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The structure and optical behaviour of the Ceylon moonstones

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1. Introduction

As is well known, the commonly occurring feldspars have a chemical composition which may be represented as a mixture of three components, namely the potash, soda and lime feldspars. The first two components resemble each other chemically but have different crystal forms, while the second and third components are isomorphous though their chemical formulae are dissimilar. The question arises whether the feldspars formed by such admixture are in all cases truly homogeneous crystals. Optical studies may well be expected to throw some light on this issue. In a recent paper in these *Proceedings*,[1] it has been shown that the iridescence exhibited by labradorite results from the segregation of the potash feldspar present in the mineral from the isomorphous mixture of the soda and lime feldspars which are its principal constituents. Tiny crystals of orthoclase are formed as the result of such segregation, but they remain dispersed in the albite-anorthite phase and diffuse the light traversing the crystal, causing it to display colours which vary with the dimensions of the crystallites and their setting with respect to the incident light-beam.

The present paper is concerned with another group of feldspars which give evidence of optical heterogeneity, namely the moonstones. The published analyses show that the potash and soda feldspars are their principal constituents, the former being usually the major and the latter the minor component; a small percentage of lime feldspar is also present. The moonstones enjoy a certain amount of favour as gems by reason of a characteristic optical effect which they display and which is generally known as "schiller". The investigation described in the paper was undertaken to ascertain the nature and origin of this effect. Though there is an extensive literature on moonstones to which both mineralogists and X-ray workers have contributed, the optical phenomena exhibited by them appear to have received comparatively little attention. The need for studying them is obvious. For, any explanation of the schiller effect which is not based on, or confirmed by, the results of such study must necessarily be regarded as dubious.

2. Materials and methods

Following the visit by one of us to Ceylon in September 1946, some forty specimens of moonstone in the rough were received as a gift from the owner of the formation near Ambalangoda in that island where they were formerly mined. A few cut specimens also accompanied them, of which one was a particularly fine gem exhibiting a blue schiller. The material thus made available to us has been utilized in the present investigation. To supplement the same, some thirty specimens of cut and polished moonstones were purchased from a dealer in Calcutta and added to our collection. It is understood that they were from the same original source at Ambalangoda. The uncut specimens have been useful, since the cleavages characteristic of feldspar which they occasionally exhibit, enable the crystallographic orientation of the specimens to be determined, a very necessary step in the investigations. On the other hand, the rough surfaces which the uncut specimens generally possess and the internal imperfections manifest in some of them are a serious inconvenience. The polished gem stones are much better in this respect, having evidently been prepared from selected material after removal of the defective portions. By immersing the specimens under study in a cell containing liquid benzene which has nearly the same refractive index as the mineral, the disturbing effects due to reflection, refraction and scattering of light at its external surfaces are minimized. The phenomena having their origin in the interior of the specimen and arising from the passage of light through it can then be conveniently studied. Suitable methods have also had to be improvised for holding the specimen inside the liquid and altering its setting with respect to the direction of the incidence of light as required, and also for determining the orientation of the crystal in such setting.

A comparison of the numerous specimens in our collection throws into relief the striking differences in their individual behaviour. A few pieces exhibit a deep blue but comparatively weak schiller. Others show a blue schiller of decidedly greater intensity. The rest show a schiller which in the different specimens passes by insensible gradations from a blue to bluish white and then to a perfect white; the intensity increases *pari passu* and in some cases is very great. It is remarkable and obviously significant that specimens from the same locality should show such large differences in their optical characters.

3. Nature of the schiller effect

It is a characteristic feature of the schiller of moonstones that the phenomenon is best seen at a particular setting of the crystal determined by the direction in which the light is incident on it and the direction in which it is observed. When either the setting of the crystal or the direction of incidence of the light is altered, the direction in which the schiller is most conspicuous also shifts. The idea generally current that the schiller owes its origin to a lamellar structure of the feldspar in which layers of orthoclase and albite alternate is based on the fact of observation just mentioned. For, such a structure would reflect light traversing the crystal in a direction varying with its angle of incidence on the planes of the lamellae. Indeed, measurements have been published[2] which claim to find the orientation of the postulated "schiller-planes" within the crystal in this way. It has even been claimed by one author[3] that examination of cleavage flakes of moonstone under low powers of the microscope when illuminated by light from a point source succeeds in revealing indications of a microperthitic or lamellar structure. Actually, however, the microscope does not reveal any noticeable structure in the typical and most interesting cases, namely the moonstones which exhibit a blue schiller. It should be remarked, further, that the coarse structure of which indications are observed under low powers of the microscope in certain other cases is *prima facie* incapable of giving rise to the optical phenomena under consideration. As already remarked, an explanation of the origin of the schiller to be acceptable should be based on the results of a detailed study of the phenomenon itself. It was with this idea that the present investigation was undertaken and it has revealed many new facts of interest. These will be set out in the proper places, but we shall proceed first to state those of which the significance in relation to the origin of the schiller is most obvious.

