A Method of Determining the Orientation of Crystal-Axes by X-Rays

By

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Abstract

A simple method of determining the orientation of crystals with the X-rays is described. In an ordinary rotating crystal spectrometer a V-shaped frame of lead wires was inserted between the crystal and the photographic plate, and was rotated with the crystal. From the positions of the shadows cast by the lead wires on the spectral lines the angle through which the crystal should be rotated to reflect these spectral lines, can be calculated. Therefore, if the wave length of the illuminating X-rays be known, the orientation of the crystallographic axes can be determined. The method is especially convenient for the thick crystal and for the crystal which has grown on the surface of a thick substance. The method is also applicable to the investigation in the arrangement of the micro-crystals on the surface of metals which have been subjected to some regular cold workings. The arrangement of the micro-crystals on the polished surface of copper was examined, and it was observed that the greater part of them were so arranged that their (111) or (100) plane is nearly parallel to the polished surface.

Introduction

For determining the orientation of crystal axes by means of X-rays the methods hitherto proposed by many investigators may be divided into two types: the Laue-spots method and the rotating crystal method. The orientation is determined from the Laue-spots pattern produced by white X-rays, with the aid of "the globe and the spherical scale" according to the method of U. Yoshida[1], and by means of the stereographic projection employed by Schiebold and Sachs[2]. If the illuminating rays contain the characteristic radiation of the target of the X-ray tube and

1 U. Yoshida; Jap. J. Phys., **4**, 133 (1927).
2 Schiebold u. Sachs; Z. S. f. Krist., **63**, 34 (1927).

if their spectra appear in the spots, they also serve as a clue for the determination, as has been experienced by T. Fujiwara[1], and by U. Yoshida and K. Tanaka[2]. Of the rotating crystal methods, those devised by Weissenberg[3], by Dawson[4], and by Davy[5] consist in rotating the crystal continuously through a certain range of angle, and in those devised by Müller[6], and by Aston[7] the crystal is made to rotate in a step-wise manner with a small step-angle. In any case the essential point is to know the angle through which the crystal should be rotated to bring forth the reflection of the characteristic radiation from a certain atomic plane of the crystal.

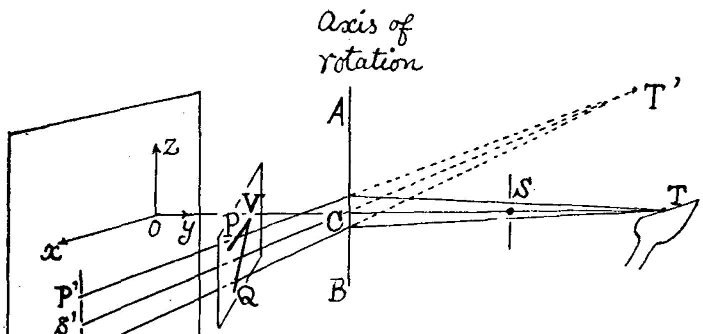
The present paper gives a rotating crystal method, using a new device to measure the angle to be rotated which would call forth the reflection of the characteristic radiation from a certain atomic plane of the crystal. With this method the writer has determined the orientation of the crystallographic axes of a thick single crystal of copper. The method is also employed to investigate the arrangement of the microcrystals on the surface of a rolled platinum foil and on the polished surface of a thick copper block.

Description of Apparatus

The arrangement used is depicted diagrammatically in Fig. 1. A specimen to be tested is so mounted at C on the table of a rotating spectrometer as to have its surface to be tested in a vertical direction. The X-rays emitted from a point source T on the target of an X-ray tube pass through a long and narrow vertical slit at S; and the X-rays thus diverged in a vertical plane illuminate the surface of the specimen to be tested in a plane parallel to the axis of rotation ACB. Behind the specimen a V-shaped frame PVQ of lead wires is so fixed to the table and is made to rotate with it that the plane containing the frame is perpendicular to the surface of the specimen to be tested, and that the bisector of the angle PVQ is always perpendicular to the rotating axis during the rotation. The X-rays reflected from the crystal are

1 T. Fujiwara; Mazda Kenkyu Jiho, Tokyo Elect. Co., **1**, 182 (1926).
2 U. Yoshida and K. Tanaka; Nature, **118**, 912 (1926).
3 Weissenberg; Z. S. f. Physik, **23**, 229 (1924).
4 Dawson; Phil. Mag., **5**, 756 (1928).
5 Davy; Phys. Rev., **23**, 764 (1924).
6 Müller; Proc. Roy Soc., **105**, 500 (1924).
7 Aston; Cambr. Phil. Soc., **23**, 561 (1927).

