X-RAY ANALYSIS OF PREFERRED ORIENTATION IN FINE-GRAINED QUARTZ AGGREGATES

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ABSTRACT

The preferred orientations of quartz in a number of small fine-grained specimens of experimentally deformed and recrystallized flint were determined with a Norelco pole figure goniometer. The device was modified by introducing small (0.5-1.0 mm.) collimators to examine restricted areas of flat specimens in both the reflection and transmission modes. Scans in the two modes were combined to yield complete pole figures. The preferred orientations in the specimens are shown to be axially symmetric and are given by intensity profiles extending 0°-90° from the symmetry axis. Profiles for as many as fifteen diffraction peaks from individual specimens are subjected to spherical harmonic analysis to obtain a complete determination of the preferred orientation (the crystallite distribution function), which is represented concisely in the form of an inverse pole figure. From the inverse pole figure the preferred orientation of any crystallographic direction (such as the [0001] direction in quartz, which is difficult to measure directly) may be generated. Similarly, the preferred orientations of positive and negative trapezohedral and rhombohedral forms (such as the pairs 3142-1342 and 1011-0111, respectively), which cannot be resolved by standard X-ray measurements, are separately determined. Earlier work with polymers and metals using these methods is reviewed. The methods are illustrated by means of two flint cylinders recrystallized during compression at high temperature and pressure; one (GB-3) is relatively homogeneous and the other (GB-11) shows marked variation in preferred orientation. There are differences in the orientations of the positive and negative forms in both specimens. The preferred orientations may be characterized as a mixture of two components: (a) parallelism of c = 0001 with the compression axis and (b) parallelism of the poles of the positive unit rhomb r = 1011 with the compression axis. The latter component dominates in the more coarsely recrystallized (hotter) regions of specimen GB-11.

INTRODUCTION

The methods described below for the determination of preferred orientations in quartz aggregates by X-ray techniques were developed as part of an experimental study of the development of preferred orientation in quartz. In the experiments, fine-grained aggregates of quartz (Dover flint and Arkansas novaculite) with initial grain size of a few microns are deformed and recrystallized at high temperature and pressure. The resulting aggregates are somewhat coarser (up to 100 μ) but are commonly too fine grained for standard optical measurements of the preferred orientation with the universal stage. It is desirable to determine the orientations of the crystals as completely as possible. U-stage techniques yield a complete determination of the orientation of a crystal only if the orientations of two or more crystallographic directions can be measured (for example, the principal optic directions in an orthorhombic crystal or the optic axis and an identifiable cleavage or twin plane in a uniaxial crystal). In quartz it is generally only possible to determine the orientation of the c-crystallographic (optic) axis. The orientations of the a-axes are unknown.

X-ray photographic techniques were used as early as 1930 to obtain information on the preferred orientations of minerals (Sander and Sachs, 1930), and more precise photographic methods have been introduced more recently (Wenk, 1963, 1965; Starkey,
In connection with the more extensive work on preferred orientations in metals, methods employing both photographic and diffractometer records have been developed (see, for example, the review by Barrett and Massalski, 1966). The photographic methods are well suited for qualitative studies, but counter methods are preferred for quantitative analysis of preferred orientations.

The first X-ray measurements of preferred orientation in the fine-grained, experimentally deformed quartz aggregates were made with an X-ray camera (Starkey, 1964), but quantitative data were not obtained from the films. The newer measurements discussed below were made with a modified commercial pole figure goniometer (Schulz, 1949), with the intensities recorded by a scintillation counter. The preferred orientations of numerous diffracting planes may be measured with high precision, and the distribution of the crystal axes and other crystallographic directions are derived from them by computation. The quantitative analysis of the data employs techniques developed in the study of metal and polymer textures (Roe and Krigbaum, 1964; Bunge, 1965a, b) which have not been applied before in the study of mineral orientations.

The methods are illustrated with results on two samples, GB-3 and GB-11 (pl. 1), from a systematic study of the development of preferred orientation in experimentally deformed flint (Green, 1966, 1967). The samples were cylinders of flint, initially 5 mm. in diameter and approximately 2 cm. long, deformed in compression by approximately 35 per cent at 780° C. and 15 kb. confining pressure in an apparatus designed by Griggs (1967); GB-3 at a strain rate of $10^{-5}$ sec$^{-1}$ and GB-11 at $10^{-6}$ sec$^{-1}$. The samples were cut in half longitudinally and a thin section for optical study prepared from one half; the cut surface of the other half was examined by X-ray in reflection and then ground to a slab 80μ thick for transmission studies. By combining reflection and transmission scans the complete pole figure could be determined. The samples show longitudinal variations of grain size and texture due to temperature gradients present during the experiments. Sample GB-3 (pl. 1A) was homogeneous in a central region of the section measuring $5 \times 5$ mm., and X-ray measurements were made in an area 3 mm. in diameter within this region. The thin section of GB-11 (pl. 1C), by contrast, shows marked gradients of texture and preferred orientation, and the preferred orientation was determined in several areas of 1.5 mm$^2$ along the length of the sample.

It is shown that the preferred orientations in the samples are axially symmetric, as might be expected from the experimental geometry, and the representation of the preferred orientation is relatively simple. The extension of these methods to coarser aggregates and samples of lower symmetry is discussed.

**TECHNIQUES AND PROCESSING OF DATA**

_X-ray technique._—Determination of preferred orientations was done with a pole figure goniometer using the technique of Schulz (1949) for reflection and that of Decker et al. (1948) for transmission. These methods have been discussed in detail by Gehlen (1960) and reviewed by Barrett and Massalski (1966). An elegant description using the reciprocal lattice is given in the

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**Plate 1.** —A, Photomicrograph of thin section of specimen GB-3. Dover flint specimen shortened 35 per cent at 780° C. and 15 kb. confining pressure at a rate of $10^{-5}$ sec$^{-1}$ in Griggs's hot-creep apparatus by J. D. Blacic. Crossed polarizers. B, Map of the thin section of GB-3. Contours represent variation in the transmission ratio $a$ (see text). Broken circle indicates the area in which X-ray measurements were made; $t-c.$ = thermocouple. C, Photomicrograph of thin section of specimen GB-11. Dover flint specimen shortened 34 per cent at 780° C. and 15 kb. confining pressure at a rate of $10^{-6}$ sec$^{-1}$; experiment by J. D. Blacic. Crossed polarizers. D, Map of the thin section of GB-11 showing variation in the transmission ratio $a$. Numbered points denote the centers of areas in which analyses were made.
An X-ray beam falling on a crystallite in an aggregate is diffracted if the conditions of Bragg's law are fulfilled. In an aggregate with grains in all possible orientations, reflections will occur at an angle $2\theta$ to the primary beam from grains with lattice planes $hkl$ inclined at an angle $\theta$ to the beam. When monochromatic radiation (in our case CuK$_\alpha$) is used, the diffracted beams from an aggregate occupy a set of nested cones about the primary beam, each cone corresponding to suitably oriented planes of constant $d$-value. In the pole figure goniometer (fig. 1) a scintillation counter, equipped with pulse-height analyzer, is kept stationary at an inclination to the primary beam $2\theta$, corresponding to the plane $hkl$ to be measured, while the specimen is simultaneously rotated about two mutually perpendicular axes to bring planes $hkl$ of all orientations in the specimen successively into the recording orientation (normal to the axis $OP$ in fig. 1). In reflection the specimen is rotated on the goniometer about axes $A$ and $B$ and in transmission about $B$ and $C$. The angles of rotation about these axes are spherical coordinates, with respect to specimen axes, of the poles of reflecting planes monitored by the counter during the scan. A set of "instrumental" angles $\alpha$, $\beta$, and $\tau$ (table 1) is used to denote the angles of rotation about the instrument axes $A$, $B$, and $C$. A set of right-handed Cartesian coordinates $a$, $b$, and $c$ is assigned to the specimen slab: $a$ is perpendicular to, and $b$ and $c$ lie in, the plane of the slab. These are the axes of reference for the pole figure.

