On the Arrangement of Micro-crystals in Aluminium Wire.

By

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(Received May 14, 1925)

ABSTRACT.

In the present experiment the arrangement of the micro-crystals in the drawn wires of Al and Cu was examined by means of X-ray patterns. And it was ascertained that the X-ray patterns were explainable by the consideration that the trigonal axes of the crystals are arranged nearly parallel to the axis of the wire, leaving the orientation in other respects rather at random. In the case of Al wire more detailed examination was made, and it was shown that the trigonal axes of the crystals were not arranged exactly parallel to the axis of the wire, and that the crystals, whose trigonal axes are inclined at about 7° to the axis of the wire, are most predominant.

Introduction.

Research into the arrangement of the micro-crystals of metals by means of X-rays has made rapid progress recently. In our laboratory Assistant Professor S. Tanaka[1] has succeeded in determining the arrangement of micro-crystals in a rolled platinum foil. In 1922 Professor A. Ono[2] looked into the question of copper wire, and suggested that the trigonal axes of the cubic crystals of copper were arranged parallel to the axis of the wire. At that time the specimen tested was cut out, in most cases, in the form of a thin plate of about 0·2 mms. in thickness from the wire, and the specimen thus prepared was tested by the X-rays from a Coolidge-tube with a tungsten anticathode.

1 S. Tanaka: These Memoirs, **8** (1925).
2 A. Ono: Mem.Coll. of Eng. Kyushu, **2** (1922).

The Experimantal Method.

(1) The Specimen employed.

In the present investigation the writer examined, in most cases, the cylindrical wires of Al itself, the diameter of which varied from 0·6 mms. to 1·2 mms. To prepare the Al. wire, the writer made three cast cylindrical bars of 7—8 mms. in diameter at first, and then these bars were drawn ca. 50 times in the same direction through circular dies of steel till the diameter of the wires became 1—0.6 mms. The velocity of drawing was about 25 meters per minute. During the process of drawing the sample was annealed twice at 550° C. for 6 hours when the wires were still fairly thick. In order to show the dies employed the two dies, No. 1 and No. 35, are indicated graphically in Fig. 1 as examples. They are not similar apparently, but the shape of the whole length of the hole of No. 1 corresponds and is similar to the part AB of No. 35. And the other dies not represented here were of shapes similar to Nos. 1 and 35 in the manner mentioned above.

The Form of Dies.

No. 1 No. 35

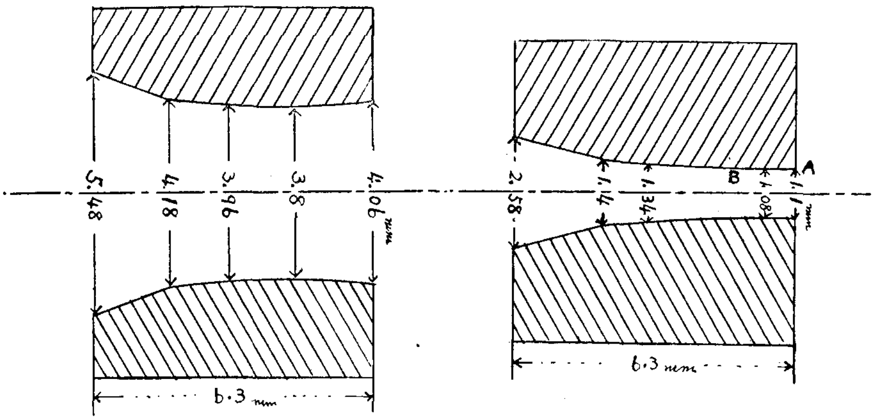


Fig. 1.

(2) Outline of the Method.

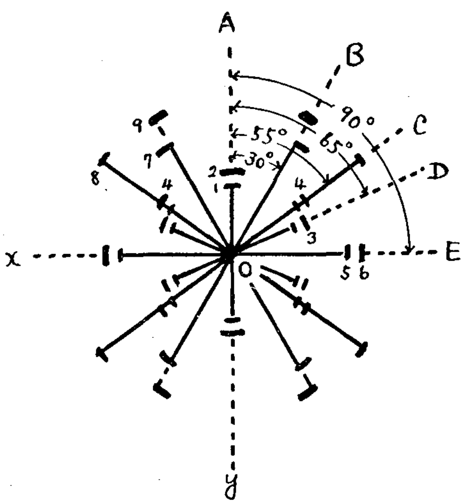
The method was essentially the same as that employed by Assist. Prof. S. Tanaka in the case of the rolled Pt. foil. The slit was a circular hole of about 1 mm. in diameter and the photographic plate was generally placed perpendicularly to the beam of the X-rays at a distance of 2·7—3 cms. from the specimen.