Fig. 1



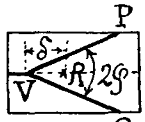


Fig. 1, *a*

absorbed by the lead wires VP and VQ on their way to a fixed photographic plate placed parallel to and behind the rotating axis, and the shadows P′ and Q′ of the lead wires are cast on the image of the spectral line on the photographic plate.

Further, a lead wire S is stretched across the long slit so as to cut off the incident ray running perpendicularly to the axis of rotation, and the shadow S′ of this lead wire is also cast on the spectral line on the photographic plate placed perpendicular to the central ray TS.

In this method, an X-ray tube with a sharply focussed target is preferable, in order to get distinct shadows of the lead wires. This purpose is, however, accomplished even with a tube the focus of which is not quite sharp, by setting it in such an orientation that the surface of its target is nearly, but not exactly, at right angles to the slit.

Calculation

From the positions of the shadows P′ or Q′ and S′ cast by the lead wires VP or VQ and S respectively on the photographic plate the angle, say ϕ, through which the crystal should be roatated in order to reflect the characteristic radiation of the target from a certain atomic plane, can be calculated as follows :— Consider a rectangular coordinates system, as shown in Fig. 1, where the foot of the central incident ray TS on the photographic plate is taken as the origin O, and the three orthogonal directions : one perpendicular to the axis of rotation of the crystal on the photographic plate, next, one parallel to the axis of rotation of the crystal on the photographic plate, and third, the direction of the central incident ray ST, are taken as the x-, z- and y-axis respectively. Let the coordinates of the two shadow points S′ and P′ be (x_0, o, z_0) and (x, o, z) respectively, and the distances of the focus T on the target, of the photographic plate and of the plane of the lead wire V-shaped frame PVQ, from the axis of rotation be a, b and d respectively.

Before the specimen is made to rotate, the frame of lead wires PVQ is so adjusted that the central beam TSC is perpendicular to the plane of the frame and intersects the bisector of its angle PVQ at the point R, as shown in Fig. 1, *a*. If we represent the distance between V and R by δ, then, at the moment when the frame is rotated through ϕ, the direction consines of the lead wire VP are $\cos\varphi\cos\phi$, $\cos\varphi\sin\phi$ and $\sin\varphi$, where φ is the half of the angle PVQ; and the coordinates of the vertex V of the frame of lead wires are $d\sin\phi-\delta\cos\phi$, $b-d\cos\phi-\delta\sin\phi$, and zero. If T′ is the average position of the image of the focus T with respect to a reflecting atomic plane, then the condition that the two straight lines T′ P′ and VP may intersect is given by

$$\begin{vmatrix} x-d\sin\phi+\delta\cos\phi & \cos\varphi\cos\phi & x+A\,x_0 \\ b-d\cos\phi-\delta\sin\phi & -\cos\varphi\sin\phi & b(1+A) \\ z & \sin\varphi & z+Az_0 \end{vmatrix}=0,$$

where $A=\dfrac{a}{\sqrt{x_0^2+b^2+z_0^2}}$.

Since x is nearly equal to x_0 we get from the above equation

$$\{(d\,x_0+b\delta)P-bQ\}\cos\phi=\{(bd-\delta x_0)P+x_0Q\}\sin\phi+R,$$

where

$$P=(1+A)\tan\varphi,$$

$$Q=A(z-z_0),$$

$$R=d\,(z+Az_0).$$

This equation gives the value of the angle ϕ.

On the other hand, if the wave length of the characteristic radiation used is known, the indices of the reflecting atomic plane of the crystal, which is responsible for the spectral line appearing on the photograph, can easily be determined from the relation

