A Simplified Analytical Determination of Cubic Crystal Orientation from Data on \{111\} Surface Traces

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The analytical treatments which have so far been developed for the evaluation of cubic crystal orientations from \{111\} surface trace data are rather complex. Another approach is produced here which is, in comparison, much simpler.

**Introduction**

Traces on a crystal surface refer to such markings as twin boundaries, slip lines, and edges of etch pits and plate-shaped precipitates which delineate the lines of intersections of certain crystallographic planes with the crystal surface. If the delineated planes are known then the disposition of the traces gives information as to what the crystal orientation could be. The attractiveness of determining crystal orientations from traces is that the technique is relatively simple, the measurement of the directions of the traces being readily performed even when crystal sizes are so small (as in polycrystalline materials) that X-ray methods are not possible. The disadvantage is that it is not always clear whether the intersections of particular crystallographic planes with the crystal surface are accurately outlined by the traces. Further, small errors in the measured directions of the traces can cause large errors in the evaluated orientation for certain orientations (Takeuchi, Honma & Ikeda, 1959). In addition, the method is applicable in the practical situation only to cases where the delineated planes are of low multiplicity, such as \{100\} and \{111\} in cubic crystals; otherwise the procedure becomes too complicated from there being too many plane variants to consider. Fortunately, the delineated planes are usually of simple form.

There are several ways of obtaining the crystal orientation after the initial trace measurements have been made. Probably the most established method is that described by Barrett & Massalski (1966). It consists of attempting to rotate the poles of the trace-delineated planes as given on a standard stereographic projection into new positions lying on diameters of the basic circle which are perpendicular to the trace directions. The new positions of the poles give the crystal orientation. The correct rotation is found by trial and error, and for this reason the method is laborious and uncertain and contributes further errors to the crystal orientation determination. The trial-and-error factor is eliminated in the semi-graphic approach of Drazin & Otte (1963) where in the case of delineated \{111\} planes of cubic crystals the locus of a \{111\} pole consistent with two trace directions taken to be of two other \{111\} plane variants is first computed and subsequently mapped out on a stereographic plot whose projection plane is the crystal surface with the traces. The intersections with this locus of the normals to the third and fourth trace directions then give the location of the corresponding \{111\} poles. This information leads to the crystal orientation. According to Drazin & Otte the method is quite quickly applied and is suitable when great accuracy is not required. The method appears to be a development of an earlier treatment by Mykura (1958).

In another approach charts or tables relating angular disposition of traces to orientation of the crystal surface are first constructed. From these, in any particular case when the trace measurements have been made, the crystal surface orientation may be quickly read off, and with this information the complete crystal orientation may be determined by further straightforward procedures. Such charts for the case of delineated \{100\} and \{111\} planes of cubic crystals have been produced by Takeuchi, Honma & Ikeda (1959). The case of \{110\} planes have been treated by Tsubaki & Nishiyama (1960). Tables associating angular trace dispositions with crystal surface orientations have also been produced for cubic crystals for the case of delineated \{100\} planes (Tucker & Murphy, 1953) and \{111\} planes (Drazin & Otte, 1964). The above charts and tables are useful when many orientation determinations are to be carried out, but the accuracy is limited by the extent of divising employed in them.

An analytical treatment (if possible) would give precise results provided of course high precision is maintained in the computation. The great advantage of an analytical approach is that it will provide equations and mathematical relationships which are readily programmed on a computer so that effortlessly rapid, exact, and repetitious orientation computations may be done. [A computerized method of successive approximations has also been proposed (Drazin & Otte, 1963) but it requires a prior knowledge of initial approximate solutions]. The analytical equations and relationships for obtaining crystal orientations from data on surface traces are obtained with relative ease in the case of delineated \{100\} planes of cubic crystals.
(Tucker & Murphy, 1953) but with great difficulty in the case of delineated \{111\} planes of cubic crystals where complex equations and relationships have been derived by Drazin & Otte (1963) and Fong (1973). It would be advantageous if for the latter case, which occurs frequently in metals, the analysis could be simplified so that it might be more readily followed and the process of precisely determining crystal orientations from trace data made easier. This then is the aim of this paper: the development of an analytical treatment considerably simpler than those of Drazin & Otte and Fong for the evaluation of cubic crystal orientations from \{111\} trace information.

