The structures of the plagioclase felspars: VI. Natural intermediate plagioclases.[1]

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Summary.—Specimens from some forty different localities over the composition range from about 20 % An to about 70 % An have been examined by X-ray single-crystal methods. The majority of these specimens show the normal intermediate plagioclase pattern characterized by weak pairs of subsidiary layer-lines of variable separation. It has been found that the separation of these layer-lines is a linear function of composition over the whole composition range. Further, this range is found to extend to more soda-rich compositions than was formerly believed, whilst the limits of the range are approximately defined by the compositions at which the separations of the subsidiary layer-lines about either the *a*- or the *b*-axes become zero.

The anomalous patterns shown by some specimens are discussed, and it is shown that most of these may be interpreted in terms of the previous geological history of the specimens.

A comparison of the present results with those obtained by previous workers in this field is made, and it is shown how the principal points of difference may be resolved. An account of the phase relationships of the low-temperature plagioclase series is given, and the problems arising from the present work are discussed.

EARLY X-ray investigation of the plagioclases (Chao and Taylor, 1940) showed that there were three main structural divisions of the series: the albite structure, the anorthite structure, and the intermediate structure. The work described in this paper has been carried out as part of a programme of research to investigate the phase relationships of the plagioclase felspars, and is concerned with the intermediate structural region. Previous papers in this series have described in detail the results of an examination of lime-rich plagioclases. In one of these publications (Gay, 1953), a convenient grouping of all possible X-ray reflections into four classes was established: class (*a*) reflections are those with $(h+k)$ even, l even; class (*b*) those with $(h+k)$ odd, l odd; class (*c*) those with $(h+k)$ even, l odd; class (*d*) those with $(h+k)$ odd, l even; all indexing being carried out in terms of the unit cell of anorthite with a 14 Å. *c*-axis. Type (*a*) reflections, the principal reflections, are common

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to all plagioclases; types (*b*), (*c*), and (*d*) are the subsidiary reflections, which distinguish the various structural arrangements. This classification will be adopted in the present publication.[1]

The most detailed examination of the intermediate structural region previously carried out has been described by Cole, Sörum, and Taylor (1951) and Sörum (1951). The diffraction pattern from an intermediate plagioclase is characterized by pairs of subsidiary reflections situated symmetrically about the layer-lines denoting the 14 Å. *c*-axis of anorthite; it appears that the type (*b*) reflections characteristic of body-centred anorthite have split into two. Doubled subsidiary reflections also appear symmetrically about the positions of the principal layer-lines in *a*- and *b*-axis photographs of these crystals. Cole *et al.* investigated the separation of these doubled subsidiary layer-lines as a function of the composition of the felspar. They expressed this separation in angular measure as a fraction of the spacing between the appropriate layers of the reciprocal lattice, successive layers being separated by 360°. δ_a, δ_b, and δ_c, defined in this way, correspond to measurements made for a reciprocal lattice based on the albite-type structure—i.e. for the anorthite structures the values of δ_a, δ_b, and δ_c will be 0°, 0°, and 180° respectively. This terminology will be employed in this paper. Cole *et al.* showed that δ_a, δ_b, and δ_c were mutually dependent, and suggested a form of variation for each of these quantities with composition. They concluded that one boundary of the intermediate structure occurred near 72 % An and that the other was near 30 % An.

It was suggested by Chao and Taylor (1940) that the main features of these patterns can be explained in terms of a regular stacking of slabs of albite-like and anorthite-like material in the direction of the *c*-axis. More recently, Sörum (1951) has proposed a refined model of this type, in which the An-rich structure is identified with body-centred anorthite. A number of points are left unexplained by these treatments, and the present work suggests some simplifications of the structural problems. It is hoped to treat these in a later publication.

The work described by Cole *et al.* was in the nature of a review of the whole plagioclase series; the work to be described in this paper represents a detailed re-examination and amplification of their results for the intermediate structure. In particular the author has rechecked the data for specimens more basic than 50 % An, and examined a number of

[1] It should be noted that a similar classification has been used by Laves and Goldsmith (1951, 1954). In their nomenclature, the classes (*a*), (*b*), (*c*), and (*d*) become (*a*), (*b*), (c_1), and (c_2) respectively.

specimens less basic than 50 % An, a region for which the results of Cole *et al.* are extremely sparse; he has also tried to determine how the transition at the sodic end of the series to the albite structure occurs, and investigated whether the separation of the subsidiary reflections could be used as a determinative method for plagioclase composition. A systematic study of the effects of heat treatment on a number of natural plagioclases in this region will be described in another paper.

Experimental methods.

Selection of material.—Untwinned and unzoned single crystals were used as far as possible. Many of the materials from which the specimens were selected had been chemically analysed; for such specimens, the grains selected were known from their optical properties to be typical of the bulk material, and it has been assumed that their composition corresponds to that of the bulk material. For specimens for which no analysis was available, optical determinations were made either from refractive indices (using the curves of Chayes, 1952) or from universal-stage measurements, usually on the fragment used for the X-ray work. Optical determinations of this kind were also made as a check on critical specimens even when their bulk composition was known from analysis. These methods were preferred to melting the crystal and determining the refractive index of the glass, principally because the crystal remains intact. The accuracy of determination is probably ± 2 % An at the worst, which is adequate for the present work, and is probably as good as can be obtained without resort to complicated experimental techniques.

