

## DOUBLE SPOTS AND THEIR ELIMINATION IN PRECESSION PHOTOGRAPHY

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### ABSTRACT

The origin of the elongation and doubling of diffraction spots in precession photography is analyzed, and four types of double spots are distinguished according to their origins: (A) parallel shift of the film, (B) tilt of the film, (C) misorientation of the crystal, and (D) nonsystematic errors. The radial and tangential components of the displacement of the diffraction spots due to the three systematic errors are derived and compared. It is possible to determine the cause of the doubling from its characteristic pattern. A new type of screen which permits only one diffraction spot per reciprocal-lattice point is presented.

### INTRODUCTION

The precession camera of Buerger (1944) is becoming the most widely used single-crystal film camera. It reproduces the reciprocal lattice in its true symmetry and undistorted, scaled-up dimensions, thus making lattice measurements and other investigations remarkably simple. Unfortunately, elongated or double spots often appear in precession photographs, and their presence disturbs the otherwise clear picture.

In some cases the elongation of the spots is due to the acicular shape of the crystal, and elongation or doubling to imperfections in the crystal structure, or to low angle boundaries. In most cases, however, the doubling is due to mechanical, orientation, or camera-setting errors, and these can be corrected either by eliminating the cause of the doubling or by using special techniques.

### THE ORIGIN OF DOUBLE SPOTS

Double spots are possible in precession photography, since every reciprocal-lattice point gets into diffracting position twice during a precession cycle—first when the sphere of reflection approaches and then when it leaves a reciprocal-lattice point. When the camera is in good adjustment and the crystal is perfectly oriented, the paths of these two diffraction beams cross at the same point on the film and leave only one mark. In case of misadjustment or misorientation, on the other hand, these two paths intersect either before or behind the film and, consequently, they leave two marks on the film which may be partially overlapping or completely separated.

The two diffraction positions of a crystal for one reciprocal-lattice point, and the corresponding paths of the diffraction beams are illustrated in Fig. 1. This illustration differs from the customary construction of the precession camera and the usual illustration of the precession principle by

keeping the film stationary and allowing the crystal and the incident  $x$ -ray beam to follow the precession motion. In the available precession cameras this is reversed; that is, the  $x$ -ray beam is kept stationary. Such diagrams, however, would make the illustration of the double diffraction extremely difficult since the film moves between the two diffractions. The illustration of the precession principle in Fig. 1 is still correct and, however impractical, such a camera could be constructed and properly be called a precession camera.

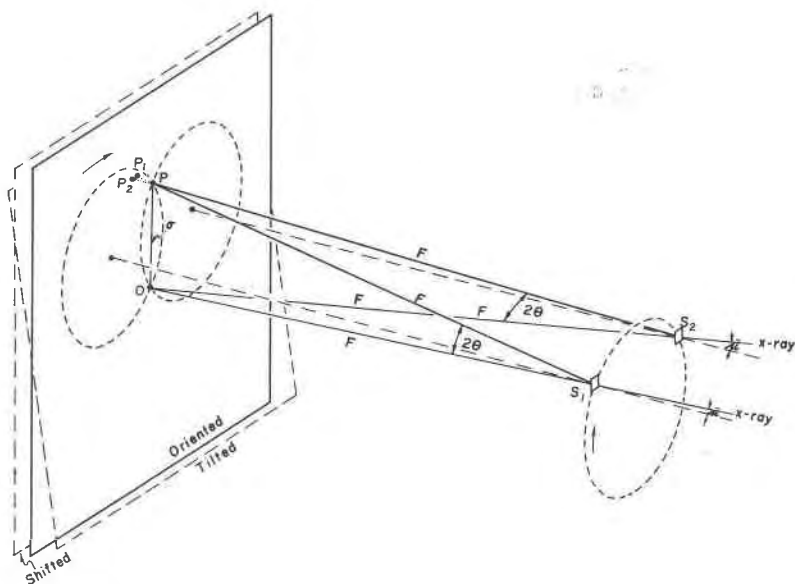


FIG. 1. Illustration of the two diffractions of a reciprocal lattice point in the precession camera and of the appearance of double spots due to a shift or tilt in the position of the film.

A. The most common and simplest cause of doubling is an error in the selection of the layer-line screen, in the setting of the  $Fd^*$  distance or of the  $\bar{\mu}$  angle. In these cases the spots will be doubled, since the film is not at the required distance from the crystal, or the layer-line screen permits the passage of diffraction beams coming from other than the desired reciprocal-lattice layer. The same effect can, obviously, be observed when the scale on the  $Fd^*$  bar or  $\bar{\mu}$  arc is marked erroneously. In all these cases the film stays parallel with the desired reciprocal-lattice plane of the crystal as it should, but is shifted by a certain amount from its proper position.

