

The diffusion haloes of the iridescent feldspars

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1. Introduction

By reason of the brilliance and variety of the optical effects displayed by it and its availability in large sizes, labradorite is easily the most spectacular amongst the iridescent feldspars. The explanation of its remarkable behaviour had long remained rather mysterious and hence it was taken up for investigation at this Institute. In a paper published in these *Proceedings*,¹ the results of our studies made with numerous specimens of this material were described and the explanation of the optical behaviour of labradorite to which they led unambiguously was also set out. In a further paper published shortly afterwards,² the optical behaviour of the Ceylon moonstones was studied and discussed. These two cases, however, do not exhaust the list of feldspars which show interesting optical effects. Professor Ito of the Mineralogical Institute of Tokyo very kindly presented us with a collection of the small but beautifully crystallised moonstones from Korea. Through the kindness of Prof. T N Muthuswamy of Madras, we have also received a collection of quite large pieces of plagioclase feldspar from Gudur in South India exhibiting a blue or bluish-white iridescence. Some kind friends also enabled one of us to visit the pegmatite areas in the Coimbatore District in South India, and to make a collection of specimens of the orthoclase feldspar exhibiting a golden-yellow schiller found in that area. Other varieties of feldspar exhibiting "Labradorescence" are described in a memoir by Boggild.³ But these were not available to us.

A feature common to all the varieties of feldspar mentioned above and studied by us is that the light entering the material and returned from its interior is not a true optical reflection but is a *diffusion of light* spread over a range of angles. The study of the features of this diffusion is of great importance for obtaining a definite idea of the structure responsible for the iridescence of the feldspar. A convenient method of carrying out such studies was devised and described by one of us two years ago.⁴ The technique adopted is to illuminate a *small area* of the specimen by a narrow intense beam of light and to receive the radiation diffused from it on a white screen held not at too great a distance therefrom. The observations have to

be made in a darkened room, and the more intense the incident pencil of light is, the more striking are the observed effects. By altering the spot on the specimen which is illuminated, or by tilting the specimen with respect to the direction of the incident pencil of light, one can quickly observe the resulting changes in the pattern of the diffused light. The disturbing effects arising from refraction and scattering at the external surface of the specimen may be eliminated completely by immersing it in a glass cell containing a transparent liquid of suitable refractive index which is traversed by the incident beam. If, however, the external surface of the specimen has been suitably cut and polished, the disturbing effects are less troublesome and such immersion is not so essential except when the polished surface is curved, in which case the effects of refraction cannot otherwise be got rid of. In all cases, the diffusion pattern may be photographed by receiving it on a sheet of bromide paper suitably held.

2. The diffusion haloes of labradorite

The summary of our paper on labradorite referred to above is reproduced below *in extenso*, since the actual facts of observation as well as their theoretical explanation are clearly stated therein.

“General considerations indicate that the phenomenon of labradorescence owes its origin to the segregation of potash feldspar as crystallites or lamellae 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."

The terms "reflections" and "reflected" used in the foregoing summary of our earlier paper are rather misleading and should be replaced by "diffusion haloes" and "diffused" respectively. Indeed, the angular spread of the blue labradorescence exhibited by the finest specimens is so large that it would be erroneous to describe the phenomenon as a reflection.

Figures 1, 2 and 3 in plate I accompanying the present paper were recorded respectively with the specimen 3, and with two different areas of the specimen 5 referred to on pages 13 and 14 of our earlier paper. The photographs display in a striking manner the distinctive features of the optical behaviour of labradorite, and would have been even more striking had they been recorded and reproduced in full colour. It would then have been evident that the two haloes seen in each figure are of identical colour and that the aventurine reflection seen midway between them is white. In figures 4, 5 and 6 of plate III and figures 6, 7 and 8 of plate IV of our earlier paper, photographs of the same two specimens as viewed from different directions were reproduced. They show that both of these specimens exhibit a visibly banded labradorescence, arising from the fact that the polished face of the specimen was nearly parallel to the crystallographic B-planes and hence that the effects due to both sets of orthoclase lamellae present in the twinning layers of the plagioclase feldspar can be observed at the same setting. In the present method of study, the light diffused by these two sets of layers in different directions is recorded simultaneously, one on either side of the aventurine reflection; the latter is produced by the plates of haematite included in the feldspar and as these have their faces parallel to the B-planes, the reflection by them indicates the position of these planes. In the case of these specimens, it is noticed that when the illuminated area is caused to traverse the bands of labradorescence, the intensity of one of the diffusion haloes fluctuates relatively to that of the other. The character and intensity of the haloes and their relative positions as recorded in the figures of plate I of the present paper thus completely demonstrate the statement (A) quoted above from the earlier paper.

