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# OPTICAL SYMMETRY FROM REFLECTIVITY MEASUREMENTS

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## ABSTRACT

Measurements of apparent uniradial reflectivities of randomly oriented grains of 14 anisotropic ore minerals at 549 m$\mu$ are given. Hematite, ilmenite, plumboferrite, niccolite, breithauptite, hollandite and famatinite can be recognized as sensibly uniaxial from the data. The first five can be recognized as optically negative; famatinite and hollandite are optically positive. Manganite, livingstonite, ferberite, arsenopyrite, stromeyerite, enargite and stibnite can be recognized as minerals of lower symmetry, and the first six of these can be classified as optically positive or negative by a suggested convention analogous to that used for biaxial transparent minerals.

The study suggests that the optical symmetries of many anisotropic ore minerals can be partly or wholly determined from reflectivity measurements in monochromatic light, and that the usefulness of reflectivity data in identification of ore minerals can thereby be greatly increased. For any uniaxial mineral, the value of the reflectivity corresponding to the ordinary ray ($R_\omega$) can be determined with an accuracy of $\pm 0.2$ percent, and this can be used as a prime basis of identification. For minerals sensibly of lower symmetry, a value of $R_m$, corresponding to $R_\beta$, the reflectivity for the Y direction of biaxial minerals, can also be determined and used in identification.

## INTRODUCTION

Improved techniques of measurement have greatly enhanced the usefulness of reflectivity in microscopic identification of ore minerals. Precise measurements offer a principal means of distinguishing isotropic minerals. Data for mean reflectivities and bireflectance of anisotropic minerals are appearing, and these properties will be especially useful when international agreement has been reached on reflectivity standards, so that internationally acceptable values for reflectivities can be established.

From theory, however, we should not be limited to just these values, for if the absolute or apparent uniradial reflectivities of a number of grains of a mineral in monochromatic light can be measured with satisfactory precision, the optical symmetry of the mineral can be partly or wholly determined. Unfortunately, a gap has existed between theory and practice, and the feasibility of symmetry determination has not been established. The present paper discusses the determination of optical symmetry from reflectivity data, as illustrated by measurements of apparent uniradial reflectivities of 14 anisotropic minerals. The recognition of $R_\omega$ and $R_m$, specific reflectivity values respectively for uniaxial minerals and minerals of lower symmetry, and their application to mineral identification are likewise discussed.

## METHOD OF STUDY

The apparatus now used in reflectivity measurements in our laboratory is based on a Leitz Epilux microscope with trinocular tube. The upright member of the tube is fitted with a 10-inch extension threaded at top to accept the photomultiplier tube housing of a Photovolt 520-M photometer. The photomultiplier tube is No. 21-C. With the tube extension, the image of a circular area of the mineral grain 20 microns in diameter is spread over an area on the photosensitive surface of the phototube approximately $\frac{1}{8}$ inch in diameter. A conical extension piece screwed into the phototube housing screens out the portions of the mineral grain lying outside the 20-micron circle. The extension tube has a slot into which a Bausch and Lomb 1×1-inch 2nd-order interference filter can be inserted. The filter used for the study here described has peak transmittance at 549 $m\mu$ and a 9 $m\mu$ half-width.

The trinocular tube is fitted with a prism. When this is inserted in the system, the image of the mineral being investigated is seen through the binocular portion of the trinocular tube. One eyepiece is fitted with a micrometer disc, the other with crosshairs. The polished section to be studied is accurately leveled. The grain to be measured is brought to the center of the field, and the micrometer scale is used to check on the limits of the image area that will be thrown onto the photomultiplier tube when the prism is withdrawn from the system. This area must be 20 microns or more in diameter and free of cracks, pits, scratches, and all other imperfections. The objective used is a 40×, n.a. 0.85 mm coated objective, carefully centered. The polarizer vibration plane is oriented parallel to the symmetry plane of the reflecting prism. The grain is precisely focused with the aid of tiny pits or other *minute* imperfections of the surface outside the 20-micron circle. *If no such imperfections are present, accurate focusing is ordinarily not possible*, and no measurement is made. The prism is now withdrawn so that light reflected from the chosen area passes directly upward to the photomultiplier tube. The deflection of the galvanometer is read as a measure of the reflectivity. If the mineral is anisotropic, then as the stage is rotated a minimum reading for the grain ($R_1$) is obtained when one of its vibration directions is parallel to the polarizer vibration direction, and a maximum reading ($R_2$) when the other vibration direction is parallel to the polarizer vibration plane. At each position, the focus is carefully checked before the final reading is taken.