**Reflection**—The specimen, a slab 2–4 mm. thick with plane parallel surfaces, is mounted normal to the rotation axis $A$ (figs. 1, 2a) so that it can be rotated in its own plane. The angle of rotation about $A$ corresponds to the azimuth $\alpha$ of poles in the plane.

**TABLE 1**

<table>
<thead>
<tr>
<th>Rotation Axis</th>
<th>Reflection</th>
<th>Transmission</th>
<th>Pole Figure Angles</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>$\alpha$</td>
<td>$\beta$</td>
<td>Azimuthal angle</td>
</tr>
<tr>
<td>$B$</td>
<td>$\beta$</td>
<td>$\beta$</td>
<td>Polar angle</td>
</tr>
<tr>
<td>$C$</td>
<td>$\tau$</td>
<td>$\tau$</td>
<td>Complement of polar angle</td>
</tr>
</tbody>
</table>

*The symbol $\phi$—a common alternative designation for $\beta$—is avoided, as it appears with another connotation in the analysis below.*

*It should be noted that this assignment of axes is made to conform with the analysis below, where $c$ is the axis of infinite-fold symmetry in axially symmetric specimens. In the analysis $c$ is the polar angle measured from $c$ and $\eta$ is the azimuthal angle measured from $a$. The transformation from instrumental angles to these coordinates is obtained from the expressions for the direction cosines $i_i$ of a unit vector in the direction $OP$:*

$$l_a = \sin \chi \cos \eta = \cos \beta_a = \sin \tau$$

$$l_b = \sin \chi \sin \eta = \sin \alpha \sin \beta_a = \sin \beta \cos \tau$$

$$l_c = \cos \chi = \cos \alpha \sin \beta_a = \cos \beta \cos \tau.$$
the pole figure (fig. 2b). The axis \( A \) and the specimen are both tilted about an axis \( B \); and the tilt angle of \( B \) corresponds to the polar angle \( \beta_R \) of poles in the pole figure. Simultaneous rotation about \( A \) and \( B \) (in our case \( B \) is tilted 5° during each full rotation of \( A \)) provides a spiral scan over the pole figure. Reflection scans go out to 70°–80° (in extreme cases out to 85°) from the center of the pole figure. There is, however, with increasing tilt angle, a decrease in intensity due to the defocusing effect and the fact that at high-tilt angles part of the beam may leave the specimen. Correction curves were obtained empirically in the present study by making reflection scans on a slab of undeformed flint without preferred orientation. (The lack of preferred orientation in the undeformed flint has been established by X-ray study of ground spheres using the method of Higgs et al. (1960) and study of variously oriented slabs with the present technique.) The normalized curves for variation of intensity with tilt \( (\beta_R) \) for a number of diffracting planes are shown in figure 3. There is a marked dependence on \( 2\theta \), in contrast to the theoretical intensity corrections derived by Feng (1965) for geometric defocusing in reflection. The empirical curves were also strongly affected by the size and shape of the collimator. Intensity corrections were found to be smallest when a small circular collimator (0.5 mm. diameter) and reflections of high \( 2\theta \) were used.

The pole density \( q_i (a, \beta_R) \) for the \( i \)th diffraction peak at \( P \) (fig. 2b) in a deformed specimen is given by:

\[
q_i (a, \beta_R) = \frac{I_{obs} - I_{bg}}{I_{undef}},
\]

where \( I_{obs} \) is the peak intensity of the deformed specimen, \( I_{bg} \) the background intensity of the deformed specimen (a constant value being used for all \( \beta_R \)), and \( I_{undef} \) the peak intensity for an undeformed specimen, corrected for background (fig. 3). The intensity (or pole density) \( q_i \) is in units that are approximately multiples of a uniform distribution. Exact normalization may be obtained only when the reflection data are combined with transmission data or the entire pole figure is otherwise determined.

**Transmission.**—The specimen, a thin section 70–90 \( \mu \) thick, mounted on the
specimen holder with Scotch Magic brand tape, is placed perpendicular to the plane of the beam and counter with the specimen normal parallel to B (fig. 4). It is rotated in its own plane around the axis B. The angle of rotation is the azimuth in the pole figure. The axis B is tilted simultaneously with the specimen about an axis C, which is also the diffractometer axis (normal to the plane of the collimator and counter). The tilt angle $\tau$ about the axis C is the complement of the polar angle in the pole figure.

![Figure 3](image-url)  
**Fig. 3.**—Empirical intensity correction curves for reflection scans. Normalized $2\theta$ scans for several planes using a specimen of undeformed flint with no preferred orientation. The $2\theta$ values (CuK$_\alpha$) radiation for planes 1010, 1011, 1120, and 2131 are, respectively, 20.9°, 26.6°, 36.5°, and 60.0°.

![Figure 4](image-url)  
**Fig. 4.**—Path of the scan in transmission, represented in spherical projection. Symbols as in fig. 2, with the exception that the specimen normal (a) is parallel to rotation axis B. a, Referred to goniometer axes; specimen axes $b$ and $c$ are in the specimen plane. Full lines represent the path in the upper hemisphere, broken lines in the lower. b, Path referred to specimen (pole figure) coordinates, upper hemisphere. A 5° spiral track is shown rather than the $21/2$° spiral generally used in transmission scans.
X-RAY ANALYSIS OF PREFERRED ORIENTATION

Rotation about C—a manual operation on the commercially available Norelco pole-figure goniometer—was automated by mounting an additional motor. In this manner a spiral scan is obtained in transmission that covers the area within 30°-40° of the periphery of the pole figure. In transmission there is an intensity correction, which is a function of tilt angle τ, due to changes in absorption and the volume of the specimen irradiated. Theoretical correction curves calculated with the formula of Decker et al. (1948) indicate that a small linear correction should be made. But transmission scans on an undeformed flint specimen with the small collimators showed that the intensities of all planes remained constant out to 40° tilt (τ). No tilt corrections were therefore made in the transmission data. For large 2θ angles (>90°), the beam hits the specimen at a flat angle so that a large area is irradiated, causing a serious defocusing effect. For this reason, planes with large 2θ angle (>90°) were avoided in transmission studies.

Instrumental settings.—The output of the diffractometer is a more or less jagged profile on the chart (e.g., figs. 9 and 18 below). The quality of the profiles is affected by the settings of the rate meter (Gehlen, 1960). The selection of the time constant influences the jaggedness of the profiles. It is desirable to use a long time constant to improve the counting statistics (and thereby smooth the profile) when the counting rate is low (due to small collimator or weak diffraction peak) or when there is a strong “crystallite effect”—maxima in the profile from individual crystallites—due to coarse grain size, small collimator, or both. However, because of the relatively rapid rotation of the specimen, the use of long time constants may result in truncation and displacement of sharp maxima with consequent distortion of the profile. To avoid this it is advisable to use the shortest time constants which yield a relatively smooth profile. The best compromise must be determined individually for each specimen and diffraction peak. A rapid manual scan of the pole figure indicates the range of intensities, so that the scale factor and multiplier may be set to utilize the full range of the recorder. It also gives an indication of the steepness of the gradients in the profile, so that the appropriate time constant (generally in the range 4-16 sec) may be selected. In general, the time constants are short for strong reflections and longer for weak reflections, but it is also necessary to use shorter time constants for weak reflections in profiles with steep topography.