If the slit of the layer-line screen is too wide or  $d^*$  of the crystal perpendicular to the investigated reciprocal-lattice layer is too short, diffraction from higher levels can register on the film. Besides the single spots of the proper level, double spots will appear which come from these higher levels. The reason for the doubling of these spots is similar to the above, that is, the film is not at the required distance to photograph these higher levels.

The doubling of spots due to the improper setting of the film has been

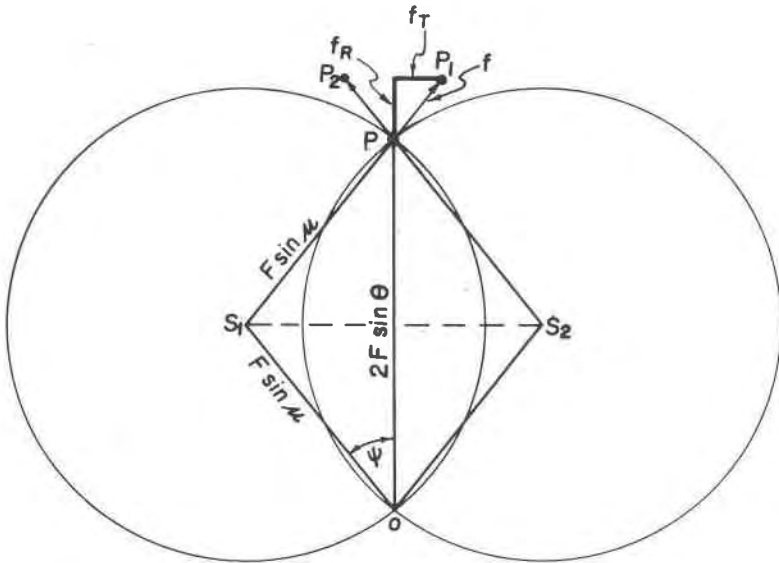


FIG. 2. Illustration of the displacement of a diffraction spot in precession photography due to a shift in the position of the film.

discussed by Buerger (1944, pp. 30–32). He derives the relationship between the error  $e$  in the setting of the film and the displacement of a diffraction spot in the radial and tangential directions with respect to the center of the film: ( $f_r$  and  $f_t$  respectively).

$$f_r = e \tan \bar{\nu} \cos \psi \tag{1}$$

and

$$f_t = e \tan \bar{\nu} \sin \psi \tag{2}$$

Where  $\psi$  is the angle  $SPO_n$  in Buerger's Fig. 15A (1944, p. 30) or  $S_1OP$  in Fig. 2 of this paper, the displacements  $f_r$  and  $f_t$  can be expressed in terms of  $\bar{\mu}$  and  $\theta$  for a zero level photograph with the aid of Fig. 2 as

$$\cos \psi = \frac{\sin \theta}{\sin \bar{\mu}} = \csc \bar{\mu} \sin \theta \tag{3}$$

and

$$\sin \psi = \frac{(\sin^2 \bar{\mu} - \sin^2 \theta)^{1/2}}{\sin \bar{\mu}}$$

or

$$= \csc \bar{\mu} (\sin^2 \bar{\mu} - \sin^2 \theta)^{1/2} \quad (4)$$

and substituting these in (1) and (2)

$$f_r = e \tan \theta \csc \bar{\mu} \sin \theta \quad (5)$$

$$f_t = e \tan \theta \csc \bar{\mu} (\sin^2 \bar{\mu} - \sin^2 \theta)^{1/2} \quad (6)$$

where  $e$  is in the reciprocal-lattice unit of the precession photograph.