The values of $R_1$ and $R_2$ obtained will be the apparent uniradial reflectivities of the grain. They may be converted to absolute values by use of a curve constructed, for the particular microscope system used, with reference to a series of standards of known reflectivities. For determination of optical symmetry, however, correction is unnecessary, since only the variation or lack of variation of $R_1$ or $R_2$ from grain to grain is significant.

A stabilized lamp circuit essentially similar to that of Bowie and Taylor (1958) is used. As a check on the stability of the lamp current and photometer, however, a reading must be taken on an isotropic standard at intervals during each series of measurements. In the present study, the standard used is a polished section of silicon for which the value of R was arbitrarily taken as 40 percent. The values reported below are therefore apparent uniradial reflectivities. Until international reflectivity standards have been established, there is no point in attempting to convert them to absolute uniradial reflectivities.

For each mineral studied, a polished section containing numerous differently oriented grains of the mineral was used. Each section contained a number of grains with perfectly polished areas in excess of 20 microns in diameter. Each surface was freshly cleaned before measurements were made, by light buffing if necessary.

## REFLECTIVITY OF UNIAXIAL MINERALS

The reflectivity of a uniaxial mineral grain in air in normally incident plane-polarized light is a function of the orientation of the polished section of the grain with respect to the crystallographic axes and the orientation of the grain with respect to the vibration plane of the polarizer. If the grain is cut $\perp c$, then its reflectivity ($R_\omega$) in air is given by:

$$R_\omega = \frac{(n_\omega - 1)^2 + k_\omega^2}{(n_\omega + 1)^2 + k_\omega^2},$$

where $n_\omega$ and $k_\omega$ are respectively the index of refraction and coefficient of absorption for the ordinary ray. If the grain is not cut $\perp c$, then it will have two vibration directions corresponding to the ordinary and extraordinary rays. When the ordinary ray vibration direction is parallel to the polarizer vibration plane, the reflectivity is given by the equation above, but when the extraordinary ray vibration direction is parallel to the polarizer vibration plane, the reflectivity of the grain is given by

$$R_{\epsilon'} = \frac{(n_{\epsilon'} - 1)^2 + k_{\epsilon'}^2}{(n_{\epsilon'} + 1)^2 + k_{\epsilon'}^2}$$

where $n_{\epsilon'}$ and $k_{\epsilon'}$ are respectively the index of refraction and coefficient of absorption for the extraordinary ray. The values of $n_{\epsilon'}$, $k_{\epsilon'}$, and hence $R_{\epsilon'}$ diverge increasingly from $n_\omega$, $k_\omega$, and $R_\omega$ as the grain section approaches parallelism with the $c$ axis. For the section $\parallel c$, the differences between $R_{\epsilon'}$ and $R_\omega$, $n_{\epsilon'}$ and $n_\omega$, and $k_{\epsilon'}$ and $k_\omega$ all reach maximum values

($n_{\epsilon'} = n_\epsilon$; $k_{\epsilon'} = k_\epsilon$). $R_\epsilon$ may be either greater or less than $R_\omega$. In transparent crystals, for which the values of $R_\omega$ and $R_\epsilon$ are determined solely by $n_\omega$ and $n_\epsilon$ in a given immersion medium ($k_\omega = k_\epsilon = 0$), the vibration direction of higher reflectivity is the direction of higher $n$. If this is the ordinary ray vibration direction, the mineral is of course optically negative (*e.g.*, Cambon, 1947, p. 46–53; Cameron, 1961, p. 171–173; 183–184). The same convention, with regard to reflectivity, can be used for absorbing uniaxial minerals, although in these minerals reflectivity is a function of $k$ as well as $n$, and the vibration direction corresponding to maximum $n$ may actually be the vibration direction for minimum $k$ (Berek, 1937, p. 60–61).

