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A NEW X-RAY DIFFRACTION TOPOGRAPHY

By Kōji FUTAGAMI* and Sigeru ŌKI**

A new technique of x-ray diffraction topography has been developed. In this technique, a Soller slit assembly composed several Soller slits of the same type is placed between a sample and a line focus of an ordinary analyzing x-ray tube, and is made to be able to perform scanning. The characteristic x-rays of $K_{\alpha 1}$ component among resulting many narrow x-ray beams are only diffracted by the sample. The mechanism and the adjustment of the apparatus are very simple. The stationary sample and plate make it easy to use various attachments such as heating and stretching devices. The topographs obtained by this technique have such high resolution for separation of dislocation images as the Lang's one. Some applications of this technique are shown.

1. Introduction

Several techniques[1] of the x-ray diffraction topography have been developed as a powerful method of direct observation of defects such as dislocations in a single crystal.

In the techniques so far developed, however, an apparatus complicated extreamly in the instrumental mechanism and adjustment such as in the Lang method[2], is required to obtain a topograph with high resolution. In the Berg-Barrett method[3][4], the previous Soller slit method[5][6] and the filter method[7], elimination of continuous x-rays and separation of the K_α doublet are not good enough to obtain dislocation images with high resolution, although the apparatuses are simplified. The technique utilizing a Soller slit was first propounded by Carlson and Wegener[5]. They made use of the Soller slit to limit horizontal divergence of x-ray beams, and did not consider separation of K_α doublet at all. Later, Andersen[6] modified this method and made topography with an oscillating narrow Soller slit, but did not succeed in clear separation of K_α doublet. Use of a grating assembly was also attempted by him[8], but little improvement was gained in resolution.

In this paper, a new method with merits of both the Soller slit method and the Lang method is described, that is, a method of a scanning Soller slit assem-

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bly which gives a topograph of high resolution by using a very simple apparatus in the instrumental mechanism and adjustment. It is evident that, in order to separate the K_α doublet of the characteristic x-rays, the x-ray beams are to be collimated by using a long, narrow Soller slit into a very small horizontal divergence so that only one of the K_α doublet can be diffracted by the single crystal to give a clear topograph. There is, however, a technical difficulty in making such a long and narrow Soller slit parallel and flat enough to obtain sufficient x-ray beams for the topography. Therefore, by aligning, at certain intervals, some short Soller slits which can be easily manufactured, there can be obtained a Soller slit assembly which is geometrically equivalent to a long Soller slit in horizontal divergence and having small loss of x-ray intensity due to passing through the assembly.

This Soller slit assembly is placed between a line focus and a sample, and is moved a little perpendicularly to the x-ray beams to scan uniformly over the sample during exposure in order to obtain diffraction topographs. Good separation of the K_α doublet is possible by choosing the slit-width and the length of the assembly appropriately, so that very high resolution is attained in this topography. The sample and the plate remaining at rest, a stretching and heating devices for instrumental attachments can be furnished easily. Some experimental results including application of these attachments are shown.

2. Principle and Construction

Generally, in order to obtain a topograph with high resolution, the separation of K_α doublet and sufficient intense intensity of $K_{\alpha 1}$ radiations which are diffracted by the sample must be required. In this new technique the separation of K_α doublet is easily achieved by placing the Soller slit assembly between the line focus and the sample and, as in the Lang method, by collimating the x-ray beams of sufficient intense intensity with small angular divergence as required. The schematic diagram of Soller slit assembly is shown in Fig. 1. It is composed of several Soller slits arranged in a line at intervals. The slits need not be arranged in contact. It is sufficient that the beams diverging from the n-th Soller slit except those for obtaining the very small divergent x-ray

