Zeitschrift: Schweizerische mineralogische und petrographische Mitteilungen = Bulletin suisse de minéralogie et pétrographie

Band: 47 (1967)

Heft: 1: Feldspäte

Artikel: Schiller and submicroscopic lamellae of labradorite : a preliminary report

Autor: Nissen, H.-U. / Eggmann, H. / Laves, F.

DOI: https://doi.org/10.5169/seals-36953

Schiller and Submicroscopic Lamellae of Labradorite

A Preliminary Report

By *H.-U. Nissen**), *H. Eggmann***) and *F. Laves**)

With 6 figures in the text

Abstract

In order to explain the nature of the lamellae (ca. 1000—2500 Å periodicity) giving rise to the "schiller" of labradorites, the composition, x-ray data and electron microscopic observations on specimens with schiller are contrasted with those lacking schiller. Schiller occurs in plagioclases with compositions from An_{48} to An_{58}, but similar lamellae were also found in andesines. All specimens with schiller have over ca. 2 mol % Or content, as do also specimens containing (at least partly) similar lamellae, with spacings too small to produce visible schiller. The spacing of the lamellae increases with An content. The structural state ("low" or "intermediate") has an influence on the lamellar spacing, and the occurrence of schiller is not restricted to the "low" structural state. The two alternating kinds of lamellae may differ in their Ca/Na proportion or, more likely, in their K content and additionally in their Al/Si arrangement. — In spite of the lamellar inhomogeneities only one set of diffraction spots is present in x-ray and electron diffraction diagrams, indicating homogeneous lattice constants of the substance.

Zusammenfassung

Um die Beschaffenheit der Lamellen (Periodizität ca. 1000—2500 Å) zu erklären, die den „Schiller" der Labradorite bedingen, werden Zusammensetzung, Röntgendaten und elektronenmikroskopische Beobachtungen von schillernden Stücken mit solchen ohne Schiller verglichen. Der Schiller ist auf die Zusammensetzungen von An_{48-58} beschränkt; indessen wurden ähnliche Lamellen auch in

---

*) Institut für Kristallographie und Petrographie, Eidg. Technische Hochschule, CH-8006 Zürich, Switzerland.

**) Institut für Allg. Botanik, Eidg. Technische Hochschule, CH-8006 Zürich, Switzerland.

Leere Seite
Blank page
Page vide

(near Ylijärvi, Finland), the area around Paul Island (Labrador, Canada) and the Tamatave area (Madagaskar); all these are old anorthositic complexes.

The approximate chemical composition of feldspars in this composition range may be determined by analysis of K and Ca. Measurements with unusually high K contents, indicating antiperthitic inclusions, occurred in all specimens and were eliminated from the microanalyser measurements before calculation of the mean compositions. Such inclusions could be observed with a high power light microscope in all specimens used for single crystal x-ray studies and could also be found, with one exception, in all precession photographs exposed for long periods. Due to the validity of Bragg's law for the case of white light reflected by the lamellar inhomogeneities (LAVES et al., 1965) the spacing of submicroscopic lamellae is roughly indicated by the schiller colour (see p. 292). X-ray powder and precession photographs were therefore made of crystal fragments showing only one schiller colour. For the powder photographs an AEG Guinier camera after Jagodzinski was used. Fragments for replica and transmission photographs were broken off the specimens as near as possible to those taken for x-ray examination. This was necessary because of the inhomogeneity of specimens as indicated by the changes in the average schiller colour.

Differences in $4\,\theta$ of the powder line pairs $(131—1\bar{3}1)$ and $(\bar{2}41—24\bar{1})$ were measured in the powder films. For a number of samples the lattice constants were calculated with the aid of the least squares lattice constants refinement program of C. Burnham, using the first 20—30 powder line spacings as input. The chemical composition was determined with an ARL electron probe microanalyser using a $15\,\mu$ beam size and synthetic plagioclase glasses as standards. Additional overall wet chemical analyses kindly supplied by Prof. Weibel often gave an Or content 1—3 mol-% higher than the microprobe data; this was probably due to the antiperthitic inclusions.

