The Light-Figure Phenomenon Revealed and Crystal Planes Developed by Etching in Tetragonal Tin Crystals and the Determination of Their Crystal Orientations by the Light-Figure Method. II : Orientation Determination by Light Figures

著者	YAMAMOTO Mikio, WATANABE Jiro
journal or publication title	Science reports of the Research Institutes, Tohoku University. Ser. A, Physics, chemistry and metallurgy
volume	7
page range	161-172
year	1955
URL	http://hdl.handle.net/10097/26694

The Light-Figure Phenomenon Revealed and Crystal Planes Developed by Etching in Tetragonal Tin Crystals and the Determination of Their Crystal Orientations by the Light-Figure Method. II

Orientation Determination by Light Figures*

Mikio YAMAMOTO and Jirô WATANABÉ

The Research Institute for Iron, Steel and Other Metals

(Received December 6, 1954)

Synopsis

The application of light figures to the orientation determination of single crystal rods of tetragonal tin has been studied and the procedure for the perfect, accurate, and rapid determination has been worked out. Full accounts are given of the geometrical relationships and etching technique required for the orientation determination, of the directly and accurately measurable ranges and kinds of orientation angles, of the stereographic representation of crystal orientations, and of the accuracy and actual examples of the determination. The perfect determination of crystal orientations can be made with an accuracy within 0.1° by the use of {001}, {100} and {101} light figures revealed by etching with concentrated aqua regia. It is shown that the orientation determination by this method agrees well with that by the X-ray diffraction method.

I. Introduction

In Part I(1), we studied the light-figure phenomenon revealed and crystal planes developed by etching in single crystals of tetragonal tin. The application of the observed light figures to the orientation determination is studied in the present part. Previously, we applied the light-figure method for the orientation determination of cubic(2), hexagonal(3), and trigonal(4) crystals with great success, and the application to tetragonal tin crystals in the present report has completed our research program of the application of the light-figure method to pure metal crystals of all types. Previously, Chalmers(5) determined orientations of tetragonal tin crystal rods with an accuracy of $\pm 1°$ by the use of light figures produced by etching with 5 percent aqueous solution of ferric chloride. According to Part I(1), his etching technique is not so suitable for the orientation work because of the longness of etching time

* The 791st report of the Research Institute for Iron, Steel and Other Metals. The original of this report written in Japanese was previously published in Nippon Kinzoku Gakkai-shi (J. Japan Inst. Metals), **17** (1953), 113.

(1) M. Yamamoto and J. Watanabé, N. K. G. (Nippon Kinzoku Gakkai-shi), **17** (1953), 68 and 78 ; Sci. Rep. RITU, **A7** (1955), 145.

(2) M. Yamamoto, N.K.G., **5** (1941), 214 ; Sci. Rep. Tôhoku Univ., **31** (1943), 191 ; Butsurigaku Kôen-shyû, **3** (1943), 193. M. Yamamoto and J. Watanabé, N. K. G., **17** (1953), 5 ; and Sci. Rep. RITU, **A7** (1955), 173.

(3) M. Yamamoto and J. Watanabé, N.K.G., **13** (1949), No. 4 ; Sci. Rep. RITU, **A2** (1950), 270.

(4) M. Yamamoto and J. Watanabé, N.K.G., **B15** (1951), 572 ; Sci. Rep. RITU, **A5** (1953), 135.

(5) B. Chalmers, Proc. Phys. Soc., **47** (1935), 733.

required (about 30 minutes) and of the insufficiency in the distinctness of light figures used, and several more excellent etching directions are available. It is to be noted that the procedure of determination adopted by him is different from ours.

In a tetragonal crystal such as white tin, the tetragonal axis [001] and the digonal axes [100] and [010], three in all, are usually taken as the principal crystallographic axes. If the geometrical axis of a specimen crystal, $\overrightarrow{OP}$, makes angles θ, α and β with the [001], [100] and [010] axes, respectively, as shown in Fig. 1, then the following relationship holds among these angles:

$$\cos^2\theta + \cos^2\alpha + \cos^2\beta = 1. \qquad (1)$$

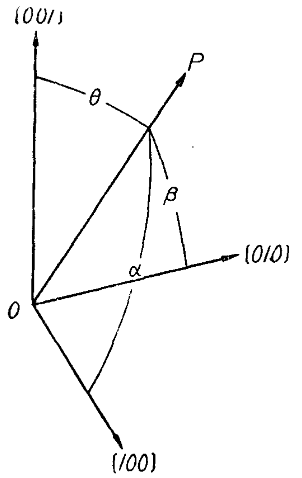

Fig. 1. An orientation in tetragonal crystal referred to the principal crystallographic axes.

Accordingly, the orientation of $\overrightarrow{OP}$ can be fixed by determining any two of θ, α, and β by the use of the {001} and {100} light figures. However, since the range of an angle which can be determined directly and accurately using a light figure is generally restricted by the inclination of the crystal plane producing the light figure to the rod axis of a specimen crystal(6), there are orientations for which the perfect determination cannot be made by the use of {001} and {100} light figures alone. In such cases, light figures other than the {001} and {100} ones, for example, the {101} light figures, must be employed. The object of the present investigation is to work out the procedure for the perfect, accurate, and rapid determination of orientations of tetragonal tin crystals by the use of light figures.

