# The Arrangement of Micro-crystals in Beaten Gold Foil.

By

**Shinsuke Tanaka.**

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ABSTRACT

The arrangement of the micro-crystals in rolled and beaten gold foil was investigated by the X-ray method. The majority of the micro-crystals in the rolled foils are so arranged that their (110) faces are parallel to the surface of rolling and the normals of the (211) faces parallel to the direction of rolling. In the case of a foil prepared by hammering for a long time, the (100) faces of the micro-crystals of the gold are arranged parallel to the surface of the foil the orientations in other respects being at random. In the intermediate stage of the hammering process, there seems to exist a comparatively persistent state, during which the micro-crystals take on roughly a fibrous structure with the normals of the (110) faces as their common axis lying nearly in the surface of the foil.

When the writer was carrying out his investigations on the arrangement of the micro-crystals in a rolled platinum foil, he found that in a gold leaf obtained from the market, the (100) faces of the micro-crystals were arranged parallel to the surface of the leaf, and he reported this fact in these memoirs.[1] A further investigation of this problem has lately been conducted and is described below.

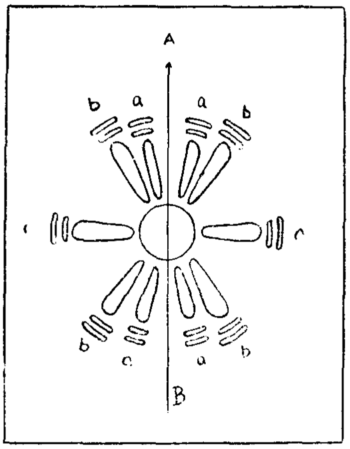
The gold leaves used in the present experiment were prepared by rolling and then hammering gold bullion: a piece of gold bullion was rolled down to a thin plate of the thickness of about 0·05mm., and then it was flattened to a very thin gold leaf by hammering. In order to prevent cracking and to keep the stretching uniform, the foils were held

(1) These Memoirs, **8**, 319 (1925).

between sheets of strong and smooth paper during the hammering process. In some cases the samples were annealed before hammering to destroy the regular arrangement of the micro-crystals caused by rolling, but this is not done in the making of gold leaf in the factory.

Using the K-radiation from the Mo. anticathode of a Coolidge tube, the writer examined the interference figures by the ordinary method of photographing the Laue spots, and the spectra of the X-rays reflected from the surface of the sample were examined by the aid of the same apparatus as was used in the previous experiment.[1]

Fig. 1′.




Two photographs corresponding to the rolled gold foils are reproduced in Fig. $1_A$ in Plate X and $1_B$ in plate XI. Fig. $1_A$ is a transmission photograph obtained under the normal in idence of the X-rays on the surface of the foil, and Fig. $1_B$ a reflection photograph. Fig. 1′ represents diagramatically Fig. $1_A$ in Plate X. AB in Fig. 1′ represents the direction parallel to that of rolling, and the radiating bands marked by a, b and c are caused by the reflection of the X-rays from the crystal faces denoted in the following Table I. The angles $\varphi$ between the radiating bands and the line AB are given in the same table.

Table I.

| Radiating bands | Indices of the faces | The angle $\varphi$ |
|---|---|---|
| a | (111) | 15°—17° |
| c | (111) | 90° |
| b | (100) | 33°—35° |

If we disregard the weak band, this distribution of the radiating bands is entirely the same as that of the rolled platinum foil given in a former paper.[1] The spectral lines in Fig. $1_B$ in Plate XI show that the X-rays are reflected from the (110) faces arranged parallel to the surface of the rolled foil. This fact is also the same as in the case of a rolled platinum foil. As is generally known, the lattice of the crystals of gold and platinum belongs to the same form as the face-centered cubic. Therefore the identity of the distribution of the radiating bands in the transmission

1 These memoirs, **8**, 319 (1925). In the first column of Table 1. page 322 of that paper the third and the fourth horizontal row must be interchanged.

photograph and that of the reflecting surface which is responsible for the spectral lines in the reflection photograph indicates that the micro-crystals in a rolled gold foil are arranged in the same manner as in the rolled platinum foil, i. e. the majority of the micro-crystals in the gold foil take such an orientation that the (110) faces are nearly parallel to the surface of rolling and that the normals to the (211) faces are parallel to the direction of rolling.