A Coolidge tube of molybdenum anticathode was excited by a transfromer, the potential applied being about 70 K. V., and the current was from 3 to 4 milliampères. The X-ray pattern was taken in the following three positions of the specimen. At first, the axis of the wire was set perpendicularly to the beam of the X-rays, and next it was made parallel to the X-ray, and at last the axis was made to incline at an angle of 45° to the beam. The duration of the exposure of the photographic plates varied from 10 to 16 hours, according to the diameter of the wires. The photographs thus taken are shown in the figures reproduced in Plates I and II.

Discussion of the Results.

From the photographs reproduced in Figs. 4, 5 of Plate I we can see that each pattern consists of concentric rings about the central spot impressed by the direct beam of the X-rays, and some bands radiating outward from the central spot. The sketch of the bands distinguished among these are shown in the next figure, Fig. 2. In this case the axis of the wire was set perpendicularly to the beam of the X-rays. The line OA in this figure is the band radiating in the direction parallel to the axis of the wire from the central spot, and OB, OC, OD and OE represent the successive bands arranged symmetrically about the vertical and horizontal lines OA and OE.

Fig. 2.



The presence of the concentric rings of strong intensity, as may be seen in the photographs, shows us that some of the micro-crystals are arranged at random about the direction parallel to the incident X-rays, and that the intensity of the α and β lines of the K series of molybdenum is much stronger than that of the continuous rays. If a definite axis of every micro-crystal in the metal be arranged in a common direction and their orientation be at random in other respects, the effect will be the same as if the same single crystal were rotated about the same axis. This will cause a radiating band of the X-ray spectrum on the photographic plate. With continuous X-rays the bands will become continuous, and if the rays contain the characteristic rays of strong intensity too, the radiating band will contain their spectrum of strong intensity on the back ground of the continuous spectrum. Therefore the presence of these bands suggests that some part, at least, of the micro-crystals in the metal are arranged with some regularity. The fact that the bands distributed rather irregularly appeared in the case of the cast Al specimen of about 7 mms. in diameter indicates the presence of some regularity in the arrangement of the micro-crystals even at the beginning of the process of drawing. But as the process of drawing advances new bands which are distributed regularly appear and develop in place of the initial ones, and at last when the wire is drawn down to about 3 mms. or more, we can not detect any trace of the initial bands. Moreover, as the positions and the arrangements of these new bands do not suffer any change by the still continued process of drawing, we may conclude that the arrangement of the micro-crystals in the wire is independent from the initial state when the wire is drawn to 3 mms. or more in the present case.

At first we must determine the indices of each atomic plane of the crystal, by whose reflection each radiating band seems to be caused. When a radiating band intersects the concentric rings the intensity is, of course, stronger at that intersection. But if we consider on one band the intensity is not the same at all its intersections with the rings. From the photographs we can readily recognize that some intersections are especially predominated by their intensities compared with the others. This predominant intersection is taken, in the following, as the position of the K_α or K_β line in the spectrum of molybdenum caused by the reflection from the atomic plane of the crystal which is responsible for producing the corresponding band. The spots of intersection of the bands and the rings thus selected are shown in Fig. 5 of plate I as examples, and they are marked by the numbers from 1 to 9 in that photograph. At first the writer determined the atomic planes from which the spots and rings are reflected. The method now employed is nothing but that used in the powder method; and the concentric rings around the central spots in the photograph may be considered as the spectrum of the K_α & K_β lines of molybdenum which are reflected from the atomic planes of various grating constants. If we denote the value of a grating constant by d, the wave length of K_α & K_β of Mo. by λ_α & λ_β respectively, the distance of the photographic plate from the sample by a and the diameter of a ring by $2\ r$, then we have the next relation between them.

$$n\lambda = 2d \sin\theta \quad \cdots\cdots\cdots \quad (1),$$

$$\tan 2\theta = \frac{r}{a} \quad \cdots\cdots\cdots \quad (2).$$

Where n is the number representing the order of the spectrum and $\theta = \frac{\pi}{2} - \gamma$ is the glancing angle. By measuring the values of r and a the value of θ corresponding to a ring will be obtained from equation (2), and with this value of θ the value of d corresponding to that ring will be found from equation (1) by assigning to λ the value of the wave length of the K_α or K_β line of molybdenum. The values of d corresponding to the various rings or to the various spots thus calculated are given in Table. 1.

Table. 1. $a = 30$ mms.