In their previous work Cole *et al.* used a number of specimens selected apparently at random from a particular source material. The composition of each of these fragments was then determined by measurement of the extinction angles on the cleavage faces. These extinction angles are quoted to 0·1°, which seems to be an overestimate of the accuracy with which such angles can be determined. Further, the compositions were then derived using the curves of Rogers and Kerr (1942). Comparison of similar curves published by other authors shows that there are appreciable discrepancies among them, and it is difficult to assess the reliability of any one set of curves. Hence it seems that the accuracy of determination of $\pm\frac{1}{2}$ % An quoted by Cole *et al.* is again an overestimate.

It is probable that the methods adopted in the present work are more satisfactory. The optical curves used appear to have a sounder basis

than those of extinction angles. Further, it is thought that the refractive indices, in particular, change very little with heat treatment. This would minimize any error due to the use of inverted and partially inverted specimens, a point mentioned by Cole *et al.*

X-ray methods.—Single-crystal oscillation and Weissenberg methods have been exclusively used. The axis of oscillation has usually been one of the principal axes, although other axes have sometimes been used for confirmatory purposes. The oscillation photographs were used for the detection and the determination of the separation of the subsidiary layer-lines, and the Weissenberg photographs for the determination of the positions of the subsidiary reflections in reciprocal space.

Measurement of the separation of the subsidiary layer-lines.—Linear measurements of the separation between subsidiary reflections were made, using either a travelling microscope or a device employing two sliding, mutually perpendicular scales (similar to that shown on p. 226, Buerger, 1942). It was found that the former was satisfactory for sharp, relatively strong spots, whilst the latter was most usefully employed on the diffuse or weak reflections. The separations in reciprocal space were then calculated from the linear measurements.

The accuracy of measurement necessarily depends on the quality of the spots, which depends in part on the composition of the specimen. Cole *et al.* claim that an accuracy of $\pm 1°$ in δ_a, δ_b, δ_c (or $\frac{1}{360}$ of the separation of the principal layer-lines) is achieved *at the worst*; the agreement between the calculated and experimental measurements of separations for axes other than the principal axes which they quote is extremely good. The present author has not been able to reproduce this accuracy; the standard deviation calculated from a number of measurements on each specimen is usually *at the best* $\pm 1°$, and often very much worse. The measurements of δ_a, δ_b, and δ_c which have been made are quoted with the appropriate standard deviation. In considering the probable accuracy of measurement it should be noted that the majority of the specimens used by Cole *et al.* were in the composition range where the subsidiary diffraction spots are strong and sharp.

Specimens.

A list of the materials used together with the bulk chemical composition and occurrence is given in table I; wherever possible, references to previous descriptions and work on the material used have been included. The specimens included in this table all fall roughly within the composition range over which the characteristic intermediate pattern is found.

TABLE I. List of specimens.

No.	Locality.	Occurrence.	An/An+Ab (Mol. %).	References.
1	Lincoln Co., Wisconsin.	Anorthosite	73	5(24); 6(23).
2	Duluth, Minnesota.	Gabbro	71	5(8); 6(22).
3	Wichita Mts., Oklahoma.	Orthoclase-quartz-gabbro	70	5(5); 6(20).
4	Chester Co., Pennsylvania.	Diabase	67	5(18); 6(18).
5	Clear Lake, Utah.	Phenocrysts	65	15(7).
6	Stillwater, Montana (EB 40).	Gabbro	64	—
7	St. John's Pt., Co. Down, Ireland.	Phenocrysts in basalt	63	12; 1; 19.
8	Shelby, N. Carolina.	Hornblende-gabbro	60	5(17); 6(16).
9	Naine, Labrador.	?	58	3(E).
10	Naine, Labrador.	?	56	—
11	New Amalfi, South Africa.	Dolerite	55	19.
12	Skaergaard, E. Greenland.	Hypersthene-olivine-gabbro	53*	25(2307).
13	Isle of St. Paul, Labrador.	?	53	15(6).
14	New Amalfi, South Africa.	Dolerite	51	20(EH20); 19.
15	Skaergaard, E. Greenland.	Middle gabbro	50*	25(1691).
16	Essex Co., New York.	Anorthosite	50	5(4); 6(9).
17	Sipoo, Uudenmaan, Finland.	?	47*	—
18	New Amalfi, South Africa.	Dolerite	46	20(EH52); 19.
19	Monte Rosso, Linosa (B.M. 1924, 914).	Crystal lapilli	46*	7; 19.
20	Skaergaard, E. Greenland.	Middle gabbro	45*	25(3661).
21	Skaergaard, E. Greenland.	Ferrogabbro	45*	25(3655).
22	Beaver Bay, Minnesota.	Diabase	44	11; 19.
23	Skaergaard, E. Greenland.	Ferrogabbro	41	25(2580); 26.
24	NW. of Agay, Esterel, France.	Andesite	40*	14; 19.
25	Skaergaard, E. Greenland.	Ferrogabbro	40*	25(1907).
26	Crestmore, California.	Granodiorite	38	5(19); 6(8); 16(16).
27	Sierra Nevada, California.	Granodiorite	38*	—

TABLE I (*cont.*)

