RAPID ALIGNMENT OF CRYSTALS IN X-RAY CAMERAS

By S. Ramaswamy and S. Ramaseshan, F.A.Sc.

(From the Department of Physics, Indian Institute of Technology, Madras-36, India)

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1. Introduction

The alignment of crystals on the Weissenberg Goniometer in the quickest possible time would always be an advantage. It is especially so in low temperature crystallography in laboratories with an inadequate supply of liquid air. Therefore, although the question of alignment has been repeatedly studied, it was re-examined with a view to finding if possible, routine procedures, that will considerably economise time. The paper presents some new results.

2. The Existing Methods

When the alignment is perfect, *i.e.*, a crystallographic axis coincides exactly with the axis of rotation, the zero layer spots are recorded on the equator in a cylindrical camera. The spots lie on a straight line on the spread out film. If the crystallographic axis is slightly inclined to the axis of rotation the spots do not lie on the equator. If the displacements of the recorded spots from the equator are measured the errors in alignment can be calculated. When the goniometer head is set such that the short arc is horizontal with its scale facing upwards, the long arc is vertical and its scale faces the observer. This is defined as the "parallel-perpendicular setting". For an error i_V in the vertical plane alone, the displacement d_1 from the equator of a reflection whose Bragg angle is θ is given by

$$d_1 = \text{R} \sin 2\theta \sin i_V \qquad (1)$$

where R is the radius of the cylindrical camera. For an error i_H in the horizontal plane alone, the displacement of this reflection from the equator is

$$d_2 = \text{R} (1 - \cos 2\theta) \sin i_H. \qquad (2)$$

If both errors are present, the resultant displacement is

$$\text{D} = d_1 + d_2 = \text{R} [\sin 2\theta \sin i_V + (1 - \cos 2\theta) \sin i_H]. \qquad (3)$$

These equations were first given by Hendershot (1937).

In the parallel-perpendicular setting the errors in the arcs will also be i_V and i_H in the vertical and horizontal arcs respectively.

From the above equations we get

$$\text{at} \quad \theta = +45^\circ \qquad D_1 = R\,[\sin i_H + \sin i_V]$$

and

$$\text{at} \quad \theta = -45^\circ \qquad D_2 = R\,[\sin i_H - \sin i_V].$$

Thus by making measurements of the displacements D_1 and D_2 at $\theta = 45^\circ$ and -45° respectively, the errors can be easily computed. Since Bragg reflections may not exist at these specific angles, unfiltered radiation is used. If two unequal exposures with the crystal rotated exactly through 180° are given on the same film, the distances at $\theta = \pm 45^\circ$ between the same reflection in the two traces give $2D_1$ and $2D_2$ from which the errors are calculated [Fig. 1 (*a*), (*b*), and (*c*)]. This procedure obviates the difficulty of locating the trace of the ideal central line [Weisz and Cole (1948)].

In a modification of this method Davis (1950) has taken the double exposure with goniometer head turned through 45° in the anticlockwise direction from the parallel-perpendicular setting [Figs. 1 (A), (B) and (C)]. This we shall call the "45° setting".

In this setting let the errors in the vertical and horizontal *planes* be i_V' and i_H'. The separation is now given by

$$2D = 2R\,[\sin i_H'\,(1 - \cos 2\theta) + \sin 2\theta \sin i_V']$$

i_V' and i_H' can be related to i_V and i_H, the actual errors in the two arcs by

$$\begin{bmatrix} \sin i_V' \\ \sin i_H' \end{bmatrix} = \begin{bmatrix} \cos 45 & -\sin 45 \\ \sin 45 & \cos 45 \end{bmatrix} \begin{bmatrix} \sin i_V \\ \sin i_H \end{bmatrix}.$$

Substituting this we get for the separation

$$2D = \sqrt{2}R\,[\sin 2\theta\,(\sin i_V - \sin i_H) + (1 - \cos 2\theta)\,(\sin i_V + \sin i_H)].$$

At $\theta = +45^\circ$ this reduces to $\qquad 2D_1 = 2\sqrt{2}R \sin i_V$

and at $\theta = -45^\circ$ this becomes $\qquad 2D_2 = 2\sqrt{2}R \sin i_H$.

Thus the errors may directly be calculated from the measurement of the separations 2D at $\theta = \pm 45^\circ$ thereby eliminating any further additions and subtractions as in the previous method [Fig. 1 (C)].