For electron-microscopic observation the samples were cut parallel to (001), i. e. approximately normal to the "schiller lamellae". This face was then etched 45 seconds in a 1% aqueous solution of HF. Technovit replicas were made immediately after drying; these were coated under vacuum with chromium (evaporation angle 45°), and afterwards a carbon layer was evaporated normal to the specimen surface. The replicas were then washed in acetone and transferred on uncoated grids. Transmission samples were crushed in an agate mortar and floated with an alcohol water solution on the carbon net foil.

RESULTS

I. Chemical composition

The calculated proportions of K-feldspar (Or), Na-feldspar (Ab) and Ca-feldspar (An) are plotted in a section of an Or-Ab-An diagram in Fig. 1. In this diagram the following features are evident:

1. Though there is some overlapping, the wavelength of the average schiller colour increases with increasing Ca content: "blue" samples vary from $An_{48.5}$ to An_{52}, "green to yellow" samples from An_{52} to $An_{55.5}$ and "orange to red" samples from An_{55} to $An_{58.5}$ (possibly $An_{59.5}$). This relation, which may be termed "Bøggild's rule", is also evident when Bøggild's optical measurements (BØGGILD, 1924) are used to determine the composition of his samples using van der Kaaden's determination graphs (TRÖGER, 1952). The averages found are: "Blue" An_{53}; "green to red" An_{55}. Bøggild's rule has been confirmed by LAVES et al. (1965) and by MELLIS and NILSSON (1965).

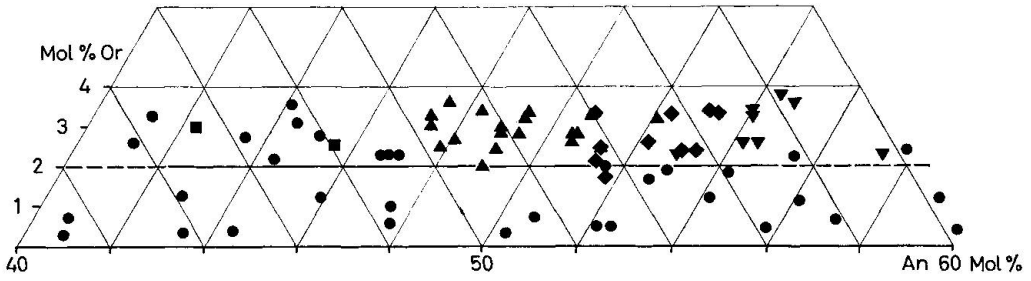


Fig. 1. Section of a K-Na-Ca feldspar triangle showing composition of specimens. Dots = specimens without schiller; squares = specimens with lamellae too densely spaced to show visible schiller ("UV-schiller"); triangles = violet to blue schiller; rhombs = green to yellow schiller; inverted triangles = orange to redbrown or redviolet schiller.

2. The Or content lies above 1.6 mol-% for all specimens with schiller. All specimens within this Ca/Na composition range which lack schiller have Or contents below 2.1 mol-%. The compositions in Fig. 1 are those of the plagioclase matrix.

3. From the diagram (Fig. 1) it appears possible that the specimens outside the schiller composition range and with more than 1.6 mol-% Or may also have lamellae of the labradorite type but with spacings too large or too small to produce *visible* schiller.

II. X-ray powder data

In Fig. 2 the powder line difference[1]) $\Delta\,\theta_{(131-1\bar{3}1)}$ is plotted against An content. It appears probable from this diagram that the degree of Al/Si ordering, as possibly indicated by $\Delta\,\theta_{(131-1\bar{3}1)}$, slightly influences the average schiller colour and therefore the spacing of the lamellae: In specimens with the same An content the spacing of the lamellae appears to be statistically smaller if the $\Delta\,\theta_{(131-1\bar{3}1)}$ is larger.