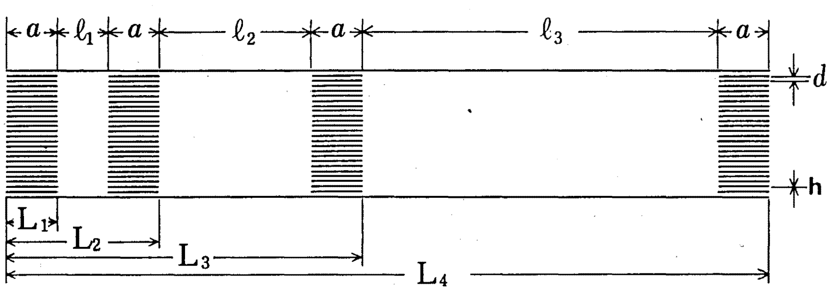

Fig. 1. Schematic diagram of a Soller slit assembly.

beams which entrance into the correspondent opening of the $(n+1)$-th Soller slit should be absorbed by the slit material.

The geometrical condition of the Soller slit assembly would be considered. When identical Soller slits are used, the maximum permissible length l_n of the n-th gap, that is, the distance from the n-th Soller slit exit to the front of the $(n+1)$-th Soller slit, is given by the following equation;

$$l_n = \frac{h}{d}\,(na + \sum_{i=1}^{n} l_{i-1}), \tag{1}$$

where h is thickness of the stainless steel sheets, d width of the space of neighboring sheets, a length of the slit in an x-ray beam direction and n number of the intervals. The whole length of the Soller slit assembly L_m is, if m Soller slits of the same type are used, given by

$$L_m = ma + \sum_{i=1}^{m} l_{i-1}. \tag{2}$$

Eq. (2) is a form similar to that found in the parenthesis in eq. (1). The parallel beams obtained by this assembly is geometrically equivalent to that obtained by a single Soller slit of length L_m and width d.

In order to arrange corresponding openings of each Soller slit of the assembly on a straight line, Soller slits of the same type are to be set on a mount with two flat plates to squeeze them from both sides. The angular divergence of the x-ray beams limited by the assembly is $\frac{2d}{L_m}$. This is the value for an ideal assembly. For actual assembly the angular divergence is smaller than the calculated value owing to the deviation from flat, parallel plane caused by the misalignment that cannot be avoided in construction. This misalignment has influence on both the resolving power and the intensity of the x-ray beams.

Each Soller slit was made by piling up many hardened steel sheets of commercial blades of 0.14 mm thickness spaced 0.10 mm. As spacers, commercial blades of 0.10 mm thickness were used. The sizes of the slits are in 22 mm length in the direction of x-ray beams and in 15 mm height. Three Soller slits were arranged according to the preceding formula, with calculated angular divergence of approximately 1.0×10^{-3} radian, in this case total length of the assembly is about 200 mm. This numerical value is a little smaller than the dispersion of MoK$_\alpha$ doublet by Si (220) plane. The value of this angular divergence is determined from the wavelength of x-rays utilized and the spacing of a reflecting plane of a sample, considering the dispersion of the K$_\alpha$ doublet. In practice, a topograph with high resolution can be obtained if the angular divergence does not exceed the dispersion of the K$_\alpha$ doublet.

Arrangement of the new technique is shown in Fig. 2. The line focus of an x-ray tube is used in a horizontal position; with a take-off angle 3~6°, the effective focus has dimensions of $10 \times 0.05 \sim 0.1$ mm^2. Topographs obtained by the Soller slit assembly fixed become a series of section ones made simulta-

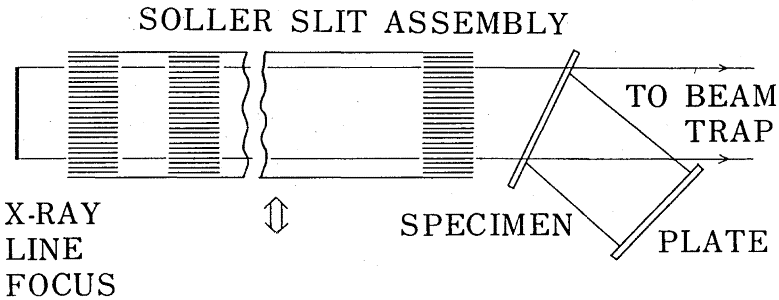

Fig. 2. Schematic diagram of the experimental arrangement (the 3S method).

