

# Simulation of X-ray Laue patterns

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An available Fortran IV computer program for the simulation of forward and back-scattered Laue patterns is described.

## 1. Introduction

High X-ray intensity from a synchrotron makes practical the analysis of certain crystal defects by X-ray topography. Furthermore, the wavelength spread of the source results in a three-dimensional sectioning of a crystal's reciprocal lattice, revealing symmetry properties associated with the crystal. This subject has been thoroughly covered in a recent textbook by Amorós *et al.* [1].

To analyse defect structures requires the Miller indices of an operating reflection to be known. To this end, several investigators have proposed different techniques. For instance, Canut-Amorós [2] produces stereographic projections from each Transmission Laue Pattern (TLP) from which the crystal orientation can be deduced while Ohba *et al.* [3] report computer-aided spot indexing for X-ray Laue patterns.

At the invitation and with the encouragement of K. Roberts of the University of Strathclyde, a computer program has been written to simulate TLP for any crystal of any orientation. This program allows direct one-to-one comparison of experimental and simulated patterns. Other investigators have allowed for image distortions when the film is not perpendicular to the X-ray beam [4]; however, the program described here has neglected such corrections.

## 2. Theory

### 2.1. General background

The computer program FSLP, written to simulate TLP, was combined with an earlier program STEREO [5] which allows:

- (a) any crystal system;
- (b) random input of orienting data; in the form of crystal directions and/or poles within any part of the Wulff net;

(c) crystal rotations of any magnitude.

The basic geometrical relationships are shown in Fig. 1. The lower right-hand corner is a schematic Wulff net indicating the relationship between the Greninger Chart parameters  $\gamma$  and  $\delta$  associated with back-scattered Laue patterns (BSLP). Any, or all, of the crystal direction vectors **DA**, **DB** and **DC** can be replaced by pole vectors **PA**, **PB** or **PC**; for a detailed description of input variables, see Ploc and Barnett [5]. FSLP will also simulate BSLP but in this instance the X-ray beam impinges upon the crystal  $180^\circ$  from the direction shown in Fig. 1. The position of the TLP (or BSLP) is also shown in the figure. Stereographic projections of the precise orientation used to produce the TLP (or BSLP) are obtainable from STEREO [5].

### 2.2. Transmission Laue patterns

A Cartesian coordinate system was constructed as in Fig. 1, i.e.,  $x$ ,  $y$  and  $z$  parallel to **DB**, **-DC** and **-DA**, respectively. From an Ewald sphere construction, it can be seen that the Bragg Law is fulfilled when the X-ray wavelength satisfies the condition:

$$\lambda = 2Zd^2 \quad (1)$$

where  $d$  is the interplanar  $d$ -spacing for the operative reflection and  $\mathbf{Z}$  is the vector dot product of the reciprocal lattice vector with **-DA**. Similarly, the  $(x_r, y_r)$  coordinates of the reflection are given by:

$$x_r = L\{g_{hkl} \cdot \mathbf{DB}\}/Z_c \quad (2)$$

$$y_r = -L\{g_{hkl} \cdot \mathbf{DC}\}/Z_c \quad (3)$$

where  $Z_c = (\mathbf{Z}\lambda - 1)/\lambda$ , and  $L$  is the specimen to film distance. The  $(x_g, y_g)$  coordinates of the point of intersection of the extended negative reciprocal lattice vector with the film are given