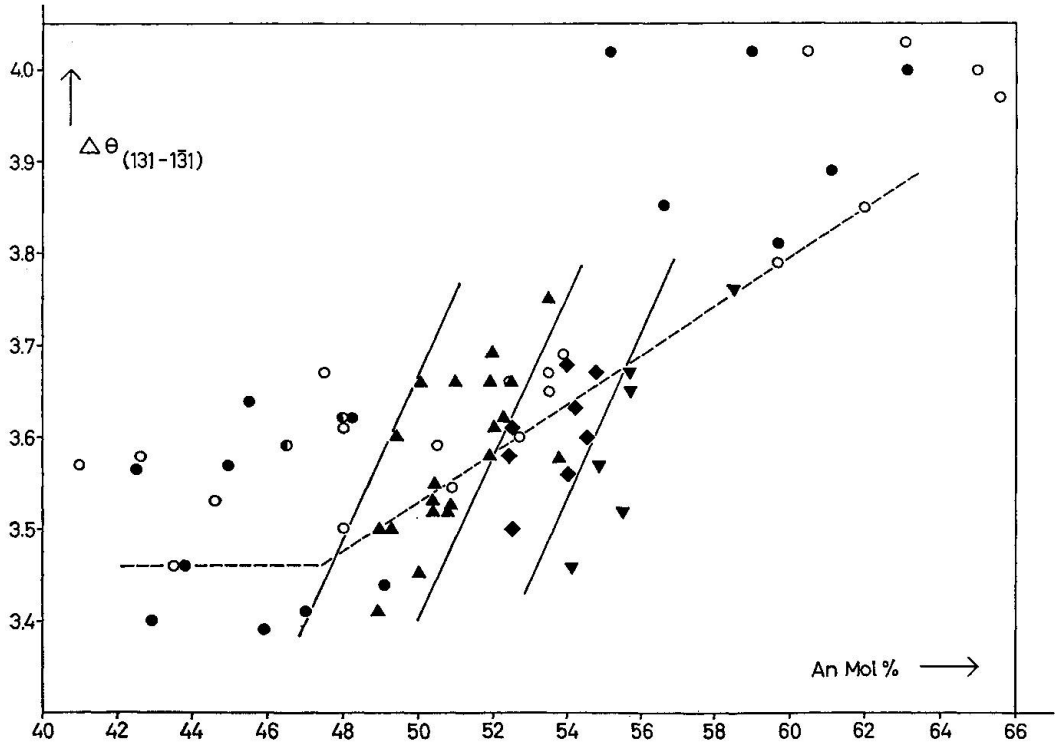


Fig. 2. Plot of $\Delta\,\theta_{(131-1\bar{3}1)}$ versus An content. Open dots = Plagioclase without schiller and <2 mol % Or content. Black dots = Plagioclase without schiller and >2 mol % Or. Other symbols as in Fig. 1. Dotted line connects minimum values of $\Delta\,\theta_{(131-1\bar{3}1)}$ for specimens with <2 mol % Or. Full lines denote approximate areas in the diagram with the same schiller colour. — Cu $K_{\alpha 1}$-radiation; camera diameter D = 114.59 mm.

Another feature appearing in Fig. 2 can be attributed to the influence of K substituting for Na: Since the values represented by the dotted line in Fig. 2 most probably represent a state of maximum Al/Si

---

[1]) The following definitions are used: $\Delta\,\theta_{(131-1\bar{3}1)} = 4\,\theta_{(131)} - 4\,\theta_{(1\bar{3}1)}$ [in degrees]; $\Delta\,\theta_{(\bar{2}41-24\bar{1})} = 4\,\theta_{(\bar{2}41)} - 4\,\theta_{(24\bar{1})}$ [in degrees].

order, $\Delta\,\theta_{(131-1\bar{3}1)}$ appears to be slightly lowered towards the corresponding values for microcline due to the high amount of K substituting for Na in specimens with schiller. Indeed, the majority of the schiller specimens fall below this curve and the largest deviations from the curve are found for crystals with very high homogeneous Or content. In a plot of $\Delta\,\theta_{(131-1\bar{3}1)}$ against $\Delta\,\theta_{(\bar{2}41-24\bar{1})}$ (Fig. 3) the specimens with Or contents over ca. 2 mol-% showing schiller and those with somewhat higher or lower An content but also "elevated" Or component deviate significantly from all other plagioclases, which show an approximately linear spread in the diagram Fig. 3. However, the specimens with elevated Or

