
0.16900E+03  0.785915E-01  0.511187E-01
0.16905E+03  0.786012E-01  0.503597E-01
0.16910E+03  0.786109E-01  0.495880E-01
0.16915E+03  0.786207E-01  0.488042E-01
0.16920E+03  0.786305E-01  0.480089E-01
0.16925E+03  0.786405E-01  0.472027E-01
0.16930E+03  0.786505E-01  0.463863E-01
0.16935E+03  0.786605E-01  0.455605E-01
0.16940E+03  0.786706E-01  0.447260E-01
0.16945E+03  0.786808E-01  0.438837E-01
0.16950E+03  0.786911E-01  0.430346E-01
0.16955E+03  0.787014E-01  0.421796E-01
0.16960E+03  0.787117E-01  0.413196E-01
0.16965E+03  0.787222E-01  0.404558E-01
0.16970E+03  0.787327E-01  0.395891E-01
0.16975E+03  0.787432E-01  0.387207E-01
0.16980E+03  0.787538E-01  0.378517E-01
0.16985E+03  0.787644E-01  0.369832E-01
0.16990E+03  0.787751E-01  0.361164E-01
0.16995E+03  0.452758E-01  0.352524E-01
0.17000E+03  0.250168E-15  0.343923E-01
```

B.6   Plotted output from the control file run

The powder pattern data can be plotted by any plotting program that accepts tab-delimited text data files. The following plots were constructed using a Macintosh plotting program called Igor^{TM}.

Figure 17: Plot of unbroadened data in file `dia.spc`.

Figure 18: Plot of pseudo-Voigt broadened data in file `dia.spc`.

88

Figure 19: Plot of unbroadened data in file `Clustdia.spc`.

Figure 20: Plot of pseudo-Voigt broadened data in file `Clustdia.spc`.

Figure 21: Plot of unbroadened data in file `FAU.spc`.

Figure 22: Plot of pseudo-Voigt broadened data in file `FAU.spc`.

Figure 23: Plot of unbroadened data in file `beta.spc`.

Figure 24: Plot of pseudo-Voigt broadened data in file `beta.spc`.

Figure 25: Plot of the "streak" data in `GaAs/GaAlAs.str`.

Figure 26: Plot of the "streak" data in `GaAs/GaAlAs.str`, expanded about $l = 4$.

B.7   Reprint.

A general recursion method for calculating diffracted intensities from crystals containing planar faults

By M. M. J. Treacy†, J. M. Newsam‡ and M. W. Deem

Exxon Research and Engineering Company, Route 22 East, Clinton Township, Annandale, New Jersey 08801, U.S.A.

A general recursion algorithm is described for calculating kinematical diffraction intensities from crystals containing coherent planar faults. The method exploits the self-similar stacking sequences that occur when layers stack non-deterministically. Recursion gives a set of simple relations between average interference terms from a statistical crystal, which can be solved as a set of simultaneous equations. The diffracted intensity for a polycrystalline sample is given by the incoherent sum of scattered intensities over an ensemble of crystallites. The relations between this and previous approaches, namely the Hendricks–Teller matrix formulation, the difference equation method, the summed series formula of Cowley, and Michalski's recurrence relations between average phase factors, are discussed. Although formally identical to these previous methods, the present recursive description has an intuitive appeal and proves easier to apply to complex crystal structure types. The method is valid for all types of planar faults, can accommodate long-range stacking correlations, and is applicable to crystals that contain only a finite number of layers. A FORTRAN program DIFFaX, based on this recursion algorithm, has been written and used to simulate powder X-ray (and neutron) diffraction patterns and single crystal electron (kinematical) diffraction patterns. Calculations for diamond–lonsdaleite and for several synthetic zeolite systems that contain high densities of stacking faults are presented as examples.

1. Introduction

1.1. General

Planar faulting is a widespread phenomenon in real crystals. It is manifested by streaking in single crystal diffraction patterns and by index-dependent peak widths and diffuse scattering in powder diffraction patterns. The presence of faulting is particularly noticeable when diffraction data are recorded at the high angular resolutions afforded by today's synchrotron and neutron diffractometers.

Interpretation of such diffraction data requires an ability to simulate the effects of stacking disorder. Although the problem of diffraction from one-dimensionally disordered systems has been treated theoretically by a number of authors over the past five decades, these earlier treatments are specialized and difficult to apply generally, particularly for complicated crystal structures.

In this paper we discuss the principal earlier treatments and then consider the one-dimensional disorder problem in terms of recursion, an approach applied independently by Michalski (Michalski 1988; Michalski *et al.* 1988) to the specific case of close-packed polytypic systems. We derive here a general recursion algorithm that does not require assumptions of layer coherence or of infinite platelet thickness, and which is applicable to any stacking configuration. We show that the recursion approach can be quite general and detail how it relates to the Hendricks & Teller (1942), Cowley (1981) and Michalski (1988) treatments. However, unlike these earlier treatments, the recursion algorithm provides an intuitively straightforward approach to the stacking disorder problem, facilitating its application to complex crystal systems.

A FORTRAN program, DIFFaX (diffracted intensities from faulted Xtals), has been written, implementing this algorithm. Although the present code is too lengthy (*ca.* 6500 lines) to discuss here in detail, the methods used to overcome some of the more pernicious numerical pitfalls are outlined. Examples are given of the application of this program to twinning in the close-packed diamond–lonsdaleite system, and to three families of zeolites that contain variable densities of growth faults.

1.2. *Historical background*

The diffracted intensity from a large sample of faulted crystals is the weighted, incoherent sum of the diffraction patterns arising from each occurring crystallite orientation and defect arrangement. It is straightforward to calculate explicitly the diffracted intensity from any one stacking configuration. However, the computational effort quickly becomes prohibitive when there are many permutations of faults to consider. Mathematical treatments of diffraction from faulted crystals seek to reduce significantly the number of terms that need be calculated explicitly to manageable proportions.

The one-dimensional problem was first tackled by Landau (1937), who gave formulae for the diffracted intensity from crystals containing arrangements of lamellar domains of variable width that are shifted relative to one another. Lifshitz (1937, 1939) extended this treatment to include different layer types and variable layer spacings. Wilson (1942, 1943) presented a difference equation method, where the probability that a given layer type will occur at a given position is related to the probabilities that the layer has occurred at previous layer positions. This treatment was successfully applied to explaining diffraction patterns measured from hexagonal cobalt, and to $AuCu_3$ containing antiphase boundaries. The difference equation approach forms the basis of the standard textbook treatment of diffraction from faulted crystals (Warren 1959, 1969) and has been widely used for studying faulting in close-packed structures.

A more general treatment, based on correlation probability matrices, was developed by Hendricks & Teller (1942), who included the effects of nearest neighbour layer–layer correlations and variable layer–layer stacking vectors. This approach was also used successfully to explain peak broadening data from layered materials such as staged graphite (Cajipe *et al.* 1989), mica and close-packed structures. A computer program using this theory, which calculates the $(00l)$ powder diffraction patterns of interstratified clay minerals (Brindley & Brown 1980), has been developed by Reynolds (1980).

Many authors have since elaborated on the difference equation and Hendricks–Teller descriptions. In particular, Kakinoki & Tomura (1965) described three

distinct approaches to solving the Hendricks–Teller matrix equations and derived explicit solutions for close-packed systems (Kakinoki & Tomura 1951, 1952; Kakinoki 1967). Independently, Allegra (1961, 1964) pointed out that the method of solution adopted by Hendricks & Teller (1942), involving a matrix diagonalization, could be circumvented and replaced by a less costly matrix inversion. In a series of papers, Jagodzinski (1949a–c, 1954) obtained solutions for various ranges of interaction, s (or Reichweite), between layers, allowing for clustering, or correlations between stacking sequences. Within this description, the probability of a given layer occurring depends on the character of those layers up to s layers removed from that given layer. His treatment distinguished between a position disorder due to stacking errors alone, and a type disorder where layers have variable atomic compositions. The case of mixed type/position disorder was also treated by Jagodzinski (1949a) for the case $s = 1$. Gevers (1952, 1954a, b) extended this work, providing formal solutions for the cases $s = 4$ and 6. However, as noted by Jagodzinski, numerical solution becomes time consuming for $s > 3$. Holloway (1969) generalized the solution for close-packed structures in a way that eliminated the need to explicitly solve the difference equations. Recently, several authors (Wellbery 1985; Berliner & Werner 1986) have reviewed and presented adaptations of the difference equation methods.

Michalski (1988) recently showed that the treatment of Holloway (1969) could be further simplified by constructing recursion relations between average phase factors for crystals beginning on each of the various layer types. He derived explicit expressions for faulted polytypic crystals (Michalski *et al.* 1988).