Let us suppose that we now measure the maximum and minimum reflectivities (corresponding to the two vibration directions) of each of a series of differently oriented grains of a uniaxial mineral. One of two cases will be found, if low values and high values for various grains are compared. In the one case, the high values ($\omega$ ray) for all grains are the same within the error of measurement, whereas the low values ($\epsilon'$ ray) are different for different grains. In this case the mineral is uniaxial negative. In the second case, the low values are the same, the high values differ, and the mineral is uniaxial positive.

## REFLECTIVITY OF MINERALS OF LOWER SYMMETRY

In transparent minerals of lower symmetry, variations in index of refraction and hence in reflectivity are commonly referred to the three coordinate ether axes X, Y, Z, with corresponding indices of refraction $n_\alpha$, $n_\beta$, and $n_\gamma$. For a section parallel to a principal plane (e.g., X-Z), the reflectivities when the X and Z ether axes are respectively parallel to the polarizer are, in air,

$$R_\alpha = \frac{(n_\alpha - 1)^2}{(n_\alpha + 1)^2}; \qquad R_\gamma = \frac{(n_\gamma - 1)^2}{(n_\gamma + 1)^2}$$

Reflectivities for the two vibration directions in the X-Y and Y-Z planes are similarly calculated from $n_\alpha$ and $n_\beta$, and $n_\beta$ and $n_\gamma$, respectively. For a section oblique to the ether axes, reflectivities for the two vibration directions are given by

$$R_1 = \frac{(n_1 - 1)^2}{(n_1 + 1)^2} \quad \text{and} \quad R_2 = \frac{(n_2 - 1)^2}{(n_2 + 1)^2}$$

where $n_1$ is the index of refraction for one vibration direction, $n_2$ for the other. If $n_1$ is the lower index, $R_1 < R_\beta < R_2$, except that a section perpendicular to an optic axis will have $R_1 = R_\beta = R_2$, a section parallel to X-Y will have $R_2 = R_\beta$, and a section parallel to Y-Z will have $R_1 = R_\beta$.

If, for each of a series of randomly oriented grains of an orthorhombic, monoclinic, or triclinic transparent mineral, we now determine in monochromatic light the reflectivities corresponding to the two vibration directions, neither the higher nor the lower set of values will be constant. For any grain, $R_1$ may have any value between $R_\alpha$ and $R_\beta$, and $R_2$ may have any value between $R_\beta$ and $R_\gamma$. Provided the values of $R_\beta - R_\alpha$ and $R_\gamma - R_\beta$ are large relative to the error of measurement, the biaxial character of the mineral should be evident from reflectivity measurements.

In orthorhombic absorbing minerals, variations in $n$ and $k$, and hence of R, are described with reference to two sets of rectangular coordinate ether axes, $x'$, $y'$ and $z'$, and $x''$, $y''$ and $z''$, both sets of axes coinciding with the crystallographic axes. The sets of axes are mathematically complex. The optical indicatrix consists of two triaxial ellipsoids having a common center (Rinne-Berek, 1953, p. 197), but the radial vectors of these ellipsoids do not have the significance of indices of refraction and coefficients of absorption. Variations in $n$, $k$ and R with ray path in an orthorhombic absorbing crystal cannot be visualized, although they can be calculated and plotted for the three principal crystallographic planes of such a crystal. In monoclinic and triclinic crystals, optical relations are even more complex.