The effects observed with moonstones exhibiting a blue schiller are of particular interest, not only because they are the finest gems, but also by reason of the unambiguous manner in which a study of them reveals the real nature of the phenomena under consideration. As with all the moonstones studied, observations show that the schiller has a maximum intensity when viewed in a particular direction, which is related to the direction of incidence of the light in such manner that the two directions—at least roughly—make equal angles with and lie in the same plane as a particular direction within the crystal which we may designate as the schiller-axis. But the schiller can by no means be described as a reflection of light by a plane or planes within the crystal normal to the schiller-axis. Actually, it has the character of a diffusion of light spread out over a wide range of angles. This is directly evident when the light beam traversing the specimen is of sufficient intensity, as the crystal then becomes itself visible in various directions by reason of the light diffused in its interior. For instance, if the specimen is set so that the light is incident along the schiller-axis, the schiller is of maximum intensity in the direction backwards towards the source. But it has a notable intensity in various adjoining directions, and it can be seen also in the opposite or forward directions, though more faintly. Viewing the schiller in any particular direction and rotating the crystal about a chosen axis, it is found that the intensity falls off quite slowly on either side of the setting at which it is a maximum. The permissible rotation of the specimen before the schiller ceases to be visible naturally depends on the intensity of illumination and the circumstances of observation. But even with a weak illumination, the angular range is so large that the phenomenon can only be described as a diffusion of light within the crystal. The spectral character of the blue schiller exhibited by the finest moonstones is equally revealing. It is a much richer blue than the blue of the first order in the Newtonian sequence: in other words, it does not permit of being explained as an interference colour due to reflection of light by thin films. On the other hand, it matches more or less perfectly with the deep blue colour exhibited in the diffusion of light by particles which are small in size compared with the wavelength of light. As is well known, this blue is due to the predominance of intensity of the shorter wavelengths in inverse proportion to their fourth power. Spectroscopic study of the blue schiller of moonstones reveals the same predominance of the shorter wavelengths and thus compels us to assume a physical origin for it of the same general nature.

On a superficial examination, the bluish-white or white schiller exhibited by the majority of moonstones may be mistaken for a reflection of light within the crystal, the direction in which it is most conspicuous altering with the setting of the crystal in a manner suggesting such an explanation. A careful study, however, reveals various features which are inconsistent with such a description. What is actually observed is that the concentration of intensity around a particular direction which is observed even with the specimens exhibiting a blue schiller becomes more accentuated in these cases. This is accompanied by an increase in absolute intensity and a change in spectral character towards a pure white. Nevertheless, the angular spread of the illumination which simulates a reflection in this manner is readily observable and extends over several degrees of arc. Further, it is accompanied by a diffusion of light which covers a still greater range of angles and the spectral character of which approaches more nearly that of the blue schiller. Even the specimens which normally exhibit a perfect white schiller exhibit diffusion of a sky-blue colour when the light enters the crystal along the schiller-axis and the diffusion is viewed perpendicular to it, or *vice-versa*.

It is evident from the foregoing recital of facts that despite the apparent large differences in optical behaviour of the individual specimens, the phenomena they exhibit are essentially all of the same general nature. We may summarise the factual position by the following statements:

I. *The schiller of moonstones is a phenomenon arising from the diffusion of light within the crystal.*

II. *The diffusion exhibits optical characters analogous to those observed in the scattering of light by small particles.*

III. *The observed facts give no support to the hypothesis of a microperthitic or lamellar structure as the cause of the schiller.*

4. Origin of the diffusion

It is obvious from what has already been stated that the schiller of moonstones is the result of optical heterogeneity in the feldspar. The most natural assumption to make in the circumstances of the case is that the heterogeneity is associated with the known chemical composition of the mineral. Its two principal constituents, namely, the potash and soda feldspars, are similar in their chemical composition, and hence might reasonably be assumed to be capable of mixing with each other in all proportions and forming a homogeneous crystal. Against this, however, must be set the fact that the two components normally crystallise in different forms, potash feldspar or orthoclase as a monoclinic crystal, and soda feldspar or albite as a triclinic crystal. X-ray study shows that though there is a general resemblance between the crystal structures of orthoclase and albite, there are specific differences, the most significant, as indicated by table 1, being the differences in the lattice spacing along the *a*-axis and in the angle α between the *b*- and the *c*-axes. It has been suggested that the factor which determines whether a feldspar crystallises with monoclinic or triclinic symmetry is the size of the ions which enter into the alumino-silicate frame-work of its structure. Since, however, the sodium and potassium ions are both monovalent and since the former are smaller in size, it should clearly be possible for the former to replace the latter in the framework of the lattice. In other words, we have good reasons for believing that it should be possible for soda and potash feldspars to mix and form single crystals in the ordinary or the macroscopic sense of the term, even when the former component is present in substantial amounts, possibly even as much as one-half of the entire mineral in its molecular proportion. Two questions, however, arise in this connection. Firstly, in what manner are the sodium ions disposed within the lattice, viz., whether they randomly replace the potassium ions or whether they cluster together in groups, thereby causing the soda feldspar to segregate itself locally from the potash feldspar. Secondly, what changes, if any, are produced in the alumino-silicate framework by the replacement and in particular, whether there are any local alterations in crystal structure produced by the clustering together of the sodium ions. We have also to consider the question of the part played by the third component, namely the lime feldspar, in these dispositions.