Cowley (1976a, b, 1981) adopted a different approach to the problem of one-dimensional disorder in infinite crystals. By inspecting the individual layer–layer interference terms in the Patterson function, he was able to separate intensity contributions from the nth layer into 'no-fault' and 'fault' terms. The no-fault terms could be summed as a simple geometric series, and the residual fault terms could be grouped as nested subseries which in turn could also be summed explicitly. The nth nested term represented a contribution to the diffracted intensity from faults at the nth layer. He noted that in certain simple cases the whole sequence could be summed in closed form. For more complex structures, the series could be terminated after a few terms, particularly if the fault probabilities were small. Because the fault and no-fault terms were separately summed, the series converges quickly for low fault probabilities. The attraction of this approach is that, with a knowledge of the fault probabilities, the interlayer stacking vectors, and the layer form factors, Cowley's equation provides a perfectly general ready-to-use solution, with no need on the part of the user to scrutinize faulting patterns to derive difference equations. This summed series approach has been effective in studying streaking in electron diffraction patterns from close-packed materials (Cowley & Au 1978) and some faulted minerals (Cowley 1976b). Despite its generality, the equation becomes numerically tedious to evaluate for large fault probabilities and for structures containing many atoms or layer types. Additionally, most faulted structures can be considered to be intermediate members of a family of materials, whose end-members might best be defined by fault probabilities of zero and unity. Such end-members are themselves perfect structures, and the definition of a single unfaulted structure is thus artificial, and the term *fault probability* becomes ambiguous. In general, it is preferable to define stacking, rather than fault probabilities, without a requirement that these probabilities be small, or close to unity.

The difference equation method, the Hendricks–Teller matrix method, and

Cowley's summed series method are related (Kakinoki & Tomura 1965; Kakinoki 1967) and represent different book-keeping methodologies for the stacking sequences and layer types. However, although all of these methods are of proven value for treating close-packed structures, the various mathematical formalisms can obscure the physical aspects of the problem for the general user, and these approaches have proved difficult to adapt for more complex structures. For example, it is only recently (Newsam *et al.* 1988; Vaughan *et al.* 1989), that detailed analyses of diffraction from faulted zeolites have been attempted.

2. Disorder in one dimension

2.1. *Diffraction described recursively*

Although, as outlined above, the problem of calculating diffraction intensity from faulted crystals has been studied by a number of earlier authors, we show here how the problem can be formulated in a simple closed recursive form, and how this description is related to the previous methods. We suppose that a crystal contains a total of N layers, and that there are M distinct types of layer. Each layer type i will have an electron density $\rho_i(\boldsymbol{r})$ about an arbitrary origin. We further suppose that there is a probability α_{ij} that an i-type layer will be followed by a j-type layer, with a vector $\boldsymbol{R}_{ij}$ connecting the two local layer origins. For a crystal which has a layer of type i at the crystal origin, followed by a layer of type j, which in turn is followed by a layer of type k, etc., the scattering potential $V_{ijkl...}^{(N)}(\boldsymbol{r})$ is

$$V_{ijkl...}^{(N)}(\boldsymbol{r}) = \rho_i(\boldsymbol{r}) + \rho_j(\boldsymbol{r} - \boldsymbol{R}_{ij}) + \rho_k(\boldsymbol{r} - \boldsymbol{R}_{ij} - \boldsymbol{R}_{jk}) + \rho_l(\boldsymbol{r} - \boldsymbol{R}_{ij} - \boldsymbol{R}_{jk} - \boldsymbol{R}_{kl}) + \dots. \quad (1)$$

The probability that any given crystal in a large sample will form such a sequence is given by the probability product $g_i \alpha_{ij} \alpha_{jk} \alpha_{kl} \dots$. The term g_i represents the *a priori* probability that a layer of type i will exist, and is found by equating the transition probabilities between type i and the other type layers

$$g_i = \sum_j g_j \alpha_{ji} \quad (2)$$

and from the normalization conditions

$$\sum_i g_i = 1, \quad \sum_j \alpha_{ij} = 1. \quad (3)$$

The kinematically scattered wave function for this sequence is given by the Fourier transform of $V_{ijkl...}^{(N)}(\boldsymbol{r})$ (the Born approximation), thus

$$\phi_{ijkl...}^{(N)}(\boldsymbol{u}) = \int V_{ijkl...}^{(N)}(\boldsymbol{r}) \exp(-2\pi i \boldsymbol{u} \cdot \boldsymbol{r}) \, d\boldsymbol{r}$$

$$= F_i(\boldsymbol{u}) + \exp(-2\pi i \boldsymbol{u} \cdot \boldsymbol{R}_{ij}) F_j(\boldsymbol{u}) + \exp(-2\pi i \boldsymbol{u} \cdot (\boldsymbol{R}_{ij} + \boldsymbol{R}_{jk})) F_k(\boldsymbol{u})$$

$$+ \exp(-2\pi i \boldsymbol{u} \cdot (\boldsymbol{R}_{ij} + \boldsymbol{R}_{jk} + \boldsymbol{R}_{kl})) F_l(\boldsymbol{u}) + \dots. \quad (4)$$

Here, $\boldsymbol{u}$ is the reciprocal space scattering vector and $F_i(\boldsymbol{u})$ is the layer form factor, the Fourier transform of $\rho_i(\boldsymbol{r})$. The diffracted intensity distribution from this sequence is $\phi_{ijkl...}^{(N)*} \phi_{ijkl...}^{(N)}$. The diffracted intensity $I(\boldsymbol{u})$ from a statistical (oriented) ensemble of crystallites is the weighted incoherent sum over all stacking permutations, that is

$$I(\boldsymbol{u}) = \sum_{i,j,k,l,...} g_i \alpha_{ij} \alpha_{jk} \alpha_{kl} \cdots \phi_{ijkl...}^{(N)*} \phi_{ijkl...}^{(N)}. \quad (5)$$

For a crystal of N layers, containing M layer types, there will be M^N stacking permutations (ignoring any degeneracies resulting from special layer symmetries, or

constraints on the character of the R_{ij}). Explicit summations of the type given by equation 5 rapidly become impractical for large N. Considerable simplification can be achieved by judiciously grouping terms. Expanding equation (5) we get

$$\begin{aligned}
I(\boldsymbol{u}) = \sum_{i,j,k,l,\ldots} & g_i \alpha_{ij} \alpha_{jk} \alpha_{kl} \ldots \{F_i^*(\boldsymbol{u}) \\
& + \exp\left(2\pi i \boldsymbol{u} \cdot \boldsymbol{R}_{ij}\right) F_j^*(\boldsymbol{u}) + \exp\left(2\pi i \boldsymbol{u} \cdot (\boldsymbol{R}_{ij} + \boldsymbol{R}_{jk})\right) F_k^*(\boldsymbol{u}) \\
& + \exp\left(2\pi i \boldsymbol{u} \cdot (\boldsymbol{R}_{ij} + \boldsymbol{R}_{jk} + \boldsymbol{R}_{kl})\right) F_l^*(\boldsymbol{u}) + \ldots\} \\
& \{F_i(\boldsymbol{u}) + \exp\left(-2\pi i \boldsymbol{u} \cdot \boldsymbol{R}_{ij}\right) F_j(\boldsymbol{u}) + \exp\left(-2\pi i \boldsymbol{u} \cdot (\boldsymbol{R}_{ij} + \boldsymbol{R}_{jk})\right) F_k(\boldsymbol{u}) \\
& + \exp\left(-2\pi i \boldsymbol{u} \cdot (\boldsymbol{R}_{ij} + \boldsymbol{R}_{jk} + \boldsymbol{R}_{kl})\right) F_l(\boldsymbol{u}) + \ldots\}.
\end{aligned} \tag{6}$$

Now, the ensemble-average scattered wave function $\phi_i^{(N)}(\boldsymbol{u})$, representing the average over all crystals containing exactly N layers which begin on i-type layers, is given by the nested series

$$\phi_i^{(N)}(\boldsymbol{u}) = F_i(\boldsymbol{u}) + \sum_j \alpha_{ij} \exp\left(-2\pi i \boldsymbol{u} \cdot \boldsymbol{R}_{ij}\right) [F_j(\boldsymbol{u}) + \sum_k \alpha_{jk} \exp\left(-2\pi i \boldsymbol{u} \cdot \boldsymbol{R}_{jk}\right) [F_k(\boldsymbol{u})$$
$$+ \sum_l \alpha_{kl} \exp\left(-2\pi i \boldsymbol{u} \cdot \boldsymbol{R}_{kl}\right) [F_l(\boldsymbol{u}) + \ldots]]]. \tag{7}$$

Inspection of equation (7) immediately yields a relation between the $\phi_i^{(N)}(\boldsymbol{u})$ and $\phi_j^{(N-1)}(\boldsymbol{u})$

$$\phi_i^{(N)}(\boldsymbol{u}) = F_i(\boldsymbol{u}) + \sum_j \alpha_{ij} \exp\left(-2\pi i \boldsymbol{u} \cdot \boldsymbol{R}_{ij}\right) \phi_j^{(N-1)}(\boldsymbol{u}),$$

with $\quad\quad\quad \phi_i^{(0)}(\boldsymbol{u}) = 0.$ $\tag{8}$

By using $\phi_i^{(N)}(\boldsymbol{u})$ and its complex conjugate $\phi_i^{(N)}(\boldsymbol{u})^*$, equation (6) can be expanded term by term, to produce the series

$$\begin{aligned}
I(\boldsymbol{u}) = & \sum_i g_i(F_i^*(\boldsymbol{u}) \phi_i^{(N)}(\boldsymbol{u}) + F_i(\boldsymbol{u}) \phi_i^{(N)}(\boldsymbol{u})^* - |F_i(\boldsymbol{u})|^2) \\
& + \sum_{i,j} g_i \alpha_{ij}(F_j^*(\boldsymbol{u}) \phi_j^{(N-1)}(\boldsymbol{u}) + F_j(\boldsymbol{u}) \phi_j^{(N-1)}(\boldsymbol{u})^* - |F_j(\boldsymbol{u})|^2) \\
& + \sum_{i,j,k} g_i \alpha_{ij} \alpha_{jk}(F_k^*(\boldsymbol{u}) \phi_k^{(N-2)}(\boldsymbol{u}) + F_k(\boldsymbol{u}) \phi_k^{(N-2)}(\boldsymbol{u})^* - |F_k(\boldsymbol{u})|^2) + \ldots.