In the general case, a section of an absorbing mineral of lower symmetry, like that of a transparent mineral, will have two vibration directions at right angles for which the reflectivities in air are respectively

$$R_1 = \frac{(n_1 - 1)^2 + k_1^2}{(n_1 + 1)^2 + k_1^2}; \qquad R_2 = \frac{(n_2 - 1)^2 + k_2^2}{(n_2 + 1)^2 + k_2^2},$$

$n_1$ and $k_1$ corresponding to one vibration direction and $n_2$ and $k_2$ corresponding to the other. Calculations from these equations, using various values of $n$ and $k$, show that so long as $k$ is small relative to $n$, the effect of absorption on values of R is small. This is believed to be the case for most absorbing minerals of lower symmetry, hence for most of them patterns of variation of $R_1$ and $R_2$ should be essentially the same as for transparent minerals.

## MEASUREMENTS

*Uniaxial minerals.* Reflectivity measurements were made for hematite, ilmenite, plumboferrite, niccolite, breithauptite, famatinite and hollandite, as examples of uniaxial minerals. Measurements for individual grains of four of the seven minerals are given in Table 1 to illustrate the pattern of reflectivities found in all seven. The error in reading the photometer depends on the position of the needle with reference to the galvanometer scale divisions, varying up to 0.1 division (roughly 0.1 percent). Total error indicated by the data and by series of measurements of the reflectivities of isotropic minerals ranges up to ±0.2 percent.

It will be noted that all grains of any one mineral show either the same high value or the same low value, within the error of measurement, in accordance with their uniaxial character. Hematite, plumboferrite and breithauptite are optically negative; hollandite is optically positive. Corresponding data (in percent) for niccolite (21 grains) are $R_\omega = 57.8$–$58.2$; mean $R_\omega = 58.0$; mean deviation of $R_\omega = 0.15$; $R_{\epsilon'} = 51.1$–$56.0$. For ilmenite (13 grains), $R_\omega = 19.7$–$20.0$; mean $R_\omega = 19.9$; mean deviation of $R_\omega = 0.07$; $R_{\epsilon'} = 16.4$–$19.4$. For famatinite (14 grains), $R_\omega = 25.8$–$26.2$; mean $R_\omega = 26.0$; mean deviation of $R_\omega = 0.08$; $R_{\epsilon'} = 26.1$–$29.5$. The data indicate that niccolite and ilmenite are optically negative, famatinite is optically positive.

TABLE 1. REFLECTIVITY MEASUREMENTS—UNIAXIAL MINERALS, IN PER CENT

| Grain No. | Hematite | | Plumboferrite | | Breithauptite | | Hollandite | |
|---|---|---|---|---|---|---|---|---|
| | $R_{\epsilon'}$ | $R_\omega$ | $R_{\epsilon'}$ | $R_\omega$ | $R_{\epsilon'}$ | $R_\omega$ | $R_{\epsilon'}$ | $R_\omega$ |
| 1 | 32.1 | 32.5 | 23.9 | 25.7 | 40.6 | 54.1 | 34.1 | 27.9 |
| 2 | 31.9 | 32.6 | 24.0 | 25.5 | 49.3 | 54.3 | 34.1 | 28.0 |
| 3 | 28.0 | 32.4 | 23.4 | 25.6 | 41.5 | 54.2 | 33.0 | 27.7 |
| 4 | 28.8 | 32.4 | 23.5 | 25.7 | 41.7 | 54.2 | 33.6 | 27.8 |
| 5 | 31.8 | 32.4 | 24.1 | 25.7 | 44.5 | 54.0 | 33.5 | 28.1 |
| 6 | 28.4 | 32.2 | 23.9 | 25.7 | 50.8 | 54.0 | 34.5 | 27.8 |
| 7 | 32.1 | 32.5 | 24.0 | 25.7 | 52.1 | 54.0 | 34.3 | 27.9 |
| 8 | 31.9 | 32.4 | 23.8 | 25.8 | 40.0 | 54.3 | 35.2 | 27.8 |
| 9 | 31.8 | 32.6 | 24.3 | 25.8 | | | 34.7 | 27.9 |
| 10 | 32.0 | 32.5 | 24.1 | 25.8 | | | 32.9 | 27.9 |
| 11 | 28.2 | 32.5 | 24.0 | 25.8 | | | 31.5 | 27.8 |
| 12 | | | 24.3 | 25.8 | | | 30.9 | 27.8 |
| Mean $R_\omega$ | 32.4+ | | 25.7 | | 54.1 | | 27.9 | |
| Mean deviation, $R_\omega$ | .09 | | .07 | | 0.12 | | 0.08 | |
| Range, $R_{\epsilon'}$ | 28.0–32.1 | | 23.4–24.3 | | 41.0–52.1 | | 30.9–35.2 | |
| Optic sign | (−) | | (−) | | (−) | | (+) | |