No. of spots	$2r$ (obs.) in mm.	θ (Calc.) in degree	Grating constant d in 10^{-8}cm.		Hull's data[1] d in 10^{-8}cm.	Hull's data[1] Indices of planes	Indices of the reflecting planes
1	16.4	7°40′	$d\alpha$	2.67			
			$d\beta$	2.39	2.34	{111}	$\{111\}_\beta$
2 & 3	19.0	8°30′	$d\alpha$	2.38	2.34	{111}	$\{111\}_\alpha$ or
			$d\beta$	2.12	2.03	{100}	$\{100\}_\beta$
4	22.2	10°10′	$d\alpha$	2.01	2.03	{100}	$\{100\}_\alpha$
			$d\beta$	1.78			
5	28.6	12°40′	$d\alpha$	1.61			
			$d\beta$	1.44	1.43	{110}	$\{110\}_\beta$
6	33.3	14°30′	$d\alpha$	1.41	1.43	{110}	$\{110\}_\alpha$ or
			$d\beta$	1.26	1.22	{311}	$\{311\}_\beta$
7	34.2	14°45′	$d\alpha$	1.39	1.43	{110}	$\{110\}_\alpha$ or
			$d\beta$	1.24	1.22	{311}	$\{311\}_\beta$
8 & 9	41.2	17°14′	$d\alpha$	1.20	1.22 or 1.17	{311} or {111}[2]	$\{311\}_\alpha$ or [2] $\{111\}_\alpha$
			$d\beta$	1.06			

In this table the spots 1, 2, 3, etc., refer respectively to those marked

1 A. W. Hull: Phys. Rev., **10**, 685 (1917); **17**, 578 (1921).

by the same figures in Fig. 5 of Plate I, or in Fig. 2; of these two figures the latter is nothing but the sketch of the former. The symbols d_α and d_β in the fourth column represent the values of d obtained by assuming that the spot denoted by the number in the same horizontal row is due to the K_α or K_β line of molybdemun. For the value of the wave length of the K_α line we have taken $\lambda_\alpha = 0{\cdot}709 \times 10^{-8}$ cm. which is the mean value of the α' and α lines of the K-series, and for that of K_β we have taken $\lambda_\beta = 0{\cdot}631 \times 10^{-8}$ cm[1]. The symbol $\{111\}^{(2)}$ in the 7th column of this table denotes that the spot is the second order spectrum caused by the crystal face $\{111\}$. In the 6th and the 7th column of Table 1, the values of d and the corresponding indices of the reflecting atomic planes obtained by Hull are represented. With some of the spots only one of the values of d_α and d_β coincides with any one of those given by Hull. But with some spots both values of d_α and d_β agree very well with the two different values of d obtained by the same author. This indicates that the same spot is caused by the superposition of the K_α and K_β lines reflected by two different atomic planes. Thus it will easily be seen from the table that the values of d obtained in the present experiment are in fair agreement with those obtained by Hull from his powder method. Moreover, it is readily recognizable, from the photograph, that the intensities of the rings and the spots are in reasonable order. Thus considering it does not seem to be unnatural to assign the indices of the reflecting atomic planes given in the last column of the table to the corresponding spots marked by the numbers given in the first column of that table. The symbols $\{111\}_\alpha$, $\{110\}_\beta$, etc., in the last column show that the corresponding spot is the spectrum of the K_α or K_β lines caused by the reflection from the atomic plane represented by the index given in the bracket.

Next, the writer determined the index of the reflecting atomic plane, which caused a radiating band, from the index of the reflecting atomic plane of the spot at the intersection of that band with the rings. Of course, there is the case where one band intersects with many rings. As to the choice of the spots which are originated from the same reflecting atomic plane as that of the corresponding band, in such a case, the spots of predominant intensity were correlated to the corresponding band. This fact has been already described. The indices of the reflecting atomic planes and the corresponding band thus determined are shown in Table 2.

1 W. Duane and Kang-Fuh-Hu: Phys. Rev., **9**, 489 (1918); **14**, 369 (1919)

Table. 2.

No. of the spots	Radiating band	Indices of the reflecting atomic planes	α (observed)
1 & 2	OA	{111}	0°
7	OB	{311} and {110}	30°
4	OC	{100}	55°
3	OD	{111}	65°
5 & 6	OE	{110}	90°

In this table, OA, OB, OC, OD, and OE are the bands represented in Fig. 2, and the angles represented by α are those between these bands and the band OA.