**Derivation of crystal orientations from \{111\} trace data**

In Fig. 1 plane $ABC$ represents the surface of a cubic crystal on which are observed \{111\} traces $AB$, $BC$, and $CA$ making angles $\hat{A}$, $\hat{B}$, and $\hat{C}$ with one another. $\hat{A}$, $\hat{B}$, and $\hat{C}$ are measurable quantities and constitute the trace information from which crystal orientations are to be evaluated.

Imagine now in Fig. 1 \{111\} planes $ABP$, $BCP$, and $CAP$ passing through traces $AB$, $BC$, and $CA$ and intersecting at a point $P$. Let:

1. $PH$ be perpendicular to plane $ABC$, $H$ being a point in plane $ABC$;
2. $\hat{P}_1$, $\hat{P}_2$, $\hat{P}_3$, $\hat{Q}_1$, $\hat{Q}_2$, and $\hat{Q}_3$ be the angles indicated;
3. $a$, $b$, $c$, $k_0$, $t_0$, and $h$ be the lengths indicated;
4. $CP=1$;
5. $0^\circ < \hat{A}, \hat{B}, \hat{C} < 180^\circ$.

For the above situation Fong (1973) has shown that either $\hat{P}_1$, $\hat{P}_2$, and $\hat{P}_3$ are all $60^\circ$, or one of them $60^\circ$ and the other two $120^\circ$. So we have:

$$\cos \hat{P}_1 = \frac{j_1}{2}, \quad \cos \hat{P}_2 = \frac{j_2}{2}, \quad \cos \hat{P}_3 = \frac{j_1 j_2}{2},$$

where

$$j_1 = \pm 1, \quad j_2 = \pm 1.$$  \hspace{1cm} (2)

We obtain then from the pyramidal figure $ABCP$ in Fig. 1:

$$a^2 = t^2 - t + 1$$ \hspace{1cm} (3)

$$b^2 = k^2 - k + 1$$ \hspace{1cm} (4)

$$c^2 = k^2 - k t + t^2.$$ \hspace{1cm} (5)

where

$$k = j_2 k_0, \quad t = j_1 t_0.$$ \hspace{1cm} (6)

From equations (3) and (4),

$$t^2 - t + 1 = (\sin \hat{A}/\sin \hat{B})^2 (k^2 - k + 1).$$ \hspace{1cm} (7)

Hence

$$t = \frac{1}{2}(1 \pm \sqrt{[4(\sin \hat{A}/\sin \hat{B})^2 (k^2 - k + 1) - 3]}).$$ \hspace{1cm} (8)

From equations (4) and (5),

$$(\sin \hat{C}/\sin \hat{B})^2 (k^2 - k + 1) = k^2 - k t + t^2.$$ \hspace{1cm} (9)

Substituting the value of $t^2$ given by equation (7) into equation (9) and noting that

$$1 + \frac{\sin^2 \hat{A} - \sin^2 \hat{C}}{\sin^2 \hat{B}} = 2 \sin \hat{A} \cos \hat{C},$$

we obtain

$$- \frac{2 \sin \hat{A} \cos \hat{C}}{\sin \hat{B}} (k^2 - k + 1) - k + 2 = (1 - k)t.$$ \hspace{1cm} (10)

On putting in the value of $t$ given by equation (8) and letting

$$x = 1 - k$$ \hspace{1cm} (11)

we obtain from equation (10):

$$\frac{4 \sin \hat{A} \cos \hat{C}}{\sin \hat{B}} (x^2 - x + 1) - x = 2 \left[ x \sqrt{[4(\sin \hat{A}/\sin \hat{B})^2 (x^2 - x + 1) - 3]} \right].$$ \hspace{1cm} (12)

This, on squaring, gathering like terms and simplifying gives

$$\sin^2 \hat{A}(4 \cos^2 \hat{C} - 1) x^4 - \sin \hat{A} \sin \hat{A} (8 \cos^2 \hat{C} - 1)$$
$$+ 2 \sin \hat{B} \cos \hat{C} x^3 + [2 \sin \hat{A} (6 \cos^2 \hat{C} - 1)]$$
$$+ \sin^2 \hat{C} x^2 - \sin (\hat{A} - \hat{C}) (4 \sin \hat{A} \cos \hat{C} + \sin \hat{B}) x$$
$$+ \sin^2 (\hat{A} - \hat{C}) = 0.$$ \hspace{1cm} (13)
Except for the special case of $\hat{C}=60$ or $120^\circ$, equation (13) is a quartic equation in $x$ whose solutions are readily obtained by established analytical methods. When $\hat{C}=60$ or $120^\circ$ the coefficient of $x^4$ vanishes and equation (13) reduces to a cubic equation which may again be readily solved analytically.