It will be seen further that the haloes appearing in plate I are of different sizes. Their colours as observed visually were also different. The haloes recorded in figure 1 had a golden yellow colour, in figure 2 a greenish-blue colour, while in figure 3 they were blue. This correlation of the spectral character of the labradorescence with the angular dimensions of the diffusion haloes is further confirmed by the series of five diffusion haloes photographed with other specimens and reproduced as figures 1 to 5 in plate II. Figure 1 which shows the largest halo was of a deep blue labradorescence, while the remaining haloes exhibited respectively a light blue, greenish-blue, yellow, and red colour. The progressive diminution in the angular spread of the diffusion haloes with the shift of the maximum intensity towards the red end of the spectrum (stated as

proposition E in the summary quoted above) is thus completely confirmed. We shall return presently to a consideration of its theoretical significance.

As is to be expected, the colour of the diffusion haloes changes with the angle of incidence of the light on the lamellae responsible for the labradorescence. Such incidence may be altered by a change in the setting of the specimen with reference to the incident light. The range of incidences attainable is however limited by reason of the refraction of light at entry into the specimen, by the inclination of the lamellae to the polished face, and also by reason of refraction of the diffused light at emergence. Thus, it is often the case that one of the haloes ceases to be observable at a particular setting of the crystal, and that a different setting is necessary to enable it to be seen. To avoid this difficulty, it is helpful to immerse the specimen in a cell containing a suitable liquid. The range of incidences attainable is thereby greatly increased and the change in colour of the halo with the angle of incidence of the light is more readily observable. Indeed, with this arrangement, the angle of incidence of the light on the orthoclase crystallites present in the alternate layers of the albite twinning may be so different that the two haloes appear with altogether different colours.

The other results (B) and (C) stated in our earlier paper, namely, the polarisation of the labradorescence and the independence of its intensity on the azimuth of incidence can likewise be demonstrated objectively by the present method of study. The polarisation of the diffused light is only complete in a direction transverse to the incident beam and to enable it to be demonstrated, it is necessary to immerse the specimen in a suitable liquid. The specimen should also be orientated within the cell in such manner that the depolarising effect of its birefringence is a minimum. A polaroid is then interposed between the feldspar and the receiving screen and rotated. The intensity of the diffusion halo undergoes large variations; the extinction observed is fairly complete for a deviation of 90° from the direction of the incident light.

The study of the diffusion haloes of labradorite thus completely confirms the findings reported in our earlier paper, and there is no need to traverse here the question of the origin of labradorescence already fully dealt with by us. It is sufficient to quote the remark made earlier that the colours of labradorite are neither a simple Tyndall diffusion nor a simple case of thin-film colours, but a special phenomenon in the elucidation of which the principles underlying both play a part. When the crystallites of orthoclase which segregate themselves from the albite-anorthite matrix are of very small size, they give rise to a Tyndall effect with the shorter wavelengths predominating and hence exhibiting a rich blue colour. On the other hand, if they are not small in comparison with the wavelength of light and are assumed on the basis of the observed facts to be lamellae having a specific orientation in the crystal, the colour of the diffracted light would be determined by their thickness while its angular extension would depend on their other dimensions. That the colour of labradorescence shows a definite correlation with the size and shape of the diffusion haloes strongly

supports the proposed interpretation. On the basis of such an explanation and the observed shape, colour and angular extension of the diffusion haloes, it is possible to make a rough estimate of the linear dimensions of the orthoclase lamellae. The figures are given below in table 1.