neously of the same sample. In order to obtain a projection topograph, the Soller slit assembly should be scanned during exposure. Scanning range is very small, equal at the least to, and in general several times, the sum of the thickness of the slit material and the opening width. In principle, the scanning motion is to be performed on a micro-slider. But owing to small range of scanning motion, topographs can be obtained also by oscillation in a double pendulum system etc[9]. In such a manner this new technique is very simple in instrumental mechanism.

The geometrical resolving power of this technique may be considered as good as in the Lang method, for the perfect separation of the K_α doublet is achieved. The detectable minimum distance[10] in crystal, are given by

$$d_P \fallingdotseq \frac{m}{k} f_w, \tag{3}$$

$$d_h \fallingdotseq \frac{m \cdot \Delta\theta}{\cos(\alpha+\theta)}, \tag{4}$$

where d_P and d_h are geometrical resolving power in the direction perpendicular to the incident plane and in the direction of the incident plane respectively, k is the distance from the x-ray source to the sample, f_w the effective size of the x-ray source in the perpendicular direction determined by the viewing angle, $\Delta\theta$ spectrum width of the $K_{\alpha 1}$ characteristic x-rays, θ Bragg angle, α the angle between the reflection plane and the normal to the crystal surface. m is the distance measured along diffracted beam from the sample to the plate, and if w is the width of total x-ray beams which irradiate the sample, the minimum of permissible length of m is given by

$$m_{min} = w \operatorname{cosec} 2\theta. \tag{5}$$

3. Result and Discussion

Some experiments have been performed to show usefulness of the new technique. This technique will be called "3S method" after the Scanning Soller Slit assembly method.

At first, a rocking curve from Si ($\bar{2}20$) with AgK_α by means of the 3S method is shown in Fig. 3. As can be seen from this result, the separation of the doublet is very good and dispersion of the doublet is about 1.3×10^{-3} radian and half width of $K_{\alpha1}$ is about 0.9×10^{-3} radian. The estimated value of the angular divergence in the horizontal direction of x-ray beams passing through the Soller slit assembly was 1.0×10^{-3} radian. But, the actual value was found about 0.8×10^{-3} radian of which value is 20% smaller than calculated one by measuring the extent of the image of section topograph of silicon single crystal and by measuring the image of opening of the Soller slits by the film placed at 500 mm apart from the exit of the assembly. The K_α doublet is perfectly separated in the 3S method in the horizontal direction, the resolving power of the direction, d_h, is determined by eq.(4). That is the same as the Lang's one. Next, we took projection topographs from the same crystal by means of both the 3S method and the Lang method, and then compared them. Photo. 1 (a) and (b) show the projection topographs gained by the 3S method and the Lang method respectively from silicon single crystal using ($\bar{2}20$) reflection with $MoK_{\alpha1}$. In this case the surface of the silicon from which x-ray beams exit is defined as (111).

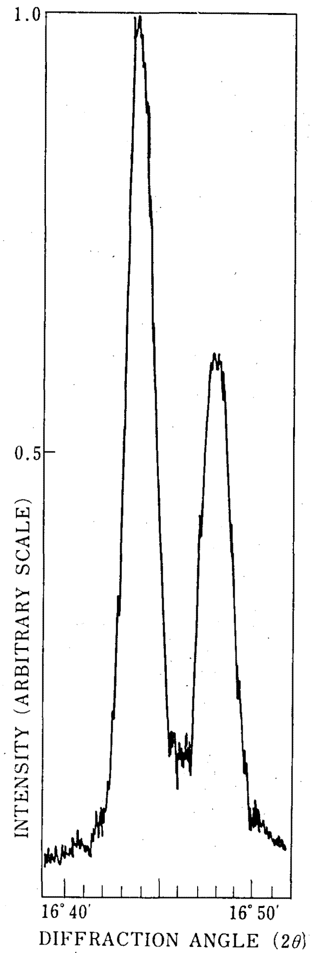

Fig. 3. The rocking curve from Si ($\bar{2}20$) with AgK_α by means of the 3S method.

