UNMIXING IN PERISTERITE PLAGIOCLASES OBSERVED BY DARK-FIELD AND PHASE-CONTRAST MICROSCOPY

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ABSTRACT

Low-temperature plagioclase feldspars in the peristerite range (2–17 wt. % anorthite) are structurally inhomogeneous mixtures of $An_{3\pm2}$ and $An_{25\pm5}$ in varying proportions. Dark-field and phase-contrast microscopy by transmitted light are used to examine this peristerite unmixing, and these techniques are described in considerable detail to emphasize their present and potential applications in mineralogic investigations. It is determined that the An_{25} phase unmixed from its An_3 host may take the form of ellipsoidal blebs less than 0.5μ in dimension. Frequent occurrence of this exsolved material along twin boundaries suggests that there may be a cause-and-effect relationship of unmixing to twinning. Schiller as a reflection and scattering phenomenon is discussed.

INTRODUCTION

The peristerite plagioclases are low-temperature feldspars ranging in composition between the limits 2 and 17 wt.% anorthite (Laves, 1954; Brown, 1960). They are structurally inhomogeneous mixtures of host $An_{3\pm2}$ and exsolved $An_{25\pm5}$ in varying proportions (Fig. 1). A thorough discussion of the x-ray and optical crystallography of the peristerites has recently been undertaken by one of us (Ribbe, 1960); however, this study does not include information on the nature and occurrence of the exsolved phase that may be revealed by the use of special microscope techniques.

X-ray diffraction has given substantial evidence of peristerite unmixing, but it has provided no clue to the size or shape of the exsolved domains. Ordinary microscope techniques have shown no perthite-type unmixing, and consequently the domains have been considered submicroscopic. However, in the course of entirely unrelated optical studies at Corning Glass Works, Corning, New York, dark-field and phase-contrast microscopy were brought to bear on the peristerites. The following is a practical description of these techniques for the interest of mineralogists and petrologists who are dealing with so-called submicroscopic inclusions and impurities. The findings in the peristerite feldspar range will be presented in conclusion as a demonstration of the value of these special microscope methods.

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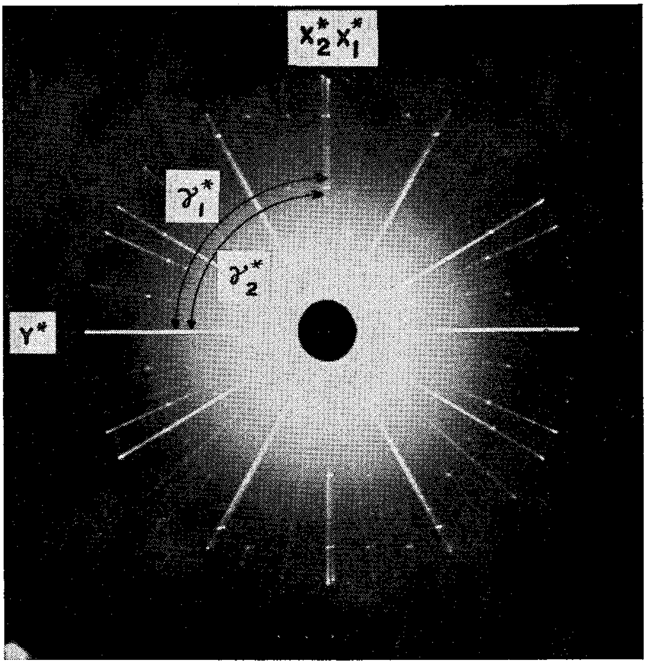


FIG. 1. A zero-level X^*Y^* precession photograph of a peristerite plagioclase (Specimen 3). The distinct X^* axes are evidence of the coexistence of two plagioclases in this single crystal. The more intense X^* axis represents the albite host, the less intense the exsolved An_{25}. Unfiltered Mo radiation.