No.	Locality.	Occurrence.	An/An+Ab (Mol. %).	References.
28	Skaergaard, E. Greenland.	Ferrogabbro	37	25(4145); 26.
29	Yosemite Valley, California.	Hornblende-bastite-quartz-monzonite	37*	...
30	Skaergaard, E. Greenland.	Ferrogabbro	37*	25(4146).
31	San Luis Obispo Co., California.	Dacite	36	5(11); 6(7).
32	Spanish Peak, California.	Gneissoid granodiorite	35	5(21); 6(6); 16(14).
33	Knoydart, Scotland.	Garnet-mica-schist	35*	...
34	Santa Maria, Guatemala (Oct. 1902 eruption).	Pumice	33*	—
35	N. Morar, Inverness-shire, Scotland.	Garnet(?)-mica-schist	32*	—
36	Macon Co., N. Carolina.	From a vein of talc-antho-phyllite-vermiculite-plagioclase cutting dunite.	30	6(28).
37	Head of Little Rock Creek, Mitchel Co., N. Carolina (97490).	Coarse crystals from pegmatite	30	15(11).
38	Hawk Mica Mine, Bakersville, N. Carolina (81065).	Crystals from pegmatite	23	15(10).
39	Hawk Mica Mine, N. Carolina (103086).	Crystals from pegmatite	22	15(9).
40	South Carolina (81822).	Greenish clear crystals from pegmatite	17	15(8).

Notes.—All compositions in this table were calculated from analyses, except those marked *, which were determined optically. The bulk compositions of specimens 27, 29, 33, and 35, and the composition of the single fragment of specimen 34 used in the X-ray work were determined by Dr. I. D. Muir.

In the reference column the unbracketed numbers refer to the list of references at the end of this paper; the numbers or letters within an associated bracket are those given to the particular specimen by the quoted author. Of the specimens for which no reference is quoted, no. 6 was received from Professor H. H. Hess, 10 from Professor C. D. Jeffries, 33 and 35 from Mr. R. St. J. Lambert; specimen 34 is to be described in a forthcoming paper by Drs. Merwin and Zies.

A number of other specimens beyond the limits of this composition range have been examined: these are not listed in the table, but relevant

aspects of the results on these specimens will be quoted in the later discussion. A full description of these results will be given in another publication.

Experimental results and discussion.

In this section it is convenient to discuss first the diffraction patterns from low-temperature intermediate plagioclases that show similar characteristic features; these will be called 'normal' patterns. The 'anomalous' patterns that do not conform to the scheme for the normal specimens are discussed afterwards.

Felspars with the normal intermediate structure give diffraction patterns showing the characteristic doubling of the type (*b*) reflections. As noted by Cole *et al.* these reflections usually occur in pairs, although sometimes one member of a pair may be absent or undetected.

It was suggested in the earlier work that the separation of the subsidiary reflections was simply related to the composition of the fragment, and that the separations about the three principal axes were not independent. In table II are listed the measured values of the angular separations δ_a, δ_b, and δ_c for as many specimens as possible. It will be noted that for a number of specimens no values have been recorded; for these no subsidiary reflections were observed. For other photographs, although the subsidiary reflections may be present, they are of such poor quality that no reliable measurement could be made. In fig. 1, δ_a and δ_b are plotted against δ_c, and in fig. 2, δ_a, δ_b, and δ_c are plotted independently against the anorthite content of the specimen. In fig. 1, it is seen that δ_a and δ_b are linearly dependent on δ_c, whilst from fig. 2 it is seen that all three quantities vary linearly with composition to within the accuracy of the experimental measurements.

TABLE II. Measurements of separations of subsidiary reflections.

No.	% An.	δ_a.	δ_b.	δ_c.	Remarks.
1	73	—	$37\pm2°$	$165\pm1°$	Diffuse (*c*) spots with intermediate pattern.
2	71	—	—	—	Diffuse (*c*) spots with body-centred anorthite pattern, i.e. anorthite D pattern (Gay, 1953).
*3	70	—	$43\pm2°$	$163\pm1°$	—
4	67	$9\pm3°$	$35\pm2°$	$157\pm1°$	—
*5	65	—	—	—	†Very diffuse single (*b*) spots of body-centred anorthite present.
*6	64	$8\pm2°$	$33\pm1°$	$159\pm\frac{1}{2}°$	† —
7	63	—	—	—	Very diffuse single (*b*) spots of body-centred anorthite present.

TABLE II (*cont.*)