The width of dislocation image is similar for both methods. In the topographs taken by the 3S method, however, a stripe pattern is seen. It is perhaps caused from scanning distance of the slit assembly. This phenomenon would be vanished by the improvement of the scanning mechanism. However, observation of dislocation images is not disturbed so much as it is. We can observe individual dislocation by both methods with the same order in the area where dislocation density is comparatively high (marked A).

Photo. 2 (a) and (b) are enlarged topographs of a part of Photo. 1, (c) and (d) are topographs of the same region taken by the 3S method using other diffraction vectors. In these topographs we can compare with individual dislocation image by both methods in more detail and it is found the 3S method can be used to take topographs for various diffraction vectors. From observations of the images of a half loop dislocation generated from Frank-Reed source and a straight dislocation, the dislocation image for perpendicular to diffraction vector, $\boldsymbol{g}$, that is, the resolving power of d_P, is a slightly inferior to the Lang case in case of the 3S method. Here, we can estimate d_P after eq. (3) using our experimental value, that is, m is 30 mm which is a little larger than m_{min}, f_w 100μ in case of take-off angle 6° and k 400 mm, then d_P is estimated 7.5μ. Since d_P of the Lang method is 1.3μ using $f_w=40\mu$, $m=300$ mm and $m=10$ mm, the 3S method is inferior to the Lang method in geometrical resolving power, too. But the width of a dislocation image can actually be observed about 8μ and recently in the Lang method a sealed off x-ray tube of which focus size is 300μ has been used for reducing the exposure time. Therefore, the inferiority of d_P in the 3S method is not a fatal demerit of the projection topography. Numerical value of d_h is smaller than 1μ for both methods. This is sufficiently good. Concerning the detailed contrast of a dislocation image, the white image can be seen at the side of the intense dislocation image (marked B)[11] and the change of the dislocation image for various diffraction vectors can be observed in topographs taken by the 3S method. Next projection topographs were taken and compared with them for a LiF single crystal by means of both the 3S method and the Lang method. $CuK_{\alpha1}$ is used because of low linear absorption coefficient for LiF. In this case, as the Bragg angle is larger than one using $MoK_{\alpha1}$, m_{min} of eq. (5) becomes smaller and the distance from the sample to the plate becomes shorter. Therefore, the resolving power for the horizontal direction becomes good. Experimental conditions in this case are following; m is 20 mm, f_w 50μ (take-off angle 2°) and k 400 mm, then d_P becomes 2.5μ.

Photo. 3 is the projection topograph of LiF gained by the 3S method under the conditions mentioned above, with (020) reflection. Resolving of images of an individual dislocation and other defects near the suface of the sample can be seen with good separation as the same order of the Lang's one. The sub-grain boundary (marked C) in the topograph can be seen clearly. Since the tilt angle of this sub-grain boundary is about twenty sec. of arc as reported by one of the authors[12], the topograph ought to be distinguished by the sub-grain boundary if the x-ray beams with sufficient parallel are used. This point suggests that the 3S method has good resolving power in the horizontal direction.

From these experiments mentioned above, it can be seen that the 3S method has good resolution for dislocation images. Then, as an example of applications and of possibility of attachments, the 3S method is applied to direct observation during compressive deformation of single crystals. LiF single crystals were used for the specimens which were made by Harshaw Chemical Company, dislocation density $4\times10^4/cm^2$ and size $5\times5\times1$ mm. $MoK_{\alpha1}$ radiations were used under the operating condition of 40 KV and 10 mA. The (020) plane was used as the reflection plane. In this case the exit surface of x-rays is defined as (100).