\end{aligned} \tag{9}$$

By using the normalization equation (2), this condenses to

$$I(\boldsymbol{u}) = \sum_{m=0}^{N-1} \sum_i g_i(F_i^*(\boldsymbol{u}) \phi_i^{(N-m)}(\boldsymbol{u}) + F_i(\boldsymbol{u}) \phi_i^{(N-m)}(\boldsymbol{u})^* - |F_i(\boldsymbol{u})|^2). \tag{10}$$

The summation over m can be simplified by rewriting the relations in matrix form. Using $\boldsymbol{\Phi}^{(N)}$ and $\boldsymbol{F}$ for the one-dimensional matrices $[\phi_i^{(N)}(\boldsymbol{u})]$ and $[F_i(\boldsymbol{u})]$, and $\boldsymbol{T}$ for the two dimensional stacking operation probability matrix $[\alpha_{ij} \exp\left(-2\pi i \boldsymbol{u} \cdot \boldsymbol{R}_{ij}\right)]$, equation (8) can be rewritten as

$$\boldsymbol{\Phi}^{(N)} = \boldsymbol{F} + \boldsymbol{T}\boldsymbol{\Phi}^{(N-1)} = \sum_{n=0}^{N-1} \boldsymbol{T}^n \boldsymbol{F}. \tag{11}$$

By using $\boldsymbol{G} \equiv [g_i F_i(\boldsymbol{u})]$, equation (10) becomes

$$I(\boldsymbol{u}) = \sum_{m=0}^{N-1} \sum_{n=0}^{N-m-1} (\boldsymbol{G}^{*\mathrm{T}} \boldsymbol{T}^n \boldsymbol{F} + \boldsymbol{G}^{\mathrm{T}} \boldsymbol{T}^{*n} \boldsymbol{F}^* - \boldsymbol{G}^{*\mathrm{T}} \boldsymbol{F}) \tag{12}$$

G^T is the transpose of G, and G^* is the complex conjugate of G. Equation (12) is at the heart of the Hendricks & Teller approach (Hendricks & Teller 1942; Kakinoki & Tomura 1965). We introduce the quantity

$$\boldsymbol{\varPsi}^{(N)} = \frac{1}{N} \sum_{m=0}^{N-1} \sum_{n=0}^{N-m-1} \boldsymbol{T}^n \boldsymbol{F}$$
$$= (1/N)\{N\boldsymbol{F} + (N-1)\,\boldsymbol{TF} + (N-2)\,\boldsymbol{T}^2\boldsymbol{F} + \dots + \boldsymbol{T}^{N-1}\boldsymbol{F}\}, \tag{13}$$

which represents the average interference term from an N-layer statistical crystal. Thus, there are N single layers which contribute only intra-layer interference terms, $N-1$ nearest neighbour layer pairs, $N-2$ next nearest neighbours, etc. The content of the braces in equation (13) is a well-known geometric series and can be readily summed to give

$$\boldsymbol{\varPsi}^{(N)} = (\boldsymbol{I} - \boldsymbol{T})^{-1}(1/N)\{(N+1)\,\boldsymbol{I} - (\boldsymbol{I} - \boldsymbol{T})^{-1}(\boldsymbol{I} - \boldsymbol{T}^{N+1})\}\,\boldsymbol{F} = (\boldsymbol{I} - \boldsymbol{T})^{-1}\boldsymbol{F}'. \tag{14}$$

I is the unit matrix having the same dimensions as T, and $(\boldsymbol{I} - \boldsymbol{T})^{-1}$ is the inverse of the matrix $(\boldsymbol{I} - \boldsymbol{T})$. Equation (14) can be rewritten in the equivalent form

$$\boldsymbol{\varPsi}^{(N)} = \boldsymbol{F}' + \boldsymbol{T}\boldsymbol{\varPsi}^{(N)}, \tag{15}$$

which emphasizes the recursive nature of the interference terms $\boldsymbol{\varPsi}^{(N)}$.

F' is a modified layer form factor, and is given by

$$\boldsymbol{F}' = (1/N)\{(N+1)\,\boldsymbol{I} - (\boldsymbol{I} - \boldsymbol{T})^{-1}(\boldsymbol{I} - \boldsymbol{T}^{N+1})\}\,\boldsymbol{F}. \tag{16}$$

By using equation (12), the normalized diffracted intensity is

$$I(\boldsymbol{u})/N = \boldsymbol{G}^{*T}\boldsymbol{\varPsi}^{(N)} + \boldsymbol{G}^T\boldsymbol{\varPsi}^{(N)*} - \boldsymbol{G}^{*T}\boldsymbol{F}. \tag{17}$$

For any N, the matrix $(\boldsymbol{I} - \boldsymbol{T})^{-1}(\boldsymbol{I} - \boldsymbol{T}^{N+1})$ in equation (16) is always bounded provided $\det(\boldsymbol{I} - \boldsymbol{T}) \neq 0$. In the special case when $\det(\boldsymbol{I} - \boldsymbol{T}) = 0$, the diffraction vector $\boldsymbol{u}$ is near a sharp spot and all of the $\boldsymbol{u} \cdot \boldsymbol{R}_{ij}$ have integer values. The infinite intensity that occurs in this case is equivalent to a delta function, with finite integrated intensity over the interval $\mathrm{d}\boldsymbol{u}$ spanning the sharp spot at $\boldsymbol{u}$. Analytic broadening of these sharp spots is possible (see §2.2 below), and thus it is found that in general, $\boldsymbol{F}' \sim \boldsymbol{F}$ as $N \to \infty$. Equation (14) then simplifies further (using equation (11) to give $\boldsymbol{\varPsi}^{(N \to \infty)} \equiv \boldsymbol{\varPhi}^{(N \to \infty)}$; that is, in an infinite crystal, the average interference term is equivalent to the ensemble-average scattered wavefunction. At the other extreme, in the limit $N = 1$, $\boldsymbol{\varPsi}^{(1)} \equiv \boldsymbol{\varPhi}^{(1)} = \boldsymbol{F}$ and $\boldsymbol{F}' = (\boldsymbol{I} - \boldsymbol{T})\boldsymbol{F}$ as would be expected if the crystal comprised only one layer.

The recursive nature of the diffraction problem is easiest to visualize for the case of a crystal containing an infinite number of layers. Equation (15) can be rewritten using the ensemble-average scattered wavefunctions $\boldsymbol{\varPhi}$ (where here, $N \to \infty$ is implicit) to give

$$\boldsymbol{\varPhi} = \boldsymbol{F} + \boldsymbol{T}\boldsymbol{\varPhi}. \tag{18}$$

Figure 1 illustrates this recursion relation graphically, for an infinitely thick crystal containing two types of layer, designated 1 and 2. Despite the infinite number of stacking permutations possible, there are only two distinct types of average sequence that can occur, namely, A sequences which begin on a type 1 layer, and B sequences which begin on a type 2 layer. Inspection of the stacking trees in figure 1 shows that these two sequences can be found embedded within one another, as outlined by the boxes in figure 1. Because of the recurrence, the ensemble-average scattered wave function from each type of sequence does not need to be calculated explicitly, as they are intimately related to each other. If $\alpha_{12} = \alpha_{21}$, then layers 1 and

Figure 1. Diagram illustrating the recursive relation between the ensemble average scattered wavefunctions from crystals which have layers of type 1 (left) and type 2 (right) at the origin. Note how the two distinct sequences, A and B, can be found embedded within one another.

2 occur in equal quantities, giving $g_1 = g_2 = \frac{1}{2}$. For this infinite structure, the ensemble-average wavefunctions are given by the simultaneous equations

$$\phi_i(\boldsymbol{u}) = F_i(\boldsymbol{u}) + \sum_{j=1,2} \alpha_{ij} \exp(-2\pi i \boldsymbol{u} \cdot \boldsymbol{R}_{ij}) \phi_j(\boldsymbol{u}), \tag{19}$$

or more explicitly

$$\phi_1(\boldsymbol{u}) = F_1(\boldsymbol{u}) + \alpha_{11} \exp(-2\pi i \boldsymbol{u} \cdot \boldsymbol{R}_{11}) \phi_1(\boldsymbol{u}) + \alpha_{12} \exp(-2\pi i \boldsymbol{u} \cdot \boldsymbol{R}_{12}) \phi_2(\boldsymbol{u}),$$
$$\phi_2(\boldsymbol{u}) = F_2(\boldsymbol{u}) + \alpha_{21} \exp(-2\pi i \boldsymbol{u} \cdot \boldsymbol{R}_{21}) \phi_1(\boldsymbol{u}) + \alpha_{22} \exp(-2\pi i \boldsymbol{u} \cdot \boldsymbol{R}_{22}) \phi_2(\boldsymbol{u}).$$

ϕ_1 and ϕ_2 are readily found by linear algebra and the diffraction intensity is given by

$$\lim_{N \to \infty} \frac{I(\boldsymbol{u})}{N} = \frac{1}{2}(\boldsymbol{F}^* \boldsymbol{\Phi} + \boldsymbol{F} \boldsymbol{\Phi}^* - |\boldsymbol{F}|^2)$$

$$= \frac{1}{2} \sum_{i=1,2} (F_i^*(\boldsymbol{u}) \phi_i(\boldsymbol{u}) + F_i(\boldsymbol{u}) \phi_i^*(\boldsymbol{u}) - |F_i(\boldsymbol{u})|^2). \tag{20}$$

Similar diagrams can be drawn for more complex structures. It should be noted that the recursion model also applies to perfect crystals, even if they have only one layer type with only one possible stacking vector.