The spots and the bands are arranged symmetrically about the vertical central line in the figures reproduced in Figs. 4 and 5, of Plate I, and also about the horizontal central line in these figures. Here it must be noted that the axis of the wire was made parallel to the vertical line in the case of these photographs.

Next, we shall consider the photographs obtained by setting the axis of the wire parallel to the X-rays. When the X-ray strikes all parts of the section of the wire, only fine concentric rings appear and we can

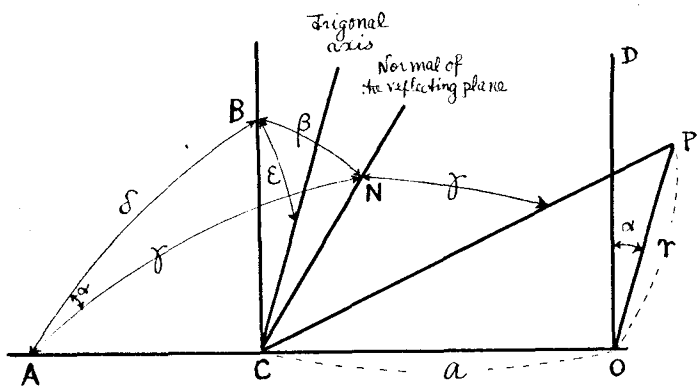


Fig. 3.

not detect any bands radiating outward from the central spot in the photographs as may be seen from Fig. 6 of Plate I. From this fact it may be inferred that the orientation of the micro-crystals is symmetrical about the axis of the wire. And it does not seem to be unnatural to suppose that a certain axis of the micro-crystals in the wire is arranged parallel to the axis of the wire.

In the annexed figure, Fig. 3, AC is the direction of the incident X-ray, CN is the normal to a reflecting atomic plane, the plane POD is the photographic plate which is set normal to the incident X-rays and P is the position of the reflected rays on the photographic plate. Let CB be the direction of the axis of the wire, then this will be the common axis of the micro-crystals in the wire. Now in the spherical triangle ABN of Fig. 3 we have the following relation.

$$\cos\beta = \cos\gamma\cos\delta + \sin\gamma\sin\delta\cos\alpha \quad \cdots\cdots\cdots\cdots\cdots\cdots \quad (3).$$

When the axis of the wire is perpendicular to the beam of the X-rays, that is when $\delta = \frac{\pi}{2}$ we get

$$\cos\beta = \sin\gamma\cos\alpha \quad \cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots \quad (4).$$

Since γ is known for the X-ray of known wave length and for a definite reflecting atomic plane, we can calculate the values of β from the observed values of α given in Table 2. Thus we can determine the inclinations of the reflecting atomic planes to the axis of the wire.

Now, let the three edges of a unit cube of the crystal be the axes of the rectangular coordinates, the direction cosines of the axis of the arrangement of micro-crystals be l, m, n and those of the normal to a reflecting atomic plane be l′, m′, n′. These are shown graphically in Fig. 4.

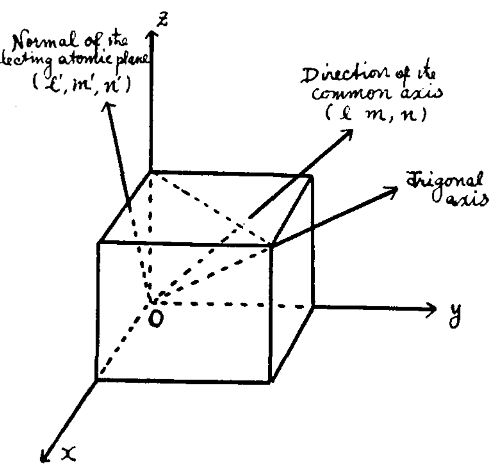


Fig. 4.

Then we get the next relation from equation (4)

$$\cos\beta = \sin\gamma \cos\alpha = ll' + mm' + nn' \quad \cdots\cdots\cdots \quad (5).$$

In the above equation, l′, m′, n′ and γ are known for any reflecting atomic plane, and for the K_α or K_β line of molybdenum, therefore, if we give the values of α observed for various reflecting atomic planes in that equation, we shall obtain the linear relations between l, m and n corresponding to every reflecting atomic plane. By solving the normal equations obtained from these linear relations the writer obtained the values of l, m and n given in Table 3.