The question raised in the foregoing paragraph have an obvious bearing on our present problem. Theory indicates that the distribution of the two component feldspars within the crystal would profoundly influence its optical behaviour. only in the particular case of a regular or orderly disposition of the sodium and potassium ions would the crystal be truly homogeneous in the

Table 1. Lattice constants

	a	b	c	α	β	γ
Orthoclase	8·45	12·90	7·15	90°	116° 3′	90°
Albite	8·14	12·86	7·17	94° 3′	116° 29′	88° 9

optical sense. Even a random disposition of the ions would give rise to an observable diffusion of light. Much more striking, however, would be the consequence of a disposition of the sodium ions in groups or clusters, in other words, of a segregation of the soda feldspar in the form of tiny crystallites dispersed through the rest of the material. The local fluctuations in chemical composition which such clustering would represent and the fluctuations of refractive index consequent thereon would give rise to a strong diffusion of light, its intensity increasing in proportion to the number of such clusters, multiplied by the square of the volume of each cluster, provided that these are of sufficiently small dimensions and that they can be assumed to be randomly distributed in the available space. Granting that the diffusion of light observed in the moonstones arises in this way, we may remark on the analogy between it and the opalescence exhibited by a mixture of two partially miscible liquids, e.g., carbon disulphide and methyl alcohol at temperatures a little higher than that at which the two components begin to separate from each other. The blue colour of the opalescence of such a liquid mixture at the higher temperatures matches perfectly with the blue schiller of the finest moonstones. As the critical solution temperature of the liquid mixture is approached, there is an enormous increase in the intensity of the opalescence and also a distinct change in its spectral character as the result of the formation of molecular clusters of rapidly increasing size, until finally a white cloud of visible droplets of liquid is released. There is thus an obvious parallelism between these phenomena and the progressive variations of the colour and intensity of the schiller noticed with different specimens in the group of moonstones.

The comparison made above between the schiller of moonstones and the opalescence of liquid mixtures should not, however, lead us to assume that the phenomena are completely similar to each other in their origin and character. So far from this being the case, many important differences between them are to be expected. It may be remarked, in the first place, that in our present problem, the local fluctuations in the composition occupy fixed positions in the lattice and there is also no reason to suppose that these positions would be distributed completely at random. In consequence, there would be specifiable phase relations between the radiations diffused from different volume elements, and this would necessarily influence the observed distribution of intensity in different directions. There is also no reason for assuming that the clusters of sodium ions in the moonstone would be of spherical shape or that they would be randomly orientated. The known character of the alumino-silicate framework of the feldspars and the differences between the crystal structures of orthoclase and albite would lead us to the opposite conclusion, namely, that the soda feldspar when it segregates and forms crystallites would tend to do so in a fashion related to the structure of the crystal. The shape, size and the orientation of the crystallites thus formed would evidently determine the intensity and spectral character of the radiation diffused by them in various directions. Hence, a notable dependence of the observed phenomena on the setting of the moonstone with respect to the direction in which the light is incident and also on the direction in which it is viewed may be expected. There is another and equally important group of differences between the present problem and that of the diffusion of light in liquid mixtures, namely, in the optical properties of the materials under consideration. The feldspars are birefringent crystals. Though their birefringence is small, we are concerned here with the *differences* in the optical polarisabilities of the medium arising from the local fluctuations in its composition, and these differences would be of the same order of magnitude as the birefringence of the feldspar. Hence, it cannot be supposed the effects under study would be of the same character for all directions of the electric vector in the light traversing the crystal. Special effects in respect of the intensity and state of polarisation of the diffused radiation are accordingly also to be expected. Altogether, it is obvious that the optical phenomena exhibited by moonstones would present a richer and more complex manifold of effects than the diffusion of light in liquid mixtures.

