

# THE AMERICAN MINERALOGIST

JOURNAL OF THE MINERALOGICAL SOCIETY OF AMERICA

---

Vol. 17

AUGUST, 1932

No. 8

---

## SOME CORRECT AND SOME INCORRECT STATEMENTS OF ELEMENTARY CRYSTALLOGRAPHIC THEORY AND METHODS IN CURRENT TEXT-BOOKS

GEORGE TUNELL AND GEORGE W. MOREY, *Geophysical Laboratory, Carnegie Institution of Washington*

In optical crystallography the following five terms are used very frequently by nearly all investigators and students: ray, wave-front, wave-normal, refractive index, and vibration direction. The meaning of each of the first three of these terms is understood and there appears to be no confusion in their use. Each of the last two terms is used in two different senses by different authors. As shown in Table I four combinations of meanings of these two terms are thus in use at the present time. The student who reads more than one book therefore almost inevitably enters a state of confusion from which it is difficult for him to emerge. The student is especially handicapped by the fact that scarcely an author (except Niggli) states his usage of both of the terms, refractive index and vibration direction, in a prominent place at the beginning of his treatment of the optical crystallography of anisotropic crystals. The usage of each author (except Niggli) must be searched for in the text. In order to assist students and others who wish to compare the various works of reference the following table has been prepared showing the usage of the terms in most of the treatises on optical crystallography.

Cogent reasons for the usage I are given by Pockels in his text-book. Pockels's definition of refractive index is that appropriate to the statement of the fundamental law of refraction as will appear in the following paragraphs. His definition of the vibration direction is that assumed in the electromagnetic theory as being, of the two possibilities, the more probable one. So far as the authors are aware

TABLE I.

The two different senses in which the term refractive index is used and the two different senses in which the term vibration direction is used in the current treatises on crystal optics.

AUTHOR	USE OF TERM REFRACTIVE INDEX	USE OF TERM VIBRATION DIRECTION
I { Pockels <sup>1</sup> Rosenbusch and Wülfing <sup>2</sup> Niggli <sup>3</sup> Johannsen <sup>4</sup> Bouasse <sup>5</sup> Duparc and Pearce <sup>6</sup>	Reciprocal of wavenormal-velocity	Vibration direction lies in wave-front and is not perpendicular to ray.
II { Groth <sup>7</sup> Dana <sup>8</sup>	Reciprocal of ray-velocity	Vibration direction is perpendicular to ray and does not lie in wave-front.
III { <i>a</i> Fletcher <sup>9</sup> <i>b</i> Tutton <sup>10</sup> Miers and Bowman <sup>11</sup>	<i>a</i> . . . . . <i>b</i> { Reciprocal of wavenormal-velocity	<i>a &amp; b</i> { Vibration direction is perpendicular to ray and does not lie in wave-front.
IV Winchell <sup>12</sup>	Reciprocal of ray-velocity	Vibration direction lies in wave-front and is not perpendicular to ray.

<sup>1</sup> F. Pockels, Lehrbuch der Kristalloptik, S. 7, 54, 62, 82, 95 (1906).

<sup>2</sup> Rosenbusch's Mikroskopische Physiographie der Petrographisch Wichtigen Mineralien, Bd. 1, Erste Hälfte, 5. Aufl., E. A. Wülfing, 1. Lieferung, S. 85, 86, 91 (1921).

<sup>3</sup> P. Niggli, Lehrbuch der Mineralogie, 1. Allgemeine Mineralogie, 2. Aufl., S. 348, 359, 373, 374 (1924).

<sup>4</sup> A. Johannsen, Manual of Petrographic Methods, Second edition, pp. 72, 74 (1918). Unfortunately Johannsen's development of the theory is inconsistent with the definitions that he states very distinctly on pages 71, 72. On pages 71, 72 under the heading "Velocity of Any Intermediate Ray in a Uniaxial Crystal" Johannsen writes:

"Let  $r$  = the velocity of the desired ray. . . .

"It is to be noted, however, that the index of refraction of the ray  $r$  is not  $1/r$ , as at first sight one might suppose, but is of a different value."

and on page 72 under the heading "Velocity of Any Intermediate Wave in a Uniaxial Crystal" he writes:

"Let  $MN=w$ , the velocity of the *wave* produced by the [parallel] rays  $r$ ,  $r$ .  
 $\epsilon_1$  = the index of refraction of the ray  $r$ .

"Since the disturbance produced by the ray  $r$  results in forming a wave whose velocity is  $w$ , the index of refraction of this wave is the index of the ray producing this velocity, or

$$w=1/\epsilon_1."$$

On page 81, however, under the heading "The Optical Indicatrix" Johannsen writes:

"If  $MA$  (or  $Mr$ ) represents the velocity of a ray of light, the normal from the vertex of its conjugate  $CM$  (or  $RN$ ) will represent its index of refraction multiplied by a constant."

