THE STRUCTURE OF LABRADORITE AND THE ORIGIN OF ITS IRIDESCENCE

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1. Introduction

LABRADORITE which is a crystalline feldspar of the triclinic class is well known for the striking iridescence or play of colours which it exhibits. The phonomenon is displayed not only by specimens from the locality which gave its name to the substance, namely the coast of Labrador, but also by those from other parts of the world. This fact and the highly distinctive character of the effects observed suggest that they are in some way connected with the nature of the feldspar itself. Hintze (1897) reviewed the earlier attempts to find an explanation of the iridescence. Later investigations published about the same time by Rayleigh (1923) and by Boggild (1924) made useful contributions to the subject; but they left unanswered the fundamental questions which arise, namely, what is the material which is responsible for the iridescence of labradorite and why is it present? In the hope that a more searching examination of the phenomena than has hitherto been made might result in finding the answers to these questions, a representative collection of specimens was acquired for the crystallographic museum of this Institute. They have been carefully studied, and the investigation has revealed basic new facts concerning the iridescence of labradorite and enabled us to reach definite conclusions regarding its origin.

2 THE CHEMICAL COMPOSITION OF LABRADORITE

Mineralogists place labradorite fairly high up in the sequence of the plagioclase feldspars running from albite to anorthite. The two end-members of the series both belong to the triclinic class of crystals, and it is a well-established fact that they are isomorphous and can mix freely in all proportions to form homogeneous crystals. To describe labradorite as a mixture of albite and anorthite, in other words of soda and lime feldspars is, however, only an idealisation and not a complete picture of the reality. The chemical analyses tabulated by Hintze (loc. cit.) show that labradorite contains also an appreciable proportion of potash feldspar. There are also other materials present as impurities in variable amounts, these being principally Fe₂O₂ and MgO. Examination of labradorite under a binocular microscope shows the

presence of inclusions of different kinds trapped at various levels within the crystal. They appear as discrete crystals with sharply defined outlines and evidently represent the impurities revealed by the chemical analysis. Their distribution within the labradorite is irregular, even in those cases in which the characteristic iridescence of the feldspar appears with remarkably uniform intensity and colour over the entire volume of the specimen. The facts stated make it highly improbable that the characteristic iridescence of labradorite is due to the presence in it of extraneous impurities or inclusions. They suggest, rather, that the phenomenon has its origin in the constitution of the feldspar itself. Our attention is thus naturally directed to the third component of the feldspar shown by the chemical analysis to be present, namely, orthoclase or potash feldspar.

3. THE LAMELLAR STRUCTURE OF THE MIXED FELDSPARS

The crystallographic data and the X-ray investigations that have been 'made (Taylor, 1934) indicate that all the feldspars possess certain structural features in common. Nevertheless, there are well-established differences amongst them, the most notable being that the feldspars in which the potash is the principal component crystallise in the monoclinic system, whereas those in which the soda and lime feldspars are the principal components form triclinic crystals. In view of its lack of isomorphism with the two other feldspars, we could scarcely expect potash feldspar to be freely miscible with them in all proportions or to form homogeneous crystals as the result of such mixture. Actually, when the results of the chemical analyses of nearly a thousand specimens of naturally occurring feldspars are plotted as points on a three-component diagram (Alling, 1921), it is found that a large part of the area of the triangle is empty and that a potash-lime series of feldspars is not represented in the diagram. The occupied area consists of a belt lying along the two sides of the triangle representing respectively a soda-potash series of feldspars and a soda-lime series, its width being greatest in the vicinity of the vertex representing pure albite and narrowing rapidly as the vertices representing pure orthoclase and pure anorthite are approached. It should be emphasised that the feldspars found in nature are not invariably homogeneous crystals. Indeed, it is well known that many of the feldspars of the soda-potash series are not of that description. Amongst the feldspars of the series in which orthoclase is the major component, only those containing a relatively small proportion of albite are truly single crystals. The others are essentially intergrowths in which the monoclinic and triclinic components have separated from each other to form lamellar structures. The lamellæ may be so coarse as to be visible to the naked eye (perthites);

they may also be so fine as to be visible only under a microscope or even so fine as to be optically irresolvable (microperthites).

In view of what has been said above, the question arises as to how the component of potash feldspar present in the soda-lime series of feldspars is distributed within the material, viz., whether it forms a homogeneous solid solution with the albite-anorthite, or whether it is segregated from the sodalime feldspar and forms crystallites of orthoclase which remain embedded in the albite-anorthite matrix and distributed through its volume. The latter alternative has to be recognised at least as a possibility. The optical behaviour of labradorite would be profoundly different according as the first or the second alternative is the correct answer. In the former case, it would be a transparent colourless crystal, assuming that there are no other inclusions or impurities present. In the second case, the difference in refractivity between the orthoclase crystallites and the albite-anorthite mass in which they are embedded is considerable. Hence their presence would render the crystal an optically heterogeneous medium, causing it to exhibit various effects when traversed by a beam of light. Since labradorite does exhibit such effects, we are entitled, at least as a working hypothesis, to adopt the second of the alternatives stated and to develop its consequences for comparison with the facts of observation.