$$\tan 2\theta = \frac{\sqrt{x_0^2 + z_0^2}}{b},$$

and Bragg's equation

$$2d \sin\theta = n\lambda.$$

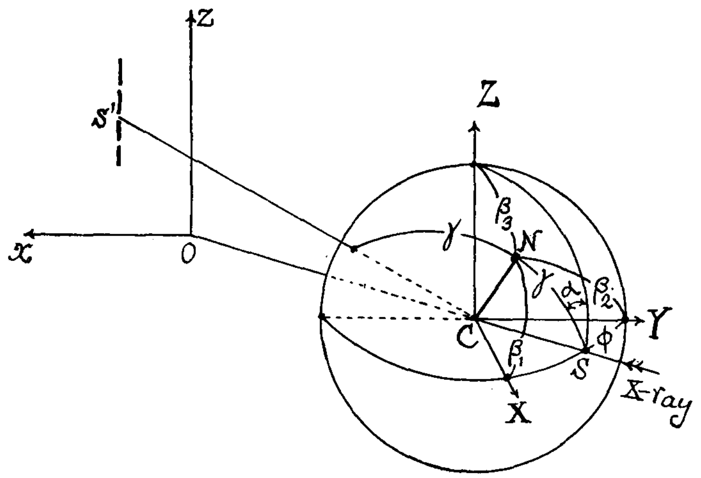
Next, take a rectangular coordinate C-XYZ fixed to the specimen as shown in Fig. 2, where the X-, Y- and Z-axes are to be taken as parallel respectively to the x-, y- and z-axes when the specimen is not rotated. Let the direction of the central beam of the X-rays be SC, then the direction cosines (l, m, n) of the normal to the atomic plane whose indices are above determined are given as follows:

$$\begin{cases} l = \cos\beta_1 = \cos\gamma \sin\phi + \sin\gamma \cos\phi \sin\alpha, \\ m = \cos\beta_2 = \cos\gamma \cos\phi - \sin\gamma \sin\phi \sin\alpha, \\ n = \cos\beta_3 = \sin\gamma \cos\alpha ; \end{cases}$$

where $\alpha = \arctan x_0/z_0,$

and $\gamma = \frac{\pi}{2} - \theta$: is the angle of incidence of the X-ray beam to the atomic plane under consideration.

Fig. 2



Thus, from a spectral line appearing on the photograph, the direction of the normal to an atomic plane referred to the axes fixed to the specimen will be determined together with its indices. Therefore, if two spectral lines, at least, appear on the photograph, the orientation of the crystal in the specimen will be perfectly determined.

Experiments

In the arrangement employed, a Müller spectrometer was utilized; and the specimen rotated, by means of clock-work, forward and backward through about twenty degrees. The distance, d, from the axis of rotation to the plane of the V-shaped frame of lead wires, and the angular opening of this frame are 17 cm. and 38° respectively. As a source of X-rays a Coolidge tube of molybdenum anticathode was used. The perpendicular distance, a, between the source point on the target and the axis of rotation was estimated as follows:— A lead plate of a given breadth was illuminated normally with the X-rays from the focus point of the target, and its shadow cast on a photographic plate placed parallel to and at a certain distance behind it; and the distance a is determined from the breadth of the lead plate and that of the shadow, the distance between the lead plate and the photographic plate and that between the photographic plate and the axis of rotation of the specimen. The value of a thus determined is 220 cm.

Table I $\begin{pmatrix} b = 4.0 \text{ c. m. s} \\ \delta = 0 \end{pmatrix}$

No. of spectral lines	x_0 in c.m.	z in c.m.	Glaning angle, θ $\theta = \frac{1}{2}\tan^{-1}\frac{x_0}{b}$ (obs.)	$\theta = \sin^{-1}\frac{n\lambda}{2d}$ (calc.)		Order of spect. lines	ϕ
1	.85	.076	6°	6°10′	$K\alpha$	3	5°20′
2	1.16	.098	8°10′	8°10′	$K\alpha$	4	7°40′
3	1.30	.107	9°	9°10′	K_β	5	8°40′
4	1.48	.123	10°10′	10°20′	$K\alpha$	5	9°30′

As a preliminary experiment the reflection from a mica sheet was examined, and the photograph taken is reproduced in Fig. 1 of Plate I;

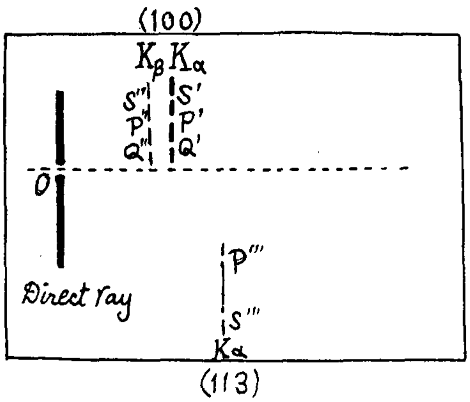
the result of the measurements from this photograph is shown in Table I. The values of the glancing angle given in the fifth column are calculated from Bragg's equation by taking 0.710 and 0.630 Å. U. respectively as the wave lengths of K_α and K_β lines of molybdenum and 9.91 Å. U. as the lattice constant of the mica. The lines and the orders assigned to the spectral lines which appeared on the photograph are given respectively in the sixth and the seventh columns of the Table.