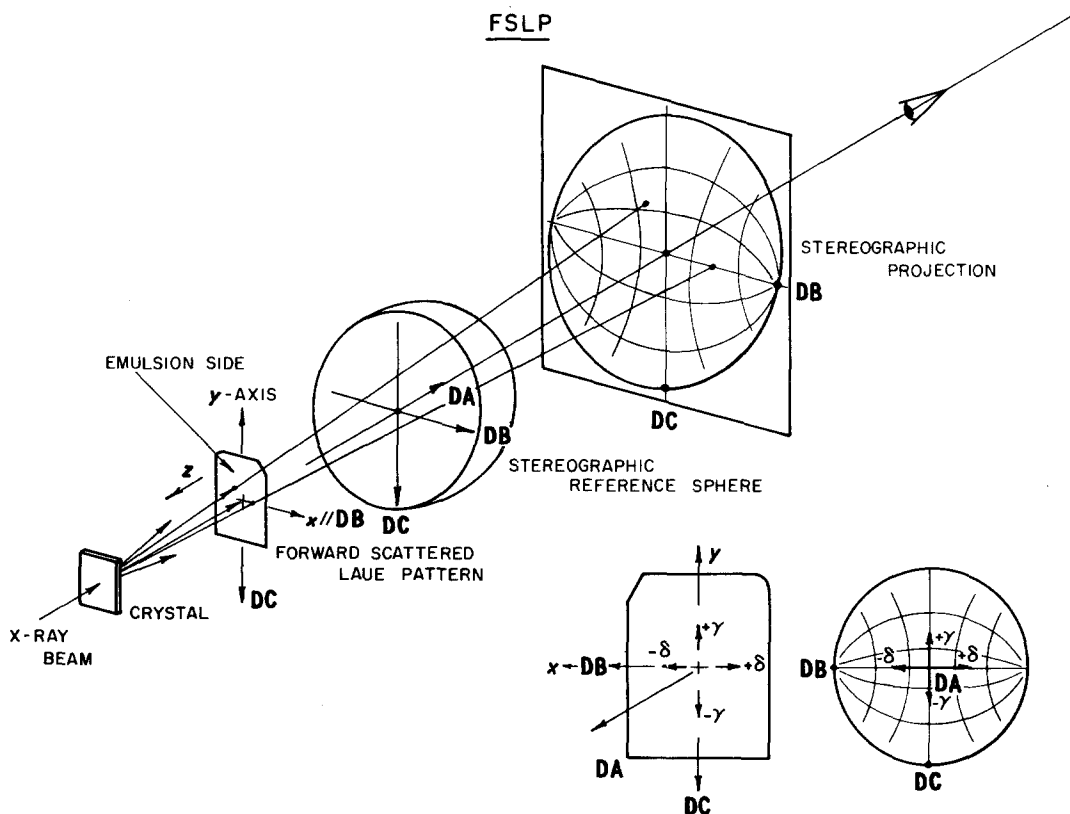


Figure 1 Relative geometry used in the program FSLP.

by Equations 2 and 3 except the parameter  $Z_c = -Z$ . FSLP calculates DA, etc. for any input data which orients the stereogram.

A simple vector diagram involving the reciprocal lattice (RL) vector and the transmitted and diffracted beams is sufficient to deduce  $(x_g, y_g)$  in terms of the film coordinates  $(x_r, y_r)$  of a reflection.

$$x_g = -Lx_r / [(x_r^2 + y_r^2 + L^2)^{1/2} - L] \quad (4)$$

$$y_g = yx_g / x_r. \quad (5)$$

In terms of traditional Grentinger chart parameters

$$\tan \gamma = y_g / L \quad (6)$$

$$\tan \delta = x_g / (L^2 + y_g^2)^{1/2}. \quad (7)$$

The delineation of the RL vector positions (negatives) on the film is useful when attempting to index the pattern analytically [6] or to locate a particular crystal pole or direction.

### 2.3. Back-scattered Laue patterns

Using the same geometry as shown in Fig. 1, the  $(x_b, y_b)$  coordinates of the back-reflected X-ray

beam are given by:

$$x_b = 2L(g \cdot DB) / [(1 - \tan^2 \sigma)(g \cdot DA)] \quad (8)$$

and

$$y_b = x_b(g \cdot DC) / (g \cdot DB), \quad (9)$$

where

$$\sigma = \cos^{-1} [(g \cdot DA)d].$$

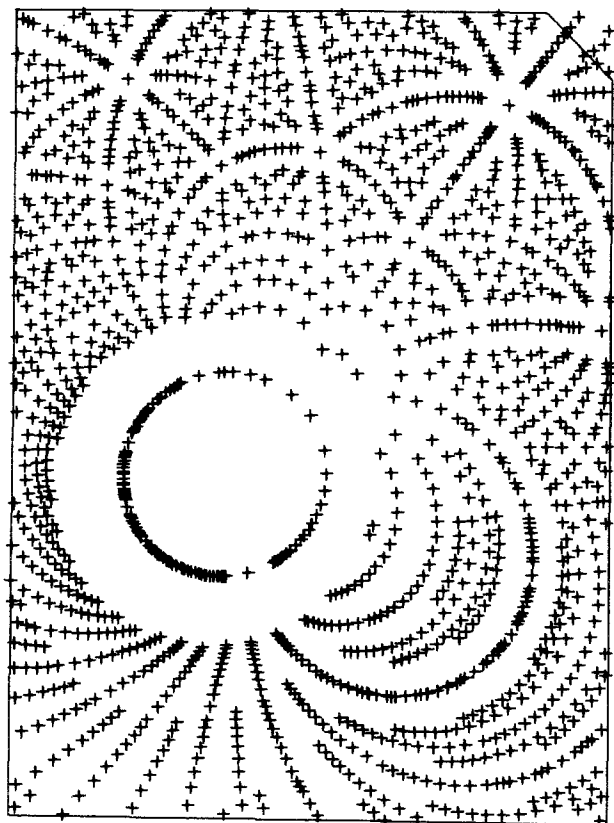
Knowing the position of the reflection on the film and the fact that, for exact diffraction, the angle between the diffracted and incident beams is twice the Bragg angle, the wavelength of the operating reflection,  $\lambda_b$ , can be calculated from:

$$\lambda_b = 2d \cos [\frac{1}{2} \tan^{-1}(XY/L)] \quad (10)$$

where

$$(XY)^2 = x_b^2 + y_b^2.$$

A cosine rather than sine term in Equation 10 results from the Bragg angle being greater than  $90^\circ$ . Grentinger charts are commonly available to determine the angular positions of the RL vectors.



SODIUM HYDROGEN MALEATE  
HYDRATE

B

C

01 = 1. 0. 0.  
02 = 2. 0. -1.  
PH11ZQ = 8.07  
PH12ZQ = 16.28  
PH11G = 101.57  
PH12G = 80.13

hkl RANGE = 24  
UPPER WAVELENGTH  
(nm) = 1.000  
LOWER WAVELENGTH  
(nm) = 0.027  
CAMERA LENGTH (cm) = 5.80

Figure 2 Simulated transmission Laue diffraction pattern for the conditions indicated. The original distance from the centre of the pattern to the corner was 7.5 cm. The directions B and C refer to DB and DC in Fig. 1.

### 2.4. Program parameters

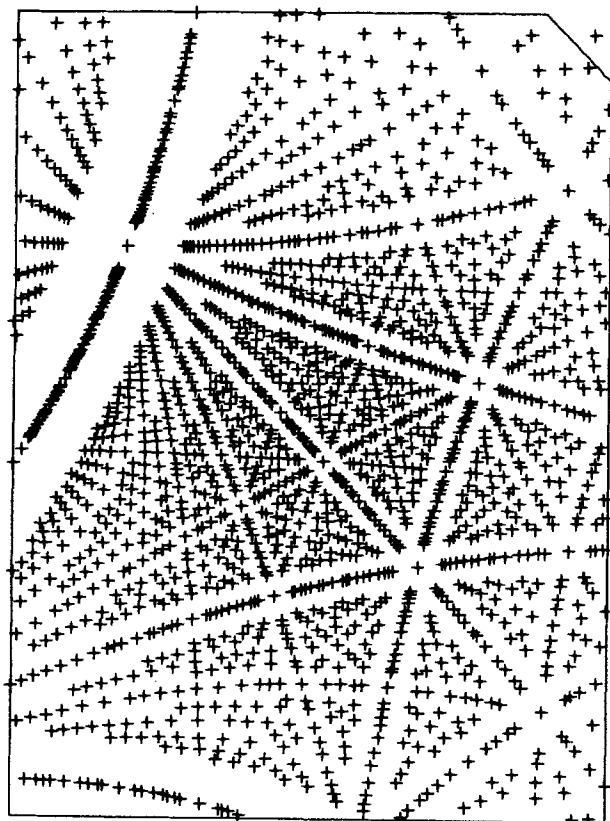
The computer program FSLP produces either a simulated forward- or back-scattered X-ray Laue pattern for any crystal system, see Figs. 2 and 3, respectively. Furthermore, a stereographic projection with the same orientation for crystal poles and/or directions can be produced by STEREO [5]. INPUT variables are listed below with their default values in parentheses.

- (1)  $h, k, l$  range (15).
- (2) L, specimen to film distance (5 cm).
- (3) RMAX, maximum dimension from centre to furthest corner of film (7.5 cm).
- (4) LLAM, lower wavelength cut-off (0.05 nm).
- (5) ULAM, upper wavelength cut-off (1.00 nm).
- (6) KVOLTS, maximum X-ray tube kilovoltage, applies to back-scattering only (20.0 kV).
- (7) UCUT, upper wavelength limit for back-scattering (0.2 nm).
- (8) NCLASS, number of classes of permissible reflections, (0).