4. THE ORIENTATION OF THE LAMELLÆ

Accepting the validity of our hypothesis, we have to ask ourselves other questions, viz., what is the shape, size and orientation of the crystallites of orthoclase? Is the setting of the crystallites arbitrary and unrelated to the structure of the albite-anorthite crystal, or does it have some definite relation to it and therefore possesses some measure of uniformity?

In seeking an answer to the questions raised, the facts which have been established by X-ray investigations on the soda-potash series of feldspars give us some useful guidance. Ito and Inuzuke (1936) found that two structures are present in the microperthites, one similar to pure potash-orthoclase, the other similar to albite. Their work has been confirmed by Chao, Smare and Taylor (1939), while a further study by Chao and Taylor (1940) revealed some finer details. For our present purpose, the principal result that emerges from the X-ray studies is that the crystal axes of the two structures present in the microperthites are either coincident, or else make small angles with each other. The latter result is not unexpected, since one of the structures is monoclinic and the other triclinic. We have, therefore, ample justification for assuming that a similar situation would exist in our present problem, in other words, that the crystal structures of the orthoclase and of the

surrounding albite-anorthite mass would be coherent. From a crystallographic point of view, therefore, we may, without serious error, consider the whole of the material as a single unit in spite of its optical heterogeneity.

In the circumstances stated, it is not unreasonable to assume that the actual shape and setting of the orthoclase crystallites would bear some recognisable relationship to the lattice structure of the enclosing crystalline mass. According to Chao and Taylor (1940), the crystal structure of labradorite is itself rather complex, sheets of albite and sheets of anorthite, each a few unit cells thick, alternating along the c-axis. In these circumstances, it is scarcely possible even to venture a guess as how the third component, viz., the monoclinic orthoclase would dispose of itself in the lattice. We have necessarily to regard this as a matter for observational study rather than for theoretical prediction. Nevertheless, as will be shown in the following sections of the paper, the theoretical ideas set out above have various specific consequences which can be tested by observation or experiment. The complete success met with in such tests is testimony to their essential correctness.

5. Some Consequences of the Twinning of Labradorite

As is well known, the plagioclase feldspars frequently exhibit multiple or repeated twinning of the albite type. The brachypinacoid or 010 face is both a plane of reflection and a plane of composition in this type of twinning; in other words, the crystal structures on either side of a composition plane are mirror images of each other. The albite type of twinning is readily recognized by the appearance of sets of parallel lines where the composition planes meet the external surface of the crystal. They are most clearly scen on the 001 faces, since these are the most perfect cleavages of the crystal and are nearly normal to the twinning planes. The visible signs of albite twinning on the 001 faces may be traced across them right up to the edges of any artificially polished surface which meets them. We shall proceed to consider how such twinning would influence the optical phenomena exhibited by labradorite.

It will be assumed that the iridescence displayed by labradorite arises from the presence in it of thin lamellæ of orthoclase which reflect a beam of light travelling through the crystal by virtue of their refractive index being less than that of the surrounding albite-anorthite mass. The direction in which the light is reflected would depend on the angle of its incidence on the lamellæ and hence also on the orientation of the latter within the crystal. This orientation is more or less precisely defined, being determined by physico-chemical considerations connected with the crystal structures of the three components of the feldspar. Further, the orthoclase lamelæ, though

optically different from the surrounding mass, are nevertheless an integral part of the crystal. Hence, if the latter is a simple albite twin, the orientations of the lamellæ in the components of the twin would not be identical but would be mirror-images of each other with respect to their composition plane. For the same reason, also, if the labradorite is a multiple or repeated twin, the orientations of the lamellæ in the 1st, 3rd, 5th, etc. twinning layers of the crystal would be parallel to each other, and likewise also the orientation of the lamellæ in the 2nd, 4th, 6th, etc. twinning layers, but the orientations of the lamellæ in the two sets of twinning layers would be different, being the mirror images of each other in the composition planes. The thickness of the orthoclase lamellæ influences their optical behaviour, including especially the colour which they exhibit. If this be the same in the 1st, 3rd, 5th, etc. twinning layers of the crystal, there is no reason why it should be different in the intervening 2nd, 4th, 6th, etc. layers. Hence, we arrive at the following inferences as necessary consequences of our theoretical views.

- A. Every specimen of labradorite which is twinned according to the albite law and exhibits iridescence, should display the latter in two distinct settings of the specimen, if the direction of incidence of the light and the direction of observation are pre-determined.
- B. For the same reason, for any given setting of the specimen and a given direction of incidence of the light on it, the iridescence should be observable in two divergent directions.
- C. The two reflections mentioned above have their origin in different parts of the crystal, namely the two differently orientated sets of components of the multiple twin.
- D. The two sets of reflections should in similar circumstances exhibit identical colours.
- E. The settings of the crystal required for observing the two independent sets of reflections should make equal angles with the setting required to obtain a specular reflection from the composition planes of the twinning.