For each real value of $x$ obtained from equation (13) the value of $k$ is given by:

$$k = 1 - x.$$  \hspace{1cm} (14)

Provided $x \neq 0$ the corresponding value of $t$ may be found from equation (10) which gives

$$t = \frac{1}{x} \left[ 1 + x - \frac{2 \sin A \cos \hat{C}}{x^2 - x + 1} \right].$$  \hspace{1cm} (15)

The case of $x = 0$, $k = 1$, arises when $\hat{C} = \hat{A}$ so that the constant and $x$ terms in equation (13) vanish. In this case the value of $t$ is obtainable from equation (8) which gives:

$$t = \frac{1}{2} \left[ 1 \pm \left( \tan^2 \hat{A} - 2 \right) \right].$$  \hspace{1cm} (16)

Both values of $t$ given by the above equation are valid as they each satisfy both equations (7) and (9) which are in fact identical when $\hat{C} = \hat{A}$ and $k = 1$ and therefore the geometry of the pyramidal figure $ABCP$ in Fig. 1. It is possible to prove that both values of $t$ lead to the same crystal orientations but this will not be shown here.

Having found $k$ and $t$ we may next ascertain the values of $j_1$ and $j_2$ from

$$j_1 = t/|t|, \quad j_2 = k/|k|.$$  \hspace{1cm} (17)

It has been shown by Fong (1973) that in Fig. 1

$$\cos^2 \hat{Q}_3 = 1 - \frac{\cos^2 ACP + \cos^2 BCP - 2 \cos ACP \cos BCP \cos \hat{C}}{\sin^2 \hat{C}}.$$  \hspace{1cm} (18)

Now,

$$\cos ACP = \frac{1 + b^2 - k^2}{2b} = 2 - k$$  \hspace{1cm} (19)

$$\cos BCP = \frac{1 + a^2 - t^2}{2a} = 2 - t.$$  \hspace{1cm} (20)

Hence

$$h = \cos \hat{Q}_3 = \sqrt{1 - \frac{(2-k)^2 \sin^2 \hat{A} + (2-t)^2 \sin^2 \hat{B} - 2(2-k)(2-t) \sin \hat{A} \sin \hat{B} \cos \hat{C}}{4(k^2 - k + 1) \sin^2 \hat{A} \sin^2 \hat{C}}}.$$  \hspace{1cm} (21)

Also we have

$$\cos \hat{Q}_2 = h/j_1 t$$  \hspace{1cm} (22)

$$\cos \hat{Q}_1 = h/j_2 k.$$  \hspace{1cm} (23)

If $(v_1, v_2, v_3)$ is the unit vector representing the crystallographic direction along $\overrightarrow{HP}$ then, as shown by Fong (1973),

$$v_1 = (-j_2 \cos \hat{Q}_1 + j_1 \cos \hat{Q}_2 + \cos \hat{Q}_3)/|2$$  \hspace{1cm} (24)

$$v_2 = (j_2 \cos \hat{Q}_1 - j_1 \cos \hat{Q}_2 + \cos \hat{Q}_3)/|2$$  \hspace{1cm} (25)

$$v_3 = (\cos \hat{Q}_1 + j_1 j_2 \cos \hat{Q}_2 - j_2 \cos \hat{Q}_3)/|2$$  \hspace{1cm} (26)

where the $(110)$ directions of the crystal parallel to $\overrightarrow{CP}, \overrightarrow{AP}$, and $\overrightarrow{BP}$ in Fig. 1 have been taken to be $[110]$, $[0j_2 1]$, and $[j_1 0j_2 j_2]$ respectively. Substituting in the values of $\cos \hat{Q}_1, \cos \hat{Q}_2$, and $\cos \hat{Q}_3$ given by equations (21) to (23),

$$v_1 = \frac{h}{|2} \left( \frac{1}{k} - \frac{1}{t} + 1 \right)$$  \hspace{1cm} (27)

$$v_2 = \frac{h}{|2} \left( \frac{1}{k} - \frac{1}{t} + 1 \right)$$  \hspace{1cm} (28)

$$v_3 = \frac{j_2 h}{|2} \left( \frac{1}{k} - \frac{1}{t} + 1 \right).$$  \hspace{1cm} (29)

Thus $(v_1, v_2, v_3)$ which gives the crystal surface orientation is known in terms of $j_1$, $h$, $k$, and $t$ which are quantities seen earlier to be determinable from knowledge of the inter-trace angles $\hat{A}$, $\hat{B}$, and $\hat{C}$.