If we beat the rolled foil to a thinner foil, then the photograph obtained by the transmission method, shows that the radiating band a in Fig. 1′ gradually approaches the band b, and that it coincides at last with the latter band as the process of beating is advanced. At this stage the radiating bands become six in all, and any neighbouring two of them make an angle of about 60°. At the same time the intensities of the parts of the bands corresponding to the continuous X-rays become weaker and the other parts of the bands corresponding to the K-radiation of the target become stronger. From the positions of these intense K lines of Mo., it was easily reconizable that the reflecting planes which produced these six bands were chiefly the (111) faces of the gold crystals. The appearance of these is shown in Figs. $2_A$, $3_A$ and $4_A$ in plate X.

The photographs of the spectra of X-rays reflected from the surface of the foils, some of which are reproduced in Fig. $2_B$, $3_B$ and $4_B$ in Plate XI, were taken at each stage of hammering. In these photographs four new spectral lines appear in addition to those observed in Fig. $1_B$ in Plate XI, and it was ascertained by calculation that they belong to the $K_\alpha$ and $K_\beta$ lines of molybdenum reflected from the (311) and (100) faces of the gold crystals. Here it must be noted that the intensity of the lines corresponding to the (100) faces becomes gradually stronger as the process of hammering is advanced.

Next the sample was again beaten down to a very much thinner foil, the thickness being reduced to about 0·001 mm. or less. The photograph obtained by the transmission of X-rays through this thin foil consists of several very faint concentric rings characteristic of the disordered arrangement of the micro-crystals with respect to the direction of the X-ray beam. The photograph obtained by the reflection method consists of two spectral lines caused by the reflection of the X-rays from the (100) faces arranged parallel to the surface of the foil. Fig. $5_A$ in Plate X and Fig. $5_B$ in plate XI are reproductions of these photographs.

Fig. $2_C$ in Plate X is a reproduction of the photograph obtained from a sample which was annealed after rolling, and it indicates, by its irregular strips, that the regularity of the arrangement of the micro-crystals is completely destroyed. With such annealed foils the writer could not observe any fine spectral lines in photographs obtained by the reflection-method. The annealed foils were hammered as in the case of unannealed foils, and at various stages of hammering the arrangement of the micro-crystals was examined by both the transmission and the reflection method. At the beginning the photographs obtained by the transmission method showed quite a different appearance from those obtained with una nealed foil as is seen in Fig. $3_C$, in Plate X; but as the process of hammering proceeded they began to resemble each other as shown in Fig. $4_C$, in Plate X. When the hammering is continued further and when the foils become very thin the transmission-photograph becomes entirely the same as the corresponding one obtained with unannealed thin foil.

The writer examined also the arrangement of the micro-crystals of the annealed and then hammered foil by the reflection-method. At the stage of hammering process corresponding to that between Fig. $3_C$ and Fig. $4_C$ in Plate X, the spectra caused by the reflection from the (311), (110) and (100) faces were recognized, these faces being arranged parallel to the surface of the foil. This fact is the same as that observed in the corresponding stage of unannealed foil. When the hammering was continued further with the annealed foil, only the spectrum caused by the reflection from the (100) faces was observed, as in the case of unannealed foil.

Here it must be noted that at no stage of the hammering could the writer find any spectral lines reflected from (111) faces on the photographs obtained by the reflection method though these faces are of the greatest atomic density, and the most efficient in reflecting the X-rays.

In the case of unannealed foil the micro-crystals in the foil whose (110) faces are arranged parallel to the surface of the sample at the beginning, seem to begin to change their positions by hammering, as stated before; and as the hammering process is still uther continued they take such orientations that their (100) faces lie parallel to the surface of the foil. It must be especially remembered that, during this process, the micro-crystals change their orientation in such a way that their (111) faces never come out parallel to the surface of the foil. We shall now consider the manner of this change of orientation of the micro-crystals.

First we shall consider the sample which gave the photographs reproduced in Figs. $4_A$ and $4_C$ in Plate X by the transmission method. This stage persists very long in the process of hammering whether the sample is annealed after rolling or not. These photographs were taken by the normal incidence of the X-rays on the surface of the foil, and they consist of six radiating bands reflected from the (111) faces of the crystals, the angle between any neighbouring two of these bands being about 60°.