Another kind of twinning which is frequently observed in the plagioclase feldspars is known as the pericline type. This species of twinning is almost invariably multiple with closely spaced components, the structures of which are derivable from each other by a rotation of the crystal about the b-axis through 180° . The composition planes of the pericline twinning in labradorite are nearly parallel to the 001 faces of the crystal, actually making an angle only 6° with them. The twinning manifests itself externally by parallel

and closely spaced striations on the 010 faces of the crystal and is thus readily distinguishable from the albite twinning where no such striations are observed.

Reasoning analogous to that set out in the case of albite twinning leads to theoretical consequences similar to those listed above as A, B, C, D and E, except that the word "pericline" is substituted for "albite" in the description of the twinning and E is modified to read as follows:—

E'. The settings of the crystal required for observing the two independent sets of reflections would be obtained, one from the other, by a rotation of the crystal about its b-axis through 180°.

6. Comparison with the Facts of Observation

Our theory thus leads us to the remarkable conclusion that precisely definable relationships should exist between the kind of twinning of the albite or the pericline type, as the case may be, which a particular specimen of labradorite exhibits and the circumstances in which it displays iridescence as well as the observable characters of such iridescence. It is scarcely conceivable that any such relationships could exist unless it is the case that the material responsible for the iridescence of the feldspar is itself an integral part of the structure of the crystal. It follows that relationships indicated by the theory, if they actually exist, are a convincing demonstration of its correctness.

The specimens of labradorite at our disposal form a fairly representative collection, both in regard to the colours exhibited and in respect of their types of twinning as identified by their external manifestations. Some of our specimens display exclusively the albite type of twinning, some exclusively the pericline type, and others show both. The material examined by us is thus sufficiently extensive to prove or disprove the deductions from the theory. Actually, the observations support the theoretical inferences in the most complete and detailed fashion.

The existence of the two sets of reflections exhibiting the same colours but originating in different parts of the crystal is readily shown and the geometric relationship between them indicated by the theory simultaneously verified by the following procedure. The observer stands with his back to a window, the light from which falls on the specimen held in his hands with its polished face turned towards him. The orientation of the specimen required to display one of the coloured reflections is first ascertained by trial. It is also easy to locate the orientation in which the specimen exhibits the specular reflection from the platy inclusions lying with their faces parallel to

the 010 planes in the crystal. The observer then ascertains by trial, the position of an axis such that if the specimen is rotated about it, first the coloured reflection and then the specular reflection successively come into view. A further rotation about the same axis and in the same direction brings the second reflection into view. If the twinning is of the albite type, the axis of the rotation needed is found to be parallel to the composition planes of the twinning: it does not coincide with either the a or the c-axis of the crystal, but makes an angle of about 70° with the former. If the twinning is of the pericline type, the correct procedure for viewing the two reflections successively is to rotate the crystal about its b-axis through 180°. But since the b-axis is not far removed from being normal to the 010 planes of the crystal, a rotation in the same direction and about the same axis as in the case of the albite twinning is found to be effective.

Except in respect of colour which is identical for both reflections, the features they display are found to be different and indeed often strikingly different. That this is due to their location in different parts of the crystal is demonstrated by the exact correspondence between the disposition of the twinning layers at the surface through which the observations are made and the features exhibited by the iridescence. Actually, it is found that the angle at which the composition planes of twinning meet the polished face has a great influence. If they meet steeply, as in the case of pericline twinning, the iridescence exhibits alternate dark and bright bands traversing the surface, these interchanging places in the two sets of reflections. the other hand, if the composition planes meet the external surface obliquely as in the case of the albite twinning, much depends on the disposition of the reflecting lamellæ in the alternate layers of the twinned crystal. If the reflecting lamellæ of one set are nearly parallel to the external face, those of the other set necessarily meet it obliquely; the reflection by the first set then appears almost uniform over the surface, while the reflection by the other set shows a banded structure. The degree of general transparency of the crystal plays a role of importance in deciding what is actually seen in any particular case; the greater the transparency, the more effective become the deeper layers in the crystal and the greater their contribution to the total observed luminosity. So much depends on the characters of the individual specimens that these details are best dealt with in describing their behaviour. This will be done later in the paper, and the particular cases illustrated in Plates I to V accompanying it will be fully commented upon.

The refractions at the external surface of a specimen occurring both at entry and at emergence of the light materially influence the settings of it required for observing the internal reflections. Except, however, when the

specimen has been inappropriately cut, the deviations in the course of the light rays produced by these refractions would not prevent the effects described above coming under observation. By immersing the specimen in a glass cell filled with a liquid having the same refractive index as labradorite, they may be completely eliminated. It is then possible to make a quantitative test of the geometrical relationship between the two sets of internal reflections indicated by the theory. Studies made in this way show that the settings at which the maximum intensity of iridescence is observed make an angle of 14° on one side or the other with that required for a specular reflection from the 010 planes of the crystal.

7. PHENOMENA OBSERVED WITH POLARISED LIGHT

Some rather spectacular effects are observed when labradorite is illuminated by a beam of light which has passed through a polaroid sheet and the characteristic iridescence is viewed through a second polaroid sheet; the setting of the labradorite in its own plane, the angle of incidence of the light on it, and the setting of the two polaroids with respect to each other may be varied in the experiment, with interesting consequences. Apart from their experimental interest, the results observed with polarised light have a direct bearing on the problem of the structure of labradorite and the origin of its iridescence which forms the subject of this paper. Indeed, they afford further confirmation of the correctness of the theoretical approach made in the paper, as we shall presently proceed to show.