II. Geometrical relationships and etching directions required for the orientation determination, the directly and accurately measurable ranges and kinds of orientation angles, and the representation of crystal orientations

A. *Geometrical relationships*

As stated above, for the perfect orientation determination of tetragonal tin crystals, it is generally necessary to use at least three kinds of light figures including the {001} and {100} light figures. According to Part I(1), the crystal planes producing distinct light figures are, except the {001} and {100} planes, the {101}, {301} and {211} planes.

Now, we will summarize geometrical relationships required for the orientation determination by the use of the {001}, {100} and {101} light figures which are contained commonly in both the first and second groups of light figures. In the following formulae, the derivation of which is given in the appendix, θ, α and β denote angles

(6) M. Yamamoto and J. Watanabé, N.K.G., **17** (1953), 5; Sci. Rep. RITU, **A7** (1955), 173.

which the rod axis of a specimen crystal makes with the [001], [100] and [010] axes, respectively, and α', β', γ' and δ' denote angles which the rod axis makes with normals of the (101), (011), ($\bar{1}$01) and (0$\bar{1}$1) planes, respectively (Fig. 2). Finally, ω is an angle between the [001] axis and the normal of a {101} plane, and is given by

$$\cos\omega = 1/\{(c/a)^2+1\}^{1/2}, \qquad (2a)$$

where c/a is the axial ratio, being equal to 0.545_6[7] for tetragonal tin.

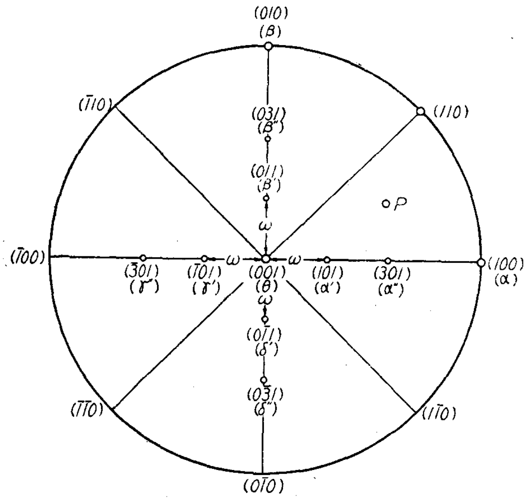

Fig. 2. Stereographic illustration of various angles which the specimen axis makes with normals of principal crystal planes.

[I] When any two of angles θ, α and β are measured, the other unknown angle γ may be calculated from Eq. (1).

[II] When angles θ and α' (or γ') are measured, angles α and β may be computed from

$$\left.\begin{aligned} \cos\alpha &= \pm[\operatorname{cosec}\omega \cos_{\gamma'}^{\alpha'} - \cot\omega\cos\theta], \text{ and} \\ \cos\beta &= [1-\operatorname{cosec}^2\omega(\cos^{2}{}_{\gamma'}^{\alpha'}+\cos^2\theta)+2\operatorname{cosec}\omega\cot\omega\cos_{\gamma'}^{\alpha'}\cos\theta]^{1/2}, \end{aligned}\right\} \quad (3)$$

where the signs $\pm$ correspond to α' and γ', respectively. It is to be noted that the calculation of α and β using the measured values of θ and β' (or δ') may be made from expressions derived from Eq. (3) by substituting α, β' and δ' in place of β, α' and γ', respectively.

[III] When angles α and α' (or γ') are measured, angles θ and β may be calculated from

$$\left.\begin{aligned} \cos\theta &= \sec\omega\cos_{\gamma'}^{\alpha'} \mp \tan\omega\cos\alpha, \text{ and} \\ \cos\beta &= [1-\{\sec^2\omega(\cos^{2}{}_{\gamma'}^{\alpha'}+\cos^2\alpha)\mp 2\tan\omega\sec\omega\cos_{\gamma'}^{\alpha'}\cos\alpha\}]^{1/2}, \end{aligned}\right\} \quad (4)$$

where the signs $\mp$ correspond to α' and γ', respectively. It is to be noted that the calculation of θ and α using the measured values of β and β' (or δ') may be made from expressions derived from Eq. (4) by substituting α, β' and δ' instead of β, α' and γ', respectively.

[IV] When angles α and β' (or δ') are measured, angles θ and β may be computed from

$$\cos\theta = \mp[(\sec\omega-\tan^2\omega\cos\omega)\cos_{\delta'}^{\beta'} - \tan\omega\cos\omega(-\cos^{2}{}_{\delta'}^{\beta'}+\sin^2\alpha)^{1/2}],$$

and

$$\cos\beta = \cos\omega[\pm\tan\omega\cos_{\delta'}^{\beta'} + (-\cos^{2}{}_{\delta'}^{\beta'}+\sin^2\alpha)^{1/2}], \qquad (5)$$

where the signs $\pm$ correspond to β' and δ', respectively. It is to be noted that the calculation of θ and α using measured values of β' and α' (or γ') may be made

(7) A. Westgren and G. Phragmen, Z. anorg. allg. Chem., **75** (1928), 80 ; E. R. Jette and F. Foote, J. Chem. Phys., 3 (1935), 65 ; M. C. Neuburger, Z. Krist., **92** (1936), 1.

from expressions obtained from Eq. (5) by putting α, α' and γ' in place of β, β' and δ', respectively.