The angular resolution in the pole figure is greatest with short time constant, small collimator and receiving slits, and slow scanning velocities. In this study the beam size and scanning velocity were limited by the sample size and goniometer speed, respectively, so that the angular resolution was dependent mainly on the selection of time constant. Thus, reduction of the crystallite effect in relatively coarse samples entailed some slight sacrifice of angular resolution.

Processing the data.—In transmission a spiral with 2½° spacing on the pole figure was used, and intensity values were read from the diffractometer records at 4°0 intervals for specimens with steep variations in intensity and at 9° intervals in specimens with small variations. The spiral in reflection had a 5° spacing. The 9° interval was invariably employed in the central region of reflection scans and also out to the limit of the scan if the topography of the record was flat. The two-digit intensities were punched on IBM cards for processing by computer. The possibility of direct digital recording of the data was investigated but was not introduced, mainly because of the high cost of a compatible system. The recording of data on diffractometer charts has some advantages: (1) Errors in specimen setting or adjustment and noise in the record due to grain effect or electronic instability are easily recognized and may be corrected manually. (2) The most economical data-sampling scheme, which depends on the topography of the intensity record,
FIG. 5.—a, Data points (intensities) from a reflection scan of 1120 in specimen GB-3, plotted in equal area projection on the lower hemisphere. b, Un-normalized pole figure obtained by smoothing the data in a. Both diagrams are computer printout, with contours added in b.
could also be determined for each specimen by visual inspection.

The first operation in the computations is correction of the data for background and geometrical effects according to the formula given above. The pole figure is represented numerically by 1,461 elements in a 41 × 49 array with center at (21, 25)—easily printed with ten characters per inch horizontally and six vertically to yield an equal-area projection of the standard 20-cm. diameter. The corrected intensities \( q_{ij} \) are entered in the array using equal area projection (fig. 5a). The recorded values only partly fill the array. The remaining elements are filled by means of a routine that computes values for the empty elements (with zero values) and also introduces short-range smoothing of the data; a weighted average \( q'_{ij} \) of the corrected intensities \( q_{ij} \) is computed over square arrays of nine elements (fig. 6):

\[
q'_{ij} = \frac{[A q_{ij} + B(q_{i,j+1} + q_{i+1,j} + q_{i,j-1} + q_{i-1,j}) + C(q_{i-1,j+1} + q_{i+1,j+1} + q_{i+1,j-1} + q_{i-1,j-1})]}{[A X_{ij} + B(X_{i,j+1} + X_{i+1,j} + X_{i,j-1} + X_{i-1,j}) + C(X_{i-1,j+1} + X_{i+1,j+1} + X_{i+1,j-1} + X_{i-1,j-1})]}
\]

where \( A = 12, B = 2, C = 1 \) are weighting factors and \( X_{ij} \) is unity if \( q_{ij} \) contains a non-zero value, and zero otherwise. This operation is repeated with weighting factors \( A = 48, B = 2, C = 1 \) to obtain a pole figure, as illustrated in figure 5b in the form of computer output and in figure 7a after contouring.

To insure continuity across the periphery of the pole-figure, intensity values at points \((\beta_r, \tau)\) within \(8^\circ\) of the periphery \( (0^\circ \leq \tau \leq 8^\circ) \) are entered twice—once on the lower hemisphere \((\beta_r, \tau)\) and once on the upper hemisphere \((180^\circ + \beta_r, -\tau)\); in projection the latter point is diametrically opposite the first and slightly outside the primitive circle. The pole figure for a transmission scan, after smoothing, is shown in figure 7b.

**Determination of the complete pole figure.**—Since the area of the pole figure covered by both reflection and transmission scans is restricted, the complete pole figure must be obtained by combining data from more than one scan. Data from reflection scans of three mutually perpendicular slabs are appropriate for large, relatively homoge-
malized so that the mean over the whole pole figure is unity; that is, the densities are expressed as multiples of that for a uniform distribution. These operations are illustrated with data for 1120 in specimen GB-3 in figure 7. The resulting normalized pole figure (7c) has contours that approximate to small circles about the compression axis $\sigma_1$, indicating that the preferred orientation is axially symmetric. An axially symmetric pole figure of this type can be characterized by a single profile extending $0^\circ$--$90^\circ$ from the symmetry axis.

If the same size of collimator is used, the area irradiated in a reflection scan is larger than that in transmission. In heterogeneous samples like GB-11, where the preferred orientation must be determined in small areas (1--2 mm. diameter), axial symmetry of the preferred orientation was assumed and the analysis was made by means of peripheral transmission ($\beta_T$) scans alone. In the coordinate system used in the analysis below, $c = \sigma_1$ and the instrumental angle $\beta_T$ is identical with the polar angle $\chi$ in the pole figure.

**Transmission profiles of axially symmetric specimens.**—The intensity values from 360° peripheral scans were read at 4° intervals, giving eighty values for each diffracting plane; only twenty of these are necessary to characterize the asymmetric unit ($\chi = 0^\circ$--90°). Making use of the symmetry, mean values were calculated from the sets of four equivalent values and a continuous profile was generated by linear interpolation between the data points. This procedure was found to be preferable to least-squares fitting of a fifth-order polynomial, which distorted profiles with steep topography.

The profile was integrated using Simpson's rule to obtain the normalized intensities:

$$q_1(\chi) = \frac{I_{\text{obs}}(\chi) - I_{\text{bg}}}{\int_0^{\pi/2} [I_{\text{obs}}(\chi) - I_{\text{bg}}] \sin \chi \cdot d\chi}.$$ 

Figure 8 illustrates the typical scatter of data points in three complete 360° scans of 1120 in specimen GB-3. The scatter is due to electronic instability (particularly when small time constants and scale factors are used), departures from axial symmetry, and misalignment of the specimen. If axial symmetry is assumed, the mean values give the best representation of the profile.

**Modifications for larger grain size.**—For fine-grained specimens (grain size <10 µ), the number of crystals irradiated by the small (~1 mm.) beam is large enough to provide satisfactory counting statistics. But if less than $10^4$ grains are irradiated, modi-
fications are necessary to bring more grains into the reflecting orientation. This is achieved with large homogeneous specimens by relatively rapid translation of the specimen in its own plane (a standard operation in the commercial pole figure device), but this procedure could not be used because of the small areas studied in our specimens. Instead of this, a rapid rocking motion,

A similar technique to improve the statistics in reflection would involve rocking the specimen around the axis B (fig. 1). It was not found necessary in the present study but is recommended for specimens with large grain size. In extreme cases the grain size problem could be overcome by simultaneous use of the rocking motion and translation of the specimen in its own plane.

![Figure 8](image-url)  
**Fig. 8.**—Intensity values at twenty-one values of $\chi$ obtained from three complete rotations in a transmission scan of 1120 in specimen GB-3. This yields, by symmetry, six values at $\chi = 0^\circ$ and $90^\circ$ and twelve at intermediate values of $\chi$. The profile was normalized using linear interpolation between the average intensity values (indicated by horizontal bars) and numerical integration. The full curve, fitted to the average values with sixteenth-order Legendre polynomials, was used in subsequent calculations.

through $\pm 5^\circ$ around the axis C (fig. 1), was introduced to provide an integration over $5^\circ$ on each side of the peripheral track. The motion was achieved by means of a small motor and cam. The effect of this integration is illustrated in figure 9, which shows a chart record obtained with and without the oscillation of the specimen. The grain effect causing the jagged profile in figure 9a is absent in figure 9b; the profile is smoother and its symmetry across the lines at $\beta_1 = 90^\circ$, $180^\circ$, and $270^\circ$ is more evident. There is, of course, some loss in angular resolution resulting from this procedure, but it is not serious in the axially symmetric specimens considered here.