Thus computation of the diffracted intensity from a crystal system requires a knowledge of the individual layer structures (from which the layer form factors $F_i(\boldsymbol{u})$ can be computed), the stacking transition probabilities α_{ij}, and the inter-layer vectors $\boldsymbol{R}_{ij}$. The normalized ensemble-average wavefunctions are completely defined by their recursive properties when the stacking operation T is applied (equations (14) and (16)). The diffracted intensity, normalized to the number of layers in the crystal, is then given by equation (17). Provided the system is being treated on a statistical basis, the recursive character of the layer stacking sequences is independent of the particular crystal system being studied, making it straightforward to implement a general computer program that works for all crystal types. The practical limitations are the speed with which the simultaneous equations can be solved, and, in the case of complex structures such as zeolites, the speed with which the layer form factors can be calculated. There is a further concern in the case where N is finite where the computation of T^{N+1} can also become time consuming, even with efficient matrix multiplication algorithms.

The possibility of layer modifications, which depend on the type of stacking transition, can be handled by replacing the F_i with an average value $\langle F_i \rangle$ in the manner suggested by Cowley (1976a)

$$\langle F_i(\boldsymbol{u}) \rangle = F_i(\boldsymbol{u}) + \sum_j \alpha_{ij} \Delta F_{ij}(\boldsymbol{u}), \tag{21}$$

where $F_i + \Delta F_{ij}$ represents the perturbed structure factor of layer i when it is followed by layer j. Such perturbations could arise from subtle strains imposed by the various neighbouring layer types. Layer–layer correlations, which will result in clustering, may be treated either explicitly from the definitions of the α_{ij}, or by representing groupings of layers as distinct layer types.

2.2. *Implementation as a computer program: practical considerations*

A general purpose FORTRAN 77 program, DIFFaX, for calculating diffracted intensity from faulted crystals, has been written by the authors. Versions of DIFFaX have been run on a CRAY X/MP14SE, a VAX 11/8350, a Silicon Graphics Personal Iris and a Macintosh II. In its present version it accepts any crystal system that contains coherent planar defects, that is, all layers must share the same in-plane a, b unit cell vectors and interconnect along c. Although the fundamental algorithm introduced above is not restricted to the case of in-plane coherence, considerable computational speedup is achieved when it is assumed that intensity is restricted to integer h, k reciprocal lattice vectors. The faulting then creates a continuum of intensity along l.

The layer form factors $F_i(\boldsymbol{u})$ are readily calculated by using the relation,

$$F_i(\boldsymbol{u}) = \sum_{p=1}^{n_i} \Omega_p f_p(\boldsymbol{u}) \exp\left(-2M_p(\boldsymbol{u})|\boldsymbol{u}|^2\right) \exp\left(-2\pi \mathrm{i}\boldsymbol{u}\cdot\boldsymbol{r}_p\right), \tag{22}$$

where n_i is the number of atoms in the layer, $\boldsymbol{r}_p$ is the position of atom p relative to the layer origin, Ω_p is the atom site occupancy and M_p is the isotropic Debye–Waller factor. With coordinates given relative to the unit cell a, b and c, $\boldsymbol{u}\cdot\boldsymbol{r}_p = h r_{px} + k r_{py} + l r_{pz}$.

Sharp peaks (i.e. peaks not broadened by the faulting) are a special concern for numerical integration algorithms since they occur at the singularities where $\det(\boldsymbol{I} - \boldsymbol{T}) = 0$. Such sharp peaks are essentially delta functions, and occur at those reciprocal lattice points $\boldsymbol{u}$ where, simultaneously, all of the $\boldsymbol{u}\cdot\boldsymbol{R}_{ij}$ have integer values. Kinematically forbidden spots occur when all the $F_i(\boldsymbol{u}) = 0$, and this may coincide with a sharp peak position.

To integrate intensities numerically in a stable fashion, it is essential to detune the sharp peaks, such that division by zero is avoided and such that the peaks develop a detectable width. A successful trick that does not distort the integrated intensities significantly is to introduce an element of uncertainty in the phases associated with the stacking vectors, that is, replace the $2\pi\boldsymbol{u}\cdot\boldsymbol{R}_{ij}$ with $2\pi\boldsymbol{u}\cdot\boldsymbol{R}_{ij} + \epsilon_{ij}$. If we suppose that there is a gaussian distribution of possible values of ϵ_{ij} with standard deviation σ_{ij}, then the average wavefunction will also be averaged over all possible ϵ_{ij}. The modified recursion relation becomes

$$\begin{aligned}
\Psi_i(\boldsymbol{u}) &= F_i(\boldsymbol{u}) + \sum_j \alpha_{ij} \frac{\Psi_j(\boldsymbol{u})}{\sqrt{(2\pi)}\,\sigma_{ij}} \int_{-\infty}^{+\infty} \exp\left(-\epsilon_{ij}^2/2\sigma_{ij}^2\right) \exp\left(-2\pi \mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{ij} - \mathrm{i}\epsilon_{ij}\right) \mathrm{d}\epsilon_{ij} \\
&= F_i(\boldsymbol{u}) + \sum_j \alpha_{ij} \exp\left(-2\sigma_{ij}^2\right) \exp\left(-2\pi \mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{ij}\right) \Psi_j(\boldsymbol{u}) \\
&= F_i(\boldsymbol{u}) + \sum_j \alpha_{ij}' \exp\left(-2\pi \mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{ij}\right) \psi_j(\boldsymbol{u}).
\end{aligned} \tag{23}$$

The outcome is that, for $\sigma_{\varphi} \leqslant 1$, α_{ij} is replaced by the slightly smaller value

$\alpha'_{ij} \approx \alpha_{ij} \ (1 - 2\sigma^2_{ij})$, and the probability summations, $\Sigma_j \alpha'_{ij}$, are slightly less than unity. This ensures that the determinant $\det (I - T)$ is always positive and non-zero. It is found that setting $2\sigma^2_{ij} = 10^{-3}$ provides sufficient detuning to stabilize the integration while maintaining acceptable integration accuracy for most purposes.

Strictly speaking, a constant value of σ_{ij} for all $u \cdot R_{ij}$ is not physically meaningful. At point u in reciprocal space, it may be interpreted locally as an uncertainty in stacking vector ΔR_{ij} through the relation $\sigma_{ij} \equiv 2\pi u \cdot \Delta R_{ij}$. The value of ΔR_{ij} will vary with diffraction vector u for constant σ_{ij}. In effect, each singularity sees a different interpretation for the uncertainty ΔR_{ij} which becomes smaller in magnitude as u gets larger. This latter property has the desirable effect that the phase term $2\pi i u \cdot \Delta R_{ij}$ does not increase to unacceptably large values at large u.

In a similar manner, a set of anisotropic layer stacking uncertainty parameters may be defined, $B_{11}, B_{22}, B_{33}, B_{12}, B_{23}, B_{31}$. These coefficients act in the same way as the anisotropic temperature factors for atom position uncertainty. Use of such stacking uncertainty parameters is potentially useful for simulating situations where layer stacking is only partially coherent along certain directions, such as in liquid crystals, layered clays or staged graphite intercalation compounds. The transition probability values are replaced by

$$\alpha''_{ij} = \alpha'_{ij} \exp\left[-\tfrac{1}{4}(B_{11} h^2 a^{*2} + B_{22} k^2 b^{*2} + B \quad l^2 c^{*2} + 2B_{12} hka^* b^* \right.$$
$$\left. + 2B_{23} klb^* c^* + 2B_{31} lhc^* a^*) \right], \quad (24)$$

where a^*, b^* and c^* are the reciprocal cell dimensions.

Calculations can be made for any type of radiation, electrons, X-rays, neutrons, etc., through the use of the appropriate atomic scattering factors $f_p(u)$. X-ray intensities are weighted in the usual way by the Lorentz-polarization factors (Warren 1969). For powder electron, and neutron diffraction pattern simulations, there is no polarization term. The geometric factor of $1/\sin(2\theta_B)$ (where θ_B is the Bragg angle) is still required to allow for the probability that a randomly oriented crystallite would be in the correct orientation for diffraction into $u = hkl$. Fuller details of these factors can be found in Warren (1969).

There are ten possible point groups that can describe the symmetry of the distribution of diffraction intensity in reciprocal space in the presence of streaking along one axis. These are, $\bar{1}$, $2/M$ (diad along streaks), $2/M$ (mirror contains streaks), MMM, $\bar{3}$, $\bar{3}M$, $4/M$, $4/MMM$, $6/M$, $6/MMM$. These are a sub-set of the standard 32 crystallographic point group symmetries. This sub-set arises because, first, we assume kinematical scattering, which implies the presence of an inversion centre at the origin (Friedel's law). Secondly, the presence of streaking along c^* prohibits the occurrence of the inclined 3-fold axes found in the cubic point groups. For powder diffraction calculations, significant computational speed-up can be achieved by establishing in advance which of the 10 point groups apply, and by restricting computational effort to an appropriate segment of reciprocal space.