[V] When angles α' and β' (or γ' and δ') are measured, angles θ, α and β may be calculated from

$$\left.\begin{aligned}
\cos\theta &= [\mathrm{cosec}\,\omega\cot\omega(\cos^{\alpha'}_{\gamma'}+\cos^{\beta'}_{\delta'})\pm\{(1+2\cot^2\omega)-\mathrm{cosec}^4\omega(\cos^{2\alpha'}_{\ \ \gamma'}+\cos^{2\beta'}_{\ \ \delta'})\\
&\quad+2\,\mathrm{cosec}^2\omega\cot^2\omega\cos^{\alpha'}_{\gamma'}\cos^{\beta'}_{\delta'}\}^{1/2}]\Big/(1+2\cot^2\omega),\\
\cos\alpha &= \pm[\mathrm{cosec}^3\omega\cos^{\alpha'}_{\gamma'}-\cot^2\omega\,\mathrm{cosec}\,\omega\cos^{\beta'}_{\delta'}\pm\cot\omega\{(1+2\cot^2\omega)-\mathrm{cosec}^4\omega\times\\
&\quad(\cos^{2\alpha'}_{\ \ \gamma'}+\cos^{2\beta'}_{\ \ \delta'})+2\mathrm{cosec}^2\omega\cot^2\omega\cos^{\alpha'}_{\gamma'}\cos^{\beta'}_{\delta'}\}^{1/2}]\Big/(1+2\cot^2\omega),\ \text{and}\\
\cos\beta &= \pm[\mathrm{cosec}^3\omega\cos^{\beta'}_{\delta'}-\cot^2\omega\,\mathrm{cosec}\,\omega\cos^{\alpha'}_{\gamma'}\pm\cot\omega\{(1+2\cot^2\omega)-\mathrm{cosec}^4\omega\times\\
&\quad(\cos^{2\alpha'}_{\ \ \gamma'}+\cos^{2\beta'}_{\ \ \delta'})+2\mathrm{cosec}^2\omega\cot^2\omega\cos^{\alpha'}_{\gamma'}\cos^{\beta'}_{\delta'}\}^{1/2}]\Big/(1+2\cot^2\omega),
\end{aligned}\right\}\quad(6)$$

where the signs $\mp$ and $\pm$ correspond to the combinations (α', β') and (γ', δ'), respectively.

[VI] When angles β' and γ' (or δ' and α') are measured, angles θ, α and β may be calculated from

$$\left.\begin{aligned}
\cos\theta &= [\mathrm{cosec}\,\omega\cot\omega(\cos^{\beta'}_{\delta'}+\cos^{\gamma'}_{\alpha'})\mp\{(1+2\cot^2\omega)-\mathrm{cosec}^4\omega(\cos^{2\beta'}_{\ \ \delta'}+\cos^{2\gamma'}_{\ \ \alpha'})\\
&\quad+2\mathrm{cosec}^2\omega\cot^2\omega\cos^{\beta'}_{\delta'}\cdot\cos^{\gamma'}_{\alpha'}\}^{1/2}]\Big/(1+2\cot^2\omega),\\
\cos\alpha &= \pm[-\mathrm{cosec}^3\omega\cos^{\gamma'}_{\alpha'}+\cot^2\omega\,\mathrm{cosec}\,\omega\cos^{\beta'}_{\delta'}\mp\cot\omega\{(1+2\cot^2\omega)\\
&\quad-\mathrm{cosec}^4\omega(\cos^{2\beta'}_{\ \ \delta'}+\cos^{2\gamma'}_{\ \ \alpha'})+2\mathrm{cosec}^2\omega\cot^2\omega\cos^{\beta'}_{\delta'}\cdot\cos^{\gamma'}_{\alpha'}\}^{1/2}]\Big/(1+2\cot^2\omega),\\
&\text{and}\\
\cos\beta &= \pm[\mathrm{cosec}^3\omega\cos^{\beta'}_{\delta'}-\cot^2\omega\,\mathrm{cosec}\,\omega\cos^{\gamma'}_{\alpha'}\pm\cot\omega\{(1+2\cot^2\omega)\\
&\quad-\mathrm{cosec}^4\omega(\cos^{2\beta'}_{\ \ \delta'}+\cos^{2\gamma'}_{\ \ \alpha'})+2\mathrm{cosec}^2\omega\cot^2\omega\cos^{\beta'}_{\delta'}\cos^{\gamma'}_{\alpha'}\}^{1/2}]\Big/(1+2\cot^2\omega).
\end{aligned}\right\}\quad(7)$$

[VII] When angles α' and γ' (or β' and δ') are measured, angles θ, α and β may be computed from

$$\left.\begin{aligned}
\cos\theta &= 2\sec\omega\,(\cos^{\alpha'}_{\beta'}+\cos^{\gamma'}_{\delta'}),\\
\text{and}\quad \cos^{\alpha}_{\beta} &= 2\sec\omega\,(\cos^{\alpha'}_{\beta'}-\cos^{\gamma'}_{\delta'}).
\end{aligned}\right\}\quad(8)$$