Since X-rays are reflected from the cleavage surface of the mica in this case the values of ϕ should be taken to be equal to those of the glancing angle, because the cleavage surfase of the mica is set parallel to the plane $\phi=0$ initially. The Table shows that the agreement between the values of θ and ϕ is roughly within the limit of experimental error.

Table II $\begin{pmatrix} b=3.8 \text{ c.m.s} \\ \delta=0 \end{pmatrix}$

No. of spectral lines	x_3 in c.m.	z_0 in c.m.	z in c.m.	Indices of reflecting atomic planes	ϕ
1	1.41	.76	.59	(100) $K\alpha$	3°
2	1.21	.69	.52	(100) K_β	3°30′
3	2.13	-2.08	-1.46	(113) $K\alpha$	13°

Fig. 3

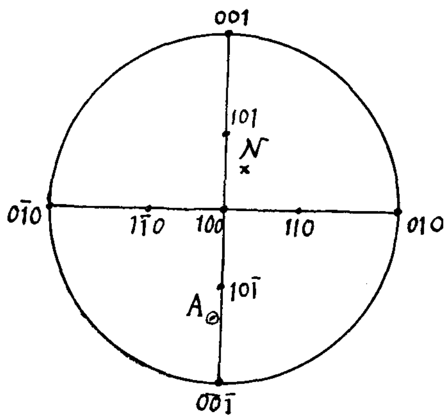


Next, the orientation of the crystallographic axes of a single crystal of copper was determined. The single crystal of copper used here was obtained by the stress-annealing-method. An annealed copper plate was elongated by about one per cent., and then it was again annealed at about 900° C. for a day or two in an evacuated furnace. By this method the writer was able to obtain copper crystals of about 1 cm. square. The orientation of one of the crystals thus prepared was determined by the above method. The photograph reproduced in Fig. 2 of Plate I was taken with such crystal of copper, and it is represented diagrammatically in Fig. 3. Table II shows the result of the measurements from this photograph. The two spectral lines on the photograph are confirmed as due to the reflection from the (100) and (113) planes. The orientation of their normals referred to the coordinate axes fixed to the specimen is calculated and shown in Table III. The stereographic representation of the orientation of the surface normal and of the axis of rotation of the specimen referring to the crystallographic axes is shown in Fig. 4.

Table III

Crystallographic axis	Angle from surface normal of specimen	Angle from axis of rotation
100	29° (28°)	62° (62°)
113	44° (46°)	133° (131°)

Fig. 4



× N : normal of the surface.
⊙ A : axis of the rotation

The orientation of the crystallographic axes of the same specimen was also determined from the Laue-spots pattern by using U. Yoshida's[1] "globe and spherical scale," and the result, which agrees well with that acquired above, is shown by the figures in parentheses in Table III. Fig. 3 of Plate I is the reproduction of the Laue-spots pattern taken with the normal incidence of the X-ray beam upon the surface of the copper plate, the distance of the photo-

1 U. Yoshida; loc. cit.

graphic plate from the specimen being 2.9 cm. in this case.

Arrangement of the Micro-Crystals on the Surface of Rolled Platinum Foil and of Polished Copper

An X-ray analysis of the arrangement of the micro-crystals on the surface of metals which were subjected to some regular cold workings has been reported by Professor S. Tanaka[1] and W. H. Bragg[2]. In Professor Tanaka's method, divergent X-rays are projected on the surface of a rolled platinum foil wound on the surface of a cylindrical wooden rod and the spectra produced by reflection from an atomic plane are obtained. By this method only is it possible to examine such a specimen as has, or can be formed into, the shape of a cylindrical surface. W. H. Bragg has also obtained sharp spectral lines reflected from certain atomic planes of platinum, gold and copper foils. But by this method it is quite impossible to know at what angular position during the rotation the specimen has reflected the spectral line. The method described before serves to fill up this defect in the method of rotating crystal. The photograph taken with a platinum foil prepared by rolling and with the polished surface of a copper block are reproduced in Fig.4 and Fig.5 of Plate I.

The platinum foil used here was the same specimen as that examined before by Professor S. Tanaka[1]; and the polished surface of copper was prepared as follows :— A rectangular block of pure copper was annealed at about 800° C. for about 10 hours in an evacuated furnace. One face of thus annealed block was polished successively with grind-stones of different grain-sizes : first with that of coarse grain, and then with those of successively finer grains ; and lastly, the surface was polished with rouge on a flat surface of pitch. All these processes of polishing were made by hand slowly and reversely in the same direction.