The presence of the radiating bands indicates that the micro-crystals in the sample have the arrangement generally known as the fibrous structure. When the micro-crystals in the foil have the same direction, defined crystallographically as common, and they are so distributed that each micro-crystal has a position produced by rotation at some angle around this common direction or "common axis," we call it a fibrous structure. Thus the determination of the direction of the fiber in the sample and the crystallographic direction of the common axis for each micro-crystal is the essential point in solving our problem. With regard to the direction of the fiber, we may determine it by taking many transmission photographs corresponding to various orientations of the sample against the incident X-ray beam. This is actually done by rotating the sample around several axes, and if we could find such an axis that no rotations around it makes any change in the photographs, this axis would be parallel to the direction of the fiber. If this axis of the fiber were known, the crystallographic direction of this axis would be determined from the distribution of the radiating bands.

Many photographs were taken in various orientations of the sample rotated about different axes parallel to the surface. But the result was not decisive, as the radiating bands were very diffuse. This is probably due to the fact that all the micro-crystals are arranged, not exactly in fibrous structure, but only roughly so, and many crystals may occupy different orientations deviating by some small angles from their mean fibrous axis. If we take into consideration the comparative irregularity of hammering as a method of working, this is of course natural. This is also in comformity with the actual observation that, at this stage of the hammering process, three different sets of spectra of X-rays are obtained by the reflection from three crystal faces, (311), (110) and (100) which are arranged nearly parallel to the surface of the foil.

It seems to be not unnatural to consider, from the experiments carried out by various authors, that some one of the normals of the prominent faces (111), (211), (110), or (100) of the micro-crystals are arranged roughly as the common axis of the fibrous structure. As stated before, the six radiating bands on the photograph are caused by the reflection of X-rays from the (111) faces. For the present, the direction of the common axis is taken provisionally to be in the surface of the foil, and by taking each normal above mentioned as the common axis,

Table 2

| Common axis | Angles between two neighbouring bands | Total number of radiating bands |
|---|---|---|
| normal of (111) | 71°, 38° | 6 |
| normal of (211) | 43°, 38°, 28° | 10 |
| normal of (110) | 70°, 55° | 6 |
| normal of (100) | 110°, 70° | 4 |

the positions of the radiating bands for one normal incidence of the X-rays to the surface of the foil were calculated and tabulated in Table 2. From this table the normal of the (110) faces seems to be suitably taken as the common axis.

Next the writer calculated the positions of the radiating bands when the normal of the (110) faces was taken as the common axis, and the X-rays were projected at various angles to this axis. The values are given in Table 3.

Table 3

| Angle between the normals of the (110) faces and the X-rays | Angle between two neighbouring bands | Total number of radiating bands |
|---|---|---|
| 20° | 230°, 130° | 2 |
| 30° | 210°, 150° | 2 |
| 40° | 200°, 160° | 2 |
| 50° | 166°, 79°, 36° | 4 |
| 60° | 170°, 65°, 60° | 4 |
| 70° | 70°, 66°, 58°, 42° | 6 |
| 80° | 70°, 60°, 59°, 56° | 6 |
| 90° | 70°, 55° | 6 |

From this table we know that when the angles between the X-ray and the common axis are not greatly different from 90°, the positions of the radiating bands are not subjected to any considerable change. If we take other normals as the common axis and calculate similarly the positions of the radiating bands, we have quite different values from those given in the above table, which can not possibly be accepted. Therefore it seems to be not unreasonable to take the normal of the (110) faces as the common axis which is arranged roughly parallel to the surface of the foil. With this as his starting point the writer could calculate the positions of the radiating bands when the sample was rotated in various directions and found them to be in conformity with actual observation, though they are not given here.

In conclusion, the author wishes to express his sincere thanks to Prof. U. Yoshida under whose direction the work was done.

Fig. 2A.

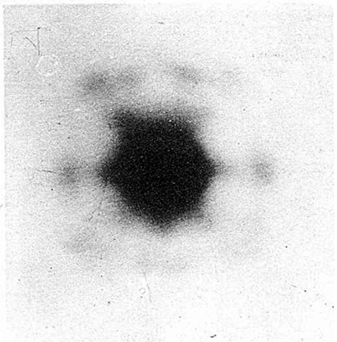


Fig. 2C.