The reflection of electromagnetic waves which occurs at the boundary between two media is a consequence of the differences in their optical polarisabilities. When both media are crystalline and anisotropic, the intensity of such reflection would, in general, depend not only on the angle of incidence and state of polarisation of the incident light, but also on how the optic axes of the two crystals are orientated with respect to the direction of the electric vector in the incident waves. The state of polarisation of the reflected light would also depend on the same circumstances. As a consequence, the observable characters of the reflected light would, in general, exhibit a complex behaviour such as is not met with in the familiar case of optically isotropic media. In the present problem, however, we are concerned with the reflection of light at the boundary between two feldspars whose structures are similarly orientated in space, and this results in a great simplification, as we shall presently notice.

The second and third columns of Table I show the maximum and minimum refractive indices of labradorite and orthoclase respectively, while the fourth column shows the differences between them. It will be seen that the

TABLE I

| | | Ng | Np | Ng- N p | Ng' | Np' | Ng'-Np' |
|-------------|-----|--------|----------|----------------|-------|---------|---------|
| Labradorite | •• | 1.5678 | 1 · 5598 | 0.008 | 1.565 | 1 - 559 | 0.006 |
| Orthoclase | • • | 1.5260 | 1.5180 | 0.008 | 1.524 | 1.518 | 0.006 |
| Differences | •• | 0.0418 | 0.0418 | 0 | 0.041 | 0.041 | 0 |

birefringence is of the same order of magnitude in the two feldspars and is quite small, while on the other hand, the differences in their refractive indices are several times larger. The fifth and sixth columns show the two refractive indices Ng' and Np' in each case for flakes lying on 010, while the seventh column gives the differences between them. It will be noticed that the lower index Np' differs little from the minimum index Np in either case, and that the differences between Ng' and Np' are identical, in other words that the birefringence in flakes parallel to 010 is very similar in the two feldspars. Taking these facts into consideration, it is evident that the optical anisotropies would have a negligible effect on either the intensity or the state of polarisation of the light reflected at the boundaries of separation, which, as we have seen, make an angle of only 14° with the 010 planes in labradorite. In other words, we may, for the present purpose, treat the feldspars as optically isotropic media having refractive indices appropriate to the circumstances of each case. The following inferences are thus justified.

- (a) The intensity of the internal reflections in labradorite would be sensibly independent of the setting of the crystal, if the angle of incidence remains unaltered.
- (b) The internally reflected light would exhibit a polarisation of the normal type; this would be sensibly perfect for the Brewsterian angle of incidence on the reflecting layers, namely 44°, in other words, when the light is deviated through 88° by such reflection.

The two results stated above and especially the complete polarisation of the reflected light in a direction transverse to the incident beam indicated by the theory furnish us with an experimentum crucis for a test of its truth. It is satisfactory to note that the studies made by us fully confirm these results. To exhibit the polarisation effect, it is necessary to immerse the specimen in a square cell filled with a liquid having the same refractive index as labradorite, and to place it so that the light entering the cell through one face and reflected within the specimen emerges through the perpendicular face. It is also necessary to arrange that the polarisation due to the internal reflection is

not modified by the birefringence of the labradorite. This may be secured by a suitable orientation of the specimen within the cell. For the same reason, it is preferable to employ a specimen that does not exhibit twinning, at least in the immediate vicinity of the area under observation. It is found that in these circumstances, the reflected light is completely quenched when observed through a polaroid set with its vibration direction in the plane of incidence.

The phenomena exhibited by labradorite when illuminated by polarised light and observed through a polariser owe their interest to the fact that they render the presence of the twinning layers adjacent to the surface as well as their inclination to the latter directly accessible to observation. The phenomena are best described when dealing with the behaviour of our individual specimens.

8. SPECTRAL CHARACTERS OF THE IRIDESCENCE

When we compare the iridescence of labradorite with the colours of thin plates, we notice certain obvious similarities as well as some striking differences. A regular colour sequence is exhibited by some specimens of labradorite, recalling those shown by soap films of varying thickness or by Newton's rings. Boggild (*loc. cit.*) describes some fine examples. The colours also change with the obliquity of observation in much the same way as the colours of thin films. The spectral nature of the colours is, however, wholly different in the two cases. We do not observe with labradorite the dark and bright bands alternating in the spectrum characteristic of the interferences of thin plates; what is actually the observed is a continuous variation of intensity in the spectrum. Particularly significant is Boggild's remark that the colours of the first order which are a prominent feature in Newton's rings are wholly absent in the case of labradorite. These begin with a blue which is far richer than the faint blue of the first order, and are followed by other rich colours and not by the white observed in the Newtonian sequence.