This is erroneous. On the basis of his definition of refractive index Johannsen should write instead that if  $MA$  (or  $Mr$ ) represents the velocity of a ray of light, its conjugate  $CM$  (or  $RM$ ) will represent its index of refraction multiplied by a constant. The same mistake is made on page 94 in respect to biaxial crystals.

<sup>5</sup> H. Bouasse, *Optique Cristalline Double Réfraction Polarisation Rectiligne et Elliptique*, pp. 4, 176, 177 (1925).

<sup>6</sup> L. Duparc and F. Pearce, *Traité de Technique Minéralogique et Pétrographique*, Première partie, pp. 41, 42, 48 (1907).

<sup>7</sup> P. Groth, *The Optical Properties of Crystals*, Translated by B. H. Jackson, pp. 100, 122 (1910).

P. Groth, *Physikalische Krystallographie*, 3. Aufl., S. 63, 78 (1895).

<sup>8</sup> E. S. Dana, *A Text-book of Mineralogy*, Third edition by W. E. Ford, p. 258 (1922).

<sup>9</sup> L. Fletcher, *The Optical Indicatrix and the Transmission of Light in Crystals*, pp. 32, 54, 55 (1892). Fletcher states that according to the most acceptable form of the solid elastic theory the vibration direction is perpendicular to the ray but that according to the electromagnetic theory it is in the wave-front and not perpendicular to the ray. Fletcher's statements appear to be clear and free from confusion.

<sup>10</sup> A. E. H. Tutton, *Crystallography and Practical Crystal Measurement*, Vol. 2, pp. 877, 878, 879 (1922).

<sup>11</sup> H. A. Miers, *Mineralogy*, Second edition, Revised by H. L. Bowman, pp. 153, 154 (1929).

<sup>12</sup> A. N. Winchell, *The Microscopic Characters of Artificial Inorganic Solid Substances or Artificial Minerals*, Second edition, pp. 77, 78, 114, 115 (1931). *Elements of Optical Mineralogy*, Fourth edition, Part I, pp. 91, 154, 155 (1931).

no reasons are given in the books of later date of publication for deviations from the usage of Pockels. Winchell's deviation in respect to the definition of refractive index leads him into a serious error. Thus he<sup>1</sup> states that:

<sup>1</sup> A. N. Winchell, *The Microscopic Characters of Artificial Inorganic Solid Substances or Artificial Minerals*, Second edition, pp. 77, 78. *Elements of Optical Mineralogy*, Fourth edition, Part I, p. 91.

"An extraordinary ray traveling in an indefinite direction, such as  $OP$ , has a velocity represented by  $OP$ , and an index represented by  $1/OP$ . Since the area enclosed by the tangents and the conjugate diameters of an ellipse is a constant (which may be assumed equal to one) it may be shown that  $1/OP$  is equal to  $RN$  of Fig. 169."

This statement is inconsistent with his later statement<sup>2</sup> that:

"the wave-normal of the extraordinary ray always occupies such a position that the index of the crystal for the extraordinary ray in the given direction is equal to the sine of the angle of incidence divided by the sine of the angle between the wave-normal and the normal to the surface. That is, if  $\epsilon'$  is the index for the extraordinary ray in a special direction and  $R$  is the angle between the wave-normal of that ray and the normal to the surface:

$$\epsilon' = \frac{\sin i}{\sin R},$$

Here  $i$  denotes the angle of incidence and  $R$  denotes the angle of refraction of the wave-normal.

Niggli,<sup>3</sup> however, well and truly says that:

"Die Wellennormale des gebrochenen Strahles liegt in der Einfallsebene. Das Verhältnis des Sinus des Einfallswinkels zum Sinus des Brechungswinkels der Wellennormalen ist dem Verhältnis der Wellennormalengeschwindigkeiten gleich.

"Nun haben wir den Brechungsindex als den reziproken Wert der Normalengeschwindigkeit definiert. Ist das Medium I leerer Raum, so ist somit der Brechungsindex  $n_2$  des zweiten Mediums für die betreffende Welle gegeben durch

$$\frac{\sin e}{\sin i} = n_2."$$

Here  $e$  denotes the angle of incidence (Einfallswinkel) and  $i$  denotes the angle of refraction (Brechungswinkel) of the wave-normal.

The statements of Winchell and Niggli are in contradiction since Winchell defines the refractive index of the extraordinary ray as the reciprocal of the ray velocity. The statement of Niggli is the correct one.