No.	% An.	δ_a.	δ_b.	δ_c.	Remarks.
8	60	$14\pm2°$	$28\pm1°$	$146\pm1°$	—
9	58	$12\pm2°$	—	$151\pm1°$	*b*-axis not observed.
*10	56	$23\pm1°$	$17\pm2°$	$141\pm1°$	Very diffuse weak reflections in addition to normal pattern.
*11	55	$16\pm2°$	$18\pm2°$	$148\pm1°$	—
*12	53	$18\pm2°$	$21\pm2°$	$145\pm1°$	—
*13	53	$25\pm1°$	$20\pm2°$	$145\pm1°$	‡ —
14	51	$20\pm1°$	$17\pm2°$	$138\pm2°$	Split (*b*) spots rather weaker than expected.
*15	50	$25\pm2°$	$21\pm2°$	$141\pm1°$	‡ —
*16	50	$24\pm2°$	$19\pm2°$	$139\pm1°$	—
17	47	$28\pm1°$	$13\pm1°$	$132\pm2°$	—
*18	46	—	—	$\sim 132°$	Split (*b*) spots barely visible.
*19	46	—	—	—	No (*b*) spots.
*20	45	$23\pm2°$	$19\pm2°$	$136\pm1°$	—
21	45	$23\pm3°$	$15\pm2°$	$136\pm1°$	—
*22	44	—	—	—	No (*b*) spots.
23	41	$27\pm4°$	$14\pm2°$	$132\pm2°$	Split (*b*) spots slightly weaker than expected.
24	40	—	—	—	No (*b*) spots.
25	40	$31\pm3°$	$9\pm2°$	$125\pm2°$	Split (*b*) spots definitely weakened, slightly diffuse.
26	38	$31\pm3°$	—	$128\pm3°$	Split (*b*) spots slightly diffuse. *b*-axis not observed.
27	38	$33\pm4°$	8°	$125\pm4°$	Split (*b*) spots diffuse.
*28	37	—	—	—	No (*b*) spots.
29	37	$32\pm3°$	$9\pm3°$	$121\pm4°$	Split (*b*) spots slightly diffuse.
*30	37	—	—	—	No (*b*) spots.
*31	36	—	—	—	No (*b*) spots.
*32	35	$30\pm5°$	$7\pm3°$	$128\pm4°$	Split (*b*) spots quite diffuse.
33	35	—	—	—	Split (*b*) spots too diffuse to be measured.
34	33	—	—	—	No (*b*) spots.
35	32§	$43\pm7°$	—	$113\pm5°$	Split (*b*) spots very diffuse.
36	30	$\sim 38°$	—	$118\pm7°$	Split (*b*) spots very diffuse.
37	30	$42\pm5°$	$\sim 4°$	$120\pm6°$	Split (*b*) spots very diffuse.
38	23	—	—	$\sim 121°$	Split (*b*) spots very diffuse.
39	22§	$\sim 43°$	—	$\sim 120°$	Split (*b*) spots extremely diffuse.
*40	17§	—	—	$\sim 100°$	Split (*b*) spots extremely diffuse.

* Indicates that more than one crystal has been examined. The values quoted for δ_a, δ_b, and δ_c are average values for all observations.

† Photographs of some specimens of these materials were taken by Dr. S. W. Bailey, University of Wisconsin.

‡ Some specimens of these materials were examined and the measurements of separations made by Mr. C. J. E. Kempster, Cavendish Laboratory, Cambridge.

§ The anorthite contents of the fragments used of these three specimens were determined from their refractive indices by Mr. J. D. C. McConnell; anorthite contents of 28 ± 3 %, 21 ± 3 %, and 23 ± 2 % were obtained for specimens 35, 39, and 40 respectively.

The quality of the type (*b*) spots is very variable. The doubled spots can be almost as sharp as the principal reflections, or extremely diffuse so that they can only be seen on very heavily exposed photographs; they can also show diffuse 'tails'. In the main, normal specimens in the composition range from about 70 % An to 40 % An show the sharp, relatively strong doubled reflections; more sodic specimens show these reflections becoming increasingly diffuse with soda content until at around 20 % An the original reciprocal points occupy very large volumes of reciprocal space. In this latter region measurements of the relative separations become increasingly more inaccurate. The doubled type (*b*) reflections which are present are derived by a systematic splitting of the type (*b*) reflections observed for body-centred anorthite. It also appears to be generally true that the intensities of the split reflections are comparable with those of the corresponding reflections in the body-centred anorthite pattern, i.e. a strong (*b*) reflection in body-centred anorthite is usually replaced by a relatively strong pair of split reflections in the intermediate pattern. It appears, however, that the intensities of the two components of any one pair are different (in agreement with Sörum, 1951, and Cole *et al.*, 1951). The intensities of corresponding (*b*) reflections measured relatively to the intensities of nearby principal reflections appear to change very little over the range 70 % An to 40 % An; it is difficult to be certain whether this remains true for the more sodic specimens owing to the rapidly changing diffuse character of the reflections. No systematic measurements of the changes in intensity of the principal reflections over the composition range have been made; it seems certain that some changes do occur that would be revealed by a detailed study, but they are small and are not immediately obvious on the photographs.

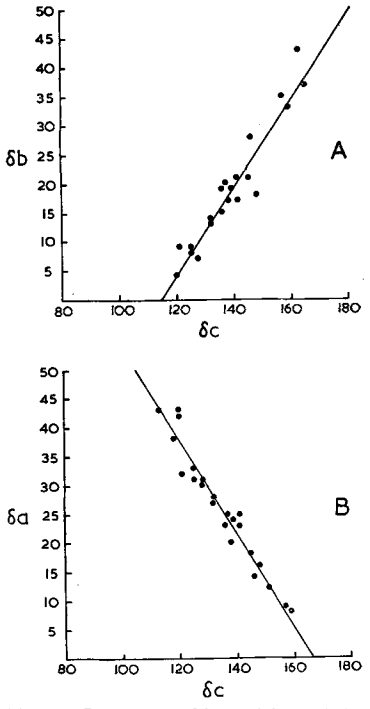

FIG. 1. Variation of δ_a and δ_b with δ_c.