Table 3.

l	m	n	ε
0·511	0·535	0·682	7°22′

In this table, ε is the angle between the trigonal axis and the calculated common axis of the micro-crystals in the wire, and it was calculated by the following equation

$$\cos\varepsilon = \frac{1}{\sqrt{3}}(l + m + n).$$

By taking various different sets of any 3 linear relations above mentioned the writer calculated also the values of l, m, n and ε, and it was ascertained that the values thus obtained are in fair agreement with those given in Table 3 within the limit of experimental errors. This shows that the distribution of the spots in the photograph can be explained by means of our previous supposition. The fact that the values of l and m are nearly equal indicates that the common axes of the micro-crystals arranged in the wire passes through the diagonal axis in the upper surface of the elementary cube represented in Fig. 4. As the angle ε is very small, as shown in Table 3, the common axes of the micro-crystals in the wire are nearly parallel to the trigonal axis of the cube.

Next the writer calculated the angles between the normals to various reflecting planes and the common axis of the micro-crystals, whose orientation with respect to the elementary cube has just been described above. The results are given in Table 4.

Next, the writer plotted the locus of the point P in Fig. 3 for various values of the wave lengths of the X-rays. From equations (2) and (3) we have:

$$r = a \tan 2\theta \quad \cdots\cdots\cdots \quad (2),$$

$$\cos\alpha = \frac{\cos\beta - \cos\gamma\cos\delta}{\sin\gamma\ \sin\delta} = \frac{\cos\beta - \sin\theta\cos\delta}{\cos\theta\ \sin\delta} \quad \cdots\cdots\cdots (3)'.$$

Table. 4.

Indices of the reflecting atomic planes	β
$\{111\}$	7° 22′
$\{11\bar{1}\}$	77° 52′
$\{1\bar{1}1\}$	69° 50′
$\{\bar{1}11\}$	66° 6′
$\{110\}$	42° 40′
$\{101\}$	32° 48′
$\{011\}$	31° 12′
$\{1\bar{1}0\}$	89°
$\{\bar{1}01\}$	83° 6′
$\{0\bar{1}1\}$	84° 20′
$\{100\}$	59° 28′
$\{010\}$	57° 53′
$\{001\}$	47° 19′
$\{113\}$	22° 5′
$\{311\}$	34° 29′
$\{131\}$	32° 52′
$\{1\bar{1}3\}$	52° 41′
$\{31\bar{1}\}$	65° 27′
$\{3\bar{1}1\}$	59° 46′
$\{\bar{1}13\}$	51° 39′
$\{\bar{1}31\}$	57° 50′
$\{13\bar{1}\}$	64° 29′
$\{\bar{1}\bar{1}3\}$	72° 34′
$\{3\bar{1}\bar{1}\}$	84° 34′
$\{\bar{1}3\bar{1}\}$	82° 54′

To find the locus we have only to find the relation between α and r for a definite reflecting atomic plane. If the value of the wave length of the X-rays and the indices of the reflecting atomic plane are given, the values of θ and β will be obtained immediately. With these values of θ & β, the values of r and α will be calculated from equations (2) & (3)′, if the values of a & δ are given. For the values of β those given in Table 4 were used, and for the value of a the actual distance between the specimen and the photographic plate was taken so that the curve thus plotted might just fit the photographic pattern. The curves represented in Fig. 5 are those applicable when $\delta = 90°$, that is, when the axis of the wire is set perpendicular to the incident rays, and the curves represented in Fig. 6 are those applicable when $\delta = 45°$. The spots and the shaded parts in these figures represent the distribution of the spots and the radiating bands in the photographs. In the case of Fig. 5 they are copied from Fig. 4, Plate I, and in the case of Fig. 6 they are copied from Fig. 1, Plate II. The dotted lines in these curves indicate that the expected bands were not actually observed. Though the reason for the absence of these bands is not clear, yet we may say that the agreement is rather astisfactory if we consider that the bands and the spots of very weak intensity will not appear clearly on the photograph.

In a preceding paragraph it was stated that the trigonal axes of the crystals were arranged nearly, but not exactly, parallel to the axis of the wire. If that be so, the normal of the atomic plane $\{\bar{2}11\}$, which is perpendicular to the trigonal axis, must be nearly perpendicular to the axis of the wire.

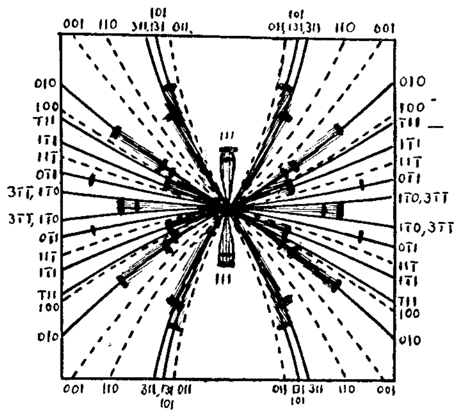


Fig. 5.