<sup>2</sup> A. N. Winchell, *The Microscopic Characters of Artificial Inorganic Solid Substances or Artificial Minerals*, Second edition, p. 83. *Elements of Optical Mineralogy*, Fourth edition, Part I, p. 96.

<sup>3</sup> *Lehrbuch der Mineralogie*, I. Allgemeine Mineralogie, 2. Aufl., S. 361. Cf. also Duparc and Pearce, *Op. cit.*, pp. 56, 57.

Another error in optical crystallography is made in the description of the phenomenon of exterior conical refraction in Winchell's "Elements of Optical Mineralogy."<sup>4</sup> This error would be considered by the present authors as an inadvertent slip were it not for the fact that it appears in two drawings, one in terms of rays and one in terms of beams, and is explicitly stated in the text in addition. Moreover, it has now gone through the second, third, and fourth editions uncorrected. The following statement written for Winchell's "Elements of Optical Mineralogy" by L. M. Scofield and N. H. Stearn appears on page 157 along with the two drawings (Fig. 185) just mentioned:

"All rays of light incident at  $O$  (Figs. 183 and 185) at right angles to a section normal to  $OR$  (the secondary optic axis) travel with equal velocities in the direction of the rays, and vibrate in various directions at right angles to their various wave-normals; though their directions of propagation are parallel, their wave-fronts are not, that of one being the circle  $II$ , of another the ellipse  $FS$ , and of all others, ellipses intermediate in curvature between these two limiting forms. . . . Upon emerging from a crystal cut normal to  $OR$  (or parallel to  $cc'$ ) these rays form a continuous hollow cone of light of which  $L'N$  and  $L'N'$  are rays at the extremity of a diameter."

Now it is necessary that a hollow cone of rays exactly the same in form as the cone  $KRG$  enter the crystal at  $O$  in order that the hollow cone  $KRG$  may emerge at  $R$ . This fact has already been stated by Pockels<sup>5</sup> in the following words:

"Äussere konische Refraction. Um die dem *Normalenkegel* entsprechende Erscheinung, die *äussere konische Refraktion*, zu beobachten, muss man dafür Sorge tragen, dass innerhalb des Kristalls sich ein Strahl parallel einer Biradialen  $OR$  fortpflanzt. Da dann in der Kristallplatte unendlich viele, den Normalenkegel bildende gebrochene Wellennormalen existieren, so müssen auch im *einfallenden* Licht unendlich viele Wellennormalen- (oder Strahlen-) Richtungen vorhanden sein, die einen bestimmten Kegelmantel erfüllen, dessen Spitze in der Eintrittsfläche liegt. Gesetzt, es falle ein derartiges Strahlenbündel, wie es bei Umkehrung des Strahlenganges aus dem Biradialstrahl hervorgehen würde, auf eine planparallele Kristallplatte, so bilden die

<sup>4</sup> Fourth edition, Part I, pp. 157, 158.

<sup>5</sup> *Op. cit.*, p. 60.

austretenden Strahlen einen ebensolchen Kegel, indem sie von demjenigen Punkte  $\mathcal{U}$  der hinteren Plattenoberfläche aus divergieren, in welchem dieselbe von dem singulären Biradialstrahl getroffen wird. (Siehe Figur 25.)"

The necessity for the incidence of a cone of rays, not merely a single perpendicular ray, is a direct consequence of the most elementary principle of geometrical optics. If the parallel-sided crystal plate be homogeneous then for every wave-normal present in the light after emergence there must be a parallel wave-normal present before incidence (the medium on both sides of the crystal plate being one and the same). Although it is not stated that the medium enclosing the crystal plate is assumed to be isotropic this is clearly the intention of the authors since they state that " $L'N$  is the ray of circular wave front, and, since its wave-normal is normal to the surface of the section, it is not refracted upon emergence," which would not be true in general if the enclosing medium were also anisotropic. Now if the enclosing medium be isotropic, such as air or an immersion liquid, then the cone of incident wave-normals must be identical with a cone of incident rays and the cone of wave-normals on emergence also must be identical with the cone of rays on emergence. Thus to obtain the cone of rays of exterior conical refraction on emergence it is necessary that a similar cone of rays enter the crystal plate.