In most of the cases the spots are not obtained at these exact angles $\theta = \pm 45°$. Garaycochea and Dresdner (1961) suggested that in such cases the Hendershot formula can be recast as

$$\frac{\sin i_V}{\frac{2D}{2R \sin 2\theta}} + \frac{\sin i_H}{\frac{2D}{2R\,(1 - \cos 2\theta)}} = 1. \tag{4}$$

Treating sin i_V and sin i_H as variables along the coordinate axes straight-lines may be drawn, one for each different spot. The intercepts along the co-ordinate axes can be calculated for each spot from its 2D and θ values. The different straight lines are expected to intersect at the same point and the co-ordinates of the point of intersection of these straight lines give the errors

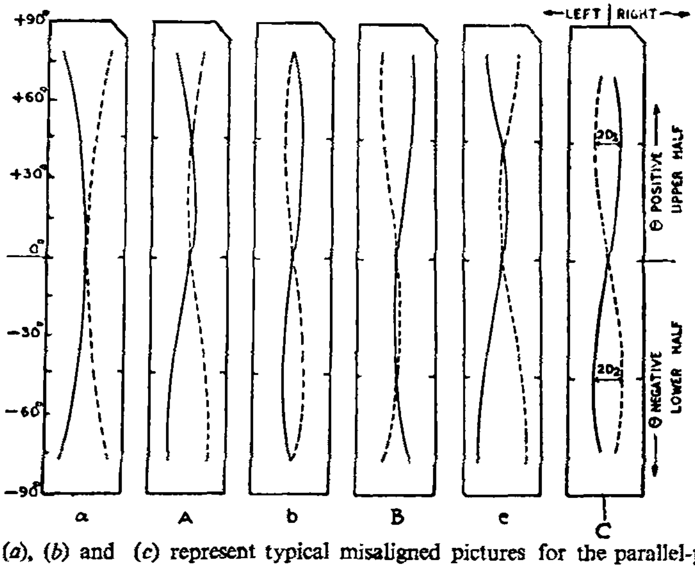

FIG. 1. (*a*), (*b*) and (*c*) represent typical misaligned pictures for the parallel-perpendicular setting of the arcs. 1 (A), (B) and (C) represent the *corresponding cases* in the 45° setting of arcs.

(*a*) and (A): Traces obtained when there is misalignment in the horizontal arc only ($i_V = 0$). It may be noted that the curve in 1 (*a*) is always a $(1 - \cos 2\theta)$ curve and in 1 (A) $2D_1 = 0$ at $\theta = +45°$.

(*b*) and (B): Traces obtained when the misalignment is in the vertical arc ($i_H = 0$). 1 (*b*) is always a sine curve and in 1 (B) $2D_2 = 0$ at $\theta = -45°$.

(*c*) and (C): Traces obtained when the misalignment is in both the arcs. The sign convention is also illustrated in 1 (C).

of the two arcs. Davis (1961) has suggested that this modification of Garaycochea *et al.* may be applied to his method also, provided the intercepts are marked along the two lines sin i_V = sin i_H and sin i_V = − sin i_H taking proper signs.

3. THE NEW METHOD

For the parallel-perpendicular setting, treating 2D and $\sin 2\theta$ as variable co-ordinates, the Hendershot equation is recast as

$$X^2 (a^2 + b^2) + Y^2 - 2bXY + 2abX - 2aY = 0. \qquad (5)$$

This is an equation to an ellipse passing through the origin. Here $X = \sin 2\theta$; $Y = 2D$; $a = 2R \sin i_H$ and $b = 2R \sin i_V$. In the case of a crystal which is misaligned in both the arcs the plot of 2D against $\sin 2\theta$ is an ellipse having its axes inclined to the axes of co-ordinates. When $i_H = 0$, the graph is a straight line inclined to the co-ordinate axes and as i_H increases the straight line opens out into an ellipse, and when $i_V = 0$ the ellipse has its axes parallel to the axes of co-ordinates. In every case the curve passes through the origin and if one has three or four points (preferably on either side of the central spot) a good ellipse can be drawn. Reading the values of the displacements corresponding to $\sin 2\theta = \pm 1$ from the graph one gets $2D_1 = a + b$ and $2D_2 = a - b$ from which the errors in the arcs can be calculated to a fair degree of accuracy.

It is found that this method is applicable also in the 45° setting used by Davis. The plot of 2D against $\sin 2\theta$ again gives an ellipse

$$X^2 (A^2 + B^2) + Y^2 - 2AY - 2BYX + 2ABX = 0$$

passing through the origin. But here $A = \sqrt{2}R (\sin i_V + \sin i_H)$ and $B = \sqrt{2}R (\sin i_V - \sin i_H)$. The advantage in working with the 45° setting of the arcs is that the calculation of the errors of the arcs is much simplified. For, if one measures the values of 2D at $\theta = \pm 45°$ one gets

$$\text{at } \theta = +45° \qquad 2D_1 = 2\sqrt{2}R \sin i_V$$

$$\text{and at } \theta = -45° \qquad 2D_2 = 2\sqrt{2}R \sin i_H.$$

It can also be seen that when $i_H = 0$ $(i_H' \neq 0)$ the displacement is zero at $\theta = -45°$ and when $i_V = 0$ $(i_V' \neq 0)$ the displacement is zero at $\theta = +45°$ and the ellipse touches the axes at these points. When both the errors are equal the values of 2D at $\theta = +45°$ and $-45°$ are also equal.