**METHODS OF ANALYSIS**

*Introductory statement.*—In preferred-orientation studies it is desirable to be able to specify completely the orientation of the crystallites with respect to the specimen. There is a fundamental difficulty in recovering this information from statistical distributions of individual lattice planes as presented in pole figures. Consider the pole figures for two lattice planes, $r = 1011$ and $c = 0001$ (fig. 10) from the same specimen. The $c$-axes of crystals, whose poles to $r$ lie at $I$ in figure 10a, must lie on the small circle $SC$ ($51^\circ 47'$ from $I$) in figure 10b, but there is an ambiguity as to their positions on the
small circle. Thus, the restriction to lie on
the small circle means that the two pole
figures are not independent of one another;
however, the ambiguity permits a consider-
able variety of compatible diagrams.

The manner in which the pole figures for
various lattice planes are related to one
another is illustrated by rotating a single
crystal to successive orientations to generate
a fabric. One can draw a series of concentric
spheres around the crystal—one for each
crystal form that is to be plotted. If the

![DT-198 10\(\bar{1}\)1](image)

**Fig. 9.**—Transmission scans \((\beta_T)\) showing the smoothing effect of a ± 5° oscillation about axis \(C\); 10\(\bar{1}\)1 profile in specimen DT-198, with average grain size 20 \(\mu\), \(a\) without and \(b\) with oscillation. Note the increased

![FIG. 10.](image)

**Fig. 10.**—\(a\), Pole figure of 10\(\bar{1}\)1 from a fine-grained metaquartzite specimen (X-ray data). The \(I\) is an
arbitrary direction in the pole figure. Contour intervals are arbitrary. \(b\), Pole figure of 0001 from the same
specimen, based on optical measurements on 200 grains. Contours are 7, 5, 3, 2, and 1 per cent per 1 per cent
area. The 0001 axes for crystals with 10\(\bar{1}\)1 in direction \(I\), fig. 10\(a\), may have any orientation on the small
circle (s.c.).
radius of each sphere is $1/d_{hkl}$, the reciprocal of the interplanar spacing, the poles to planes plotted on the various spheres will form a lattice array of points—the reciprocal lattice.

When the crystal is rotated into another orientation with respect to a fixed coordinate system, the lattice of points rotates with the crystal. The new set of points plotted on the spheres records the new orientations of the poles. The fabric is generated by rotating the crystal to a large number of orientations and at each orientation plotting the poles to the various planes on the appropriate spheres. The points on each reciprocal lattice sphere can then be contoured and projected to produce pole figures. Each reciprocal lattice sphere corresponds to a diffraction peak (Meieran, 1962). The pole figures are related to one another, since they were produced by one set of crystal orientations.

**General theory.**—To describe an arbitrary orientation of the crystal with respect to the specimen, it is convenient to fix one Cartesian coordinate system $a,b,c$ in the specimen and another system $X,Y,Z$ in the crystal and to express the difference in orientation ($\Omega$) of the two coordinate systems with the three Euler angles $\psi$, $\phi$, and $\theta$ (fig. 11a).

The orientation of the $k$th pole on the $i$th reciprocal lattice sphere is given by a unit vector $p_{ik}$ which has the coordinates $\xi_{ik}$, $\phi_{ik}$, where $\xi_{ik} = \cos \psi$ (fig. 11b). The orientation of a line with respect to the specimen axes, as represented by a point on a pole figure, is given by a unit vector $r$ which has the coordinates $\xi$ and $\eta$, where $\xi = \cos \phi$ (fig. 11c). If the orientation with respect to a crystal of a particular pole is given by $p_{ik}$ and, with respect to the specimen, by $r$, then the two sets of coordinates for the pole are related by a transformation of coordinates (cf. Wigner, 1959, eq. [A.2], p. 358, where $\alpha$, $\beta$, and $\gamma$ are identical with $\phi$, $\theta$, and $\psi$, respectively, used here) corresponding to the rotation $\Omega$.

The orientation of a large population of grains can be specified by giving the frequency distribution of orientations $w(\Omega)$—
plete orientation of grains, their usefulness is limited (McHargue and Jetter, 1960; Bunge, 1965c). If the preferred orientation of the specimen has axial symmetry and $c$ coincides with the symmetry axis, the crystallite distribution is independent of the Euler angle $\psi$ and dependent only on $\xi$ and $\phi$, so that it can be represented on a sphere. Such diagrams, called inverse pole figures (Barrett and Massalski, 1966, chaps. ix, xix, xx), show the frequency density of the specimen symmetry axis with respect to crystal axes. Inverse pole figures obtained by rotating coordinate axes of all the crystals into coincidence and plotting the distribution of the specimen symmetry axis are widely used in metallurgical research.

Methods for recovering the crystallite distribution function from a suite of pole figures using spherical harmonic analysis have been published by Viglin (1961), Bunge (1965a), and Roe (1965). Similar methods for the special case of axial symmetry were described earlier by Bunge (1959) and Roe and Krigbaum (1964).

Each pole figure may be expanded with a set of harmonics that has the same symmetry as the pole figure, that is, polyhedral harmonics (Laporte, 1948; Meyer, 1954; Bunge, 1965a, b). Let a point in the $i$th pole figure be represented by the unit vector $\mathbf{r}$ which has the spherical coordinates $\xi$ and $\eta$ (fig. 11c) and the density at this point by $q_i(\mathbf{r}) = q_i(\xi, \eta)$; $q_i(\mathbf{r})$ is normalized so that the integral of density over the unit sphere is $\int q_i(\mathbf{r}) \, d\xi d\eta = 4\pi$ (i.e., the average value of pole density is unity). Then the expansion of $q_i$ is

$$q_i(\mathbf{r}) = \sum_{i=0}^{N(l)} \sum_{n=0}^{N(l)} Q^i_{ln} K_{ln}(\mathbf{r}) .$$

The polyhedral harmonic $K_{ln}$ has the following form in the holohedral point groups of the monoclinic, orthorhombic, tetragonal, and hexagonal systems, provided a mirror plane is normal to the $b$-axis and a $v$-fold axis is parallel to the $c$-axis:

$$K_{ln}(\mathbf{r}) = \frac{(-1)^v}{\sqrt{\pi}} P^n_l(\xi) \cos u \eta \quad (u \neq 0, \ l \ even) ,$$

where $u = nv$, $P^n_l$ is the normalized associated Legendre polynomial (Jahnke and Emde, 1945, p. 110, 116), and $N(l) + 1$ is the number of linearly independent polyhedral harmonics in eq. (1) for the order $l$. For triclinic specimen symmetry, $K_{ln}$ is identical with the ordinary spherical harmonic $Y_{ln}$ as given by Edmonds (1960, eq. [2.5.29]).