General Statement on Microscopy

Bright-field, polarized-light microscopy—based on a solid cone of direct, transmitted light—is a chief method of the mineralogist. Phase-contrast and dark-field microscopy by transmitted light can often be helpful supplements, and are worthy of more use than has to date been given them. Often they are highly valuable in the detection or accentuation of very small or subtle specimen details such as inclusions, impurities, interstitial matter, bubbles, flaws, cracks, or phases unmixed from solid solution. These inhomogeneities are then seen by enhanced brightness contrast, which may result even from small differences in refractive index or optic path. Sometimes colour contrast, arising from diffraction or dispersion, can be helpful. Also, with strong brightness contrast one may detect but not delineate inhomogeneities whose sizes lie below the resolving limit of the microscope. This explains the long familiar use of the dark-field microscope in studies of colloids (Zsigmondy, 1909;

Kruyt, 1952) and the recent application of phase contrast in ultramicroscopy (Morehead, *et al.*, 1959).

The Kinds of Microscopy

Bright-field microscopy, with the added feature of polarized light, lends itself to observations dependent on birefringence, substantial contrast or relief, colour, and pleochroism. Also dispersion can sometimes be utilized, *e.g.* in the Becke-line, liquid immersion method.

Restricting the iris diaphragm of the condenser narrows the light cone and increases relief or contrast and depth of focus, but if carried very far the iris restriction impairs the brilliance and crispness of the image. Where colour contrast and brightness contrast are weak, because of little difference in optic path or in birefringence, the phase-contrast or dark-field method is often helpful.

The phase-contrast microscope accentuates small variations in optic path (thickness multiplied by refractive index) as enhanced variations in brightness contrast. This is achieved usually by illumination with a hollow cone of light. Except as deviated by the specimen being viewed or the specimen's details, the light passes through a ring-shaped section of a phase plate in the rear focal plane of the objective lens. Some of the light deviated by the specimen passes through the ring, and some passes through other parts of the phase plate. The ring either advances or retards the undeviated light one-quarter wave length with respect to the deviated light, and commonly the ring absorbs some of the undeviated light. The resulting view or image is much like that seen under the bright-field microscope when the specimen exhibits substantial absorption. For a basic understanding of phase-contrast microscopy the reader is referred to Zernike's article (Strong, 1958).

Dark-field microscopy uses a hollow cone of light sufficiently oblique to miss entrance into the objective lens except as the specimen being viewed deviates the light. The deviation consists of light scattering or diffraction at optical interfaces such as commonly are provided by grain boundaries, interstitial materials, inclusions, impurities, cracks, and flaws.

An optical interface between two transparent phases of very nearly equal refractive indices will sometimes be invisible or scarcely visible in dark-field lighting, for under this condition scattering can be nil. However, if the geometry and the orientation of the interface bear a favourable relation to the hollow cone of illumination, light can be scattered by smaller refractive-index differences than could otherwise be expected to cause light path differences. Or if the dispersions of the two phases separated by the interface differ, and white light is used, light of one colour may be selectively scattered so as to reach the observer's eye (Dodge, 1948). And regular arrays of phases, such as the lamellar unmixing in certain perthites, may behave as diffraction gratings and in white illumination deviate the whole spectrum of colours to the observer's eye.

Given a specimen having low-contrast details and poor prospects for bright-field study, one can apply phase-contrast or dark-field microscopy —sometimes the former with the greater success, sometimes the latter. Phase contrast is more likely to yield an image having both good brightness contrast and good, thorough delineation of specimen details than is dark-field microscopy. Theoretically, excellent delineation is more to be expected in phase-contrast than in dark-field microscopy, or than in bright-field microscopy when the condenser iris must be highly restricted to provide brightness contrast. This follows because the opaque stop blocks out the central spectra in dark-field lighting and the iris diaphragm trims off the outer spectra in bright-field lighting (Sommerfeld, 1954). This blocking and trimming of the light is apparently much more detrimental to the diffraction pattern making the image than is the loss of some of the deviated or diffracted light passing into the phase ring in the phase-contrast technique.