4. ROUTINE EXPERIMENTAL PROCEDURE

The goniometer head may be in the parallel-perpendicular setting or the 45° setting. A 15° oscillation picture is taken giving an exposure of 30 minutes. The crystal is rotated exactly through 180° from the first mean position and a second exposure is given for 10 minutes for the same oscillation range. As soon as the film is removed from the camera a small cut is made at the right hand top corner of the film. If the diameter of the camera is 57·3 mm. then the distance in mm. of any reflection from the central spot gives the Bragg angle θ in degrees. A transparent scale graduated in mm. with zero at the centre may conveniently be used for the measurement of the Bragg angles. A low power microscope with a graticule attached to the eye-piece may be used to measure the distance 2D directly. The following sign convention is adopted. θ is + ve when the spot is in the upper half and − ve when it is in the lower half. 2D is positive if the darker spot is on the right half and − ve if it is on the left half. [$2D_1$ is + ve and $2D_2$ is − ve in Fig. 1 C].

The graph is drawn with 2 D and sin 2θ values for three or four spots on either side of the centre and the errors are determined. Having calculated the errors in the setting it is convenient to make the necessary corrections

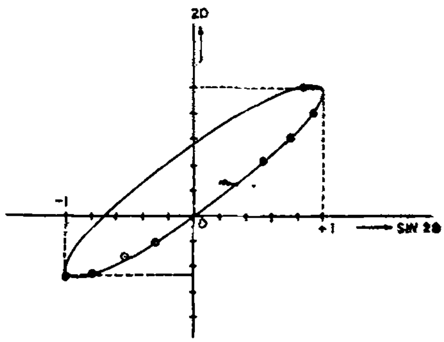

FIG. 2

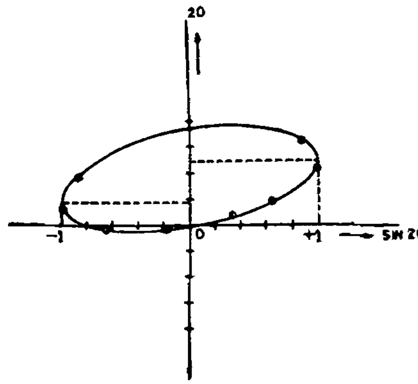

FIG. 3

FIGS. 2 and 3 refer to case 1 of the tabulation given above. Fig. 2 gives the ellipse method in the parallel perpendicular setting and Fig. 3 in the 45° setting of the arcs. 2 D values are taken from the graph at sin $2\theta = +1$ and -1.

in the arcs with the goniometer in the parallel-perpendicular setting. If the calculated error i_V along the long arc is positive then for the correction, the tip of the crystal should move down; If the error i_V is negative, the tip of the crystal should move up. If the calculated error i_H along the short arc is positive the tip of the crystal should move away from the beam; if the error i_H is negative, the tip should be moved towards the beam. When spots are not obtained near and above the value of $\theta = \pm 45°$ it is quite difficult to determine the angular errors accurately by the earlier wo methods. The straight line method suggested by Garaycochea *et al.*, and the ellipse method are useful even if there are only three or our spots on either side of the central spot.

The wet film may itself be mounted on the transparent scale and measurements made. The graph is also drawn without any further calculations. The time taken to determine the errors by this method is found to be much less than by the other methods.

The errors calculated by the four methods in five actual and different cases in this laboratory are tabulated below:

TABLE I

No.		Ellipse method		Straight line method		
		Parallel-perpendicular setting	45° setting	Parallel-perpendicular setting	45° setting	Experimental
Case 1 ..	i_V	3° 38′	3° 30′	3° 42′	3° 44′	3° 40′
	i_H	1° 21′	1° 8′	1° 30′	1° 52′	1° 20′
Case 2 ..	i_V	2° 34′	2° 30′	2° 39′	2° 36	2° 30′
	i_H	– 37′	– 38′	– 48′	– 18′	– 40′
Case 3 ..	i_V	1° 42′	1° 48′	1° 41′	1° 43′	1° 40′
	i_H	48	46	51	51	50
Case 4 ..	i_V	– 1° 54′	– 1° 51′	1° 12′	– 1° 53′	– 1° 50′
	i_H	12′	25′	30′	13′	10′
Case 5 ..	i_V	– 15′	– 25′	– 24′	– 34′	– 20′
	i_H	– 4° 15′	– 4° 36′	– 4° 12′	– 4° 36′	– 4° 40′

5. SUMMARY

The misalignment in the setting of a crystal in the Weissenberg goniometer is determined from the measurement of the displacement of the spots from the central line at $\theta = \pm 45°$ with the arcs set in the parallel-perpendicular position (Hendershot, 1937) or in the 45° position (Davis, 1950). When reflections are not present at $\theta = \pm 45°$ a graphical straight line method has been suggested by Garaycochea *et al.* (1961).

Another graphical method called the "Ellipse method" suggested in the paper does not involve too much calculation after the measurement and hence is easy in application, accurate in the determination of errors and economical in time.

6. Acknowledgement

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7. References

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