The LiF single crystal is compressed in the direction of [001] at room temperature by the compressive device. Photo. 4 (a), (b), (c) and (d) are the topographs taken by the 3S method of which compressive strains are 0, 2, 3.4 and 4.2% respectively. These strain values were determined by the turnover angle of the screw of the device, then the absolute values are not exact. During a series of the experiments of compression, the sample need not be taken off from the sample holder, because the compressive device was set up on the mount.

From the series of topographs under various strains, it is seen that the grown-in dislocations do not move under compression at room temperature. At 3.4% strain, many bands occur as seen in (c) and they are growing to $\langle 110 \rangle$ direction with increasing compression. At 4.2% strain many dislocation loops developed in the direction of [100] as seen in (d) (marked D). Any change of them can not be seen with increasing compression over 4.2% strain.

Through the above examples, it would be shown that the 3S method is useful in the direct observation of the lattice defects together with the Lang method and other techniques.

4. Conclusion

A new method of x-ray diffraction topography was developed. It has the merits of both the Lang method and the Soller slit method. Characteristics of this new technique are as follows.

(1) Resolution of dislocation images is as good as in the Lang method. The width of dislocation images is about 8μ.

(2) The instrumental mechanism and adjustment are very simple and the Soller slit assembly can be manufactured easily in a laboratory.

(3) A line focus of an analyzing x-ray tube of an ordinary x-ray generating apparatus can be used.

(4) Exposure time is short, and topography of wide range of a sample can be made at one time.

(5) A sample and a plate being fixed to obtain a projection topograph during exposure, use of various attachments is facilitated, so that a topograph of a stressed or heated sample is possible.

(6) A series of section topographs can be obtained in order to study a sample of high dislocation density or strains in a sample.

(7) It is possible that topographs of more than two samples can be made at the same time. This mechanism would be reported elsewhere.

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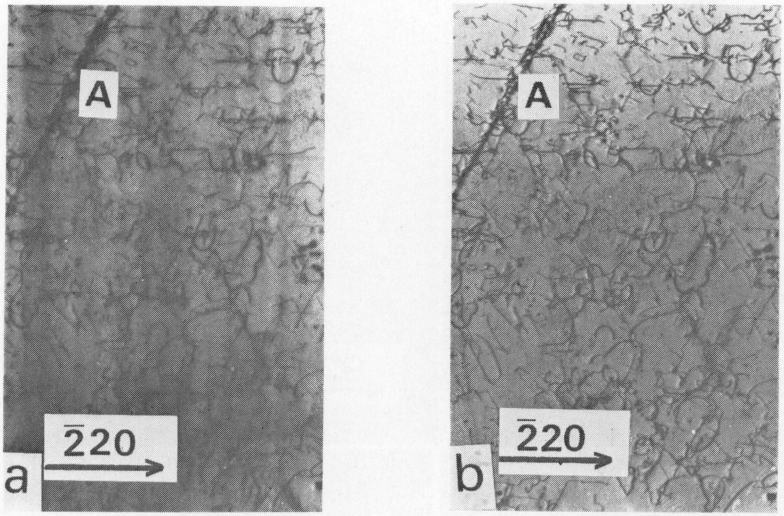

Photo. 1. Projection topographs of Si single crystal using ($\bar{2}20$) reflection with $MoK_{\alpha 1}$, $\mu t \sim 0.6$. $\times 10$
(a) Topograph by the 3S method. (b) Topograph by the Lang method. Exposure time was required 3 hours for the 3S method operated at 40 KV, 10 mA and 2 hours per 1 mm sweep for the Lang Method operated at 45 KV, 0.6 mA using Sakura nuclear plate E1 (emulsion thickness 50μ). In case of the 3S method, topographs up to 10×10 mm area of the sample can be taken by exposure of 3 hours but in the Lang method it takes about 20 hours.