The preceding paragraphs establish the changes that occur in the diffraction patterns of low-temperature series of plagioclases between about 20–25 % An and 70–75 % An. At the more basic limit there

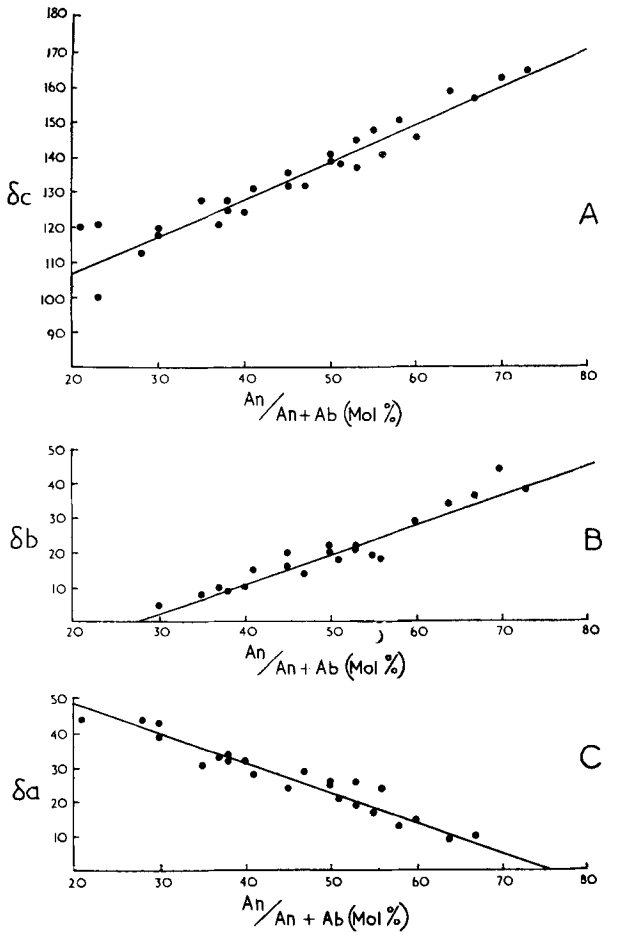

FIG. 2. Variation of δ_a, δ_b, and δ_c with chemical composition.

appears to be a sharp termination to the normal intermediate pattern somewhere in the region between 70–75 % An, after which it is apparently replaced by a body-centred anorthite pattern with single type (*b*) reflections. As the albite content increases from this limit the mutually dependent separations of the subsidiary layer-lines vary linearly with

composition over the whole range without much change in the intensities of corresponding reflections. At about 40 % An the subsidiary reflections become diffuse, the degree of diffuseness increasing with albite content. At this end of the composition range there is no sharp limit except that provided around 20 % An by the occurrence of the unmixed structure of the peristerite type (Laves, 1954). For all specimens in this composition region not peristeritically unmixed which have been examined so far the diffuse intermediate reflections have been found.

An interesting feature of the curves shown in fig. 1 is that when $\delta_a = 0$, $\delta_c \sim 167°$, and when $\delta_b = 0$, $\delta_c \sim 115°$. If these two values of δ_c are interpolated on the appropriate curve in fig. 2, they correspond to An contents of approximately 28 % An and 75 % An, i.e. roughly the limits of the region over which the intermediate structure is found. It appears that for a stable intermediate structure the separations of the reciprocal points corresponding to the doubled subsidiary type (*b*) reflections parallel to the crystallographic axes must all be finite. Further discussion of the reciprocal lattice geometry of the intermediate patterns will be given in a later paper.

Specimens 3, 4, 6, 8, 9, 11, 12, 13, 15, 16, 17, 20, 21, 26, 27, 29, 32, 33, 35, 36, 37, 38, 39, and 40 show patterns which are consistent with the normal behaviour outlined above. The diffraction patterns of specimens that do not conform to the scheme described above are dealt with next.

One group of specimens (nos. 19, 22, 24, 28, 30, 31, and 34) shows no subsidiary type (*b*) reflections; only the principal reflections are present and the pattern corresponds to that from an albite-type structure. From the results of the heat-treatment experiments to be described in another paper, such patterns are known to be indicative of a high-temperature structural state. The optical properties of some of the specimens mentioned above (those from Beaver Bay, Linosa, and Esterel) have been determined by universal-stage methods by Muir (1955). He considers that the felspars from Beaver Bay and Esterel show transitional optics lying between the low- and high-temperature values; the optical properties may be changed by heating until they are nearly the same as those of the ultimate high-temperature state. This is quite consistent with the present interpretation since it is to be expected that the high-temperature structural state (with the albite pattern), as distinct from the low-temperature state (with the normal intermediate pattern), can exist over a wide temperature range. The felspar from Linosa was selected by Köhler (1941) as one of his standard high-temperature specimens.

The patterns from specimens 5 and 7 are rather similar; the split type (*b*) reflections are not present. On heavily exposed photographs there is a weak diffuse region of blackening midway between the principal *c*-axis layer-lines; there is no apparent tendency for a separation into two discrete reflections, and the spots are like very diffuse odd layer-line reflections from body-centred anorthite. These patterns have been commented on before (Cole *et al.*, 1951; Gay and Taylor, 1953). Formerly it was thought that the material from which specimen 5 was taken showed no subsidiary reflections, but a very careful examination has shown that its pattern is similar to that of 7 in showing the single diffuse subsidiary reflections. Undoubtedly these anomalous patterns are due to some special feature of the environment of these specimens, and the patterns are similar to those found for a nearly high-temperature specimen obtained by heating a low-temperature material. The optical properties of the St. John's Point material have been commented on by Muir (1955), who described it as having transitional optics.