Table 1

| Colour of labradorescence | Thickness of the crystallites | Lateral dimension | Greatest dimension |
|---------------------------|-------------------------------|-------------------|--------------------|
| Deep blue | 0.1 μ | 1 μ | 2 μ |
| Green | 0.2 μ | 2 μ | 3 μ |
| Orange-yellow | 0.4 μ | 4 μ | 5 μ |
| Red | 0.5 μ | 6 μ | 9 μ |

The difference between the refractive indices of the orthoclase lamellae and the surrounding albite-anorthite matrix is about 0.04. On multiplying the dimensions of the crystallites by this quantity, the retardation produced in a light wave travelling in a direction normal to the lamellae is seen to be only a small fraction of the wavelength of light, and hence one could scarcely hope to be able to distinguish the individual lamellae under the microscope. The confusion due to the presence of great numbers of them and the presence of the visible inclusions which give rise to the aventurine reflection have also to be taken into consideration. They would diminish the prospect of success in observing the individual crystallites of orthoclase even with the aid of modern phase-contrast methods. On the other hand, if the crystallites are looked at edge-wise, the possibility of discovering their presence under the microscope would be greater.

3. Plagioclase feldspar from Gudur (South India)

This material of which a large collection has been made for the Museum of this Institute is an opaque white feldspar. Many of the specimens however show a distinct iridescence, varying in colour from a delicate blue to a brilliant white. Only occasionally is a hint of any other colour noticeable. As cleavage faces are frequently seen, there is no difficulty in verifying that the iridescence is noticed on the exposures of the B-face of the crystal. From the character of the iridescence, it is quite clear that the material is a plagioclase feldspar. The iridescence is seen in two different settings of the crystal with reference to the incident beam and the direction of observation; the areas which appear bright in one setting are dark in the other and *vice-versa*. The angle of rotation of the specimen needed is of the same order of magnitude as in the case of labradorite. But in other respects this feldspar behaves differently from labradorite. The colours are much less saturated

and much less varied in hue. Further, whereas in the case of labradorite the change from one setting to the other is brought about by a rotation of the specimen about a line in the B-face making an angle of 70° with the a -axis, in the present case the rotation required is about the a -axis itself. There is also visible evidence of twinning, sometimes of the albite type, sometimes of the pericline type and sometimes of both. It seems probable from what has been stated that the chemical composition of the feldspar is principally albite and that the iridescence arises from the presence of internally segregated lamellae of orthoclase. These are inclined to the B-face as in the case of labradorite but in a different azimuth altogether.

The diffusion haloes of this variety of feldspar are readily observed. They appear in pairs, one on either side of the ordinary optical reflection from the B-face. The relative intensity of the two haloes is very variable and depends upon the position of the illuminated spot on the crystal face. The angular dimensions of the haloes are rather small and the indications are that they vary with the colour, being larger for the blue and less for the bluish-white haloes. We have not however made any quantitative measurements under appropriate conditions. It has been noticed that, as in the case of labradorite, the diffusion halo is strongly polarised when it is observed in a direction perpendicular to the incident beam.

4. The diffusion halo of the Ceylon moonstones

The optical phenomenon familiarly known as schiller is exhibited by certain varieties of orthoclase feldspar, amongst which the best known are the moonstones from Ambalangoda in Ceylon of which there is a large collection in the Museum of this Institute. The optical characters of the schiller vary enormously with the individual specimens in spite of the fact that all of them come from the same area. At one extreme, we have the moonstones exhibiting a schiller of a beautiful blue colour; at the other extreme, we have those which give a schiller of great brilliance and of a perfectly white colour. In the paper which appeared earlier in these *Proceedings* and has already been referred to, the explanation of these effects was described and discussed in great detail. The conclusion reached was that the schiller is essentially a diffusion of light within the crystal and that such diffusion is a consequence of local fluctuations in the composition of the crystal. In other words, the relative proportion of the soda and potash feldspars in their composition (the latter being the principal constituent) varies from point to point. The same situation is described by the statement that the soda feldspar tends to segregate itself from the potash and form tiny crystallites which orientate themselves regularly in the lattice; by reason of the difference in refractive indices of albite and orthoclase, the crystal becomes optically heterogeneous and a diffusion of the light results. The lattice spacings of albite and orthoclase differ substantially along the a -axis and much less so along the other two axes. In

consequence, the albite crystallites may be expected to spread out principally along the *b* and *c* axes in preference to the *a*-axis. The diffusion of light would therefore tend to be non-uniform with respect to direction.