The facts stated find a natural explanation in the ideas developed in the present paper. As we have seen, the optical heterogeneity of labradorite owes its origin to the orthoclase or potash feldspar present segregating itself from the albite-anorthite. The first stage of such segregation would evidently be the formation of very small crystallites distributed throughout the volume of the crystal. Such segregation would evidently give rise to a Tyndall effect in which the shorter wave-lengths would predominate, and hence having the rich blue colour characteristic of the scattering of light by small particles. Since the particles occupy fixed positions, the diffused radiations would have specifiable phase-relations and would be capable of mutual interference,

The observed effect in any direction would depend on the disposition of the particles in space. In view of the similarities in chemical composition and crystal structure between orthoclase and albite-anorthite, it is scarcely to be supposed that such disposition would be wholly random in character. If, as may reasonably be assumed, the diffracting particles locate themselves more or less accurately in sets of planes having a specific orientation within the crystal, the optical effect resulting from the passage of a beam of light would be a diffusion, the intensity of which would be greatest around a particular direction; it may be described as a diffuse reflection by the set of planes referred to. At a later stage in the segregation, the particles would be crystallites no longer small in comparison with the wave-length of light. Assuming, for the reasons discussed earlier in the paper, that they are lamellæ having a specific orientation, the colour of the diffracted light would be determined by their thickness, while its angular extension would depend on their other dimensions. This second stage could scarcely be reached unless the dimensions of the crystallites are much in excess of those required to give interference colours of the first order in the Newtonian scale.

The foregoing remarks may be summed up by the statement that the colours of labradorite are neither a simple Tyndall diffusion nor a simple case of thin-film colour; they are a case sui generis in which the optical principles underlying both of these phenomena play a part. We may conclude this section by a brief description of some effects which we have observed and which are readily understood in the light of this statement. As both Rayleigh (loc. cit.) and Boggild (loc. cit.) have mentioned, the coloured reflections in labradorite are not sharply defined but appear spread out over a range of angles. We have noticed in our studies that the angular range of this spread is about twice as great for a blue iridescence—despite the smaller wave-lengths—as for a red or an orange iridescence with its greater wavelengths. We have noticed also that when a specimen exhibiting varied colours over its area is illuminated and is slowly turned away from the setting at which the reflection is brightest, the areas of blue iridescence continue to be visible long after the other areas are dark. Further, we have noticed in the same circumstances that the areas exhibiting these other colours show a distinct change of tint towards the blue before they are finally extinguished.

9. Some Typical Examples

In the preceding sections of the paper, references were made in general terms to the results of our observations with the specimens of labradorite at our disposal. By way of supplementing these references and to give them substance, we shall proceed to describe in some detail, the individual behaviour of a few of them.

Specimen No. 1 is our largest and finest piece. It is a slab some 24 centimeters long, 12 centimetres broad and of thickness about a centimetre at one end, thinning off to five millimeters at the other. It was obtained through the East Coast Mineral Corporation of New York, and the locality of its origin is shown on its label as the Grenfell quarry, Nepoktulegatsuk, Tabors Island, near Nain, Labrador. Though apparently dark in colour, the material is fairly transparent and exhibits a brilliant blue iridescence on both faces. One of the faces had been polished, evidently to exhibit the effect to the greatest advantage, but the other, though quite rough, showed the same colour with scarcely less brilliance. The material on careful scrutiny shows a wavy discontinuity running through it. The central part of the slab displays a bright blue colour, while the areas to the left and the right exhibit a darker and deeper blue. Except in this respect, the iridescence is seen to possess a most remarkable uniformity over the entire area of the specimen.

Fig. 1 in Plate I is a photograph of the specimen when displaying its characteristic iridescence. The illumination of the slab was a few degrees away from the normal to avoid reflection from the external surface. Fig. 2 is another photograph taken with the specimen turned to such a position that the aventurism due to inclusions lying in the 010 planes is prominently displayed. This effect varies enormously in its brightness over the area of the specimen, and is in fact almost entirely concentrated in and around the areas in which there are visible defects or breaks in the material. Fig. 3 is a photograph exhibiting the second internal reflection observed in the specimen when it is held with the light falling on it rather obliquely, in a direction running from left to right in the picture and viewed in nearly the same direction. reflection, unlike that reproduced in Fig. 1, shows a great many bands running parallel to each other across the specimen. Holding the specimen in his hand and rotating it about an axis parallel to the general direction of these bands, the observer can view successively the banded reflection, the aventurine reflection, and the uniform reflection, reproduced as Figs. 3, 2 and 1 respectively in Plate I.

Examining the edge of the slab where the 001 cleavage could be seen, it became evident that the slab was traversed in a direction parallel to the 010 planes of the crystal by a whole series of twinning bands of the albite type. It was verified that at every one of the twinning lamellæ, and exactly along the line of its meeting with the polished face of the specimen, one of the bands shown in Fig. 3, Plate I, was located. The colour of the bands was exactly the same as that of the iridescence exhibited in Fig. 1 of Plate I, namely bright blue in certain areas and a deeper blue in the other areas. The thinness of the twinning layers is evidently responsible for the fineness of the bands seen

in Fig. 3. The observer readily notices that the banded reflections are more steeply inclined to the polished face of the specimen than the twinning layers themselves, though they arise in the latter. Very faint dark lines can be seen in Fig. 1 along the intersections of the twinning bands with the polished surface of the specimen, showing thereby that in the first reflection, these regions remain dark.