Demonstration of the Kinds of Microscopy

With the help of a polished block of Corning Glass Works' uranium-bearing glass that fluoresces in tungsten light, Fig. 2 shows the paths of light for bright-field, phase-contrast, and dark-field microscopy essentially as they were employed in this study of peristerite feldspars. A

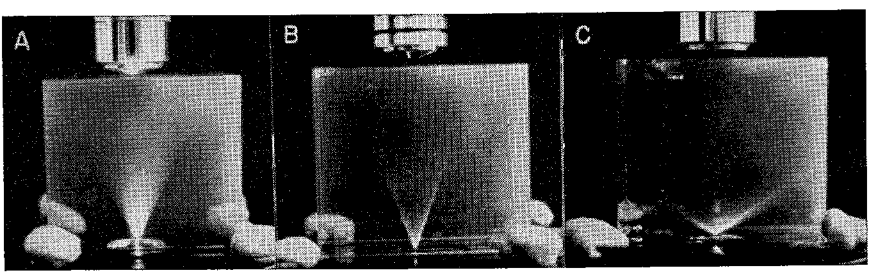
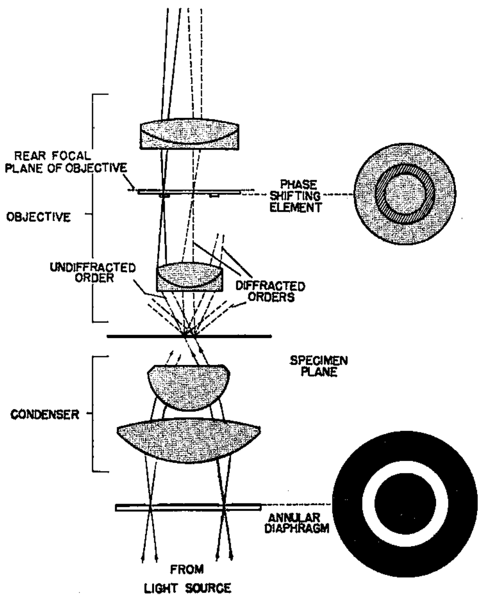


FIG. 2. Views of a fluorescent glass block on the microscope stage, and demonstrating light paths for various methods of illumination: (A) Bright-field lighting: solid cone of light for the 90× objective lens. (B) Phase-contrast lighting: hollow, acute cone of light for the 100× objective lens. (C) Dark-field lighting: hollow, obtuse cone of light for the 90× objective lens.

90× objective lens with an iris diaphragm was used for bright-field and dark-field examinations; a 100× lens with a dark-contrast phase plate was used for the phase-contrast views.

Fig. 2 shows the cones of light for the two objective lenses and the three kinds of microscopy: (A) the solid cone, polarized or unpolarized, for bright-field lighting, (B) the acute, hollow cone for phase-contrast lighting, and (C) the flattened or obtuse, hollow cone for the dark-field lighting. See also the schematic drawings, Figs. 3 and 4.



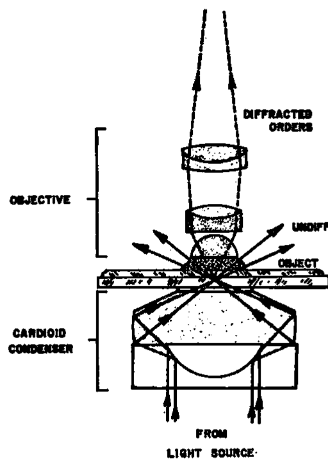


Fig. 3. Schematic drawing of phase-contrast lighting (left, after Benford, 1951). Courtesy of Bausch & Lomb, Incorporated.

Fig. 4. Schematic drawing of dark-field lighting (right, modified after Benford, 1951). Courtesy of Bausch & Lomb, Incorporated.

Fig. 5 corresponds with Fig. 2 and is a composite of polarized bright-field, unpolarized bright-field, phase-contrast, and dark-field views of a single plagioclase fragment. The polarized bright-field view alone shows the twinning, but it masks the rows of fine inclusions revealed poorly in the unpolarized bright-field view and conspicuously in the phase-contrast and dark-field views. The inclusions are thought to be blebs of plagioclase unmixed from solid solution and having 25 wt.% of the anorthite end-member. They are smaller than 0.5μ in dimension and on the average differ in refractive index from their albite host by 0.012.