Using the orthogonality property of polyhedral harmonics,

$$\mathcal{F} K^*_{ln}(\mathbf{r}) K_{ln'}(\mathbf{r}) d\mathbf{r} = \delta_{ll'} \delta_{nn'} ,$$

where $\delta_{ll'}$ is the Kronecker delta and the asterisk indicates the complex conjugate, the coefficients $Q^i_{ln}$ in equation (1) can be obtained by multiplying both sides of equation (1) by $K_{ln'}$ and integrating as

$$Q^i_{ln} = \mathcal{F} q_i(\mathbf{r}) K^*_{ln}(\mathbf{r}) d\mathbf{r} .$$

Symmetrical generalized spherical harmonics $E_{lmn}(\Omega)$ are used to expand the crystallite distribution function $w(\Omega)$ as

$$w(\Omega) = \sum_{l=0}^{M(l)} \sum_{m=-l}^{M(l)} \sum_{n=-l}^{N(l)} W_{lmn} E_{lmn}(\Omega) ,$$

where $M(l) + 1$ and $N(l) + 1$ are the number of linearly independent harmonics with respect to $m$ and $n$, respectively. The $E_{lmn}(\Omega)$ are invariant with respect to rotations in the point group of the crystal. They are also invariant with respect to rotations in the point group of the specimen. As discussed by Bunge (1965b), symmetrical generalized spherical harmonics are obtained as a linear combination of generalized spherical harmonics. It should be noted, however, that the generalized spherical harmonics used here (the $D_{mn}^l$ of Edmonds, 1960, eqs. [4.1.12] and [4.1.15], p. 55, 57) are based on the widely accepted convention for Euler angles that the second rotation is positive when a right-handed screw advances parallel to $Y$ (Edmonds, 1960, fig. 1.1, p. 7), in contrast to the $T^m_n$ of Bunge (1965b, p. 352), which is based on the con-
vvention that the second rotation is positive when a left-handed screw advances parallel to $X$ (cf. Gel'fand et al., 1963, fig. 1). The $E_{lmn}(\Omega)$ used here are equivalent to $\tilde{Y}_l^m(\hat{r})$ of Bunge (1965b, p. 354).

$w(\Omega)$ is normalized so that a uniform distribution has the value of unity, that is, the integral over all possible orientations is $\int w(\Omega) d\Omega = 8\pi^2$.

The density at any point $r$ on the $i$th pole figure is obtained from the crystallite distribution function by determining the average density of orientations in which the poles $p_{ik}$ are parallel to $r$. Thus,

$$q_i(r) = \frac{1}{2\pi v_i} \sum_{k=1}^{v_i} \int_{p_{ik}\parallel r} w(\Omega) d\Omega . \tag{5}$$

($p_{ik}\parallel r$ under the integral sign in eq. [5] indicates that the integral extends only over orientations for which $p_{ik}$ is parallel to $r$.) This corresponds to a complete rotation of $XYZ$ about this direction, hence the factor $2\pi$. The multiplicity factor of the crystal form whose poles lie on the $i$th reciprocal lattice sphere is $\nu_i$.

The integral in equation (5) has been evaluated by Bunge (1965b, p. 360). For the Euler angle conventions used in this paper one obtains

$$\int_{p_{ik}\parallel r} w(\Omega) d\Omega = \sum_{l=0}^{\infty} \sum_{m=0}^{\infty} \sum_{n=0}^{\infty} \frac{8\pi^2}{2l + 1} \prod_{k=1}^{v_i} K_{lm}^{(p_{ik})} K_{ln}(r) \tag{6}$$

where the polyhedral harmonic $K_{lm}^{(p_{ik})}$ has the diffraction group symmetry of the crystal. Since the $v_i$ poles are related by the symmetry operations of the point group and $K_{lm}(p_{ik})$ is invariant to these operations,

$$\frac{1}{\nu_i} \sum_{k=1}^{v_i} K_{lm}^{(p_{ik})} = K_{lm}^{(p_{jik})}$$

can be written as $K_{lm}^{(\hat{p})}$.

Equations (5) and (6) are relevant for pole figures from diffraction peaks that contain only one crystal form. In the case where the reciprocal lattice spheres for $J$ different crystal forms are coincident or are so close as to cause overlapping diffraction peaks, the relative intensities of reflections from the $J$ different forms must be taken into account. To do this, a relative intensity function $c_{ij}$ containing the square of the structure factor $F_{ij}$ and the multiplicity factor $v_{ij}$ of the $j$th form on the $i$th reciprocal lattice sphere (Bunge, 1961, p. 292)

$$c_{ij} = \frac{v_{ij} F_{ij}^2}{\sum_{j=1}^{J} v_{ij} F_{ij}^2} ,$$

and a composite polyhedral harmonic,

$$H_{lm}^{ij} = \sum_{j=1}^{J} c_{ij} K_{lm}^{(\hat{p}_{ij})} ,$$

is introduced so that equation (5) takes the following form:

$$q_i(r) = \sum_{l=0}^{\infty} \sum_{m=0}^{\infty} \sum_{n=0}^{\infty} \frac{4\pi}{2l + 1} \int_{p_{ik}\parallel r} w(\Omega) d\Omega \prod_{k=1}^{v_i} H_{lm}^{ij}(p_{ik}) K_{lm}(r) \prod_{k=1}^{v_{ij}} K_{lm}^{(p_{ij})} \tag{7}$$

Thus, the pole figure corresponding to any reciprocal lattice sphere or diffraction peak may be calculated if the crystallite distribution function is known. If it is necessary to determine separately the pole figures for several crystal forms that have coincident or overlapped diffraction peaks (such as 1012, 0112, 121 in quartz), this may be done using the relationship

$$q_{ij}(r) = \sum_{l=0}^{\infty} \sum_{m=0}^{\infty} \sum_{n=0}^{\infty} \frac{4\pi}{2l + 1} \int_{p_{ik}\parallel r} w(\Omega) d\Omega \prod_{k=1}^{v_i} K_{lm}^{(p_{ik})} K_{ln}(r) \prod_{k=1}^{v_{ij}} K_{lm}^{(p_{ij})} \tag{8}$$

To obtain a relation between the coefficients of the expansion of the pole figures and the coefficients of the crystallite distribution function, one multiplies both sides of equation (7) or equation (8) by $K_{l}^{(p)}(r)$ and integrates over the unit sphere. Thus,

$$Q_{lm}^{ij} = \sum_{m=0}^{M(l)} \sum_{n=0}^{N(l)} \frac{4\pi}{2l + 1} W_{lmn} H_{lm}^{ij} \tag{9}$$

and

$$Q_{lm}^{ij} = \sum_{m=0}^{M(l)} \sum_{n=0}^{N(l)} \frac{4\pi}{2l + 1} W_{lmn} K_{lm}^{(p_{ij})} . \tag{10}$$
Since $Q_{lm}^i$ in equation (9) can be calculated from experimentally determined pole figures, the coefficients of the crystallite distribution function can be determined by solving the set of linear equations (9). The number of pole figures required for a given order $I$ is greater than or equal to $M + 1$. In practice the method of least squares is used to reduce experimental errors in the data. After the coefficients of the crystallite distribution function have been determined, pole figures can be calculated using equation (8).

Restrictions for axially symmetric quartz specimens.—As the specimens analyzed in this study have axial symmetry, the analysis can be considerably simplified. Each pole figure can be represented by a line profile $q_i(\xi)$ from parallel to the symmetry axis to normal to the symmetry axis. The profile is expanded as

$$q_i(\xi) = \sum_{l=0}^{\infty} Q_l^i \hat{P}_l(\xi),$$

where $Q_l^i$, which is related to $Q_{lm}^i$ in equation (1) by $Q_l^i = (2\pi)^{-1/2} Q_{lm}^i$, is given by

$$Q_l^i = \int_{-1}^{1} q_i(\xi) \hat{P}_l(\xi) \, d\xi.$$  

The degenerate crystallite distribution function $w(\xi, \phi)$, which is a function of only two angles and can be represented on a sphere, is the inverse pole figure. Letting $\hat{p}$ be a unit vector with the spherical coordinates $\theta$ and $\phi$ with respect to $XYZ$, $w(\hat{p})$ is expanded as

$$w(\hat{p}) = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} C_{lm} K_{lm}(\hat{p}),$$

where $C_{lm} = \sqrt{4\pi/(2l+1)} W_{lm}$ in equation (4).