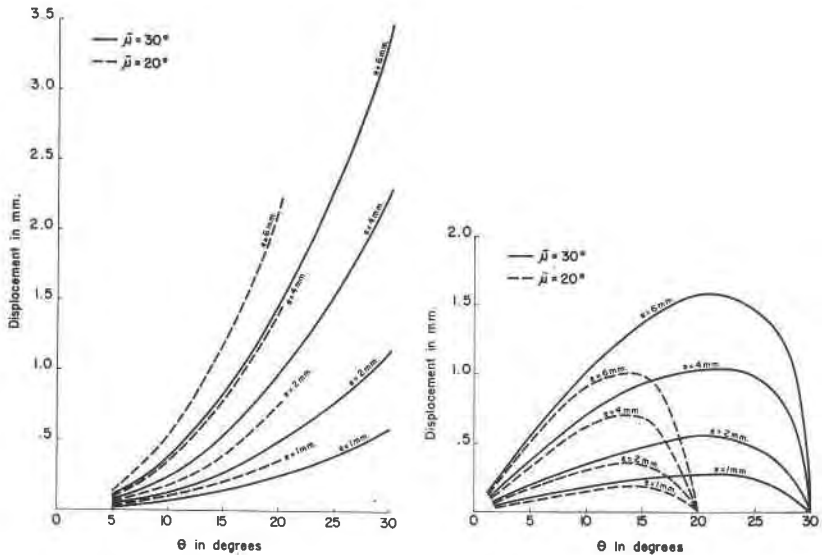


CHART 1. The radial (a) and the tangential (b) displacement of the diffraction spots due to various shifts in the position of the film.

The quantitative relationship between the shifting of the film from its proper position and the displacement of a diffraction spot (in terms of  $f_r$  and  $f_t$ ) for continuous values of  $\theta$  is given in Chart 1 for 1, 2, 4, and 6 mm. shift for a film-to-crystal distance:  $F = 6.0$  cm, both for  $\bar{\mu} = 20^\circ$  and  $30^\circ$ .

**B.** All the errors in the construction or alignment of the precession camera (with the exception of erroneous marking of the scale on the  $Fd^*$  bar and the  $\bar{\mu}$  arc) result in the destruction of the parallelism between the desired reciprocal lattice plane of the crystal and the film at certain stages of the precession cycle. For example, the misalignment of the collimator, or an angle between the rotation axis of the crystal and the horizontal

axis of the film, or a significant play in the bars connecting the crystal and the film holder columns will cause a variation in the orientation of the film with respect to the crystal during the precession motion. These errors can cause doubling of the diffraction spots, since some portions of the film are not at the proper distance from the film. The film can be regarded as being tilted with respect to the reciprocal-lattice layer around a certain tilt axis. When the reciprocal lattice vector,  $\sigma$  (in zero level photogra-

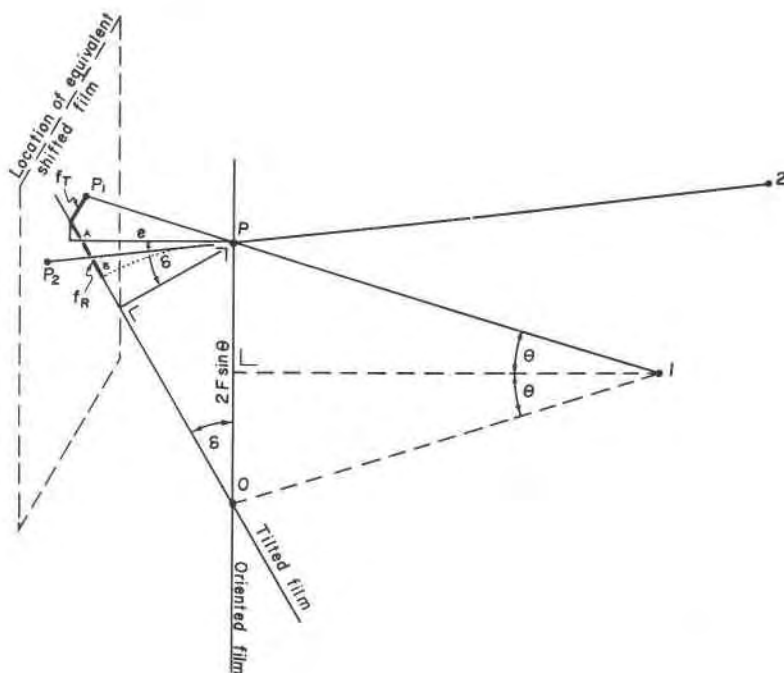


FIG. 3. Illustration of the displacement of a diffraction spot due to a tilt and of a shift of the film giving rise to identical tangential displacement.

phy) coincides with this tilt axis, there is no destruction in the parallelism, but when  $\sigma$  is  $90^\circ$  from the tilt axis, the film is tilted towards or away from the photographed reciprocal-lattice layer.