Fig. 1 and Fig. 2 in Plate II are photographs of the same specimen illuminated almost normally by a beam of light which has passed through a polaroid, and viewed nearly normally by the internally reflected light through a second polaroid. Fig. 1 represents the case in which the two polaroids are crossed and Fig. 2, the case in which they are parallel. The bands seen traversing the face of the specimen in these photographs appear and disappear periodically four times in each revolution when the specimen is turned round in its own plane. It is clear that these effects arise from the birefringence of the crystal acting jointly with the birefringence of the twinning layers running through it and meeting the surface obliquely. We get bright lines in Fig. 1 and dark lines on Fig. 2 at the places where the twinning layers meet the surface, because at these places, the birefringence in the twinning layers affects the reflected light from the maximum depth in the illuminated specimen. It may be mentioned that somewhat analogous phenomena, though much less striking, may be seen when unpolarised light is incident on the slab rather obliquely and the reflected light is viewed through a polaroid. The internal reflection itself acts as a polariser in this case.

Figs. 3 and 4 in Plate II illustrate different kinds of inclusions observed in labradorite which are responsible for the aventurine reflections.

A few words must suffice for each of our other specimens illustrated in the Plates accompanying the paper.

Specimen 2 is a typical example of multiple twinning of the pericline type. It exhibits a greenish yellow iridescence. Here the twinning layers meet the polished face of the specimen almost normally. Figs. 1 and 3 in Plate V show its two internal reflections; it is possible to make out that the dark and bright bands have interchanged places in them. Fig. 2 is a photograph of the same specimen held in such a position that neither iridescence is visible.

Specimen 3. See Figs. 4, 5 and 6 in Plate III.—These illustrate respectively the first coloured reflection, the aventurine reflection and the second coloured reflection in the specimen. The iridescence is a golden yellow, changing to green and greenish blue at oblique incidences. The banding

seen very prominently in Fig. 4 and less clearly in Fig. 6 is due to the albite twinning layers meeting the surface obliquely.

Specimen 4. See Figs. 1, 2 and 3 in Plate III.—This specimen exhibits a blue iridescence except towards the left where there are patches of opacity interspersed with areas of iridescence in which varied colours appear. The horizontal bands in the upper part are due to the pericline twinning, while the inclined bands towards the right owe their origin to the albite type of twinning. Figs. 1 and 3 illustrate respectively the two coloured reflections, and are seen to be complementary, the bright areas in one being the dark areas in the other and vice versa. Fig. 2 represents the aventurine reflection by the specimen.

Specimen 5. See Figs. 6, 7 and 8 in Plate IV.—These represent respectively, the first coloured reflection, the aventurine reflection and the second coloured reflection. The banding in Figs. 6 and 8 is due to the albite twinning, and the positions of the dark and bright bands in them are seen to be complementary to each other. Fig. 5 in Plate IV shows the same specimen viewed under incident polarised light through a second polaroid in the crossed position. It should be mentioned that this specimen exhibits very varied colours over its area; bright blue in the regions both around the dark centre and also towards its margins, with yellow, orange and red bands succeeding each other in the intervening areas.

Specimen 6. See Figs. 5, 6 and 7 in Plate II.—This is a small but beautiful piece which exhibits a peacock-blue iridescence with orange-yellow spots prominently in one setting (Fig. 5). In the other setting (Fig. 7), the iridescence shows peacock-blue patches which are much less intense; these appear in areas which are either completely dark or else rather faint in Fig. 5. Very striking also are the bright lines seen in Fig. 7 in the positions where the dark lines appear in Fig. 5. These lines represent pericline twinning bands meeting the surface steeply. Elsewhere, the specimen shows albite twinning, with one set of the iridescent laminæ almost parallel to the surface and giving the strong reflection seen in Fig. 5, while the other set is more steeply inclined to the surface and gives the feeble effect appearing in Fig. 7.

Specimen 7. See Figs. 1, 2, 3 and 4 in Plate IV.—This is rather an extraordinary piece, showing several separate crystals in different settings firmly adhering to each other. The four photographs show its iridescence under different angles of illumination and observation. The central area shows the albite type of twinning, while two others show the pericline twinning, as is evident from the sharply defined dark and bright bands crossing the

iridescent areas. The interchange in the positions of these over the area of the crystal on the left is clearly visible on a comparison of Figs. 2 and 4.

In concluding this paper, we have to express our grateful thanks to those whose help has enabled us to bring this investigation to a successful conclusion. Several of our labradorite specimens were a very generous gift from Mr. B. W. Anderson of London. Specimens of other varieties of feldspars presented to us by Professor T. N. Muthuswami of Madras, Dr. C. Mahadevan of Waltair and Mr. L. S. Krishnamurthy of Hyderabad also proved very useful in our studies. The Director of the Geological Survey in Mysore, Dr. C. S. Pichamuthu materially assisted us in getting access to the mineralogical literature on feldspar. Mr. T. K. Srinivasan, Government of India Senior Scholar in Mineralogy at this Institute, has been very helpful to us in the study of this literature.