Another type of anomalous pattern is that which is given by some of the more basic specimens, in which the diffuse type (*c*) reflections occur. Specimen 2 shows a body-centred anorthite pattern (which is not surprising since the bulk composition is in the border region) plus diffuse type (*c*) reflections and is thus the D pattern of anorthite (Gay, 1953). Specimen 1 shows the type (*c*) reflections occurring together with the split type (*b*) reflections. In a previous publication (Gay, 1953) bytownites containing approximately 70 % An and 80 % An were examined and it was found that type (*c*) reflections were absent; it was thought that specimens containing more than about 15–20 % Ab did not show these reflections. More recently Laves and Goldsmith (1954) have described specimens containing up to as much as 27 % Ab in which these reflections are still present. In two of their specimens the type (*b*) reflections were present and single, whilst in a third these reflections were split with a separation of $\delta_c \sim 167°$. This latter pattern is similar to that which is found for specimen 1. It is possible that specimen 10 shows a similar pattern, though for this specimen the diffuse reflections are so weak that it is impossible to decide with certainty the class to which they should be allocated. It seems probable that the diffuse regions on these photographs must be due to some other cause, since, as stated by Laves and Goldsmith, the (*c*) reflections for specimens between 70–75 % An are on the limit of observation.

The specimens from the layered series of New Amalfi and Skaergaard

are particularly interesting. For the New Amalfi sheet, the pattern from the most basic specimen is quite normal. The next specimen (no. 14) shows an almost normal intermediate pattern appropriate to its composition; however, if a careful examination is made it is found that when the subsidiary reflections are compared with those given by specimens from different localities but with similar compositions, they seem to be rather weaker than expected. The third New Amalfi plagioclase (no. 18) shows only a few very feeble subsidiary reflections, which, however, are present in the normal positions on the *c*-axis photographs. This suggests that the three specimens show evidence for a change of structural condition in the series, changing from a normal low-temperature material (no. 11) to a partially inverted state, which is almost that of the high-temperature structural form, for the most sodic specimen examined (no. 18). Muir (1955) has examined the optical properties of these three specimens and has reached similar conclusions; he has also been able to relate satisfactorily the state of the plagioclase to the exsolution and inversion of the associated pyroxenes. Specimens from the layered series of the Skaergaard intrusion show a similar trend. Specimens 12, 15, 20, and 21 show quite normal low-temperature patterns, but in those from specimens 23 and 25 there is a weakening of the intensities of the subsidiary reflections; the still more sodic specimens (nos. 28, 30) appear to have gone over completely into the high-temperature structural state. A complete account of X-ray and optical studies of these and other plagioclases from the Skaergaard intrusion and their relationships to the associated pyroxenes is in preparation by Muir and the present author. One immediate comment that can be made is that, although these layered series provide an extremely fine variation of felspar composition over a particular range, caution must be exercised if specimens from these (and possibly other) intrusions are to be used as samples for the determination of standard plagioclase properties.

One small anomaly which can occur in the diffraction patterns is worthy of note although it is not of fundamental importance. If the fragment used is strongly zoned, the subsidiary reflections are found to have 'tails'. The spots are present in positions corresponding to the composition of the main body of the crystal, but the tails associated with these spots show the zoning.

On some photographs, reflections not coming within the description above are seen; these will be discussed at the appropriate point in the next section, where a comparison with previous work will be made.

Comparison with previous work.

The earliest X-ray work on the intermediate plagioclases (Taylor, Darbyshire, and Strunz, 1934; Chao and Taylor, 1940) described the diffraction pattern; Chao and Taylor suggested a model by which the main characteristics of the diffraction effects could be explained. In both cases, however, only a small number of specimens was examined. A full account of the intermediate pattern was given by Cole *et al.* (1951) and Sörum (1951), using a suite of felspars provided by Professor R. C. Emmons. It is interesting to compare this later work with that described here, particularly since there are several points of difference, which will be discussed here. I am indebted to Dr. W. H. Taylor and H. Sörum for making available to me photographs taken during this previous investigation for re-examination in the light of the present work.

One of the principal points at issue concerns the variations of the separations δ_a, δ_b, and δ_c with chemical composition. It should be stressed that the magnitudes of δ_a, δ_b, and δ_c were not all recorded for all specimens by Cole *et al.*, and hence their data were less complete than those recorded here. For plagioclases containing more than 50 % An there is good agreement. For specimens with less than 50 % An Cole *et al.* tentatively suggested that the variations take the form of straight lines symmetrically about 50 % An with respect to the linear variations for the more basic specimens. Some evidence for this suggestion was provided by data from the three specimens they examined in this region. The present investigations show that this suggestion is incorrect and that the variations are linear (as far as it is possible to determine) over the whole range. It remains to explain the occurrence of the three points below 50 % An plotted by Cole *et al.* For two of these, both from the same material from Emmons's suite (no. 16 in the present work), there is no difficulty. The values recorded for one of these two (sample 4, specimen 1, Cole *et al.* numbering) are substantially in agreement with those found for a representative specimen from the same sample by the author. For the second specimen from the same sample, the author has remeasured the original photographs with the result that the previously recorded values of δ_c and δ_a lie within the standard deviation of the new measurements; they are very little different from those of the typical specimen. However, the composition recorded by Cole *et al.* for this specimen was derived from measurements of extinction angles on the cleavage faces, which are quoted as $\sim 10°$