The diffraction spots of the reciprocal-lattice points which have  $\sigma$  perpendicular to the tilt axis will, obviously, suffer the largest displacement. The relationship between the tilt of the film  $\delta$  as measured from  $\sigma_{\perp}$ , and the displacement of the spots along  $\sigma_{\perp}$  can be derived from Figs. 1 and 3. Figure 3 illustrates the identity of the  $f_i$  components of displacement for a

given shift and a corresponding tilt of the film. The shift of the film  $e$  can be expressed in terms of  $\delta$ .

$$e = 2F \sin \theta \tan \delta \tag{7}$$

therefore,  $f_t$  due to the tilt of the film is

$$f_t = 2F \sin \theta \tan \delta \tan \theta \csc \bar{\mu} (\sin^2 \bar{\mu} - \sin^2 \theta)^{1/2} \tag{8}$$

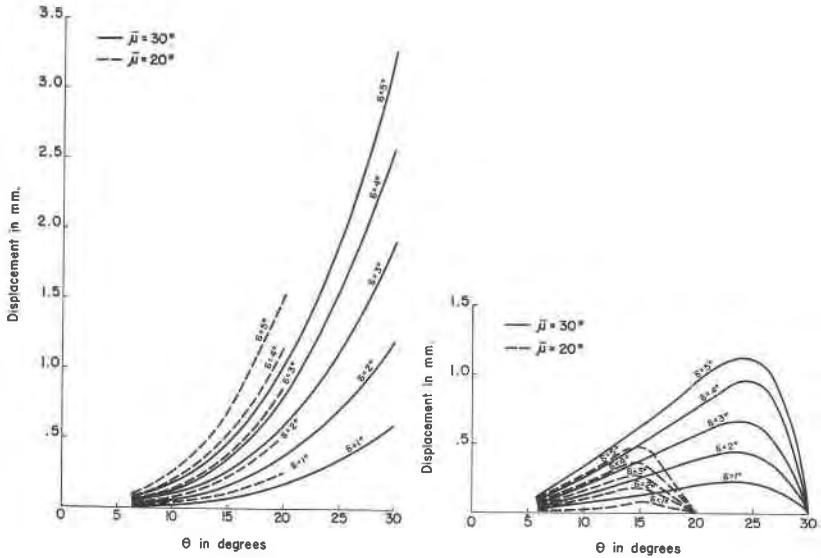


CHART 2. The maximum radial (a) and tangential (b) displacement of the diffraction spots due to tilting of the film.

We can also derive from Fig. 3 that  $f_r$  is equal to that of the shifted film divided by  $\cos \delta$  (or multiplied by  $\sec \delta$ ) plus the distance  $AB$ .  $AB$  can be expressed as

$$AB = 1/2e \sin \delta \tag{9}$$

therefore

$$f_r = 2F \sin \theta \tan \delta \tan \theta \csc \bar{\mu} \sin \theta \sec \delta + F \sin \theta \tan \delta \sin \delta$$

or

$$f_r = 2F \sin \theta \tan \delta (\tan \theta \csc \bar{\mu} \sin \theta \sec \delta + 1/2 \sin \delta) \tag{10}$$

The quantities  $f_r$  and  $f_t$  for continuous values of  $\theta$  are illustrated in Chart 2 for  $\delta = 1^\circ, 2^\circ, 3^\circ, 4^\circ, 5^\circ$ , for  $F = 6.0$  cm and for  $\bar{\mu} = 20^\circ$  and  $30^\circ$ .

C. The third and the most frequent cause of doubling of the diffraction

spots is the misorientation of the crystal. In this case the film is not parallel to a reciprocal-lattice layer, and consequently, the sought reciprocal-lattice plane is tilted with respect to the film. The misorientation of the reciprocal-lattice layer is illustrated in Fig. 4. When the crystal is well-oriented, a certain reciprocal-lattice point will be in diffracting position at  $S_1$  and again at  $S_2$ , and the resulting two diffraction paths will cross at the film. When the reciprocal-lattice layer is tilted (counter-clockwise in Fig.