SUMMARY

General considerations indicate that the phenomenon of labradorescence owes its origin to the segregation of potash feldspar as crystallites or lamellæ of orthoclase which remains embedded in an albite-anorthite matrix and give rise to optical heterogeneity. The following are derived as theoretical consequences of such segregation and have been confirmed by observations made with a representative collection of specimens.

- (A) Labradorite which is multiply twinned according to either the albite or pericline law or both, exhibits its characteristic reflections in two different settings geometrically related to each other by the twinning law. The two reflections display identical colours, but have their origins in different parts of the crystal, and this makes itself evident as a visible banding in one or the other or both of the reflections.
- (B) The light reflected transversely to the incident beam is completely polarised.
- (C) The intensity of the reflected light is independent of the setting of the reflecting layers in their own plane.
- (D) The spectral character of the reflected light would approach that of a Tyndall diffusion when the crystallites are very small. Hence the colours of the first order in the Newtonian sequence would not be observed. The spectral character and the colour sequence at the later stages of segregation would also be different from those of thin films.
- (E) The angular spread of the reflected light is greatest for the blue iridescence and less for other colours such as yellow, orange and red.

The effects arising when labradorite is illuminated and observed by polarised light are also described and explained in the paper.

Numerous photographs reproduced in five plates illustrate the observed phenomena.

REFERENCES

| 1. | Alling | Jour. Geol., 1921, 29, 193. See also Winchell, Optical Mineralogy, 1932, III Edition, Part II, p. 319. |
|----|---------------------------------------|---|
| 2. | Boggild | Math. fys. Meddel., Copenhagen, 1924, 6, No. 3. |
| 3. | Chao, Smare and Taylor | Miner. Mag., 1939, 25, 338. |
| 4. | Chao and Taylor | Proc. Roy. Soc., A, 1940, 174, 57. |
| 5. | distance property specially specially | Ibid., 1940, 176, 76. |
| 6. | Hintze | Handbuch der Mineralogie (Leipzig), 1897, Zweiter Band, p. 1512. |
| 7. | Ito and Inuzuke | Z. Kristallogr., 1936, 95, 404. |
| 8. | Rayleigh | Proc. Roy. Soc., A, 1923, 103, 34. |
| 9. | Taylor | Ibid., 1934, 145, 80. |

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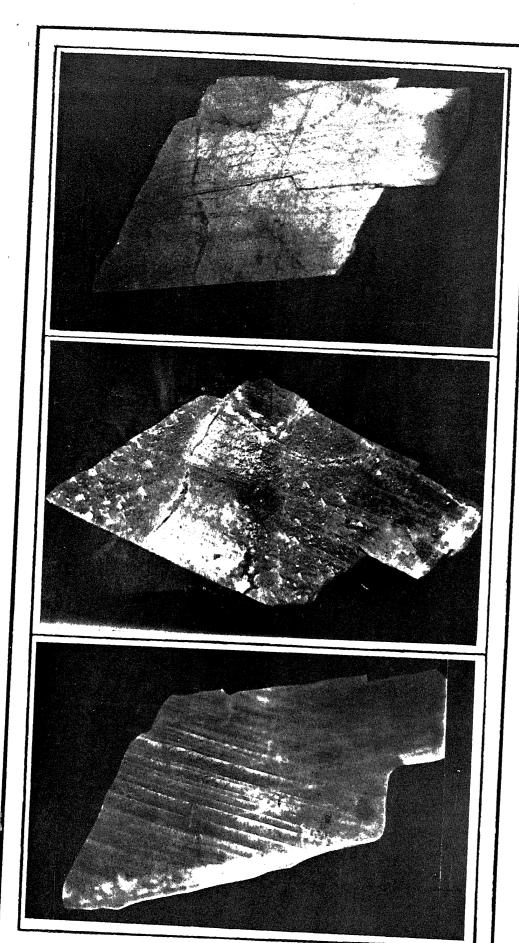