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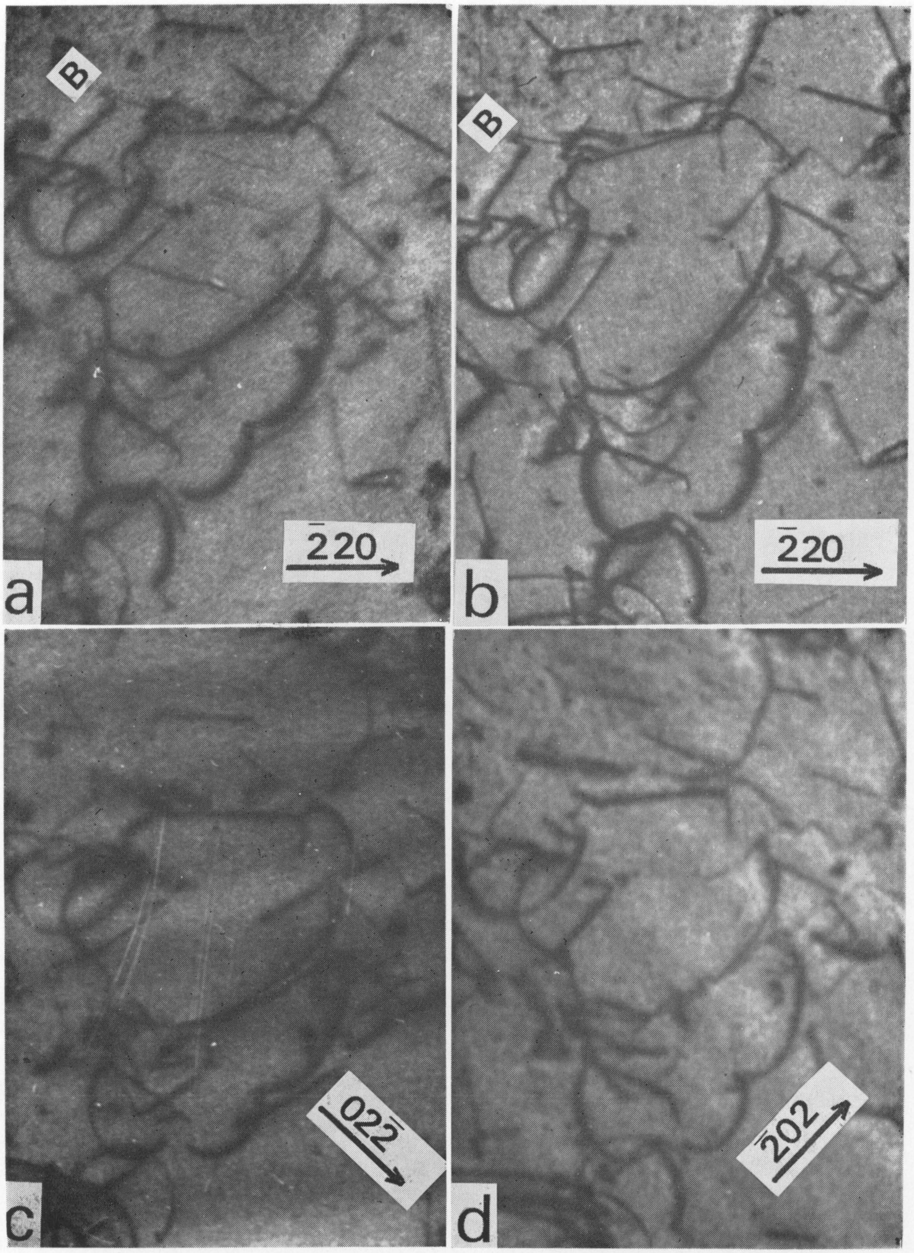

Photo. 2. Projection topographs enlarged of a part of Photo. 1. and those taken by using various diffraction vector. ×50
(a) (c) (d) are topographs by the 3S and (b) by the Lang method.