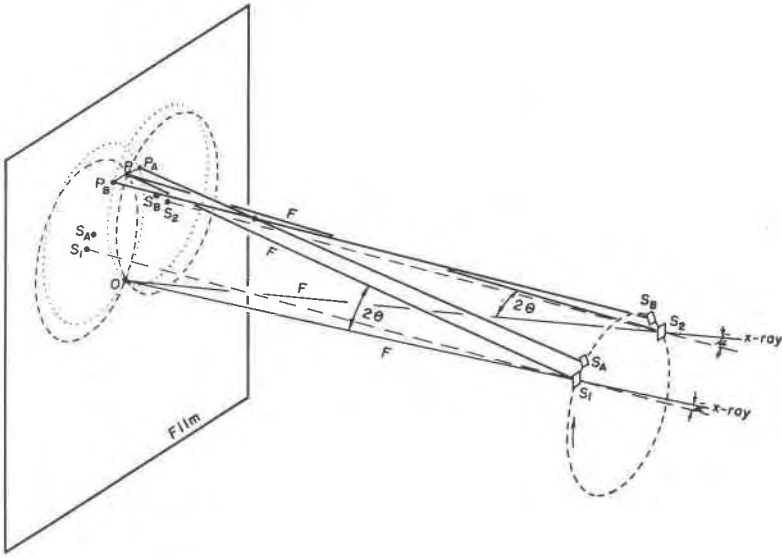


FIG. 4. Illustration of the doubling of diffraction spots due to the misorientation of the crystal

4) the first diffraction will take place at  $S_A$  rather than at  $S_1$ . This is so since at the  $S_1$  position due to the tilt of the crystal, the angle between the lattice layer and the incident  $x$ -ray beam is less than  $\theta$ . The required  $\theta$  will be fulfilled later at position  $S_A$ . Similarly the second diffraction will take place at  $S_B$  instead of  $S_2$ . The two diffraction paths from  $S_A$  and  $S_B$ , obviously, will not cross at  $P$  but before the film and leave two separate marks on the film at  $P_A$  and  $P_B$ .

The maximum displacement of the diffraction spots due to misorientation will be again for those reciprocal-lattice points which have  $\sigma$  perpendicular to the tilt axis of the reciprocal lattice. The degree of tilt can be given by  $\delta$  from  $\sigma_{\perp}$  and the relationship between  $\delta$  and the displacement of the diffraction spots along  $\sigma_{\perp}$  can be derived from Figs. 4 and 5. Figure 5 is a projection of selected elements of Figure 4 on the film.

When the incident  $x$ -ray beam is in the plane which is perpendicular to the film and contains the tilt axis, the crystal is at the position  $S_0$  in Fig. 5, and no diffraction takes place for  $\sigma_{\perp}$  reciprocal-lattice points. That is, the incident beam is in the plane of the lattice layers corresponding to  $\sigma_{\perp}$ . As the precession motion proceeds, the position of the crystal moves from  $S_0$  to  $S_1$  then to  $S_A$  and finally to  $S_{90^\circ}$ . The latter represents the case when

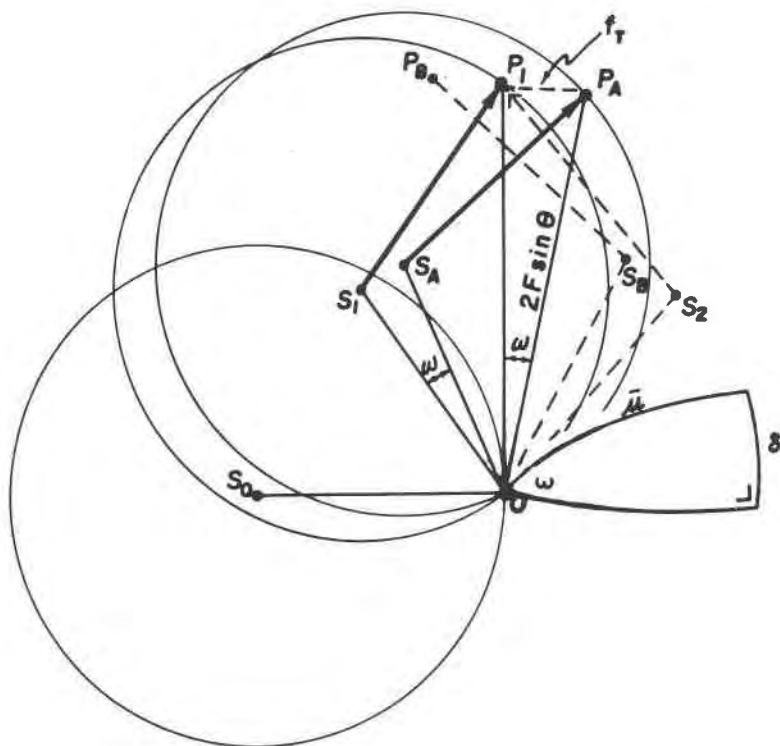


FIG. 5. Illustration of the displacement of a diffraction spot in precession photography due to misorientation. The spherical triangle illustrates the relationship between  $\bar{\mu}$ ,  $\omega$ , and  $\delta$ .

the lattice layers corresponding to  $\sigma_{\perp}$  are most inclined to the  $x$ -ray beam and the longest  $\sigma_{\perp}$  reciprocal-lattice point can diffract. The inclination of the  $x$ -ray beam at  $S_{90^\circ}$  is equal to  $\bar{\mu}$ . In other words, as the position of the crystal is rotated from  $S_0$  to  $S_{90^\circ}$ , the inclination of the  $x$ -ray beam with these lattice layers increases from  $0^\circ$  to an angle equal to  $\bar{\mu}$ .