Furthermore, if a specimen axis makes angles α'', β'', γ'' and δ'' with normals of the {301} planes, namely (301), (031), $(\bar{3}01)$ and $(0\bar{3}1)$, the same relationships as just-mentioned hold among angles θ, α, β, α'', β'', γ'' and δ'', but in this case ω is an angle between the [001] axis and the normal of a {301} plane and is given by

$$\cos\omega = 1/\{9(c/a)^2+1\}^{1/2}. \qquad (2b)$$

From these geometrical relationships it may be seen that the orientation $\overrightarrow{\mathrm{OP}}$ can be fixed perfectly if any two of θ, α, β, α', β', γ' and δ', or of θ, α, β, α'', β'', γ'' and δ'' are known, and the check and correction of the determination can be made if more than three angles are measured directly.

B. Etching directions

The etching directions[8] for revealing light figures suitable to the orientation determination were studied detailedly in Part I[1]. The results obtained are summarized in Table 1. Among the etchants given in Table 1, concentrated aqua regia

Table 1. Etching directions for the orientation determination of tin single crystals by the light-figure method. The mark ◎ denotes a distinct light figure, suitable to the determination of crystal orientation, and × an indistinct light figure or a case where no figure is observed. If the desired light figure was not observed by etching with this reagent, etch a specimen crystal preliminarily with concentrated nitric acid for 30 sec~1 min and then apply the specified etching.

No.	Etching reagent*	Sharpness and suitability of light figure produced by						Group of light figures	Optimum etching time
		{001}	{100}	{101}	{301}	{211}	{110}		
1	$HCl+CrO_3$ (95 : 5)	◎	◎	◎	◎	◎	×	I	4 min.
2a	100% aqua regia	◎	◎	◎	×	×	×	II	30 sec.
2b	50% 〃	◎	◎	◎	×	×	×	II	3 min.
3	Sat. aq. sol. of $(NH_4)_2S_2O_8$	◎	◎	◎	×	×	×	II	15 min.
4a	Sat. aq. sol. of $FeCl_3 \cdot 6H_2O$	◎	◎	◎	×	×	×	II	30 sec.
4b	50% 〃	◎	◎	◎	×	×	×	II	2 min.
5	Sat. aq. sol. of $Fe_2(SO_4)_3$	◎	◎	◎	×	×	×	II	1~2 min.
6	5% aq. sol. of $CuCl_2 \cdot 2H_2O$	◎	◎	◎	×	×	×	II	3 min.

* In concentration relative to that of "concentrated" acid or saturated aqueous solution of salt as 100 percent.

and saturated aqueous solutions of ferric chloride or of ferric sulphate are most suitable because of the very shortness of required etching-time and of the easier location of the symmetric center of the {001} light figure. For the experiments described below, we employed light figures of the second group produced by etching with aqua regia, which are shown in Fig. 3. It is to be noted that the etching with 5 percent ferric chloride solution as used previously by Chalmers[5] has a disadvantage of requiring a long time (about 30 minutes).

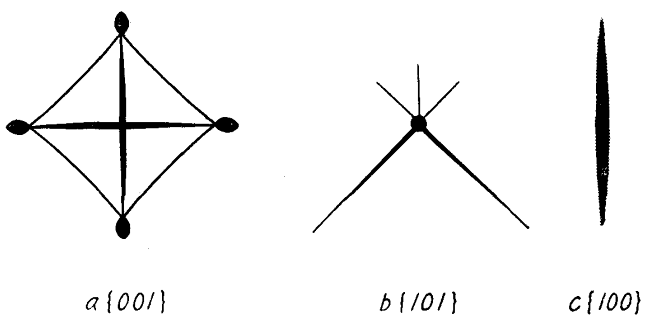

Fig. 3. Light figures of tetragonal tin crystals etched with concentrated aqua regia as used for the orientation determination.

(8) As to the conditions required for etching directions, see reference (2).

C. Directly and accuaately measurable ranges and kinds of orientation angles.

The procedure and apparatus for the orientation determination of tetragonal tin crystals are the same as that employed previously for cubic and hexagonal crystals(2,3), and so the description of them is omitted here.

As discussed previously(2,4), the factors affecting the accuracy of the orientation determination by the light-figure method are the inclination of crystal plane producing a light figure to the specimen axis and the form and distinctness of the light figure concerned. With the second-group light figures produced by etching with concentrated aqua regia, the range in which θ can be determined accurately by the {001} light figure is 90～70°, the range in which α or β can be determined accurately by the {100} light figure is 90～40°, and the range in which α', β', γ', or δ' can be determined accurately by the {101} light figure is 110～40°, as will be seen from actual examples given in section III. On the other hand, with the first-group light figures produced by etching with 95 : 5 mixture of hydrochloric acid and chromic acid anhydride, the {301} and {211} light figures are also useful besides the {001}, {100} and {101} light figures, and the range in which angles can be determined accurately by the {301} and {211} light figures are 130～40° and 105～70°, respectively, although the accurately measurable range of θ is somewhat narrower than that with the {001} light figure of the second group.