There is one other elementary feature of the description of exterior conical refraction given in Winchell's "Elements of Optical Mineralogy" that is not stated erroneously but is not brought out with the clarity required in view of the fact that several other text-books state it erroneously, for example, those of Johannsen and of Groth. This point is the following. Contrary to the statements<sup>6</sup> of

<sup>6</sup> The erroneous statements in the two text-books mentioned are as follows:

Johannsen writes: "Exterior Conical Refraction. If a section be cut from a biaxial crystal so that the two parallel faces are normal to the line  $Mp$  (Fig. 166), and a ray of light be passed along the line  $Mp$ , it will emerge in the cone formed by the perpendiculars to the planes  $u'$  and  $e'e''$ . Conversely, a cone of light  $ope$ , entering along the secondary optic axis, will pass through along the single line  $pM$ . This phenomenon is called exterior (or external) conical refraction and also was shown experimentally by Lloyd." (*Op. cit.*, p. 102.)

Groth writes: "since at this place there is a conical depression in the outer skin, an infinity of planes can be passed through the point  $M$  tangent to this skin. The normals to these planes, i.e. the rays in the air proper to the several fronts, form an acute cone. Hence, if such a cone of converging rays is made to fall on a plane-parallel biaxial crystal plate cut perpendicular to a ray-axis  $OM$ , these rays, within

Johannsen, and of Groth, the cone of rays in air emerging from the crystal plate does not have the same shape as the cone of the wave-normals inside the crystal plate corresponding to all possible rays traveling along a biradial. In Winchell's "Elements of Optical Mineralogy" it is stated that "*L'N'* is the ray of elliptical wave-front *FS*, and, since its wave-normal is not normal to the surface of the section, it is refracted away from the direction *OR*, and follows the direction determined by its wave-normal as a spherical wave upon emergence."<sup>7</sup> The student is likely to be confused, however, by the drawing in Fig. 185 in which the cone of rays (in air) outside the crystal plate has the same letters, *KRG*, as the cone of wave-normals inside the crystal plate of Fig. 183, especially since Fig. 183 bears the title "Interior and exterior conical refraction."

Now in the phenomenon of exterior conical refraction the direction of the wave-normal in air after emergence (identical with the ray in air) (*RG* of Winchell's Fig. 185) is in general (for every ray except the ray *KR*) different from the direction of the wave-normal inside the crystal plate (the latter is *RG* of Winchell's Fig. 183),<sup>8</sup> a conclusion necessitated by the law of refraction stated in the quotation from Niggli given above. It is confusing that these two different directions in Figs. 183 and 185 should bear the same letters.

It should be added that the valuable discussion of exterior conical refraction by Voigt in terms of energy transmission (also taken up in Pockels's<sup>9</sup> "Nachträge"), which explains certain observed details, does not diminish the value of the Fresnel ray surface and the explanation of exterior conical refraction derived from it, and in any event has nothing to do with the objections here raised against the treatments in the books of Winchell, of Johannsen, and of Groth, which continue in force.

---

the crystal, are all transmitted along the direction *OM* and with equal velocity, and on emerging they are refracted into a similar cone (exterior conical refraction)." (The Optical Properties of Crystals by P. Groth, Translated by B. H. Jackson, p. 141 (1910).)

<sup>7</sup> The phrases "elliptical wave-front" and "spherical wave" are not happily chosen here but the argument need not be lengthened by discussion of them.

<sup>8</sup> If *RG* in Winchell's Fig. 183 is not intended to be perpendicular to the ray-surface, i.e., not to represent the wave-normal inside the crystal plate, it was incumbent upon Scofield, Stearn, and Winchell to state this fact; in this case Fig. 183 is a very queer drawing.

<sup>9</sup> *Op. cit.*

It is not unreasonable to ask that the author of an American text-book on crystal optics present the main facts accurately, since there already exist text-books on the subject that are accurate and lucid, for example, Pockels's "Lehrbuch der Kristalloptik."<sup>10</sup> This book may be used or consulted by those who are interested only in the geometrical theory of crystal optics and are not interested in the interpretation in terms of electromagnetic theory since Pockels states in his Preface that:

"Freilich schien es mir, um die Darstellung auch den Kristallographen und Mineralogen leichter zugänglich zu machen, ratsam, die Ableitung aus den Differentialgleichungen nicht an die Spitze zu stellen, sondern sie erst, nachdem die Integralgesetze der Lichtbewegung aus einfachen Beobachtungstatistiken mit Hinzunahme naheliegender Verallgemeinerungen entwickelt worden sind, in eingeschobenen Abschnitten nachzuholen, die allenfalls auch vom Leser überschlagen werden können. Diese Darstellung der Theorie hat vor der rein deduktiven zugleich den Vorzug, sich einigermaßen der historischen Entwicklung anzuschließen, sowie die Beziehung zu den Beobachtungen stets hervortreten zu lassen."