5. Relationship of the schiller to crystal structure

Reference has already been made to the "schiller-axis" in moonstones which has the property that if light traverses the crystal along that direction, the observed schiller has the maximum intensity in the backward direction, viz., towards the light source. The observations made with our specimens seem to show that the schiller axis is not noticeably different in moonstones exhibiting a blue, a bluish-white or white schiller; it is normal to the *b*-axis of the crystal and inclined to the *c*-axis at 81°. In other words, a plate cut perpendicular to the 010 cleavages and having its faces slightly inclined to the 100 planes of the crystal and making an angle of 9° with them would show the schiller most conspicuously when illuminated and viewed normally. The question arises why the diffusion of light in moonstones exhibits the maximum intensity in these circumstances. Closely related to this effect is another phenomenon observed by us which does not appear to have been previously noticed, namely, that the diffusion of light in moonstone is not symmetric about the schiller-axis but alters in intensity and character as the crystal is rotated about that axis, the directions of incidence and observation remaining the same. So striking is this non-axiality of the diffusion that its results are evident on a simple inspection of the schiller, when a cut stone is held in the hand and viewed by the light from a window, provided the shape of the stone is not such as to obscure or complicate the situation. Figures 1 and 2 in plate I illustrate the effect, the first being a moonstone with a blue schiller and the second one with a white schiller. Both the stones had been fashioned by the lapidary into a flattened spheroidal shape, the figure axis being practically coincident with the schiller-axis, so that when illuminated and viewed from the front, one would have expected the schiller to manifest itself symmetrically about that axis. Actually, however, we see an elliptical patch of light in both cases and when the crystal is rotated, the elliptic patch turns round with it. It was verified that the longer axis of the elliptic patch was in both cases parallel to the *b*-axis of the crystal; this, as already stated, is itself normal to the schiller-axis. The effects observed with the two specimens may be described thus: the schiller has a sensible intensity over a wider range of angles in the plane containing both the *b*-axis and the schiller-axis than in a plane containing the schiller-axis and perpendicular to the *b*-axis. The same effect shows itself in a peculiar way in figure 3 in plate I which represents a moonstone which had been shaped by the lapidary into an elongated ellipsoid. Its *b*-axis was inclined to the longer axis of the ellipsoid. As a consequence, the schiller appears bent into a curve having a point of inflection at the middle.

In all the three cases described above, it is the refraction at the curved surfaces fashioned by the lapidary that enables us to observe the diffusion of light in different directions simultaneously. The same effect may be studied in a more precise fashion by immersing the specimen in benzene liquid and by a rotation about a chosen axis finding how slowly (or how rapidly) the intensity of the diffused light falls off. In every case, it is noticed that the fall of intensity is very rapid if the crystal is rotated about its *b*-axis, but relatively much slower if it is rotated, for instance, about the *c*-axis of the crystal.

The explanation of the effects described above, as well as of those already set out in section 3, is evidently to be found in a consideration of the size, shape and disposition of the crystallites of soda feldspar resulting from its segregation, since they determine the distribution of intensity of the light diffused in various directions. We proceed to consider the nature of such segregation in its relation to the structure of the crystal as a whole. We shall take it for granted that when the soda feldspar segregates, it would tend to maintain the crystal symmetry as well as the orientation of the crystal axes exhibited by the parent mass and thereby to minimise the disturbance to the crystalline order produced by the segregation. Such a result could evidently be obtained in a number of ways. The simplest would be for the soda feldspar to assume monoclinic symmetry with its axes parallel to those of the parent mass. As can be seen from table 1, this would involve only relatively small changes of the angles between the crystal axes from those of a triclinic form. An alternative would be for the soda feldspar, while retaining a triclinic structure, to mimic monoclinic symmetry by internal twinning of the pericline type in which the direction of the *b*-axis remains unaltered. In this way, the mean directions of the *a*- and the *c*-axes for the twin would be nearly the same as for the axes of the entire crystal. It is evident, however, that the latter result would be more perfectly attained, if simultaneously there is a change in the axial angles so that they approximate more closely to the values required for monoclinic symmetry. Which particular procedure is favoured would evidently be determined by the circumstances and especially by the proportion of soda feldspar present. It is not unlikely also that the lime feldspar present may influence the choice between these different alternatives.

It is evident, however, that while there is thus a variety of possibilities for the structure of the segregated soda feldspar, a change in the lattice spacing *a* must necessarily occur in every case. This is a consequence of the smaller size of the sodium ion, and it is this difference which would determine the form which the crystallites would take. For, in the grouping of the cells containing the sodium ions which results from their coming together, an extension of the groups along the *a*-axis of the lattice would *not* be favoured. The formation of such a chain would disturb or disrupt all the chains of the alumino-silicate frame-work containing potassium ions along the *a*-axis with which it is linked. *Per contra*, a formation would be favoured in which the cells containing the sodium ions link themselves to others of the same kind along the *b*-axis or along the *c*-axis or more generally in the 100 planes of the crystal. The lattice spacing parallel to the *b*-axis differs from that parallel to the *c*-axis, being nearly double of it, and hence there is no reason to anticipate that a grouping of the cells containing the sodium ions along the *b*-axis and along the *c*-axis would be favoured to an equal extent. As the *c*-spacing is smaller, we may reasonably assume that the latter would be the more favoured arrangement. We are thus led to anticipate that the crystallites of the soda feldspar segregating from the potash feldspar would tend to extend themselves principally along the 100 planes of the crystal, and that in this plane an extension along the *c*-axis would be favoured more than one along the *b*-axis.