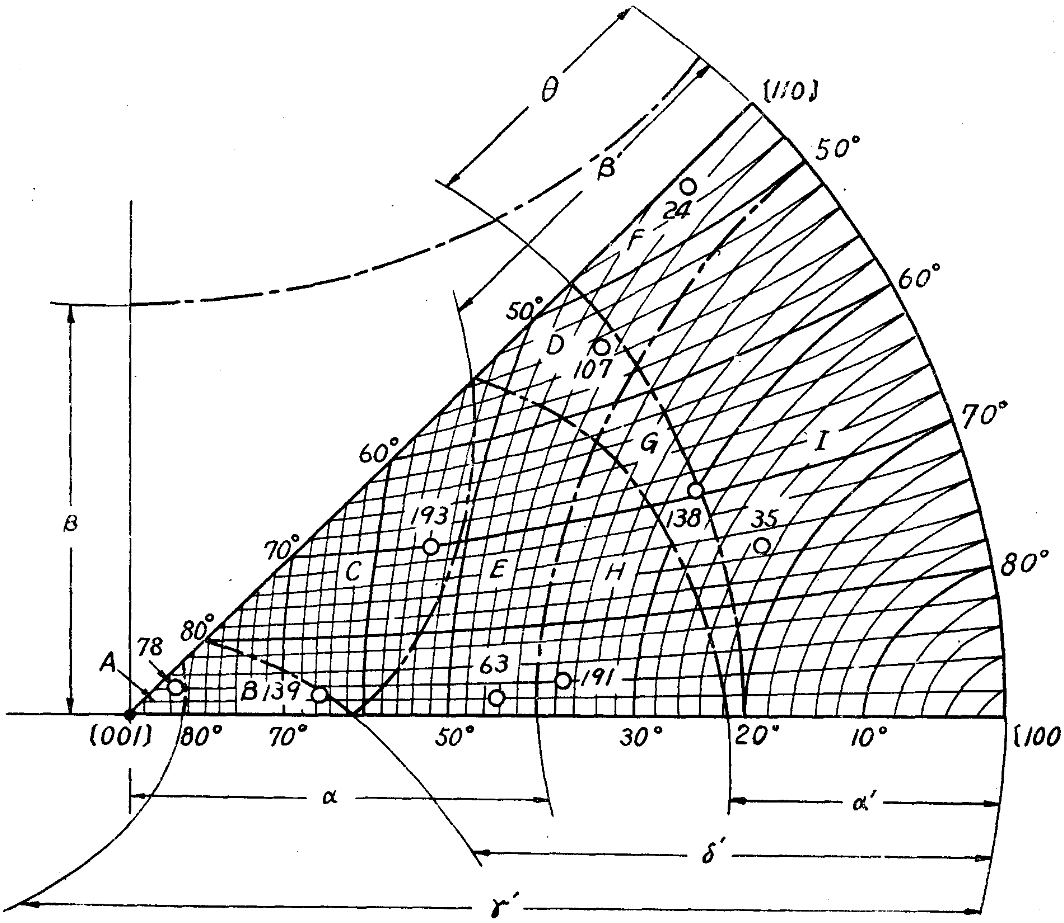

Fig. 4. Stereographic representation of the measurable ranges and kinds of orientation angles of tin crystal rods which can be determined by the use of light figures of the second group.

As just mentioned, the ranges of orientation angles which can be determined directly and accurately by light figures are limited to relatively narrow ranges, and accordingly the kind and number of orientation angles measurable with a crystal specimen vary with crystal orientation to be determined, of which the relationships are illustrated in Fig. 4 and Table 2. The (001)-(100)-(110) stereographic triangle

Table 2. Tabulation of the orientation ranges for tetragonal tin crystal rods where the kind and number of light figures applicable to the accurate orientation determination or of orientation angles accurately determinable by the use of light figures of the second group differ.

Mark of the orientation ranges	Kind and number of light figures applicable to the accurate orientation determination or of angles accurately determinable by the use of light figures of the second group		
	Number	Light figures	Orientation angles
A	2	(100), (010)	α, β
B	3	(100), (010), $(\bar{1}01)$	α, β, γ'
C	4	(100), (010), $(\bar{1}01)$, $(0\bar{1}1)$	α, β, γ', δ'
D	6	(100), (010), (101), (011), $(\bar{1}01)$, $(0\bar{1}1)$	α, β, α', β', γ', δ'
E	5	(100), (010), (011), $(\bar{1}01)$, $(0\bar{1}1)$	α, β, β', γ', δ'
F	7	(001), (100), (010), (101), (011), $(\bar{1}01)$, $(0\bar{1}1)$	θ, α, β, α', β', γ', δ'
G	5	(010), (101), (011), $(\bar{1}01)$, $(0\bar{1}1)$	β, α', β', γ', δ'
H	4	(010)· (011), $(\bar{1}01)$, $(0\bar{1}1)$	β, β', γ', δ'
I	6	(001), (010), (101), (011), $(\bar{1}01)$, $(0\bar{1}1)$	θ, β, α', β', γ', δ'

is divided into nine regions A~I, in which the number of accurately measurable orientation angles or of useful light figures vary from two to nine. Consequently, it may readily be seen that the prefect and accurate determination of orientations can be made for crystals in any orientation region and the check of determination is possible for crystals in most regions where three or more orientation angles can always be measured accurately. It is to be remarked that, for crystals in regions G and H, only an angle β among θ, α and β can be determined (by the use of the (010) light figure) and so any one more light figure other than the {001} and {100} light figures should be used for the perfect determination of their orientations, as was seen in Section IIA.