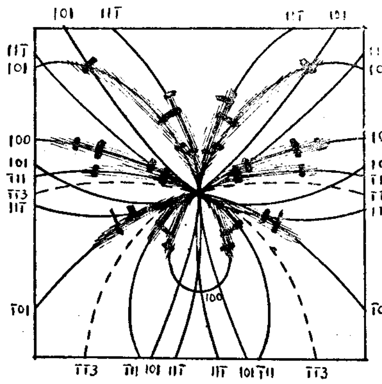


Fig. 6.

When the pencil of the X-ray covers all parts of the cross section of the wire, the photographic pattern will be the total effect from all parts of the cross section, and it will be indifferent to the orientation of the crystals situated at some definite part in the cross section. But when the beam of the X-ray strikes only one part of the cross section of the wire only the orientation of the normal to the plane $\{\bar{2}11\}$ of the crystals situated at that point will affect the nature of the pattern. In regard to this question, some experiments have already been made. Though a decisive conclusion has not yet been obtained, the experiment described below seems to have some connection with this question.

Next the wri er cut a wire, prepared by drawing in one direction only, into two pieces by sawing parallel to the central axis of the wire as is shown in Fig. 7 and Fig. 8.

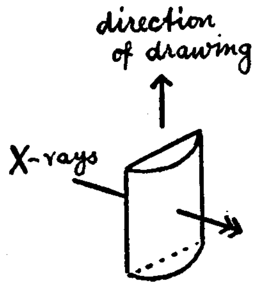


Fig. 7.

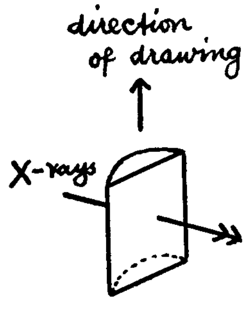


Fig. 8.

In these cases the direction of drawing is indicated by the arrows, and the X-rays were made to strike the specimens in the directions perpendicular to the axis of the wire and also perpendicular to the planes developed by cutting. In Fig. 7 the X-ray passes through the specimen from the side of the plane to the side of the curved natural surface, and in Fig. 8 the ray proceeds in the opposite direction as indicated by the arrows in the figures. The photograph reproduced in Fig. 2 of Plate II is that taken in the case of Fig. 7, and Fig. 3 of Plate II is that taken in the case of Fig. 8.

It was ascertained that the photographs thus obtained were both equal to the photograph obtained with a whole cylindrical wire, with the exception of the difference in the intensities of the bands radiating upward and downward parallel to the axis of the wire. It has already been explained that these bands are caused by the reflection from the face {111} of the crystal. In the case of Fig. 7 the band radiating downward from the central spot is much stronger in intensity than that radiating upward. In the case of Fig. 8 this is reversed. These differences of intensities may, of course, be due to the experimental arrangement; and in order to ascertain this point the writer rotated the direction of the specimen by 180° about the beam of the X-rays, keeping the other conditions entirely the same as before. The photograph thus taken showed that the order of intensities of the bands under consideration was also reversed. On these grounds it does not seem to be unreasonable to suggest that the differences of the intensities of the bands under consideration are caused by the manner in which the micro-crystals in the wire are arranged.

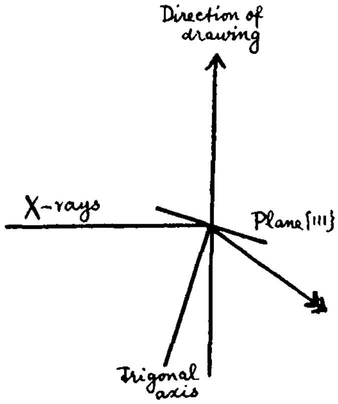


Fig. 9.

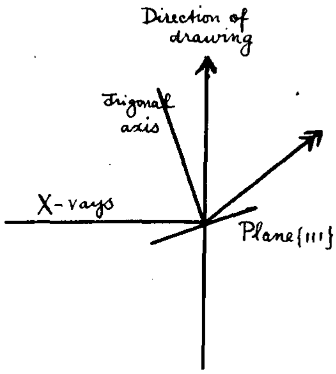


Fig. 10.

It has already been stated that most of the trigonal axes of the crystals, that is, most of the normals to the atomic plane {111} of the crystals, are not arranged exactly parallel to the axis of the wire, but are inclined at some small angle to the axis of the wire. This fact is graphically represented in Figs. 9 and 10.