*Minerals of lower symmetry.* Reflectivity data for the four orthorhombic minerals stibnite, enargite, stromeyerite, and manganite, and for the three monoclinic minerals livingstonite, ferberite, and arsenopyrite are given in Figs. 1 and 2. Each line of a figure represents, to scale, the bireflectance of a single grain. The end points of the line give the values

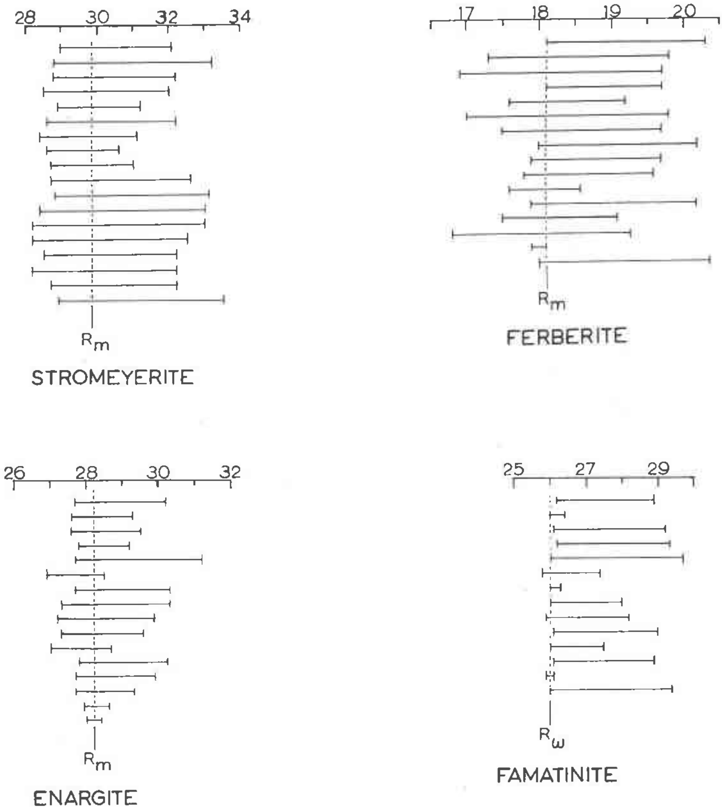

FIG. 1. Reflectivities of diversely oriented grains of stromeyerite, ferberite, enargite and famatinite. Scales give reflectivities in per cent, referred to $R_{Si}=40.0$ per cent.

of $R_1$ and $R_2$ for the grain. Even assuming an error of measurement of as much as ±0.3 percent, the measurements indicate that for none of the minerals is $R_1$ or $R_2$ constant, and that the minerals cannot be uniaxial. Those experienced in ore microscopy will recognize that this is true despite the fact that the minerals range from strongly bireflectant (*e.g.*, manganite) to weakly bireflectant (*e.g.*, arsenopyrite). For comparison, data for individual grains of famatinite are plotted alongside those for enargite in Fig. 1.