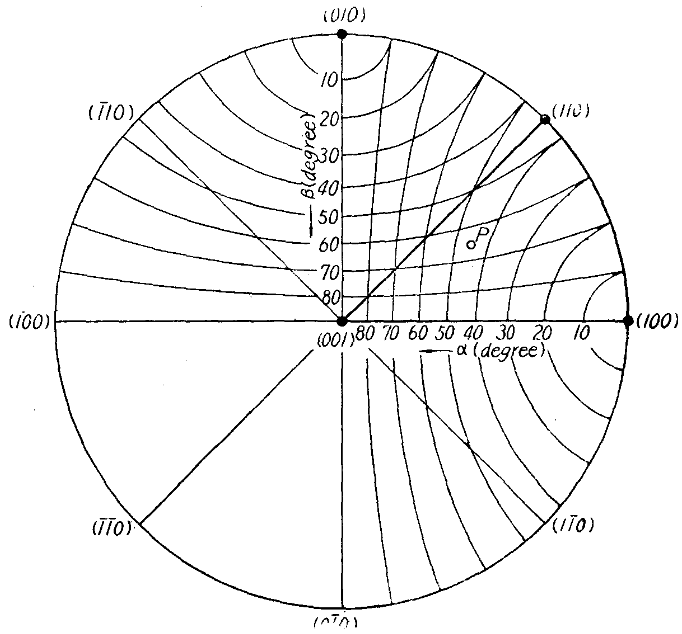

Fig. 5. Standard stereographic projection of tetragonal crystal. Plane of projection : (001).

D. *Stereographic representation of crystal orientations*

It is convinient for mutual comparison to represent the determined crystal orientations stereographically. For this purpose, we employ usually one of eight triangles as made by drawing great circles through (001), {100} and {110} poles on a standard stereographic projection projected on the (001) plane, as shown in Fig. 5. Crystal orientations are plotted on this stereographic triangle by two angles α and β which the specimen axis makes with the [100] and [010] axes, respectively. The point P in Fig. 5 represents the orientation of $\alpha=45°$ and $\beta=65°$. It is to be noted that angle θ cannot be known directly from such plotting, but it can be read by the use of the polar net or calculated by means of Eq. (1).

III. Actual examples and accuracy of the orientation determination

Single crystals of tetragonal tin used as specimens are round-bar crystals of 5 mm in diameter and several cm long, as prepared by the suction method[(9)] and by the Bridgman method[(10)]. Specimen crystals were etched with concentrated aqua regia

Table 3. An example of the actual determination of crystal orientation of a tin single crystal rod by the light-figure method. Crystal No. 35 (Diameter 4.5 mm; length 4.0 cm)

(a) The results of angle measurements.

Measured point	θ degree	β degree	α' degree	β' degree	γ' degree	δ' degree
1	73.0 72.7 73.2	74.6 74.7 74.8	45.8 45.6 45.6	67.8 67.8 67.8	100.6 100.5 100.6	82.5 82.5 82.6
2	73.1 73.1 73.0	74.7 74.8 74.6	45.8 45.6 45.8	67.6 67.7 67.6	100.3 100.2 100.2	82.3 82.4 82.4
3	72.9 72.9 73.0	74.9 74.8 75.0	45.4 45.8 45.4	67.6 67.8 67.6	100.5 100.4 100.4	82.4 82.4 82.4
Final mean value	73.0	74.8	45.6	67.7	100.4	82.4

(b) The results of orientation calculations.
(M) and (C) mark, respectively, the measured and the calculated values.

θ degree	α degree	β degree	α' degree	β' degree	γ' degree	δ' degree	Formula used for the calculation
73.0(M)	23.1(C)	74.8(M)	—	—	—	—	(1)
73.0(M)	22.3(C)	76.0(C)	45.6(M)	—	—	—	(3)
73.0(M)	23.0(C)	75.0(C)	—	67.6(M)	—	—	(3)
73.0(M)	24.1(C)	73.4(C)	—	—	100.4(M)	—	(3)
73.0(M)	23.0(C)	74.9(C)	—	—	—	82.4(M)	(3)
72.4(C)	25.2(C)	74.8(M)	45.6(M)	—	—	—	(5)
73.1(C)	23.1(C)	74.8(M)	—	67.6(M)	—	—	(4)
72.8(C)	23.3(C)	74.8(M)	—	—	100.4(M)	—	(5)
72.9(C)	23.2(C)	74.8(M)	—	—	—	82.4(M)	(4)
72.9(C)	22.7(C)	75.2(C)	45.6(M)	67.6(M)	—	—	(6)
72.8(C)	23.0(C)	74.9(C)	45.6(M)	—	100.4(M)	—	(8)
72.6(C)	23.6(C)	74.4(C)	45.6(M)	—	—	82.4(M)	(7)
72.7(C)	22.9(C)	75.4(C)	—	67.6(M)	100.4(M)	—	(7)
73.0(C)	23.0(C)	75.0(C)	—	67.6(M)	—	82.4(M)	(8)
72.8(C)	23.3(C)	74.6(C)	—	—	100.4(M)	82.4(M)	(6)