If most of the trigonal axes of the crystals are arranged in the direction represented in Fig. 9, the band radiating downward will be stronger than that radiating in the reverse direction. This corresponds to the photograph taken under the conditions represented in Fig. 7. Similarly, the difference in the intensities of the two bands under consideration obtained under the conditions in Fig. 8 will be imagined from Fig. 10. Judging from these facts, it seems probable that the trigonal axes of the micro-crystals arranged in the manner represented in Fig. 11, are most predominant.

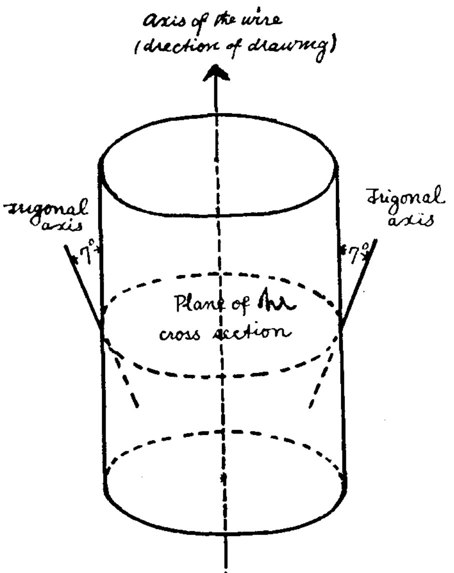


Fig. 11.

As was stated before, most of the trigonal axes of the crystals are arranged nearly parallel to the direction of the axis of the wire, that is, in the direction of drawing. But this is not exactly so, and the mean inclination of the trigonal axes of the crystals to the axis of the wire was found to be about 7°. Thus, though the trigonal axes are arranged in the manner represented in Fig. 11, the angle of inclination must be very small.

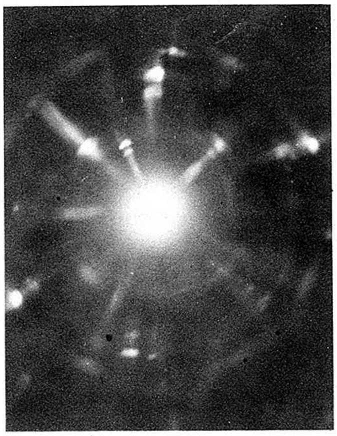
Here it must be noted that the writer has examined the effect of sawing the sample, and it was ascertained that the effect of sawing had no influence on the X-ray pattern. At first the photograph was taken with the cylindrical wire before it was cut into the two pieces of the forms represented in Fig. 7 and Fig. 8. Next, a photograph was prepared by superposing the photographs obtained with each piece. And, lastly, the two pieces of the specimen were pasted together into the original form, and a photograph was taken of this specimen. Comparing these three photographs it was ascertained that they were exactly the same. Thus it may be inferred that the effect of sawing had no effect on the X-ray pattern so far as the present experiment was concerned.

Next, the writer made a similar experiment with thin copper wires of 0·08–0·12 mms. in diameter. The photograph reproduced, as an example, in Fig. 4 of Plate II, was taken with the X-rays striking perpendicularly to the axis of the wire. Though the photograph was not very clear, it was ascertained that the distribution of the bands and the spots differed in no wise from the disposition of the bands and spots in the case of aluminium wire. So it may be inferred that the micro-crystals are arranged in the same way in a drawn wire of copper as in an aluminium wire. This result would, of course, be expected from the beginning as the crystals of these two metals belong to the same form of face-centered cubic lattice.

In conclusion, the writer's sincere thanks are offered to Prof. U. Yoshida and Assist. Prof. S. Tanaka for their kind guidance in the research. The writer wishes also to express his hearty thanks to Professor T. Mizuno for the interest he has taken in the research; and to Mr. Yamamoto of the Sumitomo Electric Wire Manufacturing Co. for his grateful gift to the writer of the thin copper wires used in the experiment.

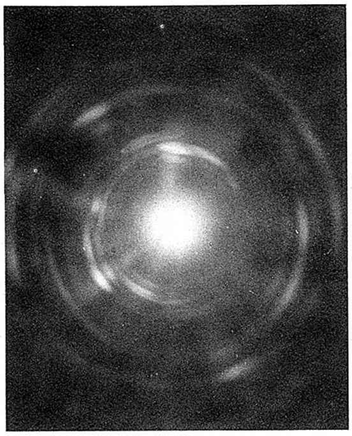
Plate I

Fig. 1



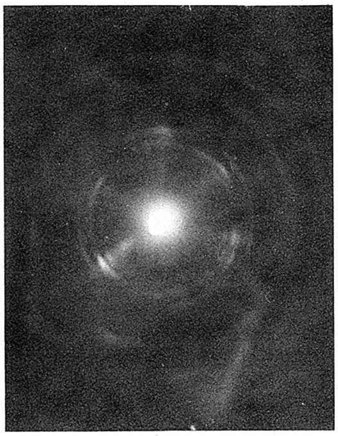
Cast Al bar.