on (010) and 2·9° on (001). Apart from any question of the accuracy of these measurements, these angles lead to compositions ~ 43 % An and ~ 35 % An, using the extinction-angle curves of Rogers and Kerr (1942) (which were used by Cole *et al.*). There is clearly some confusion or inaccuracy in the optical measurements of Cole *et al.*, and their observations on this material should be discarded.

Such considerations cannot be applied to the third specimen (sample no. 11, specimen no. 1, Cole *et al.* numbering). Several specimens of material from the same source (no. 31) have been examined and no trace of the normal intermediate reflections, which would have become a little diffuse in this composition range, has been found. This suggests that the felspar is in the high-temperature structural state. Some confirmation of this is given: by its occurrence in a dacite (a rock type in which high-temperature optics have been found (van der Kaaden, 1951); and by the work of Smith (1956) who has found that the reciprocal lattice parameters for material from the same source are intermediate between the curves of reciprocal lattice parameters for felspars from low-temperature sources (pegmatites) and synthetic felspars. It seems likely, therefore, that the few exceedingly weak reflections with $\delta_c \sim 160°$ reported for this specimen by Cole *et al.* cannot be the normal intermediate subsidiary reflections but must arise from some secondary cause; hence this point also must be disregarded.

Other specimens in this sodic region used in the present work which have been mentioned in other X-ray investigations are nos. 26 and 32. These have been examined by Cole *et al.* (numbered sample 19, specimen no. 2, and sample 21, specimen no. 1) and by Laves (1954) (nos. 16 and 14); both these investigations recorded that the patterns showed a homogeneous felspar. Both the specimens examined by the author showed the normal intermediate pattern for this composition range, i.e. one in which the type (*b*) reflections are beginning to become diffuse. It seems probable that these reflections were not observed either by Laves or by Cole *et al.* owing to insufficient exposure times. Cole *et al.* noted that *c*-axis photographs of their specimen numbered 19 showed reflections 'off the principal layer-lines, but not in positions corresponding to pairs of subsidiary layer-lines as in other intermediate plagioclases'. On re-examination, one of these original photographs shows very faintly the normal intermediate reflections; it is not clear if these were the reflections referred to by Cole *et al.*

Some specimens in the albite-oligoclase region (including some of the Emmons specimens common to both Laves and Cole *et al.*) have been

examined. In agreement with Laves, it is found that these specimens are unmixed into two phases; often, however, the detection of the unmixing is very difficult when using only oscillation methods. Re-examination of the photographs taken by Cole *et al.* of their specimens numbered 3, 9, and 22 shows that the unmixing is detectable in nos. 3 and 22. Laves notes that specimen no. 9 has 'frozen' in the process of unmixing, which probably accounts for the apparent homogeneity of the oscillation photographs.

Turning to the more basic specimens, in which diffuse type (*c*) reflections occur, re-examination of the Cole *et al.* photographs shows that they can be observed on those from specimens nos. 23 and 25 (Cole *et al.* numbering). On the pattern from no. 23 they are present but very weak, with sharp single type (*b*) subsidiary reflections; on the pattern from no. 25 they are present with doubled type (*b*) subsidiary reflections (cf. description of pattern from specimen no. 1, present investigation; this material is listed by Cole *et al.* (no. 24), but no description of the pattern is given). Material from specimen no. 25 was also used by Laves and Goldsmith (1954), who describe the occurrence of type (*c*) reflections with single type (*b*) reflections. These authors also examined materials from samples nos. 1 and 2 (Cole *et al.* numbering) and report that for specimen no. 1 they could observe type (*c*) reflections after extremely heavy exposures, whilst under similar conditions these were not shown by no. 2. These reflections are not visible on any of the photographs taken by Cole *et al.* of specimens from these materials, but this is probably due to lack of exposure.

In the course of the present work it has been necessary to take many very heavily exposed photographs. On some of these photographs extremely weak reflections not of the normal subsidiary type have been observed. It appears likely that these reflections are associated with some secondary cause due to the plagioclase structure itself, and cannot be ascribed to some external factor such as twinning, imperfect single crystal, &c. On some photographs the additional subsidiary reflections, in pairs close to and symmetrical about the principal reflections, that were mentioned by Cole *et al.* have been observed. Further, quite often associated with the strongest of the principal reflections are found diffuse 'tails' along lines of constant θ. These secondary effects have been ignored in the present study, which sought to deal with the main subsidiary reflections; some light may be thrown upon their occurrence when a satisfactory explanation of the main subsidiary reflections has been devised.

Summary and conclusions.