The usual methods of determination of the refractive indices of crystalline and other solid substances by immersion of powdered grains in a series of liquids of measured refractive index are described concisely and clearly by Larsen<sup>11</sup> and by Johannsen.<sup>12</sup> Winchell devotes relatively little space to these methods but discusses the index-variation methods in detail. The ease in application of the usual methods, which has already been proved in many places and by many workers, warrants their description in considerable detail in a text-book of crystallography. The majority of measurements of indices of refraction are made for the purpose of the identification of the substance and it is seldom necessary in such cases to vary the composition or temperature of the immersion liquid, or the wave-length of the light used; the greater speed and convenience of the simplest immersion method, by which most of

<sup>10</sup> So far as the present authors are aware the only slip found in Pockels's book thus far is a typographical error on page 23. In the sentence "(Siehe Fig. 10 *a*, *b*, welche den Meridianschnitt der Strahlenfläche für Calomel (+) und Natriumnitrate (-) darstellen.)" the letters, *a*, *b*, should appear in the reverse order.

<sup>11</sup> E. S. Larsen, *U. S. Geological Survey*, Bulletin 679, pp. 12-14, 22-24 (1921).

<sup>12</sup> *Op. cit.*, pp. 258-259.



the actual *determinative* work is done, make it the unquestionable choice in most *determinative* work. In measuring refractive indices for *record* it is desirable to obtain them with an accuracy of  $\pm 0.001$  or better, which can be accomplished by mixing the liquids, or by variation of the wave-length in the dispersion method, or by variation of both wave-length and temperature in the double variation method. It should be noted that the methods called by Winchell "immersion," "single variation" and "double variation" methods are all immersion methods; moreover, the statement that the limit of accuracy under favorable conditions of the "immersion methods" (the term being used here in Winchell's sense) is about  $\pm 0.003$  is misleading; greater accuracy than this can readily be secured by mixing the liquids until a match is obtained without variation of wave-length or temperature, although when greater accuracy than  $\pm 0.003$  is required it is sometimes more convenient to use the dispersion method or the double variation method.

Winchell's<sup>13</sup> statement: "The technique of measuring indices of refraction by this [dispersion] method is the same as that involved in using the double variation method with the single exception that all measures are made at room temperature . . ." is hardly justified by the facts; the present authors would say that the technique required in the double variation method is more complicated. Although Winchell does not claim that the double variation method is superior in point of accuracy to the dispersion method, he does state that the double variation method is more convenient. On this question there is difference of opinion. Attention should be called to the fact that a very important sacrifice, recognized as a disadvantage by Winchell's collaborator, Emmons, is made in securing the ability to vary the temperature over a range by means of Emmons's apparatus. The variation of temperature is accomplished by means of a water cell between the condenser and the preparation, and according to Emmons this makes it impossible to obtain interference figures in convergent light from the grains the refractive indices of which are being measured. Emmons<sup>14</sup> writes:

"I have been asked if interference figures can be obtained with

<sup>13</sup> The Microscopic Characters of Artificial Inorganic Solid Substances or Artificial Minerals, Second edition, p. 65 (1931), or Elements of Optical Mineralogy, Fourth edition, Part I, p. 218 (1931).

<sup>14</sup> *Am. Mineral.*, 14, 425 (1929).

the cell in place. They cannot by any method known to me. Optic axis grains are not difficult to recognize and optic normal grains though not so easy to recognize are less often needed. To measure  $n_p$  and  $n_o$  I believe the standard method is best—namely to choose the grains of highest interference colors in the  $45^\circ$  position and measure a few of them, selecting extreme values. It is to overcome this recognized limitation that I have spent considerable time attempting to adapt the universal stage to this purpose. If successful it should offer an extremely rapid, accurate and generally satisfactory procedure.”

Emmons's statement that “optic normal grains though not so easy to recognize are less often needed” deserves careful attention. Without the possibility of using the conoscope (convergent light) to obtain interference figures and unless the universal stage be used, it is impossible to recognize grains oriented perpendicular to the optic normal or to a bisectrix with certainty. Now in all cases it is highly desirable to supplement the purely statistical method of determining  $\alpha$  and  $\gamma$  ( $n_p$  and  $n_o$ ) by checking the orientations of the grains believed to exhibit  $\alpha$  and  $\gamma$ . If the mineral have no cleavage,  $\alpha$  and  $\gamma$  can be obtained by the purely statistical method but one is never sure even in this case that the lowest index,  $\alpha$ , and the highest index,  $\gamma$ , have really been obtained unless the orientation of the grains be established. In cases of minerals with good cleavage the statistical method is unreliable and the student or investigator who relies upon it exclusively in such cases will come to grief sooner or later. How many mica flakes must one examine to find one standing on edge? The case of mica is extreme; between the case of minerals with one perfect cleavage and that of minerals with no cleavage an enormous number of intermediate stages is represented among known substances. The feldspars, for example, are the most important rock-forming minerals and their determination is frequently undertaken by nearly every petrologist. One of the satisfactory methods for accomplishing this is by measurement of the *three principal refractive indices*. The cleavages of feldspars are of such character that in powder one finds that only a small proportion of the grains do not lie on a cleavage face. (For this reason Tsuboi even found it worth while to prepare data for the determination of feldspars by means of the indices exhibited by grains resting on the cleavage planes.) After a statistical search for the lowest index,  $\alpha$ , or highest index,  $\gamma$ , it is imperative in the case of