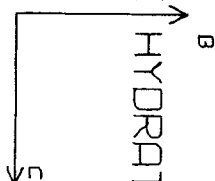
For a given space group, permissible reflections must be stated as a class (i.e.,  $hkl, hhl, hkh$ , etc.), as well as the conditions which apply (i.e.,  $h + k = nx + m$ , etc., where  $n = m = \text{integer}$ ).

### 3. Results and discussion

Fig. 4 is the printed OUTPUT corresponding to Fig. 2. Note that for the data supplied to orient the crystal relative to the X-ray beam, pole and crystal direction vector indices at A, B and C are printed (in the form of normalized real numbers). In this instance, there were no space group restrictions so every reflection within the  $h, k, l$  range from  $(\bar{24}, \bar{24}, \bar{24})$  to  $(24, 24, 24)$  was processed. The parameters  $X(D)$ ,  $Y(D)$  and  $XT(D)$ ,  $YT(D)$  are the  $(x, y)$  coordinates of each reflection relative to the pattern centre and bottom left-hand corner, respectively.  $X(-G)$  and  $Y(-G)$  are the  $(x, y)$  coordinates of where the negative pole for the given reflection intersects the film



SODIUM HYDROGEN MALEATE HYDRATE  
 O1 = 1. 0. 0.  
 O2 = 2. 0. -1.  
 PH11ZQ = 8.07      PH11G = 101.57  
 PH1ZZQ = 16.28      PH1ZG = 80.13



*hkl* RANGE = 20  
 CAMERA LENGTH (cm) = 5.00  
 LOWER WAVELENGTH CUT-OFF (nm) = 0.06199  
 UPPER WAVELENGTH CUT-OFF (nm) = 0.2000

## BACK-REFLECTION LAUE PATTERN

(relative to pattern centre). LAMBDA is the wavelength for Bragg diffraction.

Fig. 5 is a TLP of a sodium hydrogen maleate hydrate ( $\text{NaHC}_4\text{H}_2\text{O}_4 \cdot 3\text{H}_2\text{O}$ ) crystal that was supplied for indexing\*. Simulated TLP were made for the 100, 010 and 001 crystal poles and directions parallel to the X-ray beam. From these results, it was obvious that the crystal normal was near the [100] (about  $14^\circ$  away). A more exact determination of the orientation can be achieved in two ways:

(1) identification of the reflections on the experimental pattern from the simulation. Measurement of the experimental positions on the film and back calculating the position of the negative poles from Equations 4 and 5 which can then be used to set the new orientation;

(2) zones on the film can be identified from the simulation and its printout. The positions of

several reflections belonging to a zone can be used to calculate the zone axis position remembering that the ZA is perpendicular to every reflection. This procedure has been automated in our laboratory by digitizing the TLP and computer processing.

A point on the Wulff net can be characterized by two parameters,  $\phi_{za}$  and  $\phi_b$  where the former corresponds to a longitude, positive towards DC, and the latter a latitude, zero at DB and increasing to  $180^\circ$  at  $-DB$  (see Fig. 1). The angle,  $\theta$ , between any two points is given by:

$$\begin{aligned}
 \cos\theta &= \cos\phi_{1b}\cos\phi_{2b} \\
 &+ (\sin\phi_{1b}\sin\phi_{2b})(\cos\phi_{1za}\cos\phi_{2za} \\
 &+ \sin\phi_{1za}\sin\phi_{2za}). \quad (11)
 \end{aligned}$$

For zone axis calculations  $\theta = 90^\circ$  and therefore the stereographic position of the zone axis can be

\*Crystal data supplied by Jennissen and Klapper, Institut für Kristallographie, Technische Hochschule, Aachen, Germany (1981).

## Sodium hydrogen maleate hydrate

$a = 6.3840$   $b = 11.2210$   $c = 5.9450$   $\alpha = 91.517$   $\beta = 100.217$   $\gamma = 104.233$

D1 1. 0. 0.

D2 2. 0. -1.