The maximum  $\theta$  in case of  $\delta$  tilt of the reciprocal-lattice layer changes from  $\bar{\mu}$  to  $\bar{\mu} \pm \delta$ . This is  $+\delta$  in the lower half of the film in Figure 4 and  $-\delta$



in the upper half. The signs are, of course, reversed in the case of a clockwise tilt. This relationship between the tilt angle and the maximum  $\theta$  in terms of the displacement of the darkened area of the precession photograph has been pointed out by Buerger (1944) and Evans *et al.* (1949), and used in the development of their orientation-correction techniques.

The relationship between the tilt angle  $\delta$  and the corresponding change  $\omega$  in the rotation of  $S$  for a given reciprocal-lattice point can be expressed from the spherical triangle of Fig. 5 as:

$$\sin \omega = \frac{\sin \delta}{\sin \bar{\mu}} = \sin \delta \csc \bar{\mu} \quad (11)$$

Since the triangles  $S_1OP_1$  and  $S_AOP_A$  are identical, the angles  $S_1OS_A$  and  $P_1OP_A$  are both equal to  $\omega$ . The displacement between  $P_1$  and  $P_A$  for small angles of  $\omega$  can therefore be expressed as

$$f = 2F \sin \theta \sin \omega \quad (12)$$

The  $f$  of equation (12) can be regarded equal to  $f_t$  for small angles of  $\omega$  since the angle  $P_BP_AP_1$  is equal to  $\omega/2$  and the difference between  $f$  and  $f_t$  is obviously negligible. Substituting equation (11) in equation (12) we obtain:

$$f_t = 2F \sin \theta \sin \delta \csc \bar{\mu} \quad (13)$$

The radial displacement of the diffraction spot caused by the tilt of the reciprocal-lattice layer can be derived from the similar triangle  $P_AP_1P_B$  of Fig. 5 for small angles of  $\omega$

$$f_r = f_t \sin \omega/2 \quad (14)$$

and after completing the substitutions

$$f_r = F \sin \theta \sin^2 \delta \csc^2 \bar{\mu} \quad (15)$$

Chart 3 illustrates the displacement components  $f_r$  and  $f_t$  for continuous values of  $\theta$  for  $\delta = 1^\circ, 2^\circ, 3^\circ, 4^\circ, 5^\circ$ , in case of  $F = 6.0$  cm, both for  $\bar{\mu} = 20^\circ$  and  $30^\circ$ .

**D.** In addition to the above discussed systematic errors, there are other non-systematic errors which can be responsible for the elongation and doubling of the diffraction spots. The most frequent of these is the bulging of the film in the film holder. If the film is not cut properly or expands due to a change in temperature or humidity, it can bulge in the holder and destroy the parallelism between certain portions of the film and the crystal. The resulting doubling of spots is usually more pronounced around the center of the film. Random doubling of spots can be caused if the film is too small and can move in the film holder during the motion of the camera. Loose settings of the  $\bar{\mu}$  arc,  $Fd^*$  bar and other movable parts of the instrument can also result in the non-systematic doubling of spots.