Thus, there is a parallelism between the optical effects exhibited by labradorite and by the Ceylon moonstones. But there are also notable differences which are readily intelligible in the light of the different circumstances in the two cases. The Ceylon moonstones, unlike labradorite, exhibit only a single diffusion halo, the same being elliptic in shape. The angular dimensions of the halo vary enormously with the colour of the schiller. It is very large for the moonstones exhibiting a blue schiller, smaller and brighter for those exhibiting a bluish-white schiller, and smallest and brightest with those exhibiting a pure white schiller. Photographs of the haloes in these three cases have already been reproduced in the article in *Current Science* cited as reference 4. From the angular dimensions of the halo in each case, the dimensions of the crystallites responsible for them can be roughly evaluated. The figures which are the averages derived from a large number of specimens are given below in table 2.

Table 2

| Colour of schiller | Major axis | Minor axis |
|--------------------|------------|------------|
| Blue | 1.6 μ | 0.8 μ |
| Bluish white | 3.0 μ | 1.6 μ |
| White | 6.0 μ | 5.0 μ |

It may be remarked that the measurements were made with moonstones immersed in a cell of liquid and with a pencil of light traversing them in each case along the schiller axis, that is to say, along the direction in which the schiller is most prominent. Actually, the diffusion haloes may also be observed when the light traverses the moonstone in other directions. The colour and intensity of the schiller and other attendant effects alter with the angle of incidence. The study of these phenomena opens up an extensive field for research, but we do not propose to enter into the same here.

The dimensions shown in the second and third columns of table 2 are transverse to each other and to the direction of the schiller axis. The question whether crystallites of the dimensions shown could be observed directly under the microscope is one of considerable interest. A factor that needs to be considered in this connection is the difference in the refractive indices of the crystallites and of the surrounding medium. This difference for the three principal directions is respectively 0.015, 0.010, 0.010. Multiplying these by the linear dimensions shown in table 2, it is seen that the retardation in phase produced by a single crystallite with reference to the surrounding medium would lie between 0.01 μ and 0.02 μ for a blue schiller and between 0.07 μ and 0.1 μ for a white schiller. These optical

retardations are much smaller than a wavelength of light and when it is recalled that the effects of a great number of crystallites would overlap, it will be evident that there is scarcely any possibility of the individual elements responsible for the diffusion of light being distinguishable under the microscope. We are thus justified in stating that the schiller of moonstones has its origin in a heterogeneity incapable of being resolved by ordinary microscopic examination. The view put forward by some mineralogists, notably E J Spencer,⁵ that the schiller of moonstones is due to a microperthetic or lamellar structure is thus clearly untenable. The optical irregularities reported by the same author as observable under special conditions of illumination are of much larger dimensions than those noted in table 2. They obviously cannot be responsible for the observed schiller.

5. The diffusion haloes of the Korean moonstones

The chemical composition of a typical moonstone from Korea has been reported by Professor Ito.⁶ It is given as 7.32% of K_2O and 6.88% of Na_2O , while a typical Ceylon moonstone is reported to contain 11% K_2O and 3.8% Na_2O . It is not surprising in these circumstances that some differences are observed between the optical behaviour of the Ceylon and the Korean moonstones. We find the latter to fall into two categories, namely, those exhibiting a bright blue schiller and the others a bluish-white schiller. The most remarkable difference between the Ceylon and the Korean moonstones is, however, that the latter are sometimes twin-crystals and sometimes quartets. Whereas an individual crystal exhibits only a single diffusion halo, a twin crystal shows two, while a quartet shows four haloes. The first case is illustrated in figure 1 of plate III and the second case in two different settings of the moonstone in figure 2 of plate III and figure 2 of plate IV respectively. The quartet of haloes exhibited by a moonstone containing four components is illustrated in figure 1 of plate IV.