In this high-magnification study the writers found their dark-field

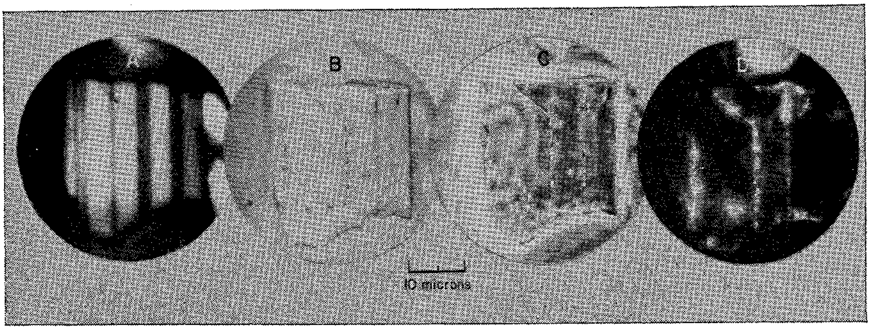


FIG. 5. A fourfold view of a single peristerite fragment, to demonstrate the value of the microscope techniques described in the text: (A) Bright-field lighting, polarized. (B) Bright-field lighting, unpolarized. (C) Phase-contrast lighting. (D) Dark-field lighting.

equipment less convenient to use than their phase-contrast equipment. For the dark-field work an oiled contact has to be maintained between the condenser and the specimen slide, and the lighting has to be aligned very precisely. Furthermore, very strong lighting is desirable for dark-field work at high powers, whereas lighting of moderate intensity suffices for phase contrast. Also, bright-field work between crossed nicols requires very strong lighting at high magnifications. The moderate light intensity needed for phase-contrast studies favours its use in photomicrography.

OCCURRENCE AND IDENTIFICATION OF THE EXSOLVED PHASE IN PERISTERITES

With dark-field and phase-contrast microscopy the writers tried to identify and reveal the distribution of An_{25} blebs in an albite matrix. The existence of the blebs was presumed from previous *x*-ray diffraction studies (Laves, 1954; Ribbe, 1960).

Of the nine plagioclase specimens microscopically examined, the six in the An_2–An_{17} range showed blebs in various abundances. As expected, Specimen 7 (Table 1) had no visible exsolved domains. Its composition is $An_{20.4}$. The two labradorites failed also to give evidence of exsolution.

To confirm that the brightness contrasts of An_{25} blebs against the An_3 host were sufficient for detection by the microscope techniques employed, the writers examined a perthitic feldspar from Larvik, Norway. Both phases (orthoclase and albite) constituting this feldspar were abundant, differed in refractive index by approximately the amount separating An_3 and An_{25} (see Table 2), and were successfully distinguished by phase-contrast and dark-field microscopy—especially by the former

TABLE 1. PLAGIOCLASE SPECIMEN DATA

No.	Locality	Calculated weight% An	Ab	Or	Source of analysis[a]	Comments
1	Auburn, Maine	5.2	94.0	0.8	CDJ	many inclusions
2	Monteagle Township, Ont.	5.7	90.3	4.0	VBM	
3	Villeneuve, Quebec	7.6	90.6	1.7	VBM	
4	Haddam, Conn.	9.5	90.5	—	CDJ	
5	Peekskill, New York	11.4	87.0	1.6	RCE	.10% impurities (chlorite)
6	Monteagle Valley, Ont.	13.3	82.8	3.9	VBM	
7	Bakersville, N.C.	20.4	75.7	3.9	CSR	very clean
8	Labrador	Both are schil-			—	Univ. of Wis. 0/43046
9	?	lered labradorites			—	Univ. of Wis. 0/43130

[a]REFERENCES: RCE—R. C. Emmons (1953); CDJ—C. D. Jeffries (1936); CSR—C. S. Ross in Emmons (1953); VBM—V. B. Meen (1933).