Optical theory indicates that as the extension of a diffracting particle or group of particles increases in a particular direction or directions, the intensity of the diffracted radiations would tend to concentrate along perpendicular directions. It follows, as a consequence of what has been stated above, that the moonstones would exhibit a schiller-axis perpendicular to the 100 planes of the crystal, in other words, normal to both the *b*- and *c*-axes of the crystal. Further, the diffusion of light would not exhibit a symmetry about the schiller-axis but would extend to larger angles in a plane containing the *b*-axis than in a plane perpendicular to it. It would probably not be a useful proceeding to discuss why the schiller-axis as actually observed is inclined at an angle of 81° to the *c*-axis of the crystal, instead of being perpendicular to it as deduced above. The considerations on which our argument is based are probably not deep enough to explain this small discrepancy between theory and observation. It is gratifying, however, to be able to record a direct observational proof of the explanation given above for the fact that the diffusion of light is not symmetric about the schiller-axis. Examination of several of our specimens discloses that the schiller exhibits distinct evidence of a fibrous structure running parallel to the *c*-axis of the crystal. A photograph showing this is reproduced in figure 4, plate I.

6. Intensity and polarisation of the schiller

Since the diffusion of light in its passage through the crystal is a consequence of the local variations in its refractive indices resulting from the changes of composition, its intensity would depend on the magnitude of these variations. The principal indices of albite are all greater than those of orthoclase, and the published data[4] show that the indices of moonstone are intermediate between them, and increase with the proportion of soda feldspar present. They are, however, in all cases, nearer the values for orthoclase than those for albite. Table 2 shows the compositions of orthoclase, of a typical moonstone from Ceylon, and of albite, and their respective refractive indices, as well as the differences between them. If we assume that the local variations in composition result in its approaching those of orthoclase and of albite respectively, the refractive indices would increase or decrease by the amounts shown in table as differences.

Table 2. Composition and refractive indices

Mineral	K_2O%	Na_2O%	CaO%	μ_g	μ_m	μ_p
Orthoclase	15·0	0·9	0·1	1·5245	1·5228	1·5192
			diff.	0·0037	0·0029	0·0024
Moonstone	11·0	3·8	0·2	1·5282	1·5257	1·5216
			diff.	0·0110	0·0071	0·0076
Albite	0·4	11·4	0·3	1·5392	1·5328	1·5292

The maximum index μ_g of orthoclase determines its optical polarisability in a direction parallel to the *b*-axis of the crystal, while the minimum index μ_p gives it for a direction nearly parallel to the *a*-axis, and the mean index μ_m corresponds to a direction in the 010 planes of the crystal nearly perpendicular to the *a*-axis, and therefore moderately inclined to the *c*-axis. In the case of moonstone, the foregoing statements would still be valid, but in the case of albite which is a triclinic crystal, they would require modification. It is evident, however, from the figures given in the table that the variations of the maximum refractive index μ_g of moonstone, in other words of the optical polarisability along the *b*-axis, resulting from the changes in composition, would be substantially larger than the changes in optical polarisability along the two other principal directions which, as stated above, are nearly parallel and perpendicular to the *a*-axis, respectively. The changes in optical polarisability in these two other directions resulting from variations of composition would, however, be of the same order of magnitude. The diffusion of light resulting from the variations in refractive index has an intensity proportional to the square of such variations. Theoretical considerations thus lead us to the very interesting result, that the schiller of moonstones should exhibit an easily observable dependence of intensity on the state of polarisation of the light incident on it, being greater when the electric vector is parallel to the *b*-axis of the crystal and less when it is perpendicular to that axis.

When the incident light is unpolarised, the same effects would manifest themselves in a different way, the state of polarisation of the diffused light being largely determined by the component parallel to the *b*-axis.