a feldspar grain that the orientation of the grain believed to exhibit the minimum or maximum index be established, either by means of its interference figure in the dispersion method or by the use of the universal stage in the double variation method as described by Emmons. In the dispersion method the establishment of the orientation of the grain by means of its interference figure involves merely the removal of the eyepiece and a glance down the tube (or if the grain be small the use of one of the numerous special eyepieces available for this purpose).

Accurate and certain results then can be obtained using the dispersion method involving the spectrometer, refractometer, and petrographic microscope, or by the double variation method involving, in Emmons's set-up, in addition to the apparatus required in the dispersion method, the special water cell and, for the reason stated above, the universal stage. An advantage of the double variation method is that all three indices of a biaxial mineral can often or usually be obtained with a single immersion mount whereas more than one mount is usually required in the dispersion method. The preparation of an additional mount requires only a few seconds work in the dispersion method, however. The use of a smaller number of liquids in the double variation method is cited by Emmons and Winchell as another advantage in comparison with the dispersion method. The decision as to the relative convenience of the dispersion method, by means of which much excellent work has been done over a period of years, and the double variation universal stage method of Emmons must be left to the test of time, although the present authors do not believe that the latter is in general as convenient as the former. It is to be noted that finer grains can be handled accurately by the dispersion method than by the double variation universal stage method of Emmons, an important point in the study of artificial materials, which are so often obtained only in a fine-grained condition. Interference figures can be obtained from grains 0.005 mm. in diameter in many cases and 0.01 mm. in nearly all cases in the dispersion method; in the double variation method Emmons<sup>15</sup> claims that grains as small as 0.03 mm. in diameter can be handled with the universal stage.

Winchell<sup>16</sup> writes: "The single variation method (also called the

<sup>15</sup> *Am. Mineral.*, 16, 554 (1931).

<sup>16</sup> *The Microscopic Characters of Artificial Inorganic Solid Substances or Artificial Minerals*, p. 65 (1931). *Elements of Optical Mineralogy*, Fourth edition, Part I, p. 217 (1931).

dispersion method) as proposed by Merwin and developed by Tsuboi, depends upon variation in wave-length (or color) to vary the index of the liquid." This is a serious misstatement of the history of the dispersion method. Merwin and Larsen<sup>17</sup> suggested a dispersion method for use with melts of sulphur and selenium. Later Merwin<sup>18</sup> used and described such a method employing a series of liquids. In the paper to which Winchell refers, Merwin did not merely propose the dispersion method but gave a complete statement of it together with an elaborate application to numerous salts for each of which each index is given for several different wave-lengths in his table of results. On the other hand, Tsuboi<sup>19</sup> did not develop the method but applied it to certain particular cases. No better statement of the facts can be given than that presented by Tsuboi himself at the beginning of his paper in the *Mineralogical Magazine*, which is as follows:

"Recently H. E. Merwin employed an improved immersion method for determining refractive indices to identify certain salts of the system  $\text{Fe}_2\text{O}_3-\text{SO}_3-\text{H}_2\text{O}$ . In the present paper is described an application of the principle of his method to the determination of plagioclases in cleavage-flakes. The following quotation is from the original statement of the method by the above-mentioned writer [Merwin]:

"In the microscopical determinations chief reliance was placed upon refractive index measurements made in standardized media. For obtaining *optical dispersion* a graphical method was used as follows. Along the right margin of a cross-section paper refractive indices from 1.520 to 1.870 were written so that readings as close as 0.001 could be made. A line was drawn across the paper through the middle at  $45^\circ$ ; along this line the refractive index  $n_D$  of each liquid was marked; above and below each such point were placed points, suitably spaced for interpolation, marking the index of the liquid for other wavelengths; points representing a given wavelength were connected.

<sup>17</sup> *Amer. Jour. Sci.*, (4), **34**, 42-47 (1912).

<sup>18</sup> *Jour. Amer. Chem. Soc.*, **44**, 1965-1994 (1922).