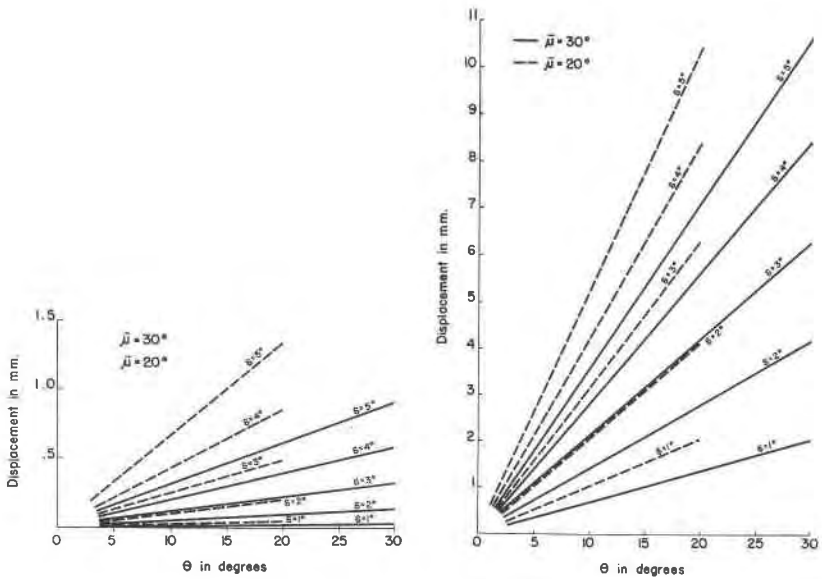


CHART 3. The maximum radial (a) and tangential (b) displacement of the diffraction spots due to the misorientation of the crystal.

### ELIMINATION OF THE DOUBLE SPOTS

The analysis of the errors responsible for the doubling of the spots can help one to determine the cause of separation and subsequently to correct the trouble. As a conclusion of the previous discussion, we can say that there are four types of errors resulting in doubling of diffraction spots:

- (A) shifting of the film
- (B) tilting of the film
- (C) tilting of the reciprocal lattice layer
- (D) non-systematic errors

The first two errors can be distinguished from the third, since in the former cases,  $f_t$  diminishes around the edge of the recorded area on the film. That is, the spots come together again, while in the third case they get farther and farther separated towards the edge of the film. The first cause can be distinguished from the second by the examination of the shape of the  $f_t$  curves. The separation of the spots is more pronounced near the center of the recorded area in the first case, whereas it is close to the edge in the second case. Non-systematic errors, on the other hand, can be recognized by the random doubling of the spots.

Once the cause of the doubling of spots has been determined, it can be corrected. These corrections, however, can be very time consuming.

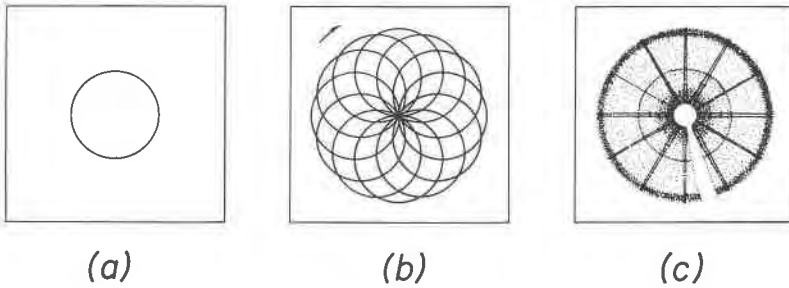


FIG. 6. The circular slit of the layer-line screen (a), the rotation of the layer-line screen with respect to the film during the precession motion (b), and a precession photograph obtained with this screen (c).

Spending time on the realignment of the camera or on the correction of mechanical faults, is, of course, justifiable. However, spending several days on the perfect orientation of the crystal, in many cases, can be of questionable value. The perfect orientation of the crystal, for example, is not essential when intensity data are collected, or when the mosaic structure, stacking faults, or the degree of order in the crystal structure is studied. A considerable amount of time can be saved when the second spot is eliminated by a special technique rather than by perfect orientation.

The circular slit of the layer-line screen of the precession camera represents the circle of reflection, and permits the passage of two diffraction beams per reciprocal-lattice point. The slit, its rotation during the precession motion, and a photograph with double spots are illustrated in Fig. 6.

The double spots can be eliminated in certain portions of the film by covering half of the circular slit of the screen. In Fig. 7a the lower half of the slit is covered, and Figs. 7b and 7c show that two circular areas along

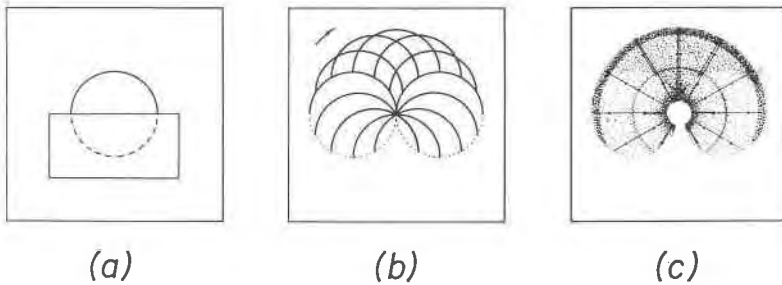


FIG. 7. The half-covered slit of the layer-line screen (a), its rotation (b), and a photograph obtained with this screen (c).

the horizontal line are swept by the circle of reflection only once, resulting in single spots, while the upper part of the film still has double spots, and the lower part is blank. By changing the position of the cover on the layer-line screen, various portions of the film can be selected to have only single spots. With this procedure the whole reciprocal-lattice layer can be recovered in single spots; however, it requires many exposures and results in several films representing one reciprocal-lattice layer.