PHI 1ZA = 8.070 PHI 1B = 101.570

PHI 2ZA = 16.280 PHI 2B = 80.390

PHI 2B Recalculated to be 80.1288

## Calculated cardinal positions

## Directions

	<i>h</i>	<i>k</i>	<i>l</i>
DA =	0.1 396 484 284	-0.0 186 770 143	-0.0 255 438 377
DB =	-0.0 720 336 459	-0.0 325 977 366	-0.1 606 111 691
DC =	0.0 489 938 921	0.0 841 875 876	-0.0 540 680 318

## Poles

	<i>h</i>	<i>k</i>	<i>l</i>
PA =	6.1 923 454 006	-4.7 660 934 648	-1.8 099 199 342
PB =	-1.2 804 068 911	-2.5 520 692 340	-5.1 339 887 960
PC =	0.8 779 935 264	9.8 327 074 964	-2.3 894 296 127

$hkl = 24$

$R_{\max} = 7.5$  cm.

Specimen to film distance = 5.80 cm.

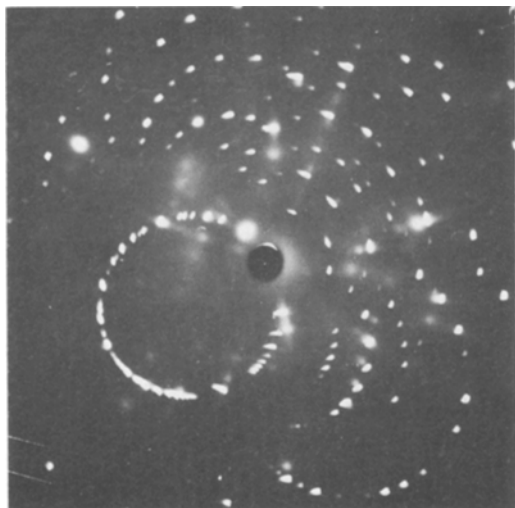
Maximum angle from zero order beam = 52.3 degrees

Lower wavelength limit = 0.02 700 nm

Upper wavelength limit = 0.10 000 nm

<i>N</i>	<i>h</i>	<i>k</i>	<i>l</i>	$\lambda$	X(D)	Y(D)	X(-G)	Y(-G)	XT(D)	YT(D)	<i>h</i>	<i>k</i>	<i>l</i>
1	-12	-20	-1	0.027 280	4.07	5.38	-7.62	-10.07	8.57	11.38	-12	-20	-1
2	-12	-20	0	0.028 447	3.97	5.95	-6.75	-10.12	8.47	11.95	-12	-20	0
3	-12	-19	-2	0.027 623	4.45	5.13	-8.24	-9.50	8.95	11.13	-12	-19	-2
4	-12	-19	-1	0.029 001	4.43	5.75	-7.36	-9.55	8.93	11.75	-12	-19	-1
5	-11	-21	2	0.027 804	2.79	5.83	-5.61	-11.72	7.29	11.83	-11	-21	2
6	-11	-20	0	0.027 753	3.43	5.28	-7.21	-11.09	7.93	11.28	-11	-20	0
7	-11	-20	1	0.028 827	3.26	5.79	-6.27	-11.11	7.76	11.79	-11	-20	1
8	-11	-19	-1	0.028 352	3.85	5.10	-7.89	-10.46	8.35	11.10	-11	-19	-1
9	-11	-19	0	0.029 674	3.74	5.67	-6.93	-10.50	8.24	11.67	-11	-19	0
10	-11	-18	-3	0.027 051	4.19	4.26	-9.61	-9.77	8.69	10.26	-11	-18	-3
11	-11	-18	-2	0.028 726	4.23	4.84	-8.58	-9.83	8.73	10.84	-11	-18	-2
12	-11	-18	-1	0.030 303	4.20	5.46	-7.60	-9.88	8.70	11.46	-11	-18	-1
13	-10	-21	3	0.027 444	2.09	5.48	-4.95	-12.99	6.59	11.48	-10	-21	1
14	-10	-21	4	0.027 916	1.79	5.80	-4.00	-12.97	6.29	11.80	-10	-21	4
15	-10	-20	1	0.027 847	2.76	5.08	-6.70	-12.12	7.26	11.08	-10	-20	1
16	-10	-20	2	0.028 775	2.54	5.51	-5.68	-12.12	7.04	11.51	-10	-20	2
17	-10	-20	3	0.029 486	2.25	5.91	-4.70	-12.32	6.75	11.91	-10	-20	3
18	-10	-19	-1	0.027 364	3.30	4.47	-8.56	-11.62	7.80	10.47	-10	-19	-1
19	-10	-19	0	0.028 751	3.19	4.97	-7.46	-11.63	7.69	10.97	-10	-19	0
20	-10	-19	1	0.029 960	3.01	5.48	-6.41	-11.65	7.51	11.48	-10	-19	1
21	-10	-19	2	0.030 951	2.76	5.96	-5.41	-11.66	7.26	11.96	-10	-19	2
22	-10	-18	-2	0.027 767	3.64	4.24	-9.36	-10.90	8.14	10.24	-10	-18	-2
23	-10	-18	-1	0.029 429	3.60	4.79	-8.23	-10.94	8.10	10.79	-10	-18	-1
24	-10	-18	0	0.030 939	3.49	5.36	-7.15	-10.97	7.99	11.36	-10	-18	0
25	-10	-18	1	0.032 247	3.30	5.93	-6.12	-11.00	7.80	11.93	-10	-18	1
26	-10	-17	-3	0.027 908	3.95	3.95	-10.16	-10.18	8.45	9.95	-10	-17	-3
27	-10	-17	-2	0.029 838	3.98	4.53	-9.00	-10.23	8.48	10.53	-10	-17	-2
28	-10	-17	-1	0.031 658	3.95	5.14	-7.90	-10.28	8.45	11.14	-10	-17	-1
29	-10	-17	0	0.033 305	3.84	5.79	-6.85	-10.33	8.34	11.79	-10	-17	0
30	-10	-16	-4	0.027 774	4.20	3.61	-10.98	-9.44	8.70	9.61	-10	-16	-4
31	-10	-16	-3	0.029 949	4.31	4.19	-9.79	-9.51	8.81	10.19	-10	-16	-3
32	-10	-16	-2	0.032 067	4.38	4.84	-8.66	-9.58	8.88	10.84	-10	-16	-2
33	-10	-16	-1	0.034 062	4.36	5.55	-7.59	-9.65	8.86	11.55	-10	-16	-1
34	-10	-15	-5	0.027 368	4.38	3.23	-11.81	-8.70	8.88	9.23	-10	-15	-5
35	-9	-20	2	0.027 413	2.09	4.77	-6.08	-13.86	6.59	10.77	-9	-20	2