(9) M. Yamamoto and J. Watanabé, N.K.G., **B14** (1950), No. 10; Sci. Rep. RITU, **A3** (1951), 165.
(10) The preparation of single crystals of tetragonal tin by the Bridman method will be reported in a separated paper.

for about 30 seconds and then mounted on the apparatus(2) for the orientation determination. Orientations were determined at two or three positions along an uniform part of the length of a crystal rod. Determinations were made thrice at each position and the mean value was taken as the final datum. An example of measurements of angles is shown in Table 3(a), which indicates that the accuracy of angle determination is within 0.1°. Table 3(b) shows, for the same crystal specimen as in Table 3(a), the results of calculation of orientation angles from any two of the measured angles by means of Eqs. (1)~(8) (excluding Eq. (2)). It can be seen that the measured and calculated values of each orientation angle are in good agreement with each other and the orientation determination can be made with an accuracy well within 1°. Other various actual examples of orientation determination are shown in Table 4 and plotted in Fig. 4, which confirm the results of consideration made in Section IIC.

Table 4. Other various examples of crystal orientations of tin single crystal rods, as determined by the light-figure method. (M) and (C) mark, respectively, the measured and the calculated values.

Crystal No.	Diameter mm	Length cm	No. of measured points	θ degree	α degree	β degree	α' degree	β' degree	γ' degree	δ' degree	Angles and the formula used for the calculation
24	4.3	3.0	2	84.8(M	44.8(M)	45.7(C)	65.1(M)	65.4(M)	101.4(M)	104.5(M)	θ and α; (1)
63	5.1	4.2	3	45.8(C)	44.3(M)	88.0(M)	—	51.4(M)	74.4(M)	53.7(M)	α and β; (1)
78	4.3	3.0	2	8.8(C)	82.3(M)	85.8(M)	—	—	—	—	α and β; (1)
107	6.0	4.0	3	68.8(C)	42.9(M)	54.8(M)	47.3(M)	53.0(M)	91.0(M)	86.8(M)	α and β; (1)
138	5.5	2.5	2	69.2(C)	29.8(C)	69.6(M)	43.4(M)	61.8(M)	96.6(M)	82.0(M)	β and α'; (5)
139	5.3	3.0	2	25.1(C)	65.2(M)	86.4(M)	—	—	53.5(M)	—	α and β; (1)
191	6.0	3.8	3	52.7(C)	37.6(C)	85.9(M)	—	55.5(M)	80.8(M)	59.9(M)	β and β'; (4)
193	6.2	3.7	3	43.4(C)	53.5(M)	69.9(M)	—	—	69.3(M)	61.8(M)	α and β; (1)

IV. Comparison of the light-figure method with the X-ray diffraction method

It is needless to say that the fundamental method for the orientation determination is the X-ray diffraction method, which, however, involves disadvantages requiring the laborious procedure and long-time consumption. We determined orientations of the same single crystal rods of tetragonal tin by the light-figure method as well as by an X-ray diffraction method and compared their usefulness for the orientation determination. The X-ray diffraction method employed was the back-reflection Laue method, and crystal orientations were determined from diffraction angles of the 010 and 130 spots of Laue patterns obtained. Experimental results are shown in Table 5, from which it may be seen that the crystal orientations as determined by the light figure method are in good agreement with those determined by the back-reflection Laue method. Thus, the accuracy of the orientation determination by the light-figure method has been found as high as X-ray diffraction method. It is to be noted, however, that we have experienced following disadvantages with the X-ray diffraction technique: (1) A long time is required

Table 5. Comparison of the orientation determination of single crystal rods of tetragonal tin as determined by the light-figure method as well as by the back-reflection Laue method. (M) and (C) denote measured and calculated values, respectively.

Crystal No.	Orientation	By the light-figure method	By the back-reflection Laue method
62	θ	80.2°(M)	79.7°(C)
	α	12.6°(C)	13.1°(M)
	β	82.2°(M)	81.9°(C)
192	θ	79.1°(M)	79.2°(C)
	α	10.9°(C)	11.2°(M)
	β	89.1°(M)	87.1°(C)

for photographing diffraction patterns, the analysis of them, and the laborious calculation, (2) the determination becomes sometimes inaccurate unless a specimen crystal is set in an X-ray camera in a definite orientation with respect to the direction of incident X-ray beam, and (3) with a few crystals of special orientations, Laue patterns can hardly appear so that the procedure must be repeated under different conditions. On the other hand, the light-figure method has none of these difficulties and is felt to be more useful for the rapid, accurate, and perfect determination of crystal orientations. The comparison of the light-figure and the back reflection Laue methods for the orientation determination will be discussed more fully in a later paper.

Appendix

Derivation of the geometrical relationships required for the orientation determination of tetragonal tin crystals by the light-figure method.