The equation relating coefficients of the expansions of the inverse pole figure and the profiles in the case of overlapped diffraction peaks is

$$Q_l^i = \sqrt{\frac{2}{2l+1}} \sum_{m=0}^{M(l)} C_{lm} H_{lm}^i,$$

and for no overlap is

$$Q_l^i = \sqrt{\frac{2}{2l+1}} \sum_{m=0}^{M(l)} C_{lm} K_{lm}(\hat{p}_i).$$

The method of least squares seeks to determine values of $C_{lm}$ which minimize the sum $S$,

$$S = \sum_{i=1}^{I} \rho_i \left[ \sum_{m=0}^{M(l)} C_{lm} H_{lm}^i - \sqrt{\frac{2l+1}{2}} Q_l^i \right]^2,$$

where $\rho_i$ is Roe's (1965) weighting factor indicating the reliability of a profile. By differentiating $S$ with respect to $C_{lm}$, and equating the result to zero, one obtains a set of normal equations,

$$\sqrt{\frac{2l+1}{2}} \sum_{i=1}^{I} \rho_i Q_l^i H_{lm}^i = \sum_{i=1}^{I} \rho_i \sum_{m=0}^{M(l)} C_{lm} H_{lm}^i H_{lm}^i,$$

which can be solved by the Gaussian reduction method for the coefficients $C_{lm}$. Profiles for the various diffraction peaks are regenerated from the inverse pole figure using equation (7) and are compared with the observed profiles to give an indication of the goodness of fit.

Several specimens with axial symmetry have been subjected to spherical harmonic analysis (Bunge, 1963; Krigbaum and Roe, 1964; Krigbaum and Balta, 1967; Wenk et al., 1967; Wenk and Kolodny, 1968). The following discussion will be restricted to quartz aggregates analyzed in this study.

Quartz has a point group symmetry of $32$—an enantiomorphous crystal class. A complete description of a preferred orientation would comprise one crystallite distribution function for left-handed crystals and another for right-handed crystals. Due to Friedel's law, quartz has the diffraction symmetry $3m(=3 2/m)$. Furthermore, in a powder diffraction pattern the positive and negative trigonal forms with the same numerical indices (e.g., $hkil$ and $hkil$) are unresolved, so that the apparent symmetry is $6/mmm$. 


The assumption of pseudohexagonal symmetry permits equation (16) to be solved with a minimum number of profiles: only five profiles are required for \( l = 22 \). No relative intensity data are required. To obtain an inverse pole figure with the symmetry \( 3 \ 2/m \), it is necessary to resolve the profiles for the positive and negative forms of the rhombohedra and trigonal scalenohedra—which have different intensities but coincident diffraction peaks. The values used for the intensities are the squares of the calculated structure factors listed by Zachariasen and Plettinger (1965). A minimum of six observed profiles is required for the sixteenth order when the diffraction group symmetry \( 3 \ 2/m \) is used for the inverse pole figure.

Analyses of flint specimens assuming \( 6/mmm \) and \( 3 \ 2/m \) symmetries are discussed below. It would be of interest to determine inverse pole figures that have the 32 symmetry of quartz to obtain information on the orientations of right- and left-handed crystals, or whether there is a preferential orientation of the positive ends of the \( a \)-axes. However, there are considerable difficulties in making such a determination. Resolution is critical because in the case of a trigonal trapezohedron there are four forms (right and left, positive and negative) for the structurally right-handed crystal and another four for the left-handed crystal that must be resolved from the composite diffraction peak. To achieve this resolution, the small intensity differences due to anomalous scattering, which according to Zachariasen (1965) are of the order of several per cent for most allowable reflections, must be used. Because of the low symmetry the number of unknown coefficients is so large that only low-order coefficients could be determined.

6 The convention used here for indexing crystal planes of quartz is that the morphological major unit rhomb \( r = 1011 \) is identified with the strongly reflecting unit rhomb \( 1011 \) (Frondel, 1962, p. 14; Lang, 1965). Thus, the sign of the index \( L \) in table 2 of Zachariasen and Plettinger (1967) must be reversed for the intensities to agree with this convention.

Computations.—Because the basic equation (16) used in this analysis is equivalent to that used by Krigbaum and Roe (1964), the computational procedures used to determine the inverse pole figure are similar to those described by them with the following exceptions: (1) Frequency densities are normalized so as to be expressed as multiples of a uniform distribution. (2) The relative intensity \( c_{ij} \) contains the multiplicity factor. (The multiplicity factor does not appear in any of Roe’s equations, although it is implicit in eq. [24] [Roe, 1965, p. 2027]. Bunge [1961] discussed the multiplicity factor in resolving overlapped reflections.) (3) The crystallographic angles for the various reflections (table 2) were taken from table 11 in Frondel (1962), with appropriate changes for the different coordinate system used here (fig. 12a). The programs were written in FORTRAN IV.

EXPERIMENTAL RESULTS

Analysis of specimen GB-3 assuming hexagonal symmetry.—It was shown above (fig. 7c), by combining data from reflection and transmission scans, that the preferred orientation in specimen GB-3 has axial symmetry, so that transmission profiles on a specimen containing the symmetry axis are sufficient for determination of the preferred orientation. Transmission profiles of fifteen reflections listed in table 2 and shown in figure 12a were used to determine the preferred orientation in the specimen. For simplicity of calculation it was first assumed that quartz has \( 6/mmm \) symmetry. The result is shown in the inverse pole figure in figure 12b. The asymmetric unit in this pole figure is a 30° sector from 0001 to 1010 and 1120. The contours show the distribution of the symmetry axis (\( a_1 \)) with respect to the crystal axes, and the diagram contains, in condensed form, the complete data on the preferred orientation contained in the observed profiles, some of which are illustrated in figure 13. The inverse pole figure is almost axially symmetric, with a diffuse maximum at (0001). Profiles for selected planes (including 0001, which was not used
as input data) regenerated from the inverse pole figure using equations (15) and (11) are shown with the measured profiles in figure 13. There is apparently satisfactory agreement between the observed and calculated profiles for the prisms and certain pseudohexagonal planes (such as 1123), but there is a marked discrepancy for the rhombohedral planes, such as 1011. The measured profile shows a maximum parallel to σ1, whereas the calculated profile has a maximum at approximately 50° to σ1, con-

<table>
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* Used to calculate fig. 20.
sistent with the concentration of 0001 parallel to σ₁. The true symmetry of α-quartz is 3\_2\_m, and the diffraction peak corresponding to 10\_1\_1 consists of the positive and negative rhombohedra (r = 10\_1\_1 and z = 01\_1\_1, respectively) with the same d-value but different intensities (in the ratio I_r/I_z = 7/3). The discrepancy could be accounted for qualitatively by assuming that the strongly diffracting planes r in crystals with 0001 inclined at approximately 40°–60° to σ₁ were preferentially oriented parallel to σ₁. To test this, the inverse pole figure was calculated using the correct diffraction symmetry for α-quartz (3\_2\_m).