Fig. 2



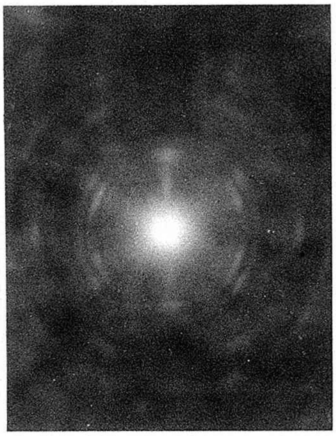
drawn 7 times, diameter 5 mms.

Fig. 3



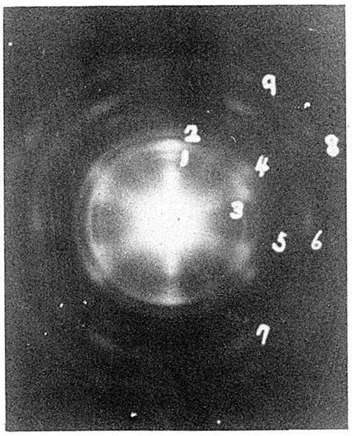
drawn 10 times,
diameter 4·3 mms.

Fig. 4



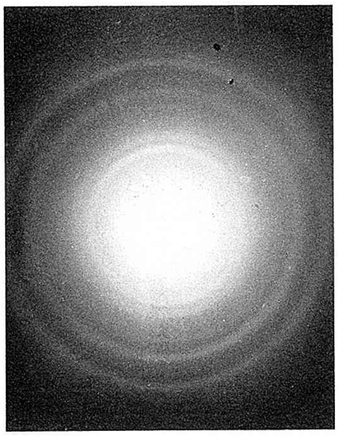
drawn 47 times,
diameter 0·97 mms.

Fig. 5



drawn 43 times,
diameter 1·1 mms.

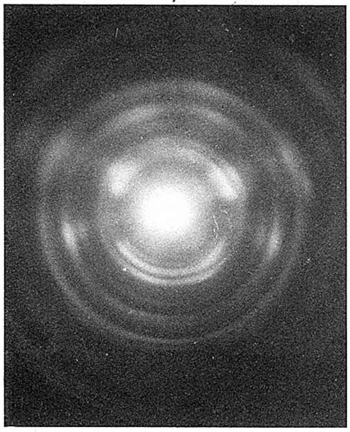
Fig. 6



diameter 0·9 mms.

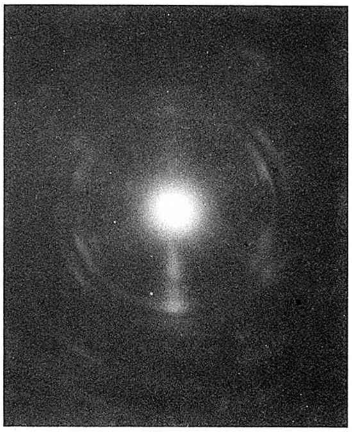
Plate II

Fig. 1



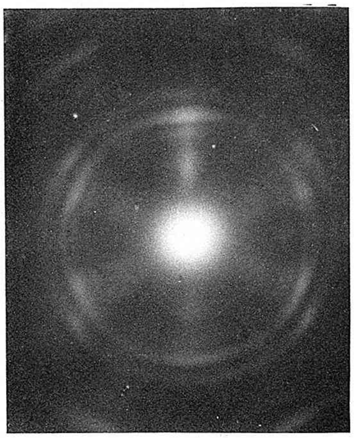
diameter 1·2 mms.

Fig. 2



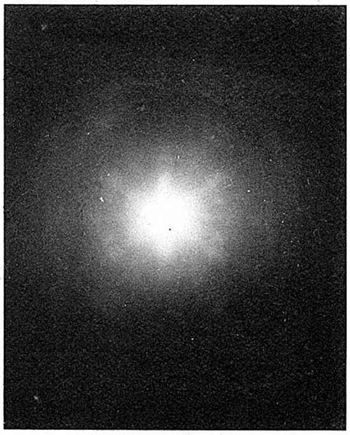
diameter 0·97 mms.

Fig. 3



diameter 1·1 mms.

Fig. 4



copper wire,
diameter 0·086 mms.