2.3. *Running DIFFaX*

The user supplies a formatted data file that describes the layer structures, cell dimensions, the inter-layer stacking vectors and stacking probabilities, as well as the radiation type and wavelength. X-rays, electrons and neutrons are accepted as radiation types. Other parameters are entered interactively in response to a series of prompts.

The user can request one of five output modes. (1) The intensity at the reciprocal lattice vector hkl. (2) The integrated intensity along l within the interval hkl_1 to hkl_2. (3) A line trace of intensities along l within the interval hkl_1 to hkl_2, with step size Δl. (4) Powder pattern intensities within the range θ_1 and θ_2 with step size $\Delta\theta$. Gaussian, lorentzian or pseudo-Voigt instrumental broadening functions can be applied to the data. (5) Single crystal patterns viewed along directions perpendicular to the c^*, or streak axis.

Results from options 1 and 2 are supplied interactively to the computer screen, and are useful for debugging data files. Options 3 and 4 produce tab-delimited text files which can be read by several common graph plotting programs. The single crystal option, 5, produces a 256×256 pixel image with 256 grey levels. This is saved as a binary file which can be displayed and edited by many commonly available image processing programs.

The powder pattern option, 4, is the most computationally intensive. To ensure that any unbroadened peaks are not missed by the comparatively coarse sampling grid, a full-adaptive 16 point gaussian quadrature integration algorithm is employed. This results in a minimum of 48 intensity samplings per $\Delta\theta$ step. This sampling rate increases in regions where the intensity changes rapidly, such as near the unbroadened peaks. The time required to compute the intensity at a reciprocal lattice point depends predominantly on the sum of two terms. The first term is the time taken to calculate the layer scattering factors which varies as the product of the number of unique layers, and the average number of atoms per layer. The second term is the time required to solve the simultaneous equations, which varies approximately as the cube of the number of layers. To ensure optimum performance, DIFFaX can automatically establish a suitable point group symmetry for the data. On a Macintosh IIfx computer the zeolite powder pattern examples, presented later in this paper, typically required about 1 hour of computation time. The total time will vary roughly as the cube of the upper scattering angle θ_2. The user has the option of disabling the full-adaptive integration mode and to request DIFFaX to guess the location of the unbroadened peaks. This can gain an approximate factor of two in speed, but with some loss of precision and the risk that some peaks may be missed altogether.

For batch jobs where many data files need to be run unattended, DIFFaX can be run non-interactively using a control file which contains the data file names and the various parameters needed to run each file. (A copy of the DIFFaX FORTRAN source code, along with instructions and example data files, can be obtained by contacting M.M.J.T.)

3. Examples

We present here, briefly, some results where the recursion algorithm has been applied to simulate peak broadening in powder X-ray diffraction patterns from several different crystal systems. The first example is from the relatively simple close-packed diamond–lonsdaleite system. The subsequent three examples are from zeolite systems. The examples are chosen to demonstrate the flexibility and generality of the recursion algorithm and, in particular, to emphasize that it is not restricted to close-packed crystal systems.

Figure 2. Montage of powder X-ray diffraction patterns calculated as a function of the probability that layers in a diamond crystal will stack in the cubic diamond sequence. Thus $\alpha = 0$ corresponds to pure lonsdaleite, and $\alpha = 1$ corresponds to pure diamond. Note the peak broadening and the disappearance of certain peaks as the stacking probability is varied between 0 and 1. The defects simulated in this sequence are analogous to growth twins. Instrumental peak broadening was simulated using a pseudo-Voigt function with $u = 0.89$, $v = -0.32$, $w = 0.08$ and $\gamma = 0.6$. The step size was $\Delta\theta = 0.01°$.

3.1. *111 twin faults in the diamond–lonsdaleite system*

Diamond is cubic with space group symmetry $Fd\bar{3}m$. Twinning can occur on (111) planes. Full twinning recurrence generates the hexagonal analogue, lonsdaleite, that has space group symmetry $P6_3/mmc$, with the hexagonal [001] axis parallel to the cubic [111] direction.

The diamond structure can be described in terms of stacking of cubic (111) layers, where successive layers are related by inversion operations, i. The carbon atoms in these layers are arranged to form puckered hexagons. The structure of the mineral lonsdaleite can also be described in terms of these (111) layers, but successive layers are related by a mirror operation, m. The two types of stacking operation can coexist in some materials.

The two layer types can be designated by the symbols $\bullet$ $\bullet$ (for type 1), and $\bullet$ $\bullet$ (type 2) that emphasize their relation to one another through a vertical mirror plane. The interlayer stacking vectors, in terms of the hexagonal unit cell vectors a, b and c (where a and b are in the plane of the layer), are

$$R_{11} = \tfrac{1}{3}(2a + b + c),$$
$$R_{12} = R_{21} = \tfrac{1}{3}c,$$
$$R_{22} = \tfrac{1}{3}(-2a - b + c).$$

The c component is the same for all four vectors.

For a random fault distribution (where there are no layer–layer correlations) the stacking transition probabilities obey the relations

$$\alpha_{11} = \alpha_{22} = 1 - \alpha_{12} = 1 - \alpha_{21}.$$

Figure 2 shows a series of powder X-ray diffraction patterns calculated as a

function of the cubic stacking probability α_{11}. The end-member with $\alpha_{11} = 1$ corresponds to 100 % inversion operations, i.e. pure diamond, and the end-member with $\alpha_{11} = 0$ corresponds to 100 % mirror operations, i.e. pure lonsdaleite. Pronounced peak broadening occurs for interim values of α_{11}. However, peaks occurring at those diffraction vectors $\boldsymbol{u}$ such that all $\boldsymbol{u} \cdot \boldsymbol{R}_{ij} = 2n\pi$ (i.e. when $h - k = 3n$), have invariant widths.

Although this is a relatively simple example of the use of DIFFaX, calculations of this type may prove helpful in characterizing synthetic polycrystalline diamond films which can contain microcrystals showing significant twinning densities (Hetherington *et al.* 1990).

3.2. *Zeolite examples*

Zeolites are aluminosilicates in which all aluminium and silicon atoms are tetrahedrally coordinated by (bridging) oxygen atoms (Newsam 1991). Zeolite frameworks generally contain cages which may be interconnected along one or more directions to form channels. Some 64 observed zeolite framework topologies are listed in the current edition of the *Atlas of zeolite structure types* (Meier & Olson 1987), and the number continues to rise steadily. Over 80 are given in the current version of *ZeoFile* (Newsam & Treacy 1991). Zeolites are generally metastable and unless stopped in time, a hydrothermal synthesis will typically proceed to a more stable, denser phase, such as phillipsite, analcite or quartz. Owing to the conditions of kinetic control under which zeolites are synthesized, they frequently contain planar faults.

Framework topologies can be categorized into families that share common structural building units somewhat larger than individual tetrahedra. In several instances, distinct zeolite structure types differ only in the manner in which successive sheets are linked along a given direction, providing possibilities for intergrowths. In practice, planar faulting is relatively common, although it varies markedly in degree from one system to another. Frequently, the differences between materials can be detected as subtle variations in peak shapes and intensities in powder X-ray diffraction spectra. The simplicity of the recursive approach to simulating diffraction means that DIFFaX lends itself readily to analysing diffraction patterns from these complex structures.

We present here a summary of studies we have carried out on three zeolite systems; faujasite (FAU)/'Breck's Structure 6' (*bss*), zeolite beta (*bea-beb*) and ZSM-5 (MFI)/ZSM-11 (MEL). The FAU-*bss* system is related to the diamond case described above, except that defects are frequently observed to be clustered. The zeolite beta system exhibits an unusual type of twinning, where sheets can connect through either 4_1 or 4_3 twinning operations. The MFI/MEL system is an example of pure type disorder where the stacking vectors for all layer types are identical. Only the layer structures differ.

3.2.1. *Clustering of 111 twin faults in faujasite/'Breck's structure 6' materials*

The structure of faujasite is cubic, with the same space group as diamond, $Fd\bar{3}m$, but with an aluminosilicate sodalite cage (or β-cage) centred at each of the lattice points (carbon atom positions in diamond). Furthermore, there are shared oxygen atoms at the (111) layer connections.

As for diamond, (111) twin faults can occur and they are frequently observed to be clustered in the FAU/*bss* system. A Reichweite of 1 (where layers 'remember' how the preceding layer was stacked) can be handled by increasing the number of

Structure type A layer   Structure type A' layer

Type 1 layer   Type 2 layer   Type 3 layer   Type 4 layer

Cubic stacking   Hexagonal stacking

Typical faulted sequence

Figure 3. Diagram illustrating how sheets of sodalite cages can connect in a crystal to form localized regions of predominantly hexagonal (*bss*), or predominantly cubic (FAU) material. In this example, sheets retain a memory of how they are connected to the preceding layer (shown here as shaded layers), and this may influence the connectivity to the next layer. Four distinct layer types are required to model this situation where the Reichweite is 1. The view here is along [1$\bar{1}$0] in the cubic setting.