As may be seen in Figs. 4 and 5, Plate I, we have the spectral lines containing the shadows cast by the lead wires. The result of the measurement from the photographs are represented in Tables IV and V. The values of ϕ and θ for each corresponding line in these Tables are nearly equal within the limit of experimental error. As the surface of

1 S. Tanaka; These Memoirs, **8**, 319 (1925).
2 W. H. Bragg; Nature, **115**, 639 (1924).

Table IV

Rolled Platinum Foil $\begin{pmatrix} b=4.0 \text{ c.m.s} \\ \delta=0 \end{pmatrix}$

No. of spectral lines	x_0 in c.m.	z in c.m.	Indices of reflecting atomic planes	Glancing angle, θ		ϕ
				$\theta=\frac{1}{2}\tan^{-1}\frac{x_0}{b}$ (obs.)	$\theta=\sin^{-1}\frac{\lambda}{2d}$ (calc.)	
1	2.24	.205	(110) $K\alpha$	14°30′	14°50′	13°30′
2	1.94	.175	(110) K_β	12°30′	12°30′	12°

the specimen, which is examined, is set parallel initially to the plane $\phi=0$ in both cases, the agreement between the values of ϕ and θ indicates that the atomic plane of the crystal under consideration is parallel to the surface of the specimen. Thus it may be concluded from the result obtained that the majority of the micro-crystals are so arranged that the (110) plane in the case of the rolled platinum foil and the (111) and (100) planes in the case of the polished surface of copper are almost parallel to the examined surface of the specimens.

Table V

Polished Surface of Copper $\begin{pmatrix} a=21.0 \text{ c.m.s} \\ b=4.0 \text{ c.m.s} \\ \delta=0.2 \text{ c.m.s} \end{pmatrix}$

No. of spectral lines	x_0 in c.m.	z in c.m.	Indices of reflecting atomic planes	Glancing angle, θ		ϕ
				$\theta=\frac{1}{2}\tan^{-1}\frac{x_0}{b}$ (obs.)	$\theta=\sin^{-1}\frac{\lambda}{2d}$ (calc.)	
1	1.35	.210	(111) $K\alpha$	9°	9°50′	10°
2	1.59	.215	(100) $K\alpha$	11°	11°20′	12°

The result obtained with the rolled platinum foil is entirely the same as that obtained by Professor S. Tanaka. As to the examination of the polished surface of copper it is entirely of a preliminary nature; and though the final conclusion on the effect of polishing is still far from the scope of the present experiment, yet the result obtained suggests, at least, that we must be very cautious in examining microscopically the crystalline structure of the polished surface of metals.

Lastly, it must be noted that the method described before is especially preferable for the examination of the crystalline arrangement near the surface of a heavy substance.

In conclusion the writer wishes to express his sincere thanks to Prof. U. Yoshida under whose kind guidance and encouragement the present experiment was carried out.

Plate I

Fig. 1

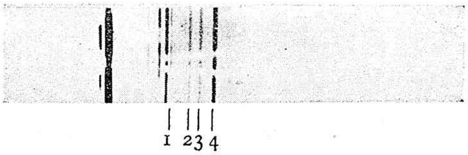


Fig. 2

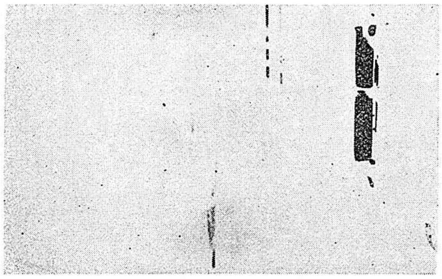


Fig. 3

Axis of rotation

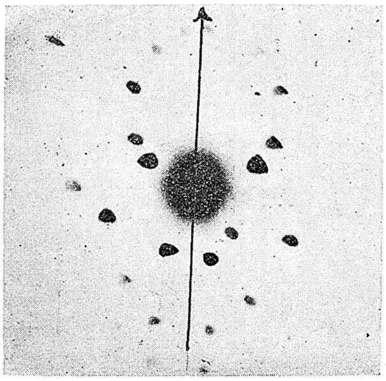
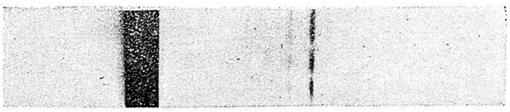
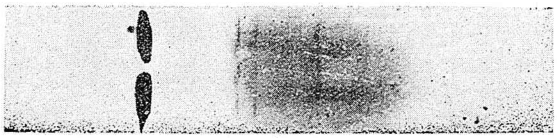


Fig. 4



Rolled Pt foil

Fig. 5



Polished Cu surface