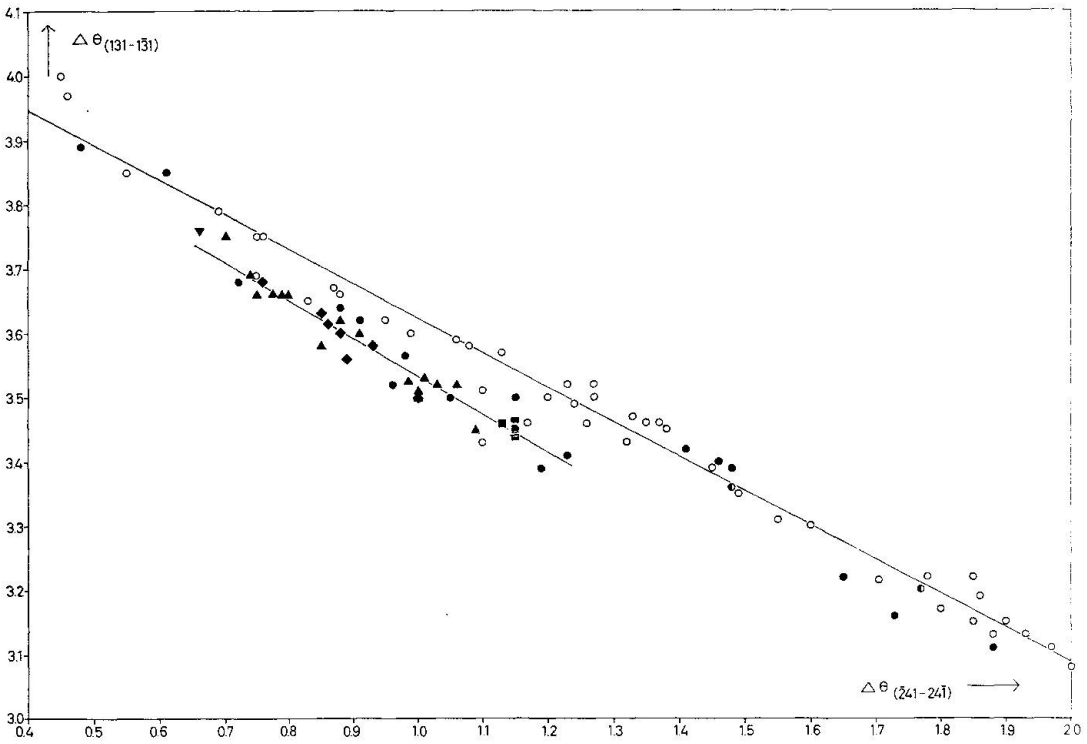


Fig. 3. Plot of $\Delta\,\theta_{(131-1\bar{3}1)}$ versus $\Delta\,\theta_{(\bar{2}41-24\bar{1})}$. For explanation of symbols see Figs. 1 and 2. — Cu $K_{\alpha 1}$-radiation, camera diameter D = 114.59 mm.

content in the bytownite and oligoclase range do only in part show a similar shift from the normal values. This may indicate an abnormality in the lattice geometry of the specimens with "schiller lamellae" which is only in part due to the influence of the substituting K.

Table 1 shows representative lattice constants of labradorites with blue and with red schiller as compared with a specimen of corresponding composition but without schiller. Significant irregularities of γ^* values in labradorites were already found by Brown (1960, Fig. 5). He also

Table 1. *Lattice constants of labradorites*

	S 429 blue schiller $An_{52.5}$ $Or_{2.5}$	S 1411 orangered schiller $An_{55.5}$ $Or_{2.6}$	Labradorite without schiller An_{56} (COLE et al., 1951)
a	8.163 ± 0.019 Å	8.181 ± 0.007 Å	8.1353 Å
b	12.856 ± 0.007 Å	12.870 ± 0.005 Å	12.7883 Å
c	7.105 ± 0.009 Å	7.114 ± 0.005 Å	7.1542 Å
a*	0.1366 ± 0.0003 $Å^{-1}$	0.13628 ± 0.00009 $Å^{-1}$	0.13643 $Å^{-1}$
b*	0.07796 ± 0.00005 $Å^{-1}$	0.07788 ± 0.00003 $Å^{-1}$	0.07795 $Å^{-1}$
c*	0.1572 ± 0.0001 $Å^{-1}$	0.15703 ± 0.00006 $Å^{-1}$	0.15725 $Å^{-1}$
α	93.52° ± 0.05°	93.59° ± 0.03°	94.23°
β	116.18° ± 0.09°	116.21° ± 0.04°	116.52°
γ	89.80° ± 0.06°	89.80° ± 0.03°	89.71°
α^*	86.17° ± 0.05°	86.10° ± 0.04°	86.14°
β^*	63.78° ± 0.09°	63.75° ± 0.04°	63.86°
γ^*	88.49° ± 0.06°	88.46° ± 0.04°	88.49°

reported a range of composition near that of labradorite in which a line connecting γ^* values for plagioclases in the low structural state is approximately horizontal, i. e. differs in inclination when compared with other compositional ranges.