distinct layer types to 4. Layers 1 and 3 are both structurally of type ● ●, and layers 2 and 4 are structurally of type ● ●. Layer 1, however, is preceded by a layer of type ● ●, whereas layer 3 is preceded by a layer of type ● ●. Similarly, layer 2 is preceded by type ● ●, and type 4 is preceded by type ● ●. In the hexagonal setting, the stacking vectors are

$$R_{11} = R_{31} = \tfrac{1}{3}(2a+b+c),$$
$$R_{44} = R_{24} = \tfrac{1}{3}(-2a-b+c),$$
$$R_{43} = R_{12} = R_{23} = R_{32} = \tfrac{1}{3}c.$$

The stacking transitions α_{13}, α_{14}, α_{22}, α_{34}, α_{21}, α_{33}, α_{41} and α_{42} cannot occur, by definition. The probability of a fault in an inherently cubic stacking sequence, α_{11} ($\equiv \alpha_{44}$), and the probability of a fault in an inherently hexagonal stacking sequence, α_{23} ($\equiv \alpha_{32}$), need not be equal or correlated. When both are small, the material comprises large blocks of unfaulted cubic and hexagonal stackings. However, when both are close to unity, the faults tend to be uniformly distributed, with stacking types tending to repel one another. When the sum of the two probabilities is close to unity, the faulting resembles the random case presented above for diamond–lonsdaleite. Figure 3 illustrates how these four layer types describe clustered faulting in the FAU/*bss* system.

Figure 4 shows detailed comparisons between measurement and calculation for

Figure 4. Comparison of synchrotron powder X-ray diffraction data (X10B NSLS) (a, c, e) and calculation (b, d, f) for three different materials of the FAU–*bss* family. Instrumental peak broadening was simulated using a pseudo-Voigt function with $u = 0.1$, $v = -0.036$, $w = 0.009$ and $\gamma = 0.6$. The step size for the simulations was $\Delta\theta = 0.01°$.

three specific materials. Figure $4a, b$ are for ECR-35, where the fit is for $\alpha_{11} = 0.85$ and $\alpha_{23} = 0.6$. Figure $4c, d$ are for an ECR-30 material with $\alpha_{11} = 0.2$ and $\alpha_{23} = 0.8$ (implying the faulting is essentially random). Figure $10e, f$ are for a material synthesized using the 18 crown ether recipe of Delprato *et al.* (1990). The material comprises mostly hexagonal (mirror) stackings. The best fit occurs for $\alpha_{11} = 0.06$, $\alpha_{23} = 0.94$ (implying the faulting is essentially random) and for a thickness of the platelet crystallites along $[001]_{hex}$ of about 200 layers, which corresponds to about 0.35 μm.

3.2.2. *Zeolite beta*

Structurally, zeolite beta comprises one topologically distinct layer type, which occurs in four distinct orientations in the crystal, each related by a $\pm 90°$ rotation about a common c axis. The two tetragonal (chiral) end member structures have purely 4_1 ($+90°$ with a $\frac{1}{4}c$ shift) or 4_3 ($-90°$ with a $\frac{1}{4}c$ shift) stacking operations. When

Layer symbol ⌐ (or ⌐)

Layer symbol ⌐ (or ⌐)

Layer symbol ⌐ (or ⌐)

Layer symbol ⌐ (or ⌐)

Figure 5. [100] view of the framework of a typical zeolite beta crystallite. Viewed along this direction, only every other fault produces a detectable switch in the stacking direction. The unobserved faults are stacking either into, or out of, the page.

we take into account possible structural distortions, and their influence over how subsequent layers may stack, eight distinct layer types are required to describe a system with a Reichweite of 1, where the handedness of the previous transition influences the next transition. For convenience, we represent these eight layers here by the symbols ⌐⌐L⌐ and ⌐L⌐⌐. These symbols represent the four mutually rotated layers and their enantiomorphs. For end-member polymorph A (*bea*) with space group $P4_122$, the four layers stack such that a layer of type ⌐ is followed by a layer of type ⌐ (related to the first, through a $+90°$ right-handed rotation), followed by a layer of type L (again, related to the previous layer through a $+90°$ right-handed rotation) which in turn is followed by a type ⌐ layer. Thus, for a perfect right-handed crystal, the stacking repeats in a ⌐⌐L⌐⌐⌐L⌐ ... sequence. Likewise, the left-handed enantiomorph, space group $P4_322$, will stack in the sequence ⌐L⌐⌐⌐L⌐⌐ .... On the other hand, polymorph B (*beb*), space group $C2/c$, can stack as ⌐L⌐L⌐L ..., or ⌐⌐⌐⌐⌐⌐ ..., etc. For proper connectivity, layers cannot attach to other layers which are related by $0°$ or $180°$ rotations, i.e. layers of structure type ⌐ cannot attach to layers of type ⌐ or L , only to layers of type ⌐ or ⌐. Similar restraints apply to all other layers. A typical faulted sequence in zeolite beta is shown in projection in figure 5.

An alternative, more compact way of describing faults in zeolite beta would have been to assume that the stacking sequence attempts to revert to a preferred left- or right-handed sequence, such that errors in handedness tend to be corrected after the fault. Thus, in the limit of zero faulting, the crystal is pure right-handed, and in the limit of maximum faulting, the crystal comprises uninterrupted left-handed stacking.

Figure 6. Montage of calculated powder pattern plots for zeolite beta as a function of stacking probability. The probability here represents the chance that the stacking is part of a chiral sequence, either left- or right-handed. This description requires a Reichweite of 1, i.e. that each layer remembers how the previous layer stacked. Instrumental peak broadening was simulated using a pseudo-Voigt function with $u = 0.89$, $v = -0.32$, $w = 0.08$ and $\gamma = 0.6$. The step size was $\Delta\theta = 0.01°$.

This description requires only four distinct layers, which we label 1, 2, 3 and 4, and which are topologically equivalent to the layer types ⌐–L and ⌐ (which are assumed to be identical to layer types ⌐–Γ and ⌐). In this description, the crystal is racemic only when the fault probability is 0.5.

Figure 6 shows a montage of simulated powder X-ray patterns for this system ranging from fault densities of 0.0 (achiral *beb*) to 1.0 (chiral *bea*, although with equal mixtures of left- and right-handed connected crystals). The fault probabilities follow the relation

$$\alpha = \alpha_{12} = \alpha_{23} = \alpha_{34} = \alpha_{41} = \alpha_{58} = \alpha_{65} = \alpha_{76} = \alpha_{87},$$

$$1-\alpha = \alpha_{18} = \alpha_{25} = \alpha_{36} = \alpha_{47} = \alpha_{52} = \alpha_{63} = \alpha_{74} = \alpha_{81}.$$

The stacking vectors connecting the centroids of the basic repeat unit in each layer, (described relative to the tetragonal cell of the *bea* polymorph) are

$$R_{12} = R_{36} = R_{52} = R_{76} = -\tfrac{1}{3}b + \tfrac{1}{4}c,$$

$$R_{23} = R_{47} = R_{63} = R_{87} = \tfrac{1}{3}a + \tfrac{1}{4}c,$$

$$R_{34} = R_{58} = R_{74} = R_{18} = \tfrac{1}{3}b + \tfrac{1}{4}c,$$

$$R_{41} = R_{65} = R_{81} = R_{25} = -\tfrac{1}{3}a + \tfrac{1}{4}c.$$

Figure 7 shows a direct comparison between data and the best calculated fit. This occurs for probability $\alpha = 0.44$, and with unit cell dimension $a = b = 12.44$ Å, and $c = 26.41$ Å. This stacking probability is in good agreement with that obtained from electron micrographs.

3.2.3. *Intergrowths of zeolites ZSM-5 (MFI) and ZSM-11 (MEL)*

ZSM-5 and ZSM-11, the end-members of the pentasil family of zeolites, like Breck's structure 6 (*bss*) and Faujasite (FAU), share a common building unit. As for the FAU/*bss* system, in ZSM-5 (and FAU) consecutive layers are related by inversion

Figure 7. Comparison of calculated powder X-ray diffraction spectrum with data for a typical zeolite beta sample. The fit is best for $\alpha = 0.44$, implying that the left- and right-handed stacking operations are near-random, with a slight bias toward the c-glide stacking operation (i.e. a right–left–right...stacking sequence) rather than a 4_1 or 4_3 screw stacking operation (i.e. right–right–right...or left–left–left... stacking sequences).

Figure 8. Diagram illustrating the frameworks and connectivities of the four layer types which may occur in an intergrown MFI/MEL material. Note that all stacking vectors are equal in this instance.

operations, whereas in ZSM-11 (and *bss*), consecutive layers are related through mirror operations. Figure 8 shows in projection, the frameworks of the four layer types necessary to fully describe the MFI/MEL system. All four layers are topologically identical, but are oriented differently. Layers 1 and 2 are related by a vertical mirror, whereas layers 1 and 3 are related by an inversion. Layers 2 and 4 are also related by an inversion.

Unlike the FAU/*bss* and the *bea*/*beb* examples, the stacking vectors between the four layer types are identical in the ZSM-5/-11 system. Streaking in the diffraction patterns arises purely from type disorder, with no contribution from position disorder. The stacking vectors for both ZSM-5 and ZSM-11 are $(0, 0, \frac{1}{2}c)$, compared with $(\pm\frac{2}{3}a, \pm\frac{1}{3}b, \frac{1}{3}c)$ for the FAU system. The FAU stacking vectors have non-zero lateral components, and therefore the stacking of mirror-related layers will require a change in sign of these components. The ZSM-5/-11 stacking vectors have zero lateral components, thus the stacking vectors for mirror-related layers are identical.