Specimens 1, 4, 8, and 9 were contributed by the University of Wisconsin Museum.

Specimens 2, 3, and 6 were contributed by the Royal Ontario Museum of Geology and Mineralogy.

Specimens 5 and 7 were contributed by R. C. Emmons, University of Wisconsin.

method. The Norwegian material lent itself less favourably to dark-field lighting than did the peristerites, with their small, nearly spherical blebs, because its unmixing took the form of lamellae of thicknesses near 0.5μ.

With dark-field lighting, size and geometry are important factors. Because dark-field is an edge-lighting technique, a minute, rounded grain becomes essentially a point source of light and may contrast brilliantly with a larger fragment whose more widely separated and irregular boundaries deviate and concentrate the diffracted orders of light much less effectively. Thus the tiny blebs that occur in most

TABLE 2. DIFFERENCES BETWEEN REFRACTIVE INDICES OF DOMAINS IN PERTHITES AND IN LOW- AND HIGH-TEMPERATURE PERISTERITES

Domain composition	Refractive index (n_D) γ	β	α	Source
Low-temp. Or	1.5245	1.5228	1.5192	
Ab	1.5392	1.5328	1.5292	Raman, *et al.* (1950)
Difference:	.0147	.0100	.0100	
Low-temp. Ab	1.5379	1.5314	1.5274	
An_{25}	1.5486	1.5447	1.5408	Chayes (1952)
Difference:	.0107	.0133	.0134	
High-temp. Ab	1.535_6	1.534_2	1.527_3	J. R. Smith (1957)
An_{25}	1.548_5	1.545_8	1.541_1	
Difference:	$.012_9$	$.011_6$	$.013_8$	

peristerite grains are rather brightly contrasted against the host material. Their shape, which is more clearly delineated by phase contrast, is commonly spherical or ellipsoidal.

Hematite, a not uncommon inclusion in sodic plagioclase, had to be excluded as a possible alternative explanation in the process of identifying these blebs. Peristerites containing hematite visible in bright-field lighting were examined by dark-field and phase-contrast methods. The dark-field view showed the hematite colour even in very small inclusions, and the brightness contrast with the host feldspar was extreme. This contrast was even more clearly pronounced in the phase-contrast view, where the

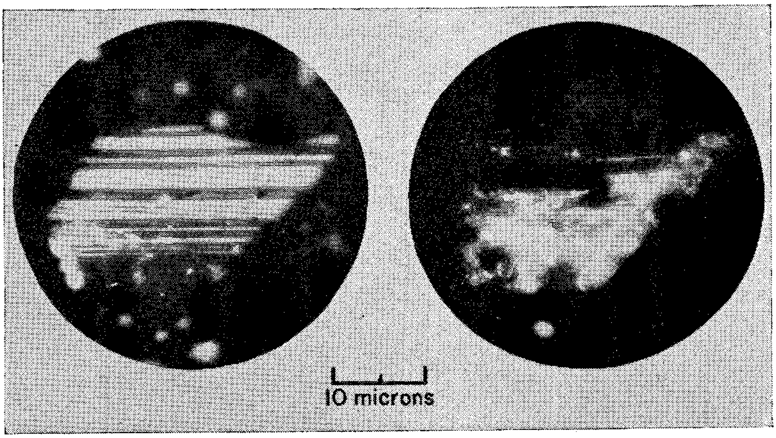


FIG. 6. A twofold view of a single peristerite fragment (Specimen 5): (A) Polarized bright-field lighting, showing twinning and a NE-SW fracture near the right-hand boundary of the grain. (B) Dark-field lighting, showing clouds of blebs in random distribution and occasional lineation of distribution along twin composition faces.

high-index hematite appeared as brilliant as polished metal. In addition, the most spectacularly unmixed peristerite (Specimen 5: Figs. 6, 7-B, 7-C) is known from chemical analysis (Emmons, 1953) to contain less than 0.08% FeO and Fe_2O_3. Perhaps the most convincing argument that the blebs are not hematite is that they remain through the extended heat-treatment discussed below. It is well known that Fe_2O_3 will go into solid solution with the feldspar upon heating and will not exsolve again upon cooling (Anderson, 1915).