6. The Coimbatore moonstones

The material with which some observations were made and are reported here was collected from the Kangayam area of the Coimbatore District in South India. From its observed crystallographic characters, it would appear that it is an orthoclase feldspar. Some specimens are translucent with a pale brownish tint. Other examples are however forthcoming which are much more transparent and exhibit a pale greenish hue. These specimens have a perfect C-cleavage, and plates of the material parallel to that cleavage can therefore be obtained suitable for optical investigation. A very interesting property of the greenish moonstones is they exhibit a schiller which in many cases has a golden yellow colour, while in

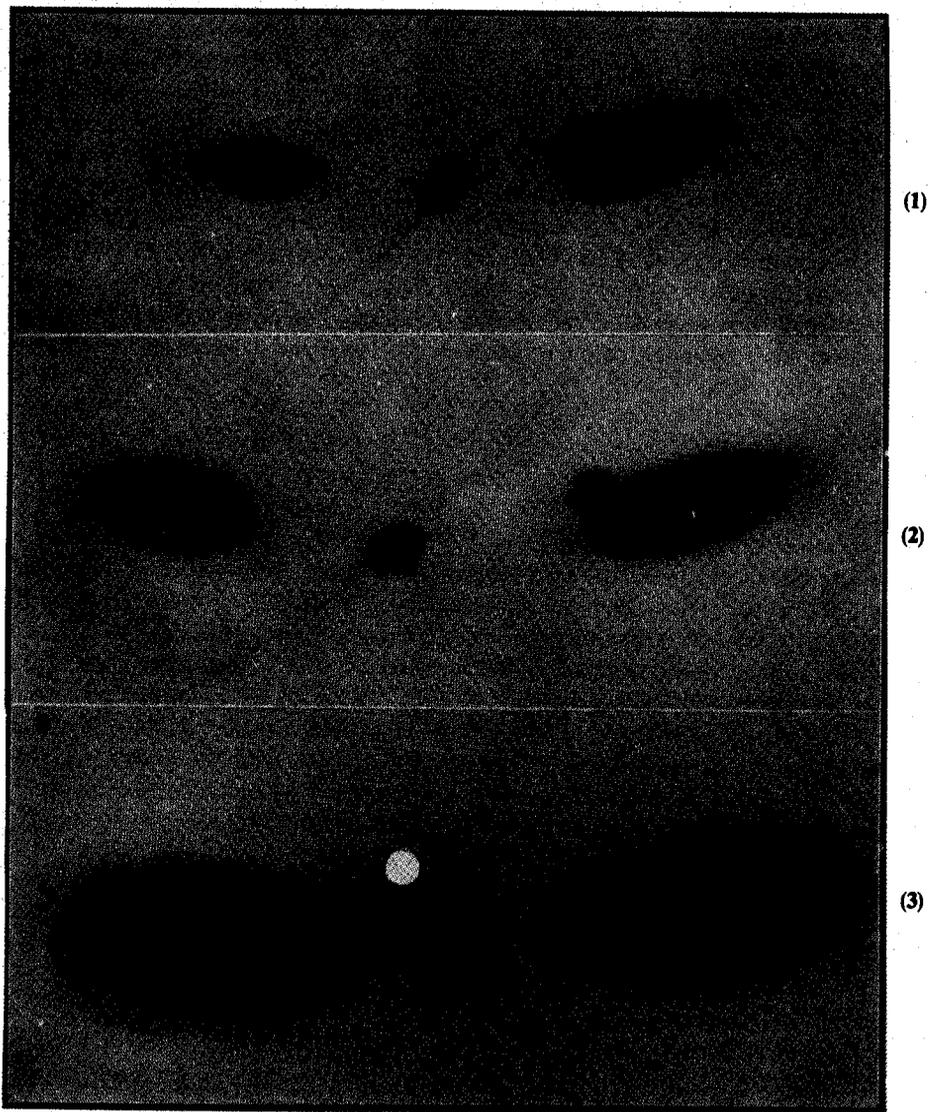
others it is yellowish-white. We have not as yet made any quantitative studies, but from general inspection, it would seem that the position of the schiller axis for this variety of moonstone is approximately the same as for the Ceylon moonstones. The character of the diffusion halo is however notably different. The halo in the present case is rather small in area and exhibits the same colour as the schiller itself. A special feature is the appearance of a bright cross whose arms pass through the diffusion halo and extend in directions perpendicular to each other. We have also noticed that when the moonstone is set so that the characteristic schiller appears in a direction transverse to the incident light, the diffusion halo is almost completely polarised. Another remarkable property of this moonstone is the polarised extinction of light which it exhibits in certain circumstances. This effect is of the same general nature as that described on pages 135 and 136 of our earlier paper on the Ceylon moonstones,² but is much more conspicuously observable.