Analysis of GB-3 assuming trigonal symmetry.—In this analysis the intensity profiles (fig. 14) were treated as composites of profiles of the positive and negative forms, with different intensity contributions and different distributions, as discussed above. In addition to these composite profiles of planes with identical d-values, some of the diffraction peaks used contain forms with similar d-values (such as the peak at 2θ = 55°, with contributions from 2022, 0222, 1013, and 0113). These profiles were also resolved into their components in the analysis. The inverse pole figure obtained is shown in figure 12c, and the composite profiles generated from the inverse pole figure are in good agreement with the measured profiles (fig. 14). The profiles for the positive and negative rhombohedra, which have been resolved from the six composite rhombohedral profiles recorded, show

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**Fig. 12.—Inverse pole figures for specimen GB-3.**

- **a**: Asymmetric unit of spherical (equal area) projection showing the orientations of planes measured. The unit in the 3\_2\_m group lies between the mirror planes (solid lines).
- **b**: Inverse pole figure derived with the assumption of 6/mm\_m symmetry using fifteen measured profiles truncated at the sixteenth order. The figure is repeated by reflection across the 11\_0\_0 plane.
- **c**: Inverse pole figure derived with 3\_2\_m symmetry using fifteen measured profiles, shown in part in fig. 14.
- **d**: Inverse pole figure derived with 3\_2\_m symmetry using the eight least-weighted profiles. Figures b and c are truncated at the sixteenth order, d at the twelfth order.
marked differences. The profiles for the positive rhombohedra are characterized by a maximum parallel to $\sigma_1$ and one normal to $\sigma_1$; those for the negative rhombohedra by a maximum at intermediate angles. With decreasing inclination of the rhomb (i.e., as $\theta_{ij}$ goes to 0°), the profiles grade into a single maximum parallel to $\sigma_1$; and as $\theta_{ij}$ approaches 90°, they grade into a single maximum normal to $\sigma_1$. There is a marked tendency for poles of the positive unit rhomb $r = 1011$ to be parallel to $\sigma_1$, as was postulated above. This is reflected in the inverse pole figure (fig. 12c) by the strong concentration of $\sigma_1$ extending from $c(0001)$ to $r(1011)$ and the minimum near $z(01\bar{1}1)$; the negative values near $01\bar{1}1$ are, of course, artifacts of the computation and are discussed below. The mathematical analysis thus appears to provide resolution of planes that are not resolved in the direct X-ray measurements.

Reproducibility and accuracy of the results.—It is important to establish the reproducibility of the preferred orientations represented by the inverse pole figures and to determine the number of measured profiles necessary to provide an accurate representation of a preferred orientation. A number of tests were performed on data from GB-3 and other specimens to elucidate these points.

Fig. 13.—Selected profiles measured in transmission in GB-3 (solid lines) and profiles regenerated from the inverse pole figure (fig. 12b) with $6/mmm$ symmetry (broken lines). Units of density on the ordinate are multiples of a uniform distribution. The $x$ is measured from the compression axis $\sigma_1$. Note the disparity between measured and calculated profiles of $10\bar{1}1 + 01\bar{1}1$. 

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FIG. 14.—Profiles for fourteen of the fifteen diffraction peaks used in the analysis of GB-3 and the profiles of these and 0001 regenerated from the inverse pole figure (fig. 12c). Solid heavy lines are measured profiles; light dashed curves are individual forms resolved in the analysis; and heavy dashed lines are the composite calculated profiles.
The quality of the measured profiles used in the calculations is variable. This depends on a number of factors: (a) The area of the specimen irradiated is a function of $2\theta$ angle; in transmission the size of the irradiated area increases with increasing $2\theta$. The profiles are only compatible, therefore, if the specimen is fine grained and statistically homogeneous over a relatively large area ($1.5 \text{ mm}^2$ for transmission with a 0.5-mm. collimator). (b) For weak reflections, determination of the background is more difficult and larger errors arise than for strong reflections. Taking account of these factors, the profiles were weighted in the calculations, profiles of strong peaks with low to medium $2\theta$ being weighted more heavily. Eight of the “poorest” profiles were selected from the fifteen measured, and an inverse pole figure was calculated with these as the input data (fig. 12d). It is qualitatively similar to that obtained from all fifteen profiles (fig. 12c). This demonstrates that the inverse pole figure is not determined or unduly distorted by the influence of the “best” and most strongly weighted profiles. It also indicates that it is a reliable representation of the preferred orientation and that, for this type of preferred orientation, considerably less than fifteen profiles are necessary to obtain the preferred orientation.

The number of measured profiles required for the calculation of a reliable inverse pole figure depends mainly on the complexity of the preferred orientation and the order of the harmonics necessary to represent it adequately, but also to some extent on the distribution of the diffracting planes used to obtain the profiles. The latter should preferably be distributed uniformly over the area of the inverse pole figure; it is inadvisable to select planes of similar crystallographic orientation (such as 3142, 2131, 3141). It is clearly useful for resolution of the equivalent positive and negative planes to select peaks with marked intensity differences between the positive and negative forms.

The minimum number of profiles $I$ required to solve the simultaneous equations (16) for the $l$th order is greater than the theoretical value of $(l/v+1)$, where $v = 3$ for trigonal crystals, because experimental errors in the data lead to numerical instabilities. These instabilities are indicated by anomalous values for the high-order coefficients. They are eliminated when equation (14) are overdetermined by a factor $f$ (the ratio of knowns to unknowns $= I/[\ell/v + 1]$) greater than 1.5 for the coefficients of highest order $L$. To compare the stability of profiles expanded with the series in equation (11), but truncated at different orders $L$, it is useful to determine a mean factor of overdetermination:

$$\bar{f} = \frac{IL}{2\sum_{l=4}^{L} \left( \frac{l}{v} + 1 \right)},$$

where $l$ is even and $l/v$ is truncated to a whole number. A good example of numerical instability is seen in the 0001 profiles of GB-11 (fig. 19) calculated with the hexagonal program from four measured profiles. The twelfth- and sixteenth-order profiles both exhibit instability. The value of $f$ is the same for $L$ in both cases ($f = 1.3$), but $f$ is greater for the twelfth-order profile ($f = 2.2$) than for the sixteenth-order profile ($f = 1.9$). The lower limit of stability in terms of $f$ may be taken as approximately 2.5.

The order of the harmonics necessary to represent the preferred orientation faithfully depends on the topography of the inverse pole figure. For flat topography with concentrations up to twice the value for a uniform distribution, the eighth order is adequate; for concentrations up to six times uniform, the twelfth or sixteenth order is recommended, and for higher concentrations, orders greater than twenty are necessary. If the orders are not high enough, the polynomials cannot represent the details in the profiles with accuracy; sharp peaks are cut off even when a sufficient number of profiles is used to avoid numerical instability. The use of low orders also causes termination errors arising from truncation of the infinite series of spherical harmonics.
Krigbaum and Roe (1964, p. 743) illustrate a method of estimating quantitatively the series termination error. Termination effects are illustrated for two specimens, DT-459 and DT-460, with very sharply peaked 0001 profiles (Wenk, et al., 1967), in figure 15. Termination effects are similar to those in Fourier series in crystal structure determination and give rise to artificial peaks in the vicinity of real peaks in the profiles. It was found empirically that twelfth-order harmonics adequately represent most moderate preferred orientations, and for these eight input profiles may be regarded as a lower limit in the trigonal case.

A puzzling feature of the inverse pole figures is the physically impossible negative area in the region of $\alpha = 01\overline{1}1$, which appears consistently in most of the aggregates with different orientations of the positive and negative rhombohedra so far studied. It cannot be explained as a termination error (as in the case given by Krigbaum and Roe, 1964), because there are no significant differences in the pole figures of GB-3 obtained by truncating the series of harmonics at the eighth, twelfth, and sixteenth orders. The intensity ratios used for forms contributing to a single diffraction peak are those for a perfect crystal; the use of an extinction cor-

![Figure 15](image-url)
rection (Zachariasen and Plettinger, 1965) increases the negative areas in the inverse pole figures. The negative areas can be eliminated by suitable adjustment of the intensity ratios. Since deformation may change the relative intensities, there is some justification for making these adjustments. Adachi (1967) uses an iteration process, with repeated least-squares solutions for the relative intensities and the coefficients $C_{im}$ of the inverse pole figure, to obtain the best set of relative intensities. It is anticipated that a similar method could be used to refine the inverse pole figures for quartz and to decrease or eliminate the areas of negative concentration.