A small amount of powdered (-50μ) Specimen 5 (see Table 1) was heat-treated for 65 days at temperatures ranging between 1045 and 1070° C in an air-atmosphere electric furnace. The blebs had not disappeared after this long heating, and this leads to the following alter-

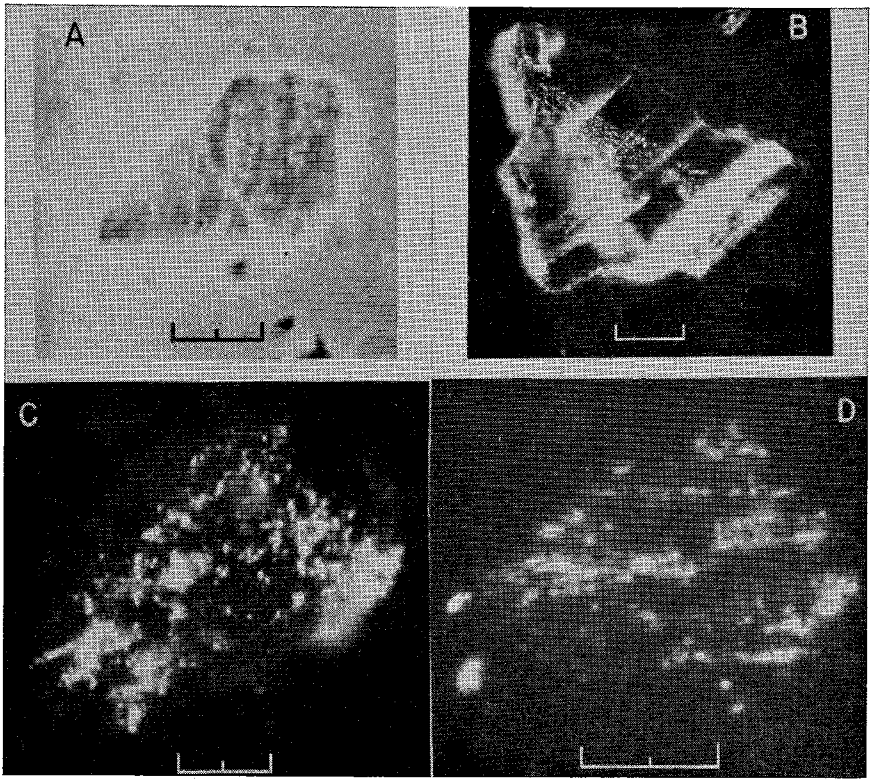


FIG. 7. Views of peristerite fragments using special microscope techniques: (A) Specimen 3. Phase-contrast lighting. (B) & (C) Specimen 5. Dark-field lighting. (D) Specimen 6. Dark-field lighting. All scales are 10μ from end to end.

natives. The first is that a much longer heat treatment is required for true homogenization, which includes the diffusion of Al-Si between the phases. Goldsmith (1952) emphasizes the strength of the Al and Si tetrahedral bonds and the extreme sluggishness of their diffusion over substantial distances. The second is that the blebs are something other than An_{25} and must be considered inclusions. A liquid, a gas, or another mineral, perhaps quartz, are possibilities. The near equality of the refractive indices of host and bleb rules out a great number of materials. If the inclusions were quartz, the condition of similar refractive indices would be fulfilled, but one would expect that this would show up as excess silica in the chemical analyses. Emmons (1953) calculates 3.1% excess silica for Specimen 5, but this would hardly be sufficient to account for the number of blebs observed.

There is, then, some uncertainty in the identification of the blebs as exsolved An_{25}, due to the failure of the blebs to disappear with heat treatment. But it is to be noted that the refractive-index contrast of the high-temperature forms of Ab and An_{25} is somewhat greater than the contrast of the low-temperature forms (Table 2); therefore, if heating had only served to disorder completely the Al-Si *within* the host Ab and the exsolved An_{25} domains, but not *between* them (as required for true homogeneity), the blebs should remain visible and schiller ought also to persist. Much longer heating or perhaps heating in a water-enclosed system is probably required to eliminate blebs and schiller. However, until this further evidence is available, it is assumed that the indirect evidence for An_{25} forms an adequate case for the discussion at hand.