Over a large composition range the low-temperature plagioclases are characterized by the normal intermediate pattern. In such patterns, in addition to the main type (*a*) reflections, subsidiary reflections, often occurring in pairs, replace the single subsidiary type (*b*) reflections found in basic plagioclases. The angular separations of these subsidiary reflections vary linearly with composition over the whole composition range. At the basic limit ($\sim$ 70–75 % An) δ_a is approximately zero, whilst the sodic limit ($\sim$ 20–25 % An) appears to occur at the composition for which δ_b becomes zero; more sodic specimens show peristeritic unmixing. The subsidiary reflections are relatively sharp over the region from 70 % An to 40 % An; rough estimates of the intensities of corresponding reflections show that they change very little (if at all) over this range. Within the range from about 40 % An to 20–25 % An the subsidiary reflections become more diffuse the more sodic the felspar.

It is possible to use the separation of the subsidiary reflections to determine the plagioclase composition. Probably the δ_c variation would be the most suitable to use, although the *c*-axis is usually the most difficult to locate. On specimens between 70 % An and 40 % An it is probable that with careful work δ_c could be determined to $\pm 1°$; using the present curve this would lead to an accuracy of about ± 1 %. This is probably better than all but the most careful optical work. It should be realized, however, that in such determinations any other factors (e.g. the orthoclase content of the plagioclase) which may affect the positions of the subsidiary reflections are not taken into account. Below 40 % An, where the reflections become diffuse, it would be considerably less accurate than this. It seems hardly likely that this method would be of much petrological use, since although it offers greater accuracy than routine optical measurements it is rather more difficult to carry out and of necessity is performed on only one crystal. It should be noted, however, that the method is independent of the state of inversion of the felspar provided, of course, that the subsidiary reflections can still be observed; this will be fully dealt with in a later publication.

Whilst the majority of the specimens examined showed the normal intermediate pattern, other specimens give anomalous patterns, which may be ascribed to the different structural conditions of the felspars, arising probably from their particular geological environments; some supporting optical data have been quoted and further evidence will be given in a later paper on the heat treatment of these felspars.

Low-temperature plagioclase felspars appear to show a very limited region of existence for the homogeneous low-albite structure, probably over a composition range from An_0 to An_{1-5}. All low-temperature specimens (i.e. the completely stable structural forms) lying in composition between An_{1-5} and An_{70-75} are unmixed. Those between An_{1-5} and An_{20-25} are unmixed into soda-rich and calcium-rich components,

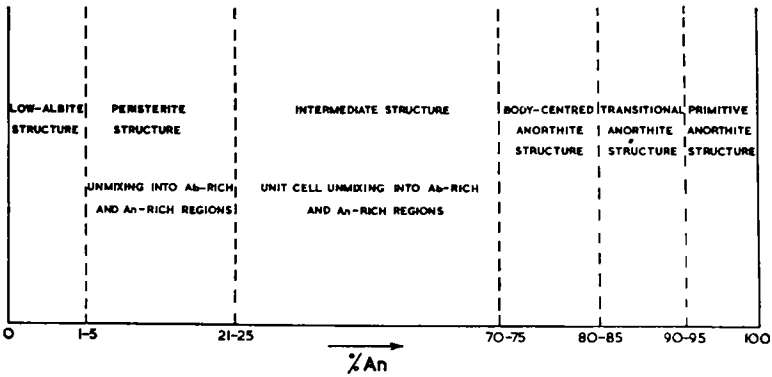

FIG. 3. Schematic representation of the structural types distinguished in low-temperature plagioclase felspars.

usually on a sub-microscopic scale. Nevertheless, the X-ray patterns from the two phases can be detected (Laves, 1954; Gay and Smith, 1955). If the explanation of the intermediate diffraction patterns in terms of an ordered superlattice of soda-rich and calcium-rich regions is accepted, it may be said that the intermediate structures between An_{20-25} and An_{70-75} are unmixed; for these, however, the unmixing is on a unit-cell scale, and is of a different character from that in the albite-oligoclase region. Bytownites and anorthites in the low-temperature state can show the body-centred, transitional, and primitive anorthite arrangements depending apparently on their composition. A schematic representation of the probable composition ranges of the different structural modifications distinguished in low-temperature plagioclases is shown in fig. 3.

The calcium-rich phase of unmixed albite-oligoclase is of interest, for so far it has only been detected as a component phase in two-phase specimens. All specimens which have been examined in the 15–30 % An region have shown either the normal intermediate pattern or the dual pattern of a two-phase specimen. This suggests that the peristerite and

intermediate structures occupy adjacent fields in the An–Ab series, and that the calcium-rich component of the peristerites cannot form a relatively large homogeneous crystal. It seems likely that the calcium-rich and soda-rich regions in the normally accepted model of the intermediate structure can be identified with body-centred anorthite (Sörum, 1951) and the calcium-rich component of the unmixed albite-oligoclases. For low-temperature materials the region of stable separate existence of body-centred anorthite is in doubt since it now appears that the transitional anorthite D pattern occurs for specimens more sodic than was suspected earlier. Traces of the diffuse type (*c*) reflections characteristic of the transitional anorthite structure have been found on intermediate patterns, suggesting that the intermediate and transitional anorthite structures may have a field of coexistence.

Further work is proceeding to elucidate the changes occurring at the boundaries of the intermediate structure and to establish the relationships between the two two-phase regions, together with an examination of very sodic specimens. Succeeding publications will describe the heat treatment of the intermediate plagioclases and the structural significance of the present work.

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