As is well known, the light diffused in an optically isotropic medium exhibiting local variations of refractive index and traversed by an unpolarised beam of light would be plane-polarised in a transverse direction. Since the schiller of moonstones is also a diffusion of light of the same general nature, we should expect it to manifest similar polarisation effects. In observing and interpreting them, it has to be borne in mind that the crystal is itself birefringent. Hence, the radiation diffused in its interior and traversing it before emergence would undergo changes in its state of polarisation which have to be taken into consideration. This is easier when the setting of the crystal with respect to the direction of incidence of the light and the plane in which the diffused radiations are observed are appropriately chosen. As has already been remarked, the state of polarisation of the light diffused within the crystal would be influenced by the direction of the electric vector with respect to the *b*-axis of the crystal. In other words, it would exhibit a dependence on the setting of the crystal very conspicuously. There is still another point which requires to be noted; not merely the principal refractive indices but also the directions of electric vibration which they refer to are different in orthoclase and in albite. It is therefore not to be expected that the diffused radiations would, in general, be perfectly polarised in a transverse direction as in the case of optically isotropic fluctuations of refractive index, even when the setting of the crystal has been appropriately chosen. Finally, we have to remember that the extension in space of the volume elements within the crystal diffusing the light is by no means always negligibly small. In the case of the moonstones exhibiting a white schiller with a large intensity, the diffusing elements are undoubtedly of appreciable size. The state of polarisation of the light diffused by such elements would necessarily be influenced by this fact.

7. Comparison with the case of labradorite

The figures given in table 2 enable us to appreciate the reason for the observed differences in the optical behaviour of moonstone and of labradorite. The differences in the refractive indices of the mixture of albite and anorthite which principally constitute labradorite and the refractive indices of the orthoclase crystallites which segregate from them are greater than the average of the fluctuations in refractive indices in the case of moonstone by a numerical factor of about 7. The intensity of the light diffused by volume elements of equal size would be therefore some fifty times greater for labradorite than in moonstone. The brilliancy of the effects exhibited by labradorite despite the fact that the orthoclase which segregates in it is relatively small in quantity is thus readily understood. The circumstances in labradorite are clearly also favourable for the segregated crystallites to appear throughout the mass with a tolerable uniformity of size, a condition necessary for the manifestation of vivid colour in the later stages of segregation. On the other hand, in the case of the moonstone, the materials under consideration, viz., the soda and potash feldspars, are present in comparable quantities and are more or less freely miscible with each other. In these circumstances, we may expect the segregated crystallites to vary widely in their sizes. Thus, except in the earliest stages when all the crystallites are small and therefore give a diffusion of a blue colour, a manifestation of vivid colour is not to be expected in the case of moonstones. There is still another striking difference between the two cases which is indicated by theory and confirmed by observation. The intensity of the diffusion in the case of labradorite is sensibly independent of the direction of the electric vector inside the crystal, and the diffused light, in consequence, exhibits a polarisation of the same character as the diffusion in optically isotropic media. As has already been remarked, a different situation is expected and actually found in the case of the moonstones.

8. Polarised extinction of light

There is another group of effects observed with moonstones which are of interest and importance for the elucidation of their structure. As we have seen in the foregoing pages, the diffusion of light in its passage through the moonstone is a consequence of the local variations in composition and the variations in refractive index resulting therefrom. Apart from the effects arising in this way, the Ceylon moonstones are transparent monocrystals which do not exhibit any body-colour. Hence, any loss of light which appears in the passage of a light beam through the moonstone may reasonably be attributed to the deviation of the incident energy as diffused radiation. In other words, the energy appearing as transmitted light is the incident energy less the energy diffused in various directions. It follows that the extinction of light resulting from its passage through the crystal would exhibit various features which are the counterpart of those observed in diffusion. Besides thus representing the integrated effect of the diffusion, the extinction effects usefully supplement the information given by a study of the diffusion phenomena. As examples of the correlations found between diffusion and extinction in our studies may be mentioned the following instances. The moonstones in our collection, though derived from the same source, show a wide divergence in the colour and intensity of the schiller they exhibit. The extinction phenomena likewise show a similar wide range of behaviour. Again, as the angle which the direction of the incident beam makes with the schiller-axis of a moonstone is gradually increased from 0° to 90°, large changes are observed in the angular distribution of the diffused radiation and in its aggregate intensity; the nature of these changes is related to the colour and intensity of the schiller of the specimen under study. The extinction phenomena likewise show remarkable changes when the angle between the direction of the incident light beam and the schiller-axis is altered, these changes altering in character, as we proceed from the moonstones with a blue-schiller to those with a bluish-white and a white schiller respectively. The most interesting features of the extinction of light are the colour and the polarisation observed in various circumstances even when the incident light is unpolarised. These features are found to be closely connected with the spectral character and the polarisation exhibited by the diffusion of light and discussed earlier in the paper.