For stibnite, stromeyerite, livingstonite, ferberite and arsenopyrite, the order of grains from top to bottom is the order in which they were measured. In each case, the polished section was traversed by means of a mechanical stage, and $R_1$ and $R_2$ of each suitably polished grain were measured as the grain was intersected. Selection of grains for measurement was therefore random. The same is true for manganite, except that

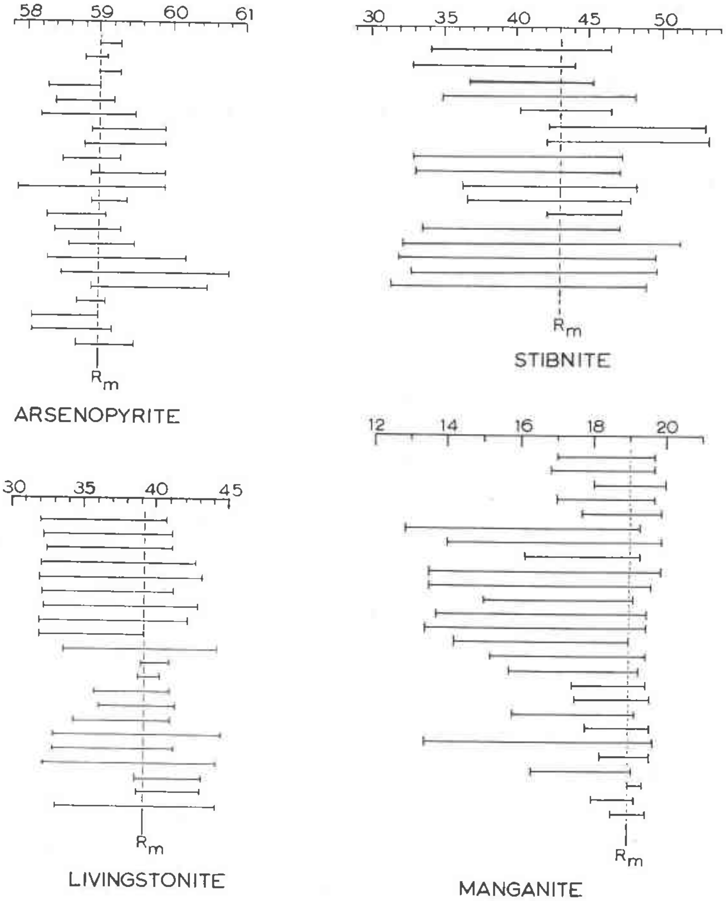

FIG. 2. Reflectivities of diversely oriented grains of arsenopyrite, stibnite, livingstonite and manganite. Scales give reflectivities in per cent, referred to $R_{Si}=40.0$ per cent.

the last five grains measured were deliberately selected as grains of low bireflectance. Except for enargite and stromeyerite, measurement of the first half dozen grains measured was sufficient to indicate non-uniaxial character. For enargite, $R_1$ and $R_2$ were determined for 27 grains; the data plotted are for grains selected as illustrating the range of values. The range of $R_1$ for the first 12 grains encountered was only 0.6; the reason may be a certain degree of preferred orientation. It is evident, however, that some caution must be used in interpreting measurements of a limited number of grains of a mineral.

The most interesting feature of the measurements plotted in the figures, apart from the clear indication that they give of non-uniaxial character, is that for every mineral the pattern of variation of $R_1$ and $R_2$

from grain to grain is the same as would be expected if the minerals were transparent. Each grain of manganite for example (Fig. 2), shows a value of $R_1$ that is equal to or less than $19.1 \pm 0.1$, and a value of $R_2$ that is either equal to or greater than $19.1 \pm 0.1$. If the mineral were transparent, this value would be taken as the $R_\beta$ value. Further, since this value is evidently closer to the maximum value for the mineral ($R_\gamma$) than to the minimum value ($R_\alpha$), the mineral would be indicated as optically negative. The reflectivity of manganite is determined by $k$ as well as $n$, but inasmuch as it is orthorhombic there should still be minimum, intermediate, and maximum values of R corresponding to the three coordinate ether axis directions (parallel to $a$, $b$ and $c$). If we designate these values of R as respectively $R_p$, $R_m$, and $R_g$, then $R_m$ corresponds to $R_\beta$ in transparent minerals. There would seem no objection to using also the same convention of optic sign as for transparent minerals, and designating manganite as optically negative at $\lambda = 549$.