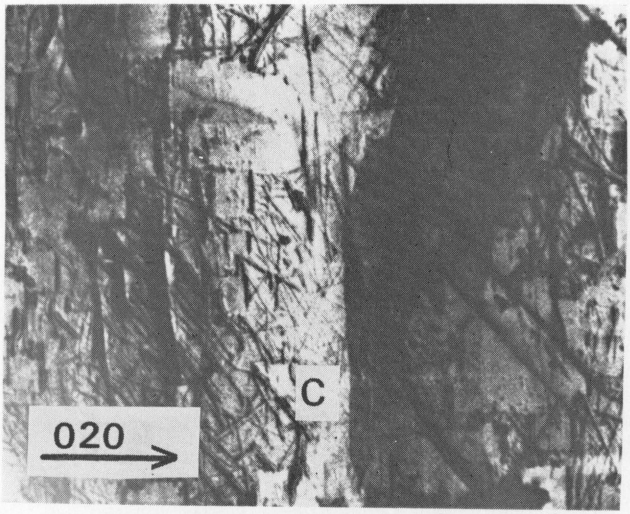

Photo. 3. Projection topographs taken by the 3S method of a LiF single crystal using (020) reflection with $CuK_{\alpha 1}$, $\mu t \sim 1.5$. $\times 30$
Exposure time was required 5 hours, using Sakura nuclear plate El (emulsion thickness 50μ).

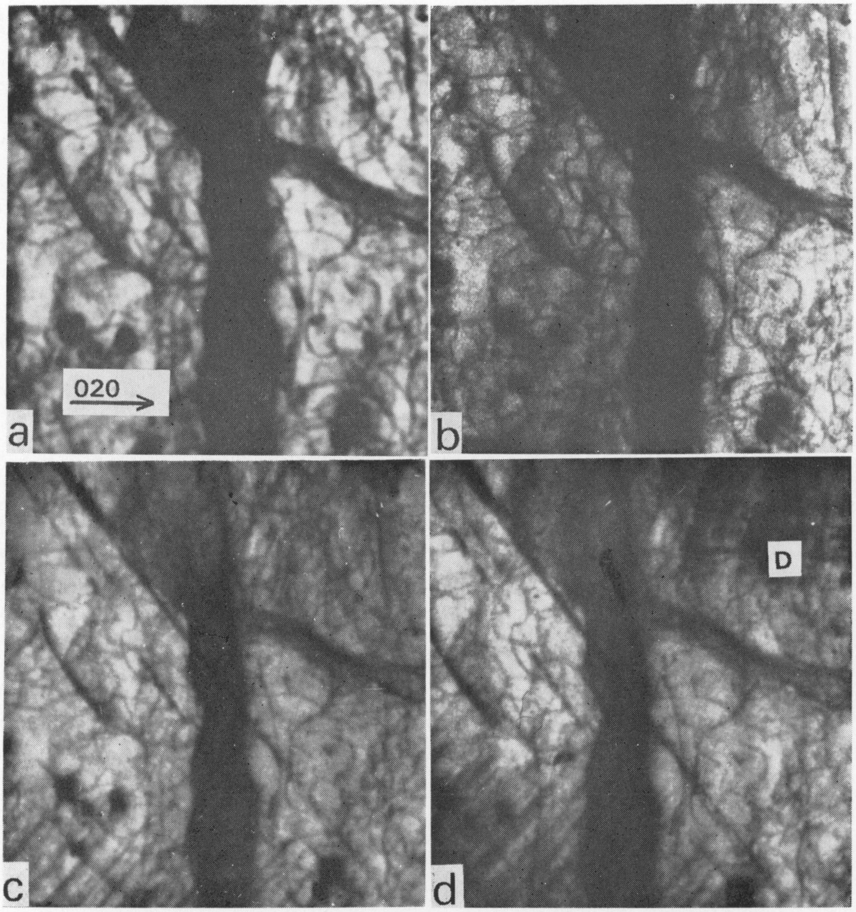

Photo. 4. Projection topographs taken by the 3S method for internal changes in the deformed LiF single crystal by compression. ×33
(a) before compression, (b) strain 2%, (c) strain 3.4%, (d) strain 4.2%.
Compression was applied at the top and bottom on the photograph.