Through application of the microscope techniques here described, certain additional information concerning the peristerites was gained. The most rudimentary is the size of the blebs. They appear in spherical or ellipsoidal form in dimensions ranging downward from 0.5μ (5000 Å). Not uncommonly observed in dark-field lighting were bluish clouds of sub-micron blebs associated with larger, more brilliant domains of An_{25} (Fig. 6B). It is presumed that there is a lower limit of domain size in the vicinity of 0.02μ, because x-ray diffraction patterns are generally sharp and clear, except as discussed by Brown (1960) for cases of incomplete unmixing. It is probable that a multitude of smaller belbs cannot be detected with the optical equipment used and the rather weak light source.

Unfortunately it is not possible to distinguish positively which of the peristerite phases, host or bleb, has the higher refractive index.[2] Phase-contrast microscopy is normally capable of supplying this information, but perfect focus on a bleb is necessary. The minute size of the blebs makes for unreliable results.

The occurrence of blebs of exsolved plagioclase is very often in lines (Figs. 5, 6, 7A, 7D) or planes (Fig. 7B), although patches of randomly distributed blebs are the most commonly seen. It is likely that the unmixing of An_{25} from a solution occurs at points of lattice defects and impurities, fractures, and cleavages. Fractures are known to localize blebs (Fig. 6B, the northeast-southwest line at the right); but interpretative caution is advised, for air or vacuum can provide extreme refractive-index contrast with the host and thus cause brightness contrast in the field of view.

The tendency for unmixing to occur on the composition face of the

[2]It is, therefore, debatable whether the domains are in fact Ab and not An_{25}—a point of some academic interest. Because the vast majority of peristerites examined have a ratio of Ab to An greater than 1, it is assumed herein that Ab is the host material.

albite twins—crystallographic (010)—suggests a cause-and-effect relation between twinning and exsolution. There is structural adjustment at composition faces that may tend to serve as dislocation and impurity traps and that may nucleate the exsolution phase. It is proposed that twinning precedes exsolution for these reasons: (1) the random distribution of the blebs (Figs. 6B, 7) is seen much more frequently than the planar distribution, (2) a substantial percentage of the twin boundaries contain no visible blebs (Figs. 5, 6B), and (3) Brown's observation that his ($0\bar{8}1$) lamellae are younger than the twinning (see discussion below and Brown, 1960, p. 334).

The light-blue to white schiller occasionally shown by peristerite plagioclases is additional evidence of the relationship that exists between the exsolved phase and its host. Peristerite schiller is visible only through the (010) cleavage and generally parallels ($0\bar{8}1$) (Bøggild, 1924). Brown (1960) has stated evidence for a reflecting ($0\bar{8}1$) schiller plane, showing in thin section very thin lamellae of a discontinuous nature and of parallel orientation along ($0\bar{8}1$). It is clear that this crystallographic plane is an important one in schiller formation, and the most obvious assumption is that schiller then is a reflection phenomenon.

However, there are strong arguments against schiller as exclusively a reflection from planes of a size very much greater than the wave length of light. Indeed, it would seem that in the case of the usually pale blue or white of peristerite schiller, diffuse reflection from particles of a size near or somewhat larger than that of a wave length of light, and scattering from particles much smaller than the wave length, combine to produce the schiller. The intensity of the blue colour of the schiller would increase with the increasing proportion of scattering particles to reflecting particles, and white schiller would be visible if the exsolved phase were predominantly larger than, say, 4000 Å.

The schiller of peristerites varies from grain to grain and specimen to specimen, and more often than not is absent from grains known to be unmixed from *x*-ray photographs. When schiller is absent it is probable that the unmixed An_{25} is in domains so small that scattering is unobservedly weak.