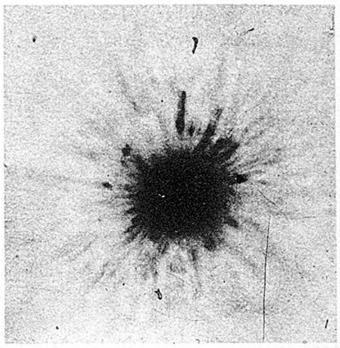


Fig. 1A.

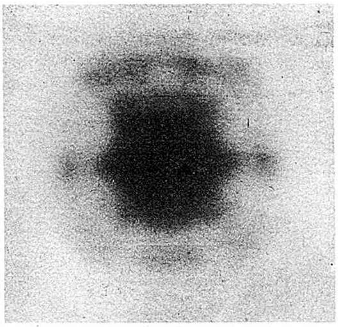


Fig. 3A.

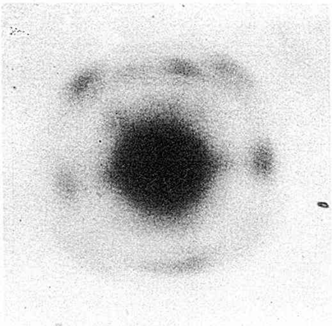


Fig. 3C.

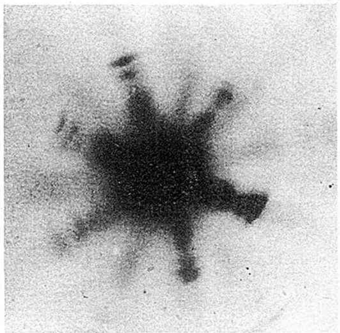


Fig. 5A.

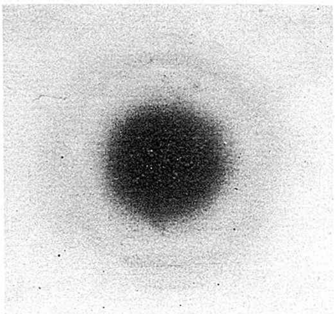


Fig. 4A.

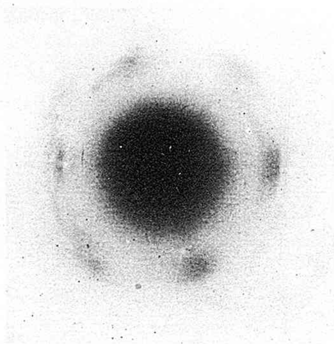
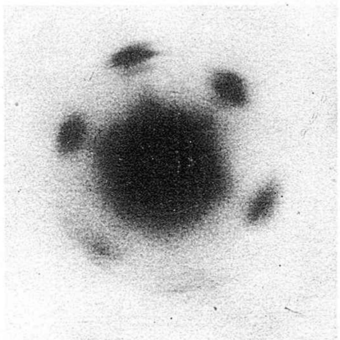
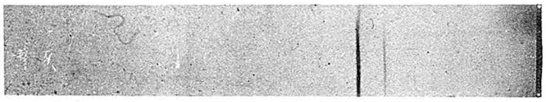


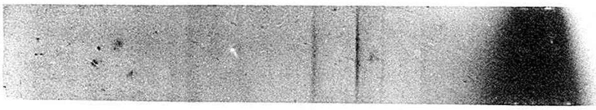
Fig. 4C.





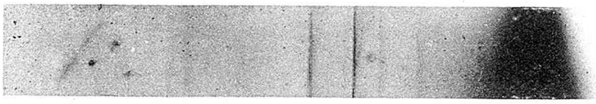
(110)

Fig. 1B.



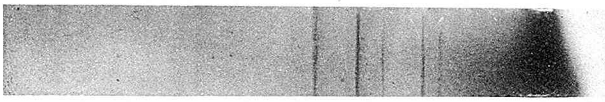
(311) (110) (100)

Fig. 2B.



(311) (110) (100)

Fig. 3B.



(311) (110) (100)

Fig. 4B.

(100)

Fig. 5B.