III. Electron microscopy

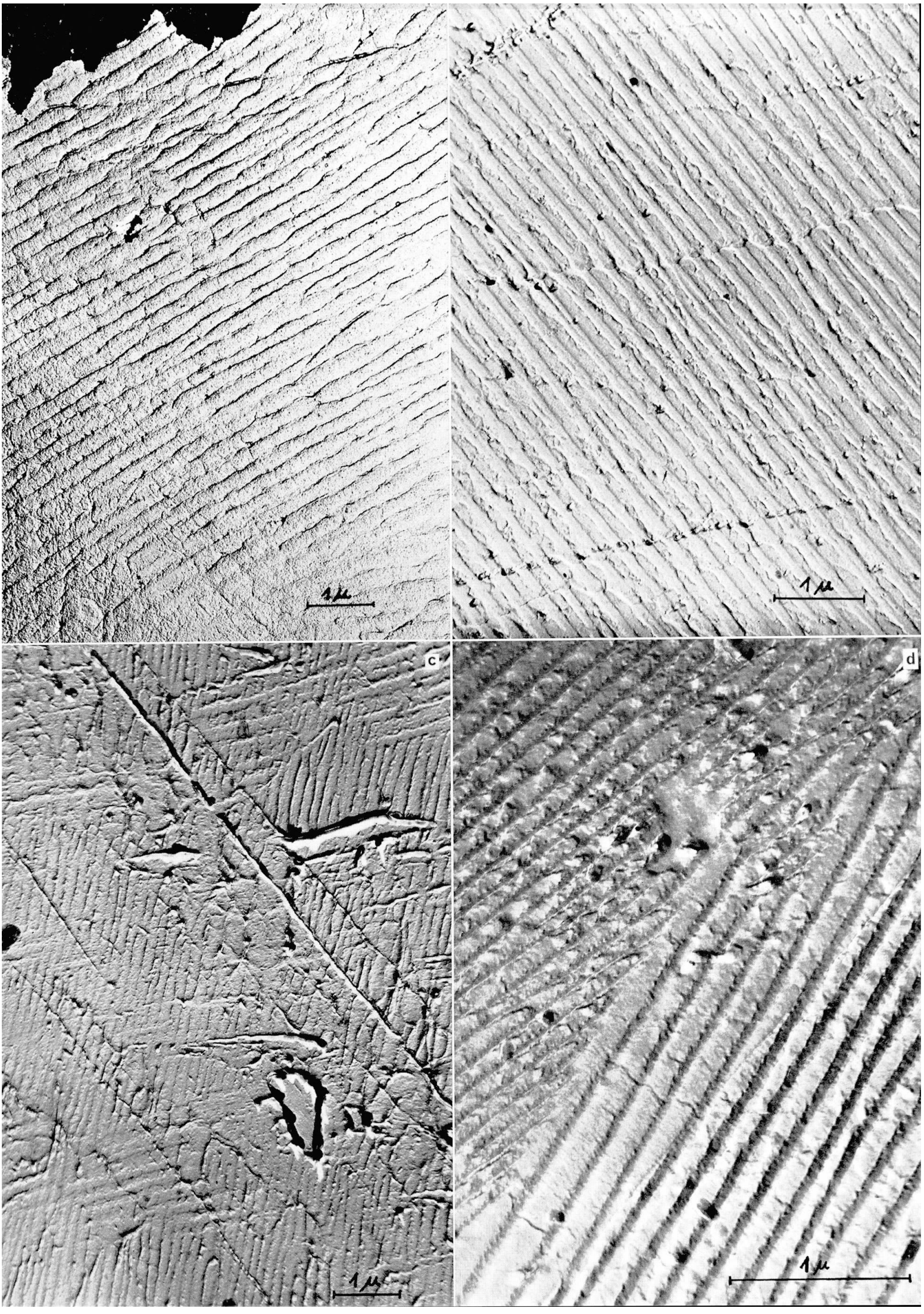
The lamellae responsible for the schiller can be seen in the electron microscope using either transmission preparations or etched replicas. Once the lamellar nature had been verified with both methods[2]) (LAVES et al., 1965) mainly replicas were made; this method has the advantage that planes normal to the lamellae can be used for replication.