<sup>19</sup> *Min. Mag.*, **20**, 108-122 (1923); *Jour. Geol. Soc. Tokyo*, **32**, 1-6 (1925). An abstract of the latter paper (Tsuboi's paper is in English) is given by L. J. Spencer (*Min. Abs.*, **3**, 197 (1926-1928)) and may be consulted by those to whom the Journal of the Geological Society of Tokyo is not accessible.

'Two (or more) refractive indices of a salt under investigation were found with the aid of a monochromatic illuminator, and placed on the plot. The dispersion was read from a straight line through these points. . . .'

Moreover, extensive application of this method was made by Eskola<sup>20</sup> prior to the publication of the work of Tsuboi just discussed. Eskola says:

"In the determination of the refractive indices I had the advantage of using the improved immersion method as worked out by Merwin. This method involves an improvement in accuracy as well as in completeness, making it possible to determine at the same time dispersion as well as refractive indices. One determines directly, using a monochromatic illuminator, the wavelengths for which the refractive indices to be measured match those of two or more members in the set of refractive liquids. The dispersions of the whole series of liquids used having been determined and expressed graphically, it now remains simply to locate, on the diagram, the points determined and to read the refractive index for any wave-length desired."

In regard to the nomenclature of the immersion method it may be pointed out that the phenomena observed under the microscope with central illumination of powdered grains immersed in a liquid, sometimes called the "Becke line," are improperly so called;<sup>21</sup> the expression, "Becke line," was coined by Salomon<sup>22</sup> to designate the phenomena associated with total reflection at a vertical interface,

<sup>20</sup> *Amer. Jour. Sci.*, (5), 4, 331-375 (1922).

<sup>21</sup> Cf. the following statement by F. E. Wright: "The method of central illumination is frequently called the Becke-line method while that of oblique illumination is designated the Schroeder van der Kolk method. The papers by Becke and Schroeder van der Kolk were of great importance in emphasizing the significance of these methods in microscopical petrography; but in view of the fact that both methods had been described and applied by Maschke and Thoulet one or two decades earlier and also that in ordinary microscope work (biological, etc.) these methods have long been used and called by the above descriptive terms—central and oblique illumination—it would seem better that these terms be employed in preference to the above. The term Becke-line should be retained, however, for some of the relative refringence determinations in rock thin sections; but in refractive index determinations of crystal grains by central illumination the Becke-line constitutes only part of the phenomena observed, the determination being based primarily on the relative convergence or divergence of the light transmitted through the mineral grain." *Jour. Wash. Acad. Sci.*, 5, 104 (1915).

<sup>22</sup> W. Salomon, *Z. Kryst. Min.*, 26, 182 (1895).

which differ in theoretical interpretation from the phenomena obtained with central illumination of irregular grains; the latter are properly known as the central illumination effect.

Winchell, Dana, and some other authors of text-books discuss the symmetry properties of crystals describing three elements, namely, centers, axes, and planes of symmetry. Thus Winchell<sup>23</sup> introduces the student to the subject of crystal symmetry with the following statement:

"All the faces of a crystal, as well as all the constituent atoms, are arranged in accordance with certain elements of symmetry, which are fixed in their position for a given crystal, and determine, not merely its external form, but also the distribution of all the internal physical characters. . . . There are three types of symmetry commonly recognized in crystals, namely, symmetry with respect to (1) a point, (2) a line, (3) a plane. One, two, or three of these types of symmetry may be present in a given crystal.

"A crystal is symmetrical with respect to a point when for each face and edge on one side of the point (or center) there is a similar face and edge directly on the other side of the center. . . .

"A crystal is symmetrical with respect to a line (or axis) when a rotation of less than  $360^\circ$  about this line causes the crystal to occupy exactly the same position in space as at first. If a rotation of  $180^\circ$  produces the first repetition of position, there are two repetitions in a complete rotation, and the line is said to be an axis of two-fold or binary symmetry. . . .

"A crystal is symmetrical with respect to a plane when for each face or edge on one side of the plane there is a similar face or edge directly opposite on the other side, so that one side is the mirror image (in the given plane) of the other."