Variation of preferred orientation in an inhomogeneous specimen (GB-11).—The value of the inverse pole figure as a concise method of representing completely preferred orientations with axial symmetry becomes more evident when several preferred orientations are to be compared. The variation of the preferred orientation in the inhomogeneous specimen GB-11 is illustrated by means of the inverse pole figures in figure 16. A photomicrograph of a thin section of the specimen is shown in plate 1C. The heterogeneity of the fabric is demonstrated by the contour map (pl. 1D) of a photometrically determined transmission ratio $a$, which reflects the variation in preferred orientation of optic axes $[0001]$ in the specimen (Baker, 1965). Those areas in which $a$ is greater than unity indicate a tendency for parallelism of the optic axes $[0001]$ with the compression axis $\sigma_1$, whereas in those areas with $a$ less than unity the $[0001]$ axes tend to be inclined at angles close to $45^\circ$ from $\sigma_1$. The numbered spots in plate 1D are the centers of regions, approximately 1.5 mm. in diameter, where transmission scans were made. An inverse pole figure was calculated with twelfth-order harmonics from seven profiles at each of the eight localities (fig. 16). The variation in $0001$ profiles along the specimen, shown in the block diagram (fig. 17), is consistent with the photometric data: at position 7, $a = 1.5$ and $0001$ shows a maximum parallel to $\sigma_1$; at position 3, $a = 0.9$ and the strongest concentration of $0001$ is at $35^\circ$ from $\sigma_1$.

The series of inverse pole figures shows a tendency for either $0001$ or $1011$ to be aligned parallel to the compression axis. The $1011$ component is strongest in the center of the specimen (position 3) where the temperature was highest during the experiment and the increase in grain size is most marked. The $0001$ component is more strongly developed in the cooler recrystallized portions, where the grain size is comparable to that of the starting material. These gradations in preferred orientation are of considerable importance for the study of the mechanisms of development of orientations, and the spatial resolution of our X-ray method (achieved by use of small collimators) is indispensable for the study of such inhomogeneous specimens.

Determination of the c-axis profile.—Although it was shown above that the c-axis pole figure alone is an inadequate description of the preferred orientation in a specimen, the c-axis distribution is still of primary importance, as it provides the only basis for comparison of the X-ray results with the abundant optical data on preferred orientation of quartz in the geological literature. It is extremely difficult to measure the basal reflections $0003$ directly in quartz, as the intensity is low and the peak has a $2\theta$ angle very close to the very strong $1122$ reflection. Direct measurements of $0003$ profiles have been obtained on some specimens (Wenk et al., 1967), but these are subject to large errors because of the difficulty in determining the background accurately. For this reason, measured $0003$ profiles were not used in the present analysis, and the c-axis profiles are derived from the inverse pole figure. A measured $0003$ profile for GB-3, given as a chart tracing, is presented in figure 18. There is considerable uncertainty in the background, but the measured curve resembles that computed...
FIG. 16.—Inverse pole figures calculated with 3 2/m symmetry from data at positions 1 through 8 in specimen GB-11 (pl. 1c, d), from seven profiles, truncated at the eighth order.
from the inverse pole figure (fig. 18b) in form.

It should be possible to calculate the c-axis profile with a minimum of input data with the hexagonal (6/mmm) program if profiles of only hexagonal and pseudohexagonal planes are used. Four measured profiles (e.g., 1120, 1010, 1122, 1123) will all twelve measured profiles using the trigonal program. Thus, it appears that if only the 0001 profile is required, it may be determined with the minimum amount of data if hexagonal and pseudohexagonal planes are measured and hexagonal (6/mmm) symmetry is assumed in the calculation.

Fig. 17.—Series of 0001 profiles obtained from the inverse pole figures of GB-11 in fig. 16. Ordinate and “contours” on the top surface of the block are in multiples of a uniform distribution.

yield the eighth- and possibly the twelfth-order harmonics. To test this, twelve profiles were obtained from an additional location close to position 3 in GB-11 (designated 3a). The calculated 0001 profiles are shown in figure 19. The eighth- and twelfth-order profiles calculated from the four measured profiles listed above with the hexagonal program compare well with the sixteenth-order profile obtained from

**DISCUSSION**

Progress in obtaining a theory of the development of preferred orientation in minerals during deformation has been severely hampered until now by our inability to determine the preferred orientations in small and relatively fine-grained experimental samples with sufficient accuracy. Optical (U-stage) methods are inapplicable when the grain size is less than 30–50 μ and,
Fig. 18.—a, Measured profile of 0003 from a transmission scan in specimen GB-3. The approximate background is at B. b, The profile (fig. 14a) calculated from the inverse pole figure is shown for comparison.

Fig. 19.—The 0001 profiles calculated from data for a region near position 3 in GB-11. The sixteenth-order profile from the trigonal program is based on twelve measured profiles. The remaining profiles are calculated with the hexagonal program using only four of the measured profiles.
in any case, yield only the preferred orientation of the 0001 (optic) axes. The X-ray photographic methods used hitherto were not sufficiently quantitative. The new methods described above have removed these limitations. The small grain size of most of the specimens permitted the use of very small collimators so that the preferred orientations could be determined in 1-mm² areas of thin slices. In the coarser specimens, the number of diffracting crystals was increased without an increase of the irradiated volume by oscillation of the sample through 10° during the scan. The oscillation technique was used only in peripheral transmission scans in this study, but it may also be used in spiral scans in both transmission and reflection. Although the technique reduces the resolution and has a smoothing effect on the pole figures, it is very effective in removing the grain (or “crystallite”) effect in samples of marginal grain size.

The methods of spherical harmonic analysis used in this work are modified from methods of earlier workers (notably Bunge, 1965a, b, and Roe, 1965) but are applied for the first time to the study of mineral orientations. They are very powerful in that they (a) constitute a rigorous and effective means of relating statistical distributions of different diffracting planes in the crystals of an aggregate and (b) allow resolution of the preferred orientations of positive and negative forms of quartz crystals; these cannot be resolved by direct X-ray measurements. The latter development should be of great significance in elucidating the mechanisms by which quartz acquires its preferred orientations.

The programs developed for quartz may be used without change for other hexagonal and trigonal minerals, such as graphite (6/mmm), ice (6/mmm), calcite, brucite, siderite, hematite, and corundum (all 3 2/m), and possibly for minerals of related classes, though to recover fully the symmetry in some classes (as for dolomite, 3) some modification is necessary.

The axial symmetry of the preferred orientations in the specimens is advantageous in both the X-ray analysis and the computations: the preferred orientation of a diffracting plane may be represented by a single profile from 0° to 90° from the symmetry axis, instead of a complete pole figure; the crystallite distribution function is expanded with polyhedral harmonics rather than symmetrical generalized spherical harmonics, and may be fully represented in a single spherical projection, such as the inverse pole figure. Most natural preferred orientations of quartz and other minerals show symmetry lower than axial (orthorhombic and monoclinic being common), and the extension of these methods to the determination of natural preferred orientations in rocks presents a challenging problem. The analysis may be generalized, however, as outlined in the description above, to cover specimens with more general symmetry. The operations involve a considerable increase in computation and may require an increase in the number of pole figures for a given order of harmonics over the axially symmetric case considered here. The representation of the crystallite distribution function is also more complex and can be achieved only by serial sections or projections. The latter entail a considerable loss of information (Bunge, 1965c). We are now extending our investigation to this problem, as a more complete description of natural preferred orientations is desirable, and indeed necessary, for an exact comparison of fabrics of naturally and experimentally deformed rocks.

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