A consequence of there being type disorder only is that, relative to the FAU/*bss*, and *bea*/*beb* systems, certain peaks lose intensity rapidly as a function of stacking

Figure 9. Montage of calculated powder pattern plots for intergrowths of ZSM-5 and ZSM-11. Low values of α correspond to pure ZSM-5 (MEL) and high α values correspond to pure ZSM-11 (MEL). Because all stacking vectors are equal, there is no position disorder, only type disorder due to subtle structural differences between layers. Note that there is little of the dramatic peak broadening that occurs in the other systems where position disorder is present. However, peak intensities can change rapidly, and are not a linear function of the fault probability. Instrumental peak broadening was simulated using a pseudo-Voigt function with $u = 0.1$, $v = -0.036$, $w = 0.009$ and $\gamma = 0.6$. The step size was $\Delta\theta = 0.01°$.

probability, and peak broadening tends to be subtle, as can be seen for the series of powder X-ray diffraction patterns simulated over the range of fault probability shown in figure 9. Of particular importance is the observation that peak intensities do not vary linearly with the degree of disorder. All of the most marked changes to peak intensities occur near $\alpha_{13} \approx 0$ and $\alpha_{12} \approx 0$ for the pure MEL and MFI end members.

4. Conclusions

The recursion model of diffraction provides a powerful, intuitive way of envisioning the diffraction process from crystals, particularly from those containing defects. The simplicity of the recursion algorithm has enabled the development of a general-purpose computer program, DIFFaX, which can be used to calculate intensities from any type of crystal system that contains any type of coherent planar defect. DIFFaX has been used successfully on a wide variety of structure problems, particularly zeolite systems. Powder X-ray diffraction patterns, simulated as a function of stacking transition probabilities, provide a database against which measured patterns can be compared, a facility that is already proving useful in directing zeolite synthesis efforts.

Appendix A 1. Relationship between the recursion equations and the Hendricks–Teller Equation

The relation to the Hendricks–Teller equation can be established by expanding equation (12) in m and rearranging

$$I(\boldsymbol{u}) = \sum_{m=0}^{N-1} ((N-m)\,\boldsymbol{G}^{\mathrm{T}*}\,\boldsymbol{T}^n\boldsymbol{F} + \text{c.c.}) - N\boldsymbol{G}^{\mathrm{T}*}\boldsymbol{F}$$

$$= \sum_{m=1}^{N-1} ((N-m)\,\boldsymbol{G}^{\mathrm{T}*}\,\boldsymbol{T}^n\boldsymbol{F} + \text{c.c.}) + N\boldsymbol{G}^{\mathrm{T}*}\boldsymbol{F}, \tag{A 1}$$

where c.c. stands for complex conjugate.

Redefining our matrices as

$$\langle H_{ij} \rangle = g_i\,\delta_{ij} \quad \text{and} \quad \langle V_{ji} \rangle = F_i F_j^* \exp\left(-2\pi\mathrm{i}(h(R_{xi}-R_{xj}) + k(R_{yi}-R_{yj})\right),$$

equation (A 1) can be rewritten as

$$I(\boldsymbol{u}) = \sum_{m=1}^{N-1} ((N-m)\,\mathrm{tr}\,\boldsymbol{V}\boldsymbol{H}\boldsymbol{T}^n + \text{c.c.}) + N\,\mathrm{tr}\,\boldsymbol{V}\boldsymbol{H}, \tag{A 2}$$

where tr is the sum of the diagonal terms of a square matrix, sometimes referred to as the spur. Equation (A 2) is the form of the Hendricks–Teller equation discussed by Kakinoki & Tomura (1965) and Allegra (1961).

Appendix A 2. Relation between the recursion model and the Cowley equation

The relation with Cowley's solution (1976a) can be established by combining equation (8) and equation (9) to form the nested series

$$\frac{I(\boldsymbol{u})}{N} = \{\sum_i g_i F_i^*(\boldsymbol{u})\,[F_i(\boldsymbol{u}) + \sum_j \alpha_{ij}\exp\left(-2\pi\mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{ij}\right)[F_j(\boldsymbol{u})$$

$$+ \sum_k \alpha_{jk}\exp\left(-2\pi\mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{jk}\right)[F_k(\boldsymbol{u}) + \sum_l \alpha_{kl}\exp\left(-2\pi\mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{kl}\right)[F_l(\boldsymbol{u}) + \ldots]]]]$$

$$+ \text{c.c.}\} - \sum_i g_i |F_i(\boldsymbol{u})|^2. \tag{A 3}$$

If we isolate terms involving no faults, the R_{ii} transitions, from terms involving faults, all other R_{ij} transitions where $i \neq j$, we find that for a large number of layers, N, the no fault terms can be collected and summed, as in

$$1 + \alpha_{ii}\exp\left(-2\pi\mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{ii}\right) + \alpha_{ii}^2\exp\left(-4\pi\mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{ii}\right) + \ldots \alpha_{ii}^n\exp\left(-2n\pi\mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{ii}\right) + \ldots$$
$$= 1/(1 - \alpha_{ii}\exp\left(-2\pi\mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{ii}\right))$$

and equation (A 3) becomes

$$\frac{I(\boldsymbol{u})}{N} = \left\{\sum_i \frac{g_i F_i^*(\boldsymbol{u})}{1 - \alpha_{ii}\exp\left(-2\pi\mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{ii}\right)}\left[F_i(\boldsymbol{u}) + \sum_{i \neq j} \frac{\alpha_{ij}\exp\left(-2\pi\mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{ij}\right)}{1 - \alpha_{jj}\exp\left(-2\pi\mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{jj}\right)}\left[F_j(\boldsymbol{u})\right.\right.\right.$$

$$\left.\left.\left. + \sum_{j \neq k} \frac{\alpha_{jk}\exp\left(-2\pi\mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{jk}\right)}{1 - \alpha_{kk}\exp\left(-2\pi\mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{kk}\right)}\left[F_k(\boldsymbol{u}) + \sum_{k \neq l} \frac{\alpha_{kl}\exp\left(-2\pi\mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{kl}\right)}{1 - \alpha_{ll}\exp\left(-2\pi\mathrm{i}\boldsymbol{u}\cdot\boldsymbol{R}_{kk}\right)}\left[F_k(\boldsymbol{u}) + \ldots\right]\right]\right]\right]$$

$$+ \text{c.c.}\right\} - \sum_i g_i |F_i(\boldsymbol{u})|^2, \tag{A 4}$$

which is Cowley's equation (1976a), with the substitution $\alpha_{ii} = (1 - \Sigma_j \alpha_{ij})$. Although this form is also recursive as the nested terms are similar, but begin on different layers, it is less convenient to implement numerically than are equations (14) and (17).

Appendix A 3. Relation to Michalski's recurrence treatment

Michalski (1988) has independently pointed out that the stacking of layers in a statistical crystal contains recurring patterns. Starting from the perspective of the difference equation method developed by Holloway (1969), Michalski noted that there exists a recurrence relation between the average phase factors from layers separated by n spacings, and the average phase factors from layers separated by $n-1$ spacings (Michalski 1988, equation (18)). In terms of the nomenclature used here, where we include the scattering amplitude from each layer, this is equivalent to noting that the interference terms between layers separated by n layers, φ^n, and the interference terms between layers separated by $n-1$ layers, φ^{n-1}, are related, i.e.

$$\varphi^n = T^n F = T(T^{n-1}F) = T\varphi^{n-1}. \tag{A 5}$$

In this paper we develop this notion further by demonstrating that the average interference terms from the whole crystal can also be expressed in recursive form. The average interference terms from an N-layer crystal $\Psi^{(N)}$ can be expressed in terms of the φ^n-using equation (13)

$$\Psi^{(N)} = \frac{1}{N} \sum_{m=0}^{N-1} \sum_{n=0}^{N-m-1} T^n F = \frac{1}{N} \sum_{m=0}^{N-1} \sum_{n=0}^{N-m-1} \varphi^n. \tag{A 6}$$

In Michalski's treatment, N is assumed to be large, thus the recurrent terms φ^n build up the ensemble-average scattered wavefunction Φ

$$\Psi^{(N \to \infty)} = \Phi = \sum_{n=0}^{\infty} \varphi^n. \tag{A 7}$$

As shown in equations (15) and (18), the $\Psi^{(N)}$ (and the Φ) can themselves be expressed in recursive form, which greatly simplifies the theory, as these quantities can be used directly in equation (17) to provide the diffracted intensity.

References

Allegra, G. 1961 A simplified formula for the calculation of the X-ray intensity diffracted by a monodimensionally disordered structure. *Acta crystallogr.* **14**, 535.

Allegra, G. 1964 The calculation of the intensity of X-rays diffracted by monodimensionally disordered structures. *Acta crystallogr.* **17**, 579–586.

Berliner, R. & Werner, S. A. 1986 Effect of stacking faults on diffraction. The structure of lithium metal. *Phys. Rev.* B **34**, 3586–3603.

Brindley, G. W. & Brown, G. 1980 *Crystal structures of clay minerals and their X-ray-identification.* London: Mineralogical Society.

Cajipe, V. B., Heiney, P. A. & Fischer, J. E. 1989 *In situ* study of staging disorder in cesium-intercalated graphite. *Phys. Rev.* B **39**, 4374–4385.

Cowley, J. M. 1976a Diffraction by crystals with planar faults. I. General theory. *Acta crystallogr.* A **34**, 83–87.

Cowley, J. M. 1976b Diffraction by crystals with planar faults. II. Magnesium fluorogermanate. *Acta crystallogr.* A **32**, 88–91.

Cowley, J. M. 1981 *Diffraction physics*, pp. 388–400. New York and London: North-Holland.