The following further details may be mentioned by way of supplementing the foregoing brief remarks. The moonstones which exhibit a blue schiller when the light is incident along the schiller-axis show a pale yellow colour by transmitted light, evidently the result of greater extinction of the smaller wavelengths in the incident light. Examination through a polariser shows that this extinction is more marked for a vibration direction along the *b*-axis of the crystal than for a perpendicular direction. This effect is evidently the counterpart of the greater intensity of the schiller for a vibration along the *b*-axis. As the schiller-axis is turned away from coincidence with the incident light beam to a perpendicular direction, the schiller becomes steadily weaker in intensity, and the extinction likewise tends to zero. Moonstones with a strong white schiller show rather different phenomena. When the specimen is held with the schiller-axis parallel to the incident light, the polarisation in the backward direction parallel to the *b*-axis is hardly observable, evidently because the crystal, in that position, throws back nearly all the light that it receives. The transmitted light is very weak and shows a yellow colour; it is found to be distinctly polarised with the *b*-axis as the direction of smaller intensity. Moonstones with a white schiller also show a remarkable type of polarised extinction when the light traverses them in a direction nearly perpendicular to the schiller axis. Here again, the setting of the *b*-axis of the crystal with respect to the direction of incidence of the light is found notably to influence the completeness of the extinction and the perfection of the polarisation of the weakly transmitted light. The special polarisation effects observed in these circumstances are rather reminescent of those noticed in the light transmitted by a pile of glass plates held obliquely. But the diffusion of a blue colour simultaneouly observed in a transverse direction and the dependence of the phenomena on the setting of the *b*-axis with respect to the plane of incidence serve to remind us that the analogy with the behaviour of a pile of glass plates is only superficial.

9. The X-ray evidence

The crystal structure of moonstones has been very extensively investigated by X-ray workers. We shall not here attempt anything like a complete review of the literature, but will content ourselves with referring to some of the more important investigations[5,6 7] in the field. It is evident that for a satisfactory correlation of the optical behaviour of a moonstone with the facts revealed by an X-ray examination of its structure, it is necessary that the same specimens should be studied by both methods, and that the optical evidence should be as complete and detailed as the X-ray evidence with which it is sought to be compared. It is essential, for instance, to have quantitative data for the intensity and spectral character of the diffused light and their variations with the setting of the crystal and the direction of observation. Only then it would be possible to estimate the size, shape and orientation of the crystallites of the soda-feldspar which diffuse the light, and to find whether the X-ray evidence supports those findings or at least is not in disagreement with them. We may, however, permit ourselves to make a few comments. The X-ray evidence does indeed support the finding that the soda-feldspar in moonstones tends to segregate from the potash feldspar. Particularly significant are the results of a recent investigation reported in a book[8] by Professor Ito in which the Weissenberg X-ray diagrams of a Korean moonstone containing 7·3% of K_2O, 6·9% of Na_2O, and 0·7% CaO are reproduced and discussed. Though the data are not strictly comparable with those of the Ceylon moonstones, they show clearly that segregation of the soda-feldspar occurs in such a manner as to make a near approach to the structure and symmetry of the parent crystal. We may remark also on the observation by Chao, Smare and Taylor that certain specimens of moonstones show a faint schiller but give no trace of any additional X-ray reflections; there is also an observation of Chao and Taylor that in certain cases, the supplementary X-ray reflections observed are diffuse. Ito has also remarked that in a number of photographs obtained by him, the Weissenberg diagram consists of bands rather than discrete spots. It is obvious that if the segregated crystallites are sufficiently small and numerous, they could give an observable diffusion of light, but that in the X-ray patterns, the reflections given by them would be smeared out just as they would be in the case of very finely divided colloidal particles. Whether this is the explanation of the facts observed, or whether there is actually some indefiniteness in the crystal structure of the segregated soda-feldspar is a matter for further investigation. There is clearly a need for further studies in which the optical and X-ray behaviour of the specimens are examined with equal thoroughness. The position as it stands now may be conservatively stated thus: the X-ray data do not contradict the evidence regarding structure presented in this paper on the basis of their optical behaviour; on the other hand, they support the main conclusions put forward in the paper.*

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*B V Thosar (*Philos. Mag.*, October 1945) described some observations on the spectral character, polarisation and intensity of the schiller of moonstone on the basis of which he concluded that the phenomenon is a reflection of light at the twinning planes of a microcline structure which he assumed it to possess. This view is in such complete contradiction with the facts regarding the optical behaviour of moonstone, to say nothing of the X-ray evidence, that we do not propose to traverse it in detail. Thosar's observations lack certain particulars needed to give them scientific significance. But so far as they go, they are fully explicable in the light of the considerations advanced in the present paper.

10. Concluding remarks

To avoid overloading the present paper, we have had to confine ourselves to a rather terse statement of the experimental facts on the basis of which the explanation of the schiller of monostone given in the paper has been developed. Much other material regarding the angular distribution of the intensity of the diffused light, its spectral character and its state of polarisation in various circumstances was gathered in the course of our studies. Some of this material was of a quantitative character, as for instance, the determination of the state of polarisation of the schiller for various settings of the crystal and for various angles of observation. Other observations, though of a qualitative character, were of scientific significance, as for instance, the fact that a "residual blue" is exhibited by moonstones giving a white schiller when the latter is extinguished by observation through a polariser. Detailed studies have also been made of the extinction of light in moonstone and of its spectral character and state of polarisation in various circumstances. To have set out all these results in detail here would have diverted attention from the main theme of the paper, namely, the genesis of the schiller as an optical consequence of the local fluctuations in its chemical composition and refractive index. We hope to be able to follow up the present communication by a paper in which the experimental facts of the subject are set out in greater detail.