As an introduction to the subject of crystal symmetry this statement is open to the following serious objection. Axes (rotations), planes (reflections), and centers (inversions) do not constitute a possible minimum set of symmetry elements in terms of which the exterior symmetry of crystals can be described. This can be accomplished by means of either (I) axes (rotations), planes (reflections), and axes of alternating symmetry (rotary-reflections);

<sup>23</sup> The Microscopic Characters of Artificial Inorganic Solid Substances or Artificial Minerals, Second edition, pp. 11, 12 (1931) or Elements of Optical Mineralogy, Fourth edition, pp. 3, 4 (1931).

or (II) axes (rotations), centers (inversions), and rotary-inversions.<sup>24</sup> Set I is that used by Fedorov, Tutton,<sup>25</sup> and Groth.<sup>26</sup>

Tutton<sup>27</sup> writes that:

“Pierre Curie, whose name is so familiar to us in connection with the discovery of radium, had previously made it clear that as regards the exterior symmetry of crystals the whole of the 32 classes could only be fully accounted for by admitting as elements of symmetry not only the well-known planes and axes of symmetry, but also a combined plane and axis of alternating symmetry, that is, a new element of symmetry involving a combined rotation about an axis and reflection across a plane, both operations being completed before the second crystal face is arrived at.”

This element is termed a rotary-reflection by Wyckoff.

If only axes, planes, and centers be admitted as elements of exterior symmetry, then only one-half of the faces of the general form of the tetartohedral (tetragonal bisphenoidal) class of the tetragonal system can be accounted for. E. S. Dana<sup>28</sup> rightly states that the general form of this class includes 4 faces. The Miller symbols of these faces are:  $(hkl)(\bar{k}h\bar{l})(\bar{h}k\bar{l})(kh\bar{l})$ . Dana further rightly states that this class has one binary axis of symmetry, no plane of symmetry, and no center of symmetry. The diagram of Dana further indicates correctly that no horizontal axes of symmetry are present. Winchell and Dana only introduce axes, planes, and centers as elements of exterior symmetry and therefore according to their method the general form of this class should only include two faces and it should be identical with the hemimorphic (monoclinic sphenoidal) class of the monoclinic system; it should then not belong to

<sup>24</sup> The problem here at issue is the statement of the minimum number of symmetry elements necessary to fix the number of faces in the general form and the positions of these faces. The minimum number of symmetry elements at the same time automatically generates all of the remaining symmetry elements of each class. A second and distinct problem is the statement of *all* of the symmetry properties of each of the 32 symmetry classes. The second problem of course cannot be answered without the use of all of the symmetry elements of both Set I and Set II.

<sup>25</sup> Crystallography and Practical Crystal Measurement, Vol. 1, pp. 128-130, p. 135 (1922).

<sup>26</sup> Physikalische Krystallographie, 3. Aufl., S. 311, 312 (1895).

<sup>27</sup> *Op. cit.*, Vol. 1, p. 566.

<sup>28</sup> A Text-book of Mineralogy by E. S. Dana, Third edition by W. E. Ford, p. 90 (1922).

the tetragonal system at all, a fact already pointed out by Rogers.<sup>29</sup> In reality the minimum symmetry requirement of this class is a four-fold axis of rotary-reflection (or a four-fold axis of rotary-inversion) and it does belong to the tetragonal system.

The present authors cannot admit that Winchell and Dana are justified in starting the student off with the idea that axes, planes, and centers of symmetry will serve to explain the exterior symmetry of crystals, especially since the sets of symmetry elements used by the recognized authorities, which are adequate, are not more complicated than the inadequate set described by Winchell and Dana.<sup>30</sup>

Niggli<sup>31</sup> takes Set I as the fundamental elements of symmetry and then shows that a two-fold axis of rotary-reflection gives rise to a center of symmetry and is equivalent to it. Thereafter he uses centers of symmetry in addition to axes, reflections, and rotary-reflections.

Wyckoff<sup>32</sup> gives an excellent brief derivation of the 32 classes from each of the Sets I and II.

<sup>29</sup> A. F. Rogers, *Proc. Amer. Acad.*, **61**, 168 (1926). This statement of Rogers holds true whether one adopts his derivation of the thirty-two symmetry classes or that of Fedorov, Tutton, and Groth. Rogers's derivation given in his paper just cited requires the use of all five symmetry elements of Sets I and II, but in the opinion of the present authors he has not advanced any objection that invalidates the method of Fedorov, Tutton, and Groth.

<sup>30</sup> Dana himself recognized that his method encountered some difficulty as compared with the method in which the symmetry elements of Set I are used; thus Dana wrote: "This method [the method in which the symmetry elements of Set I are used] is not followed here since, though having certain theoretical advantages, it is likely to confuse the student meeting the problems of crystallography for the first time." *A Text-book of Mineralogy*, Second edition, p. 10 (1898).

<sup>31</sup> *Lehrbuch der Mineralogie*, 1. Allgemeine Mineralogie, 2. Aufl., S. 25, 29, 30, 31, 33 (1924).

<sup>32</sup> *The Analytical Expression of the Results of the Theory of Space Groups*, Second edition, pp. 11-15 (1930).