Figure 4 Part of FSLP printout corresponding to Fig. 2. Each reflection can be identified from its (*x*, *y*) coordinates.



**Figure 5** The experimental transmission Laue pattern requiring indexing (corresponds to Fig. 2).

found from Equation 11 for any two reflections belonging to a zone. Knowing the Miller indices and spatial positions of two zone axes, the crystal/X-ray orientation is known. In this manner, Fig. 2 was constructed.

In attempting to establish the exact crystal orientation two possibly common errors became obvious; the film is not perpendicular to the X-ray beam and secondly, the specimen-to-film distance is not accurately set. Both these errors have a significant effect on procedures 1 and 2 above. The former error cannot be compensated for by the procedures described here, however, the latter can. Knowing the Miller indices of the reflections or zone axes, since their stereographic positions depend on  $L$  (see Equations 2 and 3), a series of inter-reflection or inter-direction angles can be calculated from Equation 11 as a function of  $L$  and compared to those calculated from the crystal formula.

On many occasions, it is difficult to obtain an exact match because the variables listed in Section 2.4 are not accurately known, the film is skewed to the X-ray beam and because reflection intensities are dependent on several geometrical conditions. As in the case of Fig. 2, it is often found helpful to allow all reflections ( $NCLASS = 0$ ) to contrib-

ute to the simulated TLP and work from symmetry. However, when all conditions are known and set, exact one-to-one correspondence has been achieved. In fact, with standard crystals such as silicon, the voltage of the X-ray tube or synchrotron can be closely determined by examining the reflections present (i.e. decrease LLAM until all reflections are accounted for). The possibility also exists for determining space groups by comparing the simulated results ( $NCLASS \neq 0$ ) with experiment.

#### 4. Conclusions

The computer program FSLP has produced accurate simulated TLPs and BSLPs. Formulae are given for refining the crystal/X-ray beam relative orientation; however, caution must be used to set the experimental conditions as accurately as possible. The computer program FSLP operates on a CDC 6600 and CDC Cyber 175 both in the author's laboratory and in the Institut für Kristallographie, Technische Hochschule, Aachen, Germany (K. Roberts, on attachment), and is available upon written request.

#### Acknowledgement

The author wishes to thank K. Roberts of the University of Strathclyde for suggesting this project, for his encouragement and his several synchrotron TLPs.

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*Received 9 August  
and accepted 1 September 1982*