Summary

Labradorite, the moonstones from Ceylon and Korea and other varieties of feldspar exhibiting iridescence have been investigated, and in all cases it has been found that such iridescence is the consequence of a diffusion of light within the material and not a true optical reflection. By a suitable technique, the diffusion haloes can be photographed and from their geometric positions, colour and angular dimensions, important conclusions can be drawn regarding the structures responsible for the iridescence. The plagioclase feldspars usually show two haloes geometrically related to each other according to the twinning law. The Ceylon and Korean moonstones differ in this respect; while the former invariably shows only one halo, the latter exhibits one, two or four according to the number of components present in the individual crystal.

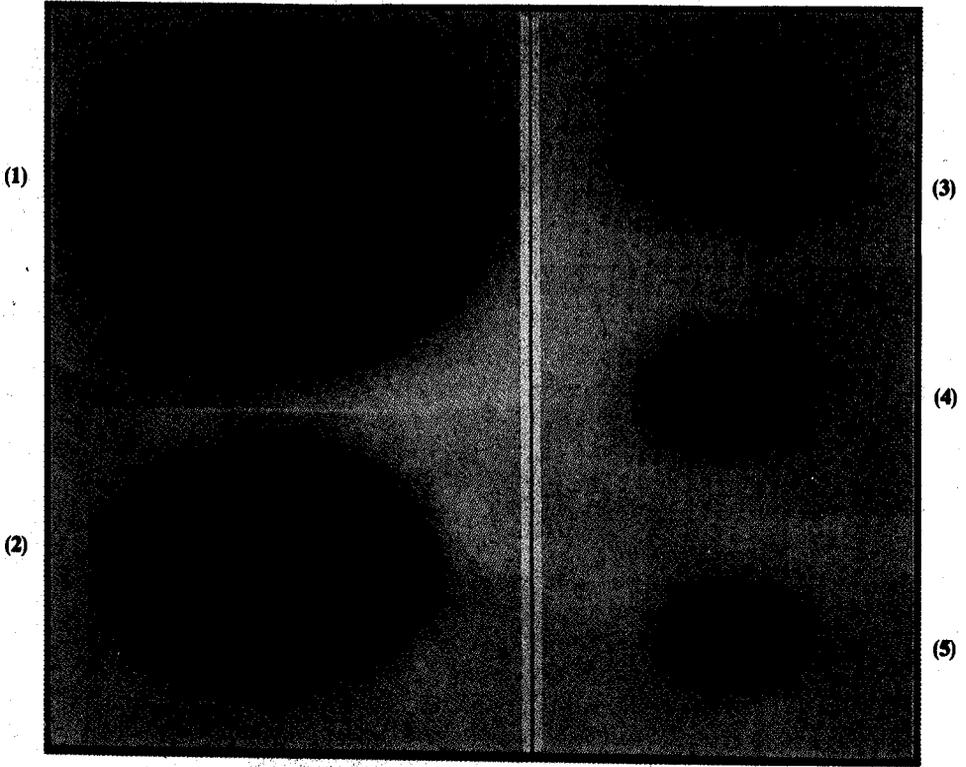
References

1. C V Raman and A Jayaraman, *Proc. Indian Acad. Sci.*, 1950, 32A, 1.
2. C V Raman, A Jayaraman and T K Srinivasan, *Ibid.*, 1950, 32A, 123-40.
3. Boggild, *Math. Fys. Meddl.*, Copenhagen, 1924, 6, No. 3.
4. C V Raman, *Curr. Sci.*, 1951, 20, 85-87.
5. E Spencer, *Miner. Mag.*, 1930, 22, 291.
6. Tei-Ichi Ito, *X-Ray Studies on Polymorphism*, Marusen Co., Tokyo, 1950.