Cowley, J. M. & Au, A. Y. 1978 Diffraction by crystals with planar faults. III. Structure analysis using microtwins. *Acta crystallogr.* A **34**, 738–743.

Delprato, E., Delmotte, L., Guth, J. L. & Huve, L. 1990 Synthesis of new silica-rich cubic and hexagonal faujasites using crown-ether-based supramolecules as templates. *Zeolites* **10**, 546–552.

Gevers, R. 1952 Desordre unidimensionnel dans SiC et influence sur les intensities diffractées des Rayons X. *Acta crystallogr.* **5**, 518–524.

Gevers, R. 1954*a* X-ray diffraction by close-packed crystals with 'growth stacking faults' assuming an '*n*-layer influence'. *Acta crystallogr.* **7**, 493–494.

Gevers, R. 1954*b* The diffraction of X-rays by close-packed crystals containing 'growth stacking faults' and 'deformation or transformation stacking faults'. *Acta crystallogr.* **7**, 337–343.

Hendricks, S. & Teller, E. 1942 X-ray interference in partially ordered layer lattices. *J. chem. Phys.* **10**, 147–167.

Hetherington, A. V., Wort, C. J. H. & Southworth, P. 1990 Crystalline perfection of chemical vapor deposited diamonds films. *J. Mater. Res.* **5**, 1591–1594.

Holloway, H. 1969 Diffraction by faulted close-packed lattices: an analytic solution for systems without long-range correlation of stacking symbols. *J. appl. Phys.* **40**, 4313–4321.

Jagodzinski, H. 1949*a* Eindimensionale Fehlordnung in Kristallen und ihr einfluss auf die Röntgeninterferenzen. I. Berechnung des Fehlordnungsgrades aus den Röntgenintensitäten. *Acta crystallogr.* **2**, 201–207.

Jagodzinski, H. 1949*b* Eindimensionale Fehlordnung in Kristallen und ihr Einfluss auf die Röntgeninterferenzen. II. Berechnung der Fehlgeordneten dichtesten Kugelpackungen mit Wechselwirkungen der Reichweite 3. *Acta crystallogr.* **2**, 208–214.

Jagodzinski, H. 1949*c* Eindimensionale Fehlordnung in Kristallen und ihr Einfluss auf die Röntgeninterferenzen. III. Vergleich der berechnung mit experimentellen Ergebnissen. *Acta crystallogr.* **2**, 298–304.

Jagodzinski, H. 1954 Der Symmetrieeinfluss auf den allgemeinen Lösungsansatz eindimensionaler Fehlordnungsprobleme. *Acta crystallogr.* **7**, 17–25.

Kakinoki, J. 1967 Diffraction by a one-dimensionally disordered crystal. II. Close-packed structures. *Acta crystallogr.* **23**, 875–885.

Kakinoki, J. & Tomura, Y. 1951 Intensity of X-ray diffraction by a one-dimensionally disordered crystal. (I) General derivation in the cases of the 'Reichweite' $s = 0$ and 1. *J. Inst. Polytech. Osaka City Univ.* B **2**, 1–9.

Kakinoki, J. & Tomura, Y. 1952 Intensity of X-ray diffraction by a one-dimensionally disordered crystal. (II) General derivation in the case of the correlation range $s \geqslant 2$. *J. Inst. Polytech. Osaka City Univ.* B **3**, 1–33.

Kakinoki, J. & Tomura, Y. 1965 Diffraction by a one-dimensionally disordered crystal. I. The intensity equation. *Acta crystallogr.* **19**, 137–147.

Landau, L. 1937 The scattering of X-rays by crystals with variable lamellar structure. *Phys. Z. SowjUn.* **12**, 579–585.

Lifshitz, I. M. 1937 On the theory of scattering of X-rays by crystals of variable structure. *Phys. Z. SowjUn.* **12**, 623–643.

Lifschitz, I. M. 1939 Correlation and scattering of X-rays in solid solutions. *J. exper. theor. Phys. U.S.S.R.*, **9**, 491–511. (In Russian).

Meier, W. M. & Olson, D. H. 1987 *Atlas of zeolite structure types*. London: Butterworths.

Michalski, E. 1988 The diffraction of X-rays by close-packed polytypic crystals containing single stacking faults. I. General theory. *Acta crystallogr.* A **44**, 640–649.

Michalski, E., Kaczmarek, S. & Demianiuk, M. 1988 The diffraction of X-rays by close-packed polytypic crystals containing single stacking faults. I. Theory for hexagonal and rhombohedral structures. *Acta crystallogr* A **44**, 650–657.

Newsam, J. M. 1991 Zeolites – a graduate-level overview. *Solid state chemistry* (ed. P. Cheetham & A. K. Day), vol. 2. Oxford University Press. (In the press.)

Newsam, J. M. & Treacy, M. M. J. 1991 ZeoFile – a hypercard stack of zeolite structure types. *Zeolites.* (Submitted.)

Newsam, J. M., Treacy, M. M. J., Koetsier, W. T. & de Gruyter, C. B. 1988 Structural characterization of zeolite beta. *Proc. R. Soc. Lond.* A **420**, 375–405.

Reynolds, R. C. 1980 Interstratified clay minerals. In *Crystal structures of clay minerals and their X-ray identification* (ed. G. W. Brindley & G. Brown). London: Mineralogical Society. (Also, the NEWMOD program is available from R. C. Reynolds, Department of Earth Sciences, Dartmouth College, Hanover, New Hampshire 03755, U.S.A.)

Vaughan, D. E. W., Treacy, M. M. J., Newsam, J. M., Strohmaier, K. G. & Mortier, W. J. 1989 Synthesis and characterization of zeolite ZSM-20. In *Zeolite synthesis* (ed. M. L. Occelli & H. E. Robson), ACS Symp. Ser. no. 398, pp. 544–559. Washington: American Chemical Society.

Warren, B. E. 1959 X-ray studies of deformed metals. *Prog. Met. Phys.* **8**, 147–202.

Warren, B. E. 1969 In *X-ray diffraction*, pp. 275–312. Reading, Massachusetts: Addison-Wesley.

Wellbery, T. E. 1985 Diffuse X-ray scattering and models of disorder. *Rep. Prog. Phys.* **48**, 1543–1593.

Wilson, A. J. C. 1942 Imperfections in the structure of cobalt II. Mathematical treatment of proposed structure. *Proc. R. Soc. Lond.* A **180**, 277–285.

Wilson, A. J. C. 1943 The reflexion of X-rays from the 'anti-phase nuclei' of $AuCu_3$. *Proc. R. Soc. Lond.* A **181**, 360–368.

Index

adaptive integration, 38
`AXIAL`, 12

beta, 62–74
binary files, 43
brick wall, 45

cell dimensions, 11
chiral stacking, 62
clustering
    of faults, 55
compiling DIFFaX, 43
control file, 25, 40–42, 77–82
Cray, 44

`data.sfc`, 15, 25, 27, 36
Debye-Scherrer
    peak broadening, 13, 46–48
Debye-Waller
    temperature factors, 16, 20, 52
`DETUN`, 50
detune, 37, 49–52
`dia.dat`, 82
`dia.dmp`, 28, 82–84
`dia.sadp`, 34
`dia.spc`, 86–87
`dia.str`, 30
`dia.sym`, 28, 84–86
diamond, 54–58
`DIFFaX.f`, 43, 49, 50
`DIFFaX.inc`, 43
`DIFFaX.par`, 43, 49
dump file, 27, 82

`EXPLICIT` stacking, 18

"Fats-Waller"
    temperature factors, 20, 52
faujasite, 58–62
files
    binary, 43
    `control.dif`, 25, 40–42, 77–82
    `data.sfc`, 15, 25, 27, 36
    `DIFFaX.f`, 43, 49, 50
    `DIFFaX.inc`, 43
    `DIFFaX.par`, 43, 49
    dump, 18, 27, 42, 82–84
    naming conventions, 42
    selected area diffraction, 31, 42
    spectrum, 42
    streak, 29, 42
    structure data, 2–25, 54–77

    symmetry, 28, 42, 84–86
Friedel's law, 11, 12
full-width half-maximum, 7
FWHM, *see* full-width half-maximum

GaAlAs, 74–77
GaAs, 74–77
GaAs/GaAlAs, 74–77
Gauss-Legendre integration, 38
`GAUSSIAN`, instrumental broadening, 7, 40

headers
    `INSTRUMENTAL`, 6
    `STACKING`, 18
    `STRUCTURAL`, 11
    `TRANSITIONS`, 19

Image program, 34
`INFINITE`
    layer width, 13, 48
    number of layers, 19
`INSTRUMENTAL`, 6
instrumental broadening, 7
    `GAUSSIAN`, 7, 40
    `LORENTZIAN`, 8, 40
    `NONE`, 7, 9
    `PSEUDO-VOIGT`, 9, 40
    parameters
        u,v,w, 7–9
        σ, 9
integrated intensity, 30
intensity
    along a streak, 29
    at a point, 28
    integrated, 30
    powder pattern, 34
    selected area diffraction, 31

Laue symmetry, 11
layer
    atom data, 14
    finite width, 13, 46
    infinite width, 13, 48
    symmetry
        `CENTROSYMMETRIC`, 14, 40
        `NONE`, 14
    types, number of, 13
    widths, 13, 48
lonsdaleite, 33, 54, 55
`LORENTZIAN`, instrumental broadening, 8, 40

MacDIFFaX, 25, 27, 36–37