In the technique presented here the layer-line screen is replaced by a rotating screen which has only a semi-circular slit instead of the circular slit. One end of the semi-circular slit is kept in the precession axis of the

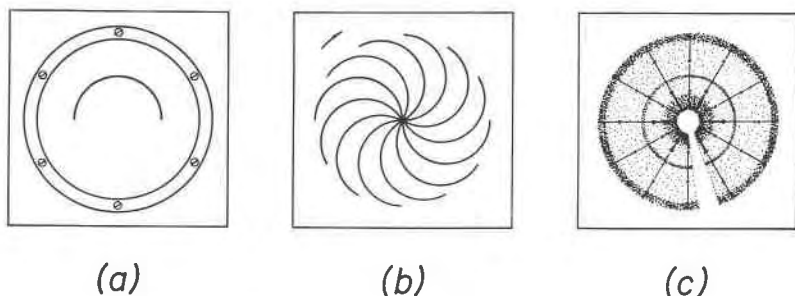


FIG. 8. The semi-circular slit of the double-spot eliminating screen (a), its rotation (b), and a photograph obtained with this screen (c).

camera by a bar attached to the stationary collimator post (the bar also acts as direct-beam catcher) while the slit itself, representing half of the circle of reflection, rotates and sweeps over the reciprocal-lattice layer. Since the rotation of the slit is synchronized with the precession motion, each reciprocal-lattice point gets into diffracting position only once during a precession cycle (Figs. 8a and b). With this screen a complete reciprocal-lattice layer can be recorded with single spots using only one film (Fig. 8c). The plate with the semicircular slit can be replaced by other similar plates with various slit radii.

It should be kept in mind, however, that if the crystal is not in perfect orientation (or the film is shifted or tilted) the spots may be significantly displaced. Consequently, translation measurements should not be attempted on photographs taken with the double-spot eliminating screen except, of course, for approximate information or for the indexing of the spots. The intensity of the spots, on the other hand, can be accepted, since the path-length of the diffraction beam did not change due to the misorientation of the crystal. If the doubling of the spots is due to shifting or tilting of the film, there is a slight change in the path-length of the

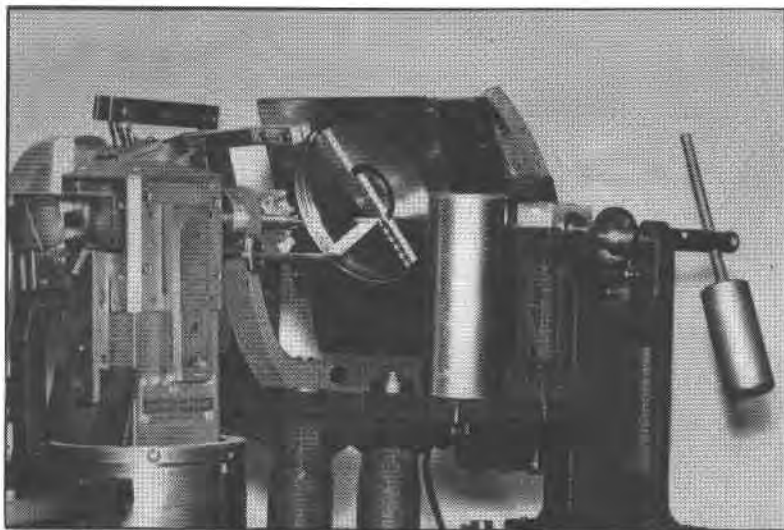


FIG. 9. Photograph of the double-spot eliminating screen mounted on a precession camera.

diffraction beam; however, since air has a very low absorption coefficient, the change in the intensity is negligible. For example, a  $5^\circ$  tilt in the film corresponds to less than  $\frac{1}{3}\%$  change in the intensity of the diffraction spots.

Figure 9 is a photograph of the double-spot eliminating screen mounted on a precession camera. Detailed blueprints of this screen are available on request from the author.

The double-spot eliminating screen was developed in the process of crystallographic research supported by the National Science Foundation.

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