Fig. 1

F16. 2

Frg. 3

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A. Jayaraman

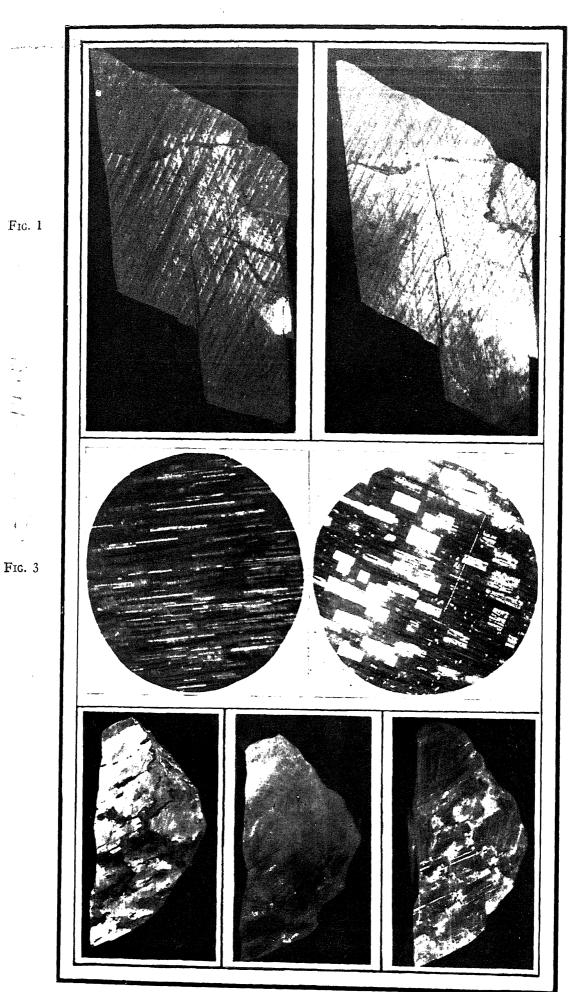


Fig 2

Fig. 4

Fig. 5

Fig. 6

Fig. 7

A. Jayaraman

Fig. 1

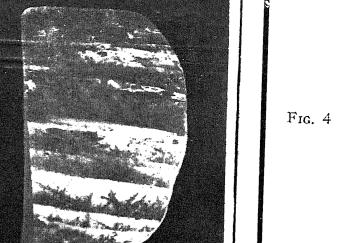
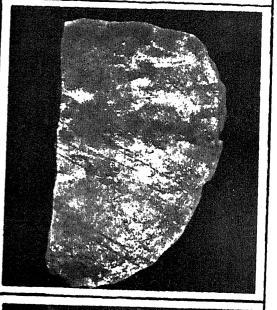
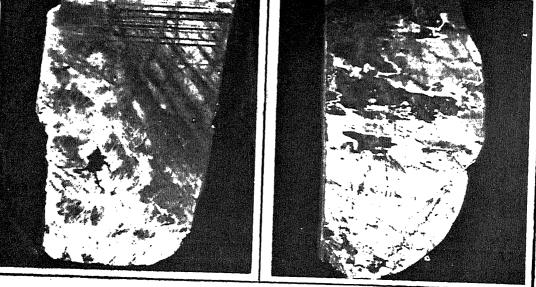


Fig. 2



F16. 3



Frg. 5

Frg. 6

F1G. 3

Fig. 1

Fig. 2

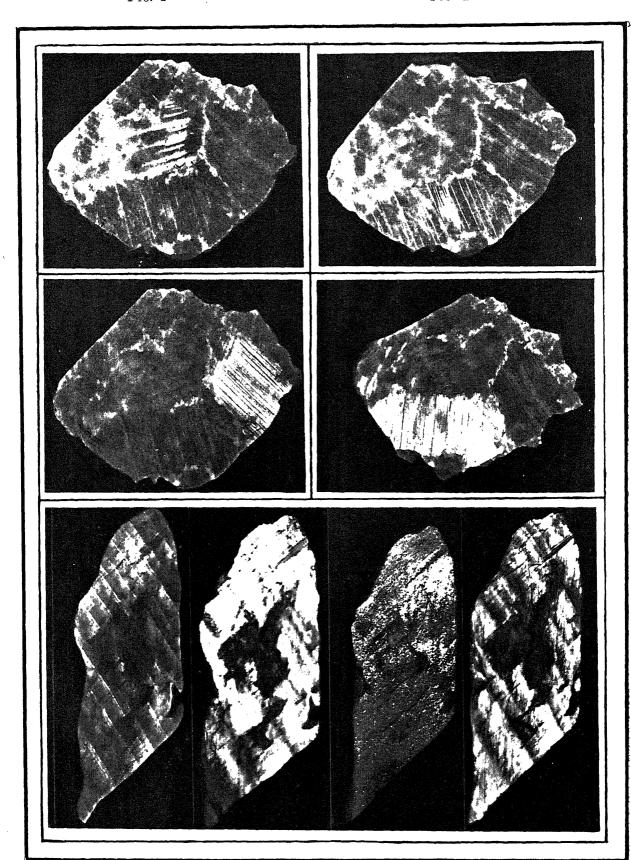


Fig. 5

Fig. 6

Fig. 7

Fig. 8

Fig. 4

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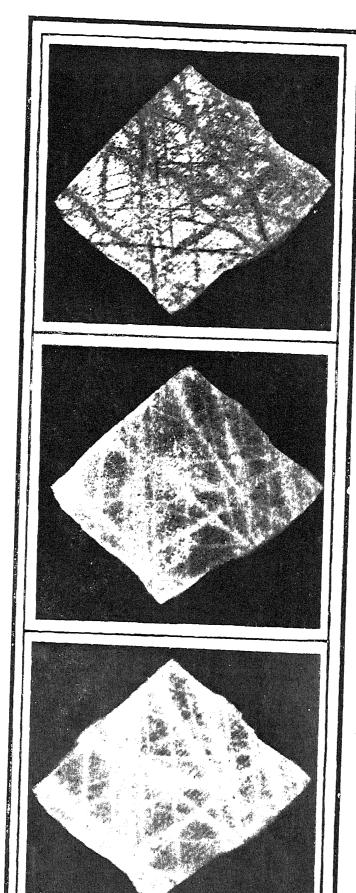


Fig. 1

Fig. 2

Pig. 3