Since the principal crystallographic axes [001], [100] and [010] in a tetragonal lattice meet at right angle with each other and {101} planes belong to the {001}-{100} (or the <100>) zones, there exist following angular relationships among the normal directions of {101} planes and principal crystal axes:

	(101)	(011)	($\bar{1}$01)	(0$\bar{1}$1)
[001]	ω	ω	ω	ω
[100]	$90°-\omega$	90°	$90°+\omega$	90°
[010]	90°	$90°-\omega$	90°	$90°+\omega$

Then, if the [001], [100] and [010] axes are taken as the Z, X and Y axes of a rectangular coordinate system, the direction cosines of the normals of {101} planes referred to this coordinate system are as follows:—

Crystal orientation	Direction cosines		
(101)	$\sin\omega$	0	$\cos\omega$
(011)	0	$\sin\omega$	$\cos\omega$
($\bar{1}$01)	$-\sin\omega$	0	$\cos\omega$
(0$\bar{1}$1)	0	$-\sin\omega$	$\cos\omega$

From angular relationships between the direction $\overrightarrow{OP}$, of which the direction cosines are (l, m, n), and normals of {101} planes, following equations are derived:

$$\left.\begin{aligned} n &= \cos\theta, \\ l &= \cos\alpha, \\ m &= \cos\beta; \end{aligned}\right\} \quad (9)$$

$$\left.\begin{aligned} l\sin\omega + n\cos\omega &= \cos\alpha', \\ m\sin\omega + n\cos\omega &= \cos\beta', \\ -l\sin\omega + n\cos\omega &= \cos\gamma', \\ -m\sin\omega + n\cos\omega &= \cos\delta', \end{aligned}\right\} \quad (10)$$

and then Eq. (10) may be written as

$$\cos\alpha\sin\omega + \cos\theta\cos\omega = \cos\alpha', \quad (11a)$$

$$\cos\beta\sin\omega + \cos\theta\cos\omega = \cos\beta', \quad (11b)$$

$$-\cos\alpha\sin\omega + \cos\theta\cos\omega = \cos\gamma', \quad (11c)$$

$$-\cos\beta\sin\omega + \cos\theta\cos\omega = \cos\delta'. \quad (11d)$$

Among the direction cosines of direction $\overrightarrow{OP}$ holds the relationship $l^2+m^2+n^2=1$, so that Eq. (1) may be obtained readily from Eq. (9).

Eqs. (3) and (4) may be obtained by seeking $\cos\alpha$ or $\cos\theta$ as a function of $\cos\theta$ and $\cos\alpha'$ or of $\cos\alpha$ and $\cos\alpha'$ from Eqs. (11a~d) and then by solving quadratic equations produced by substituting each solution into Eq. (1).

The substitution of Eqs. (11a~d) into Eq. (1) yields quadratic equations. Solving each quation with respect to $\cos\theta$ and substituting the obtained solution into Eqs. (11a~d) yield Eq. (5).

From Eqs. (11a~d) $\cos\theta$ is elliminated by the substraction or addition of any two expressions. Then, the substitution of the obtained equation together with Eqs. (11a~d) into Eq. (1) yields a quadratic equation, the solutions of which are Eqs. (6), (7) or (8).

Summary

The application of the light-figure method to the orientation determination of single crystal rods of tetragonal tin has been investigated and the procedure for the perfect, accurate, and rapid determination has been worked out. Orientations of tetragonal crystals can be fixed perfectly by the specification of angles θ, α and β which the geometrical axis makes with the [001], [100], and [010] axes, respectively. θ, α and β may be determined directly by means of the (001), (100) and (010) light figures, respectively. The accuracy in the orientation determination by light figures, however, is influenced by the inclination of a crystal plane producing the light figure to the rod axis, so that in some cases any one more light figure other than the {001} and {100} light figures, namely {101} or {301} or {211} light figure, and one of the {001} and {100} light figures must be used for the perfect and accurate determination. So, the necessary geometrical relationships held among θ, α, and β as well as α', β', γ' and δ', which can be determined by the {101} or {301} light figures, were derived.

The ranges of orientation angles to be determined accurately by the use of the {001}, {100} and {101} light figures produced by etching with concentrated aqua regia (the most suitable etching for orientation determination) are $90^\circ > \theta > 70^\circ$, $90^\circ > \alpha$ and $\beta > 40^\circ$ and $110^\circ > \alpha'$, β', γ' and $\delta' > 40^\circ$. Consequently, the kind and number of accurately measurable orientation angles or of useful light figures vary with orientation to be determined, of which the relationships are shown in Fig. 3 and Table 1. The (001)-(100)-(110) stereographic triangle is divided into nine regions, in which any two or more angles of θ, α, β, α', β', γ' and δ' are always determinable and thus the perfect and accurate determination can be made with crystal of any orientation.

The procedure and accuracy of the orientation determination by light figures were illustrated with reference to various actual examples and then compared with an X-ray diffraction method. It has been shown that the light-figure method can be made with an accuracy as high as that of the back-reflection Laue method and that the former method is rather more excellent in its simple and rapid procedure than the latter one.

In conclusion, it is to be noted that a partial fund required for this work was furnished from the Grant in Aid for Fundametal Scientific Researches of the Ministry of Education.