The crystal orientations consistent with the three trace directions $AB$, $BC$ and $CA$ in Fig. 1 may now be evaluated along identical lines with those of Fong (1973). Choosing for our frame of reference the right-handed rectangular coordinate system $OXYZ$ with axis $OX$ parallel to $\overrightarrow{CA}$ and axis $OZ$ parallel to $\overrightarrow{HP}$ in Fig. 1 we thus find that any crystallographic direction representable by a unit vector $u$ is given in the reference system $OXYZ$ by the unit vector $U$ where

$$U = Mu$$  \hspace{1cm} (30)

$$M = \begin{pmatrix}
\frac{v_3 + j_2 v_2}{w_1} & \frac{v_3 - j_2 v_1}{w_2} & \frac{-v_1 - v_2}{w_3} \\
\frac{j_1 w_1}{w_1} & \frac{j_1 w_2}{w_1} & \frac{j_1 w_3}{w_1} \\
\frac{v_1 w_1 - 1}{w_1} & \frac{v_2 w_1 + 1}{w_1} & \frac{v_3 w_1 - j_2}{w_1} \\
\frac{-j_2}{w_1} & \frac{j_2}{w_1} & \frac{j_2}{w_1}
\end{pmatrix}.$$  \hspace{1cm} (31)

Thus a point overlooked...
For each real value of $x$ obtained by solving equation (13) the transformation equation (30) gives two possible crystal orientations which are mirror reflexions of each other in the crystal surface. Since equation (13) may yield as many as four real values of $x$ there may be as many as four pairs of mirror-image orientations which could give rise to three observed {111} trace directions as has been previously noted in the treatments of Mykura (1958), Drazin & Otte (1963), and Fong (1973). The fourth {111} trace direction may be used to reduce the number of possibilities of crystal orientation to one pair of mirror images for this trace direction will be that of $(1 \, 1 \, j_2)$ and the angle $\theta$ which it makes with the trace direction $\overrightarrow{CA}$ (measured clockwise about $\overrightarrow{HP}$) for each possible pair of mirror orientations may be determined with the help of equation (30) which gives

$$\theta = \tan^{-1} \left[ \frac{1}{j_1} \left( \frac{j_2 v_3 - v_1}{v_3 - j_2 v_1} \right) \right] + 90^\circ. \quad (32)$$

Using equation (32) to compute $\theta$ for each of the possible pairs of mirror-image crystal orientations and matching these computed $\theta$ values with the actually observed value we will be able to distinguish the pair of mirror orientations which are in accord with all four observed trace directions. This is the limit of solution possible with a single surface trace analysis. To further identify the right orientation from this pair of mirror orientations a second surface study will have to be undertaken or the single surface study will have to be compounded with some other observations such as ascertaining the directions of inclination of etch pit faces.

**Conclusion**

In the present treatment the steps to be taken to evaluate crystal orientations from {111} surface traces may be summarized as follows.

(i) Consider first three trace directions making angles $\hat{A}$, $\hat{B}$, and $\hat{C}$ with one another as in Fig. 1 and solve the following polynomial equation for $x$:

$$4(k^2 - k + 1) \sin^2 \hat{A} \sin^2 \hat{C}$$

(ii) For each real value of $x$ obtained determine the corresponding values of the quantities $k, t, j_1, j_2, h$ and the crystal surface orientation $(v_1, v_2, v_3)$ from:

$$k = 1 - x$$

$$t = \frac{1}{x} \left[ 1 + x - \frac{2 \sin \hat{A} \cos \hat{C}}{\sin \hat{B}} (x^2 - x + 1) \right], \quad \text{if } x \neq 0$$

$$t = \frac{1}{2} \left[ 1 + \frac{1}{(\tan \theta \hat{A} - 2)} \right], \quad \text{if } x = 0$$

$$j_1 = \frac{t}{\sqrt{t}}$$

$$j_2 = k |k|$$

Using equation (32) to compute $\theta$ for each of the possible pairs of mirror-image crystal orientations and matching these computed $\theta$ values with the actually observed value we will be able to distinguish the pair of mirror orientations which are in accord with all four observed trace directions. This is the limit of solution possible with a single surface trace analysis. To further identify the right orientation from this pair of mirror orientations a second surface study will have to be undertaken or the single surface study will have to be compounded with some other observations such as ascertaining the directions of inclination of etch pit faces.