In conclusion, we have to thank Mr G L Punnyasoma whose generous gift of moonstone specimens from Ambalangoda encouraged us to undertake the present research.

11. Summary

The paper describes the optical behaviour of the moonstones from Ambalangoda in Ceylon which exhibit a wide diversity in the colour and intensity of their schiller. The studies made show that the schiller is a diffusion of light within the material which is macroscopically a monocrystal but exhibits pronounced local variations in its composition and refractive index. Though the potash and soda feldspars mix together when they crystallise, the soda component tends to segregate and form tiny crystallites of which the size, shape and orientation determine the angular distribution of the diffused light, its spectral character, intensity and state of polarisation in various circumstances. The crystallites tend to appear with their greatest extension nearly parallel to the 100 planes of the crystal, the direction of the *c*-axis being preferred more than that of the *b*-axis. The distribution of intensity of the diffused light accordingly does not exhibit a radial symmetry with respect to the direction of its maximum intensity, and this lack of symmetry is conspicuously observable. The intensity of the schiller and its state of polarisation are also markedly influenced by the orientation of the electric vector

within the crystal, as a consequence of the variations of refractive index being much greater along the *b*-axis of the crystal than in perpendicular directions. The diffusion of light in moonstone is accompanied by a marked extinction, which exhibits an almost perfect polarisation in certain circumstances. An explanation is given of the striking differences in the optical effects displayed by moonstone and by labradorite in spite of the general similarity in their physical origin.

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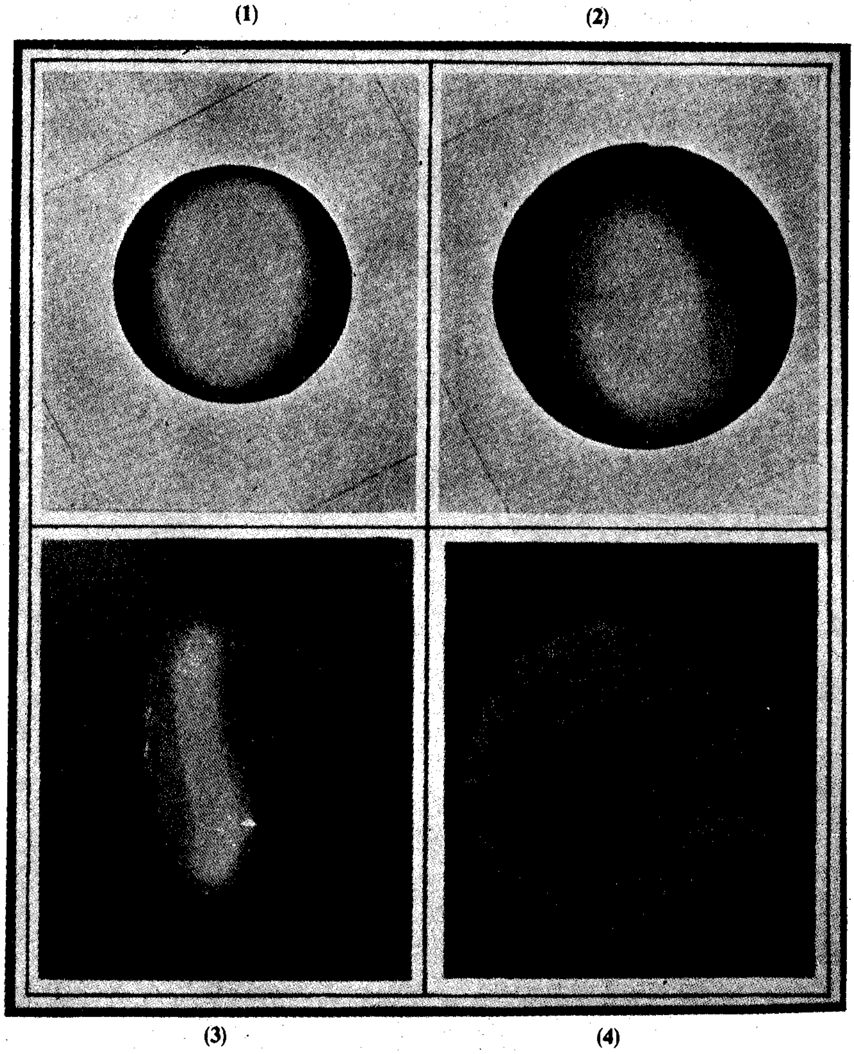

Figures 1–4

Plate I