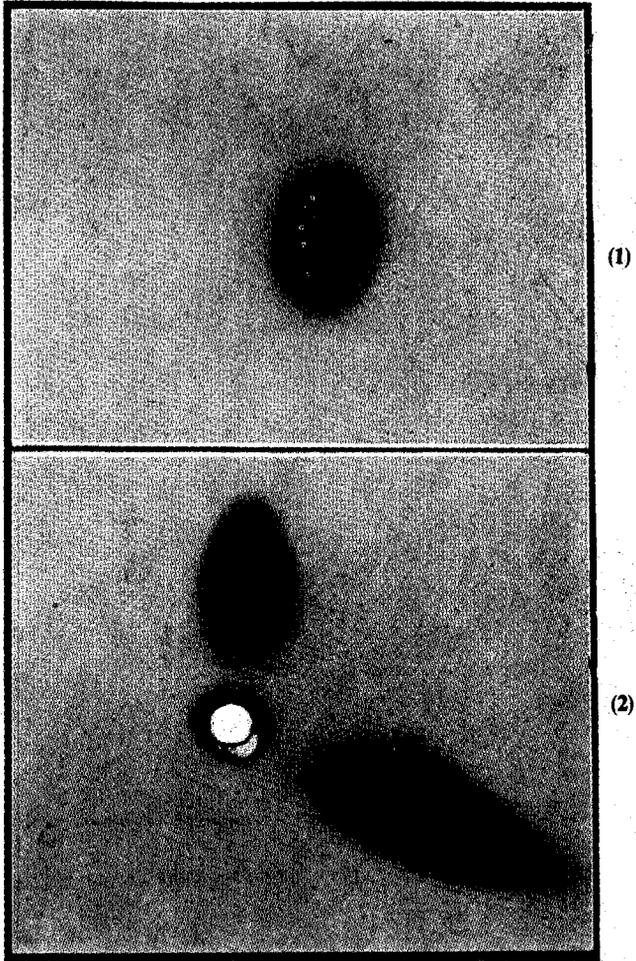
Figures 1-3. Diffusion haloes of labradorite.

Plate I.



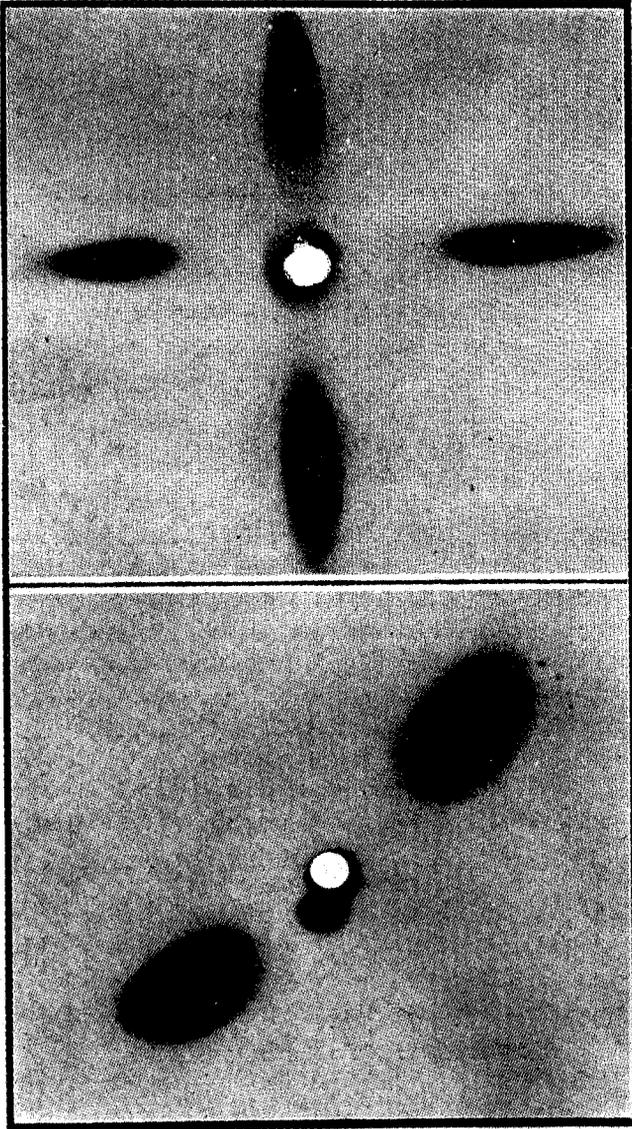
Figures 1-5. Diffusion haloes of labradorite.

Plate II



Figures 1 and 2. Diffusion haloes of the Korean moonstones.

Plate III



Figures 1 and 2. Diffusion haloes of the Korean moonstones.

Plate IV