(iii) For each surface orientation obtained compute the angle $\theta$ which the fourth {111} trace direction would make with the trace direction $\overrightarrow{CA}$ (Fig. 1) measured clockwise about $\overrightarrow{HP}$, $\theta$ being given by

$$\tan \theta = \frac{j_1 (j_2 v_3 - v_2)}{v_3 - j_2 v_1}.$$
identifying the correct pair of mirror orientations). They will certainly provide for simpler calculation procedures. Nevertheless the present treatment has not that special advantage of the treatment of Fong where, provided there are four sufficiently precise \{111\} trace directions, a more expeditious solution arises from the fact that two quartic equations in a common unknown \(y\) (the counterpart of \(x\) in the present work) could then be established by considering first one set of three trace directions and then a second set from the four available trace directions. In this way \(y^4\), \(y^3\), and \(y^2\) may be eliminated between the two quartic equations and a linear equation in \(y\) results so that \(y\) is readily and uniquely obtained without having to solve a quartic equation.

References


Skew-Reflection X-ray Microscopy of the Vapor-Growth Surface of an \(\text{Al}_2\text{O}_3\) Single Crystal

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The most commonly used geometry for the Berg-Barrett X-ray microscopy uses the zero-layer reflections as described by Newkirk. It can be shown that non-zero-layer reflections, skew-plane reflections, can be used equally well to obtain X-ray micrographs. The analysis of the stereographic representation of the skew-reflection geometry demonstrates the many usable reflections and gives the conditions for minimum image distortion. In these X-ray micrographs the contributions to diffraction contrast from shadowing and sub-boundaries can be identified. An estimate of the height of steps occurring on the crystal surface can also be made.

Introduction

Armstrong (1965) has shown that the Berg–Barrett X-ray technique can be utilized to give information on both the surface topography and the dislocation substructure threading the underlying crystal volume of a vapor-condensed zinc crystal. Zero-layer reflections, as originally described by Newkirk (1959), were employed to obtain the Berg–Barrett micrographs. Because growth processes determine the crystal-surface orientations and it is desired that these surfaces not be damaged by cutting and polishing procedures, there is no guarantee that suitable zero-layer reflections will be available to study the ‘as-grown’ surface features. The macroscopic vapor-growth surface of an \(\text{Al}_2\text{O}_3\) crystal grown along the [0001] direction is such an example (Farabaugh, 1972). For these reasons non-zero-layer (Juleff, Lapierre & Wolfson, 1966; Turner, Vreeland & Pope, 1968), i.e. skew reflections, have been employed here to investigate the surface topography and the dislocation substructure of this crystal. The study has involved the following items: a stereographic-projection method for determining favorable skew reflections; an assessment of the surface shadowing or enhancement of diffracted X-ray intensity which occurs at surface steps; and, an analysis of image distortion which occurs within skew reflections.

Experimental details

The \(\text{Al}_2\text{O}_3\) crystal was produced by a vapor-transport technique previously described by Parker & Harding (1970). The crystal was grown at 1750°C at a total pressure of 4 torr according to the reaction:

\[
2\text{AlCl}_3(g) + 3\text{CO}_2(g) + 3\text{H}_2(g) \rightarrow \text{Al}_2\text{O}_3(s) + 6\text{HCl}(g) + 3\text{CO}(g).
\]

Under these conditions, the average deposition rate was 80 mg h\(^{-1}\) cm\(^{-2}\). A growth period of 30 h resulted in a crystal 20 mm long and 15 mm in diameter. The crystal-growth surface was found to be macroscopically composed of numerous steps and terraces. Measurements by optical goniometry and optical microscopy led to identification of the terraces as being nearly parallel to (0001) planes and the steps to be nearly parallel to \{10\1\} planes; thus the step edges run parallel to \langle1120\rangle. The largest steps were estimated to be of 50 \(\mu\)m in height.