For stibnite, the value of $R_m$ is indicated as $43.0 \pm 0.6$. $R_m - R_p$ is approximately equal to $R_g - R_m$, hence the optic sign is indeterminate within the limits of error. For enargite, the value of $R_m$ lies between 28.0 and 28.4; the indicated optic sign is positive. For stromeyerite, the value of $R_m$ is only roughly indicated, but the optic sign is positive.

For the three monoclinic minerals, despite possible complexities due to absorption, the pattern of variation of $R_1$ and $R_2$ is the same as for the orthorhombic minerals. For ferberite and livingstonite, one may speculate that this is because $n$ is large relative to $k$, hence optically the two minerals are nearly transparent, but this is much less likely for aresnopyrite, a mineral of high reflectivity. For practical purposes, however, the important thing is that a value of $R_m$ can be recognized for each mineral. Based on the ratio of $R_m - R_p / R_g - R_m$ in each case, livingstonite can be designated as optically negative at $\lambda = 549$, the others as optically positive.

## APPLICATIONS TO IDENTIFICATION OF ANISOTROPIC MINERALS

The foregoing data cover only 14 minerals. However, both weakly and strongly anisotropic minerals are included, hence the data suggest that most minerals can be recognized either as sensibly uniaxial or nonuniaxial from careful reflectivity measurements of randomly oriented grains in polished sections. If a mineral is sensibly uniaxial, it can be classified as optically positive or optically negative in somewhat the same sense as transparent uniaxial minerals. If the mineral is sensibly of lower symmetry, a diagnostic value of $R_m$ can be determined. Further, if $R_m$ is clearly closer to $R_p$ than to $R_g$, an unknown mineral can be classified as optically positive, whereas if $R_m$ is closer to $R_g$, the mineral can be classified as optically negative, it being understood that the significance of sign is not the same for absorbing minerals of lower symmetry as for transparent minerals.

It is not to be expected that optical symmetry will be determinable for all anisotropic minerals from reflectivity data, barring a marked improvement in precision of measurement. First, the method will be inapplicable to minerals of extremely low bireflectance, such as chalcopyrite. Second, if $R_m-R_p$ or $R_g-R_m$ for a mineral of lower symmetry is less than the error of measurement, the mineral will be sensibly uniaxial. Despite these limitations, however, the data have a bearing on tables of values of reflectivities and other optical properties of anisotropic ore minerals. Except as noted above, selection of grains was random. The figures indicate that for uniaxial minerals of measurable bireflectance, the value of $R_\omega$ can be rapidly determined, likewise the optic sign. To find the true value of $R_\epsilon$, a search for grains of maximum anisotropism must be made, and inasmuch as strength of anisotropism is a function of the phase difference produced by the mineral as well as the ratio $R_\omega:R_\epsilon$, a sizeable number of grains must be measured. On the other hand, measurement of 4 or 5 grains showing various degrees of anisotropism will give both $R_\omega$ and the optic sign. For purposes of rapid identification, it is possible to divide uniaxial minerals into two groups according to the optic sign, and to arrange them in each group in order of increasing $R_\omega$.

For the minerals of lower symmetry, there are two possible schemes of arrangement according to reflectivity. One is an arrangement in order of increasing mean reflectivity, defining this term as $R_p+R_g/2$. To obtain the true value for a particular mineral, a grain so oriented as to show maximum bireflectance must be found. The present results suggest an alternative; namely, arrangement in order of increasing $R_m$. As $R_m$ can be found by a series of measurements of diversely oriented grains encountered at random in traversing a section, its determination should ordinarily be faster than the determination of mean reflectivity. The use of $R_m$ as a basis for arrangement of minerals of lower symmetry in tables therefore merits consideration.

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