The preceding argument can be developed from a purely theoretical viewpoint (Jenkins & White, 1957, pp. 455–8 and 505–7). However, the same explanation has been advanced with full experimental confirmation by Raman, Jayaraman, & Srinivasan (1950) for the case of moonstones. Since the peristerites are analogous to the moonstones in optical properties (Ribbe, 1960), and since the present study confirms the existence of particles of the correct size to cause the observed schiller, it would appear that the picture is complete. But the reason why the preferred direction of schiller is parallel to $(0\bar{8}1)$ is yet to be determined. It is beyond the scope of the present work to discuss this further. However, what has been deduced about the nature of the exsolution phase in peristerites from a consideration of the schiller phenomenon further supports the identification of the observed blebs as An_{25} in an albite matrix.

Conclusions

Phase-contrast and dark-field microscopy by transmitted light have been presented as powerful tools in mineralogical research, which is increasingly concerned with the minutiae and the "submicroscopic" in its search for petrogenic indicators. The peristerites have been examined by those methods and found to duplicate the crystallographic model that had been mentally but never visually constructed from the less direct evidence of *x*-ray diffraction and schiller. Peristerite schiller is discussed as a reflection and scattering phenomenon whose orientation is observed, but not understood.

Acknowledgments

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References

Anderson, O. (1915): On aventurine feldspar, *Am. J. Sci.*, **40**, 361–397.

Benford, J. R. (1951): *The theory of the microscope*, Bausch & Lomb Optical Co., Rochester, N.Y.

Bøggild, O. B. (1924): On the labradorization of the feldspars, *Det Kgl. Danske Videnskabernes Selskab*, **6**, 1–79.

Brown, W. L. (1960): The crystallographic and petrologic significance of peristerite unmixing in the acid plagioclases, *Zeits. Krist.*, **113**, 330–344.

Chayes, F. (1952): Relation between composition and indices of refraction in natural plagioclase, *Am. J. Sci., Bowen volume*, 85–106.

Dodge, N. B. (1948): The dark-field color immersion method, *Am. Mineral.*, **33**, 541–549.

Emmons, R. C., ed. (1953, reprinted 1956): Selected petrogenic relationships of plagioclase, *Geol. Soc. Am., Mem.*, **52**.

Goldsmith, J. R. (1952): Diffusion in plagioclase feldspars, *J. Geol.*, **60**, 288–290.

JEFFRIES, C. D. (1936): II. Optical studies of feldspars, *Unpublished Ph.D. thesis*, Univ. of Wisconsin, Madison.

JENKINS, F. A., & WHITE, H. E. (1957): *Fundamentals of optics*, McGraw-Hill Book Co., New York.

KRUYT, H. R., ed. (1952): *Colloid science*, Elsevier Publishing Co., Amsterdam.

LAVES, F. (1954): The coexistence of two plagioclases in the oligoclase compositional range, *J. Geol.*, **62**, 409–411.

MEEN, V. B. (1933): A description of a few plagioclases, *Univ. Toronto Stud., Geol. Ser.*, **35**, 37–45.

MOREHEAD, F. F., *et al.* (1959): *Symposium on microscopy*, Amer. Soc. Testing Materials, Philadelphia.

RAMAN, C. V., JAYARAMAN, A., & SRINIVASAN, T. K. (1950): The structure and optical behavior of the Ceylon moonstones, *Proc. Ind. Acad. Sci.*, **32A**, 123–140.

RIBBE, P. H. (1960): An *x*-ray and optical investigation of the peristerite plagioclases, *Am. Mineral.*, **45**, 626–644.

SMITH, J. R. (1957): Optical properties of heated plagioclases, *Ann. Rep. Dir. Geophys. Lab.*, 188–190.

SOMMERFELD, A. (1954): *Optics*, Academic Press, New York.

STRONG, J. (1958): *Concepts of classical optics*, W. H. Freeman & Co., San Francisco.

ZSIGMONDY, R. (1909): *Colloids and the ultramicroscope*, John Wiley and Sons, New York.

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