Fig. 4, a—d shows a selection of replica electron micrographs from which the following observations can be made:

1. The labradorites consist of two kinds of lamellae, A and B, one of which is more deeply etched by the HF solution (Fig. 4, a—b).

2. There is a linear relationship between the An content and the lamellar periodicity (a + b), where a and b are the average thicknesses of the two kinds of lamellae (Fig. 5, cf. LAVES et al., 1965).

[2]) Further work on this problem is necessary and planned.


1 μ
1 μ
c
1 μ
d
1 μ

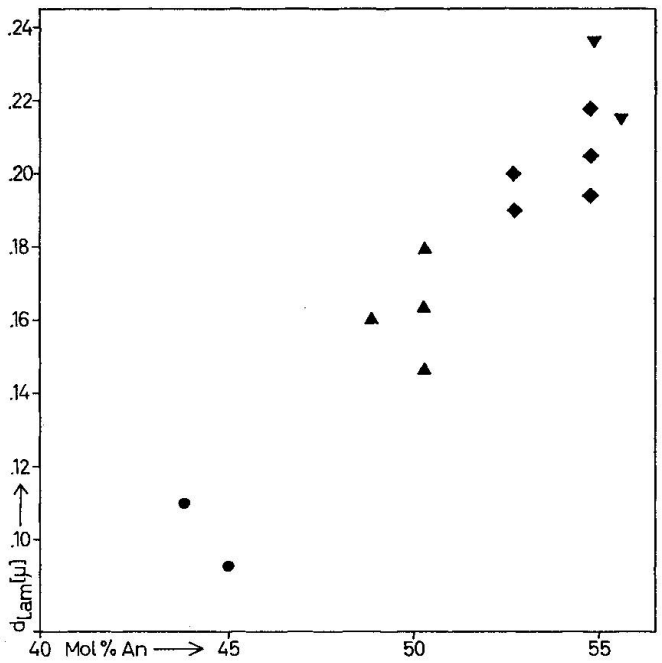


Fig. 5. Plot of lamellar periodicity (in μ) versus mol % An measured in replicas. Symbols as in Fig. 1.

3. The periodicity (a + b) varies within the field of an electron micrograph (Fig. 4d). This is compatible with the broad maxima in the diagrams of intensity versus λ of the schiller (SCHRÖDER, 1964). Frequently two lamellae unite and, near the bifurcation, show especially strong thickness variations.

4. The proportion a/b may vary strongly within single specimens or even areas of one replica preparation. However, if different specimens are compared, a regular change of a/b with An content is evident as reported earlier (LAVES et al., 1965).

5. Occasionally two sets of lamellae with different spacing and lamellar proportion a/b are seen on replicas (Fig. 4c). Similarly, KOREKAWA (1966, personal communication) has found in one specimen "supersatellites" corresponding to two systems of lamellar inhomogeneities (see also JAGODZINSKI and KOREKAWA, 1965), and BØGGILD (1924) described

---

Fig. 4. Replica electron micrographs of labradorites with schiller. a: blue schiller; b: red schiller, both from Labrador. c: two lamellar systems and (?) crypto-antiperthitic exsolutions forming a third system of lines. Blue schiller, Madagaskar. d: boundary of small twin. Green schiller, Ylämaa, Finland. Note irregularities of twin boundary and apparent subunits of schiller lamellae.

three different positions of "schiller planes" (Fig. 6), two of which may occur in the same specimen.

6. Frequently extremly fine twins are indicated by a sudden change in the position of the lamellae (Fig. 4d). The angles between the lamellae and the trace of the twin as seen in the replicas are compatible with twinning after the albite law, which is also found in most of the precession photographs.

The "schiller lamellae" are frequently seen in transmission samples because most cleavage fragments settle with (001) normal or nearly normal to the electron beam, so that the lamellae are parallel to it. All low structural state plagioclases investigated in the composition range of the schillering specimens with an Or content less than 1.6 mol-% showed no such regular lamellae in either transmission or replica electron micrographs. However, some anorthosite bytownites (ca. An_{75}) showed regular lamellae in transmission samples with spacings over 0.6 μ and the lamellae found in bytownites from amphibolites of the Valle d'Ossola (JÄGER and HUTTENLOCHER, 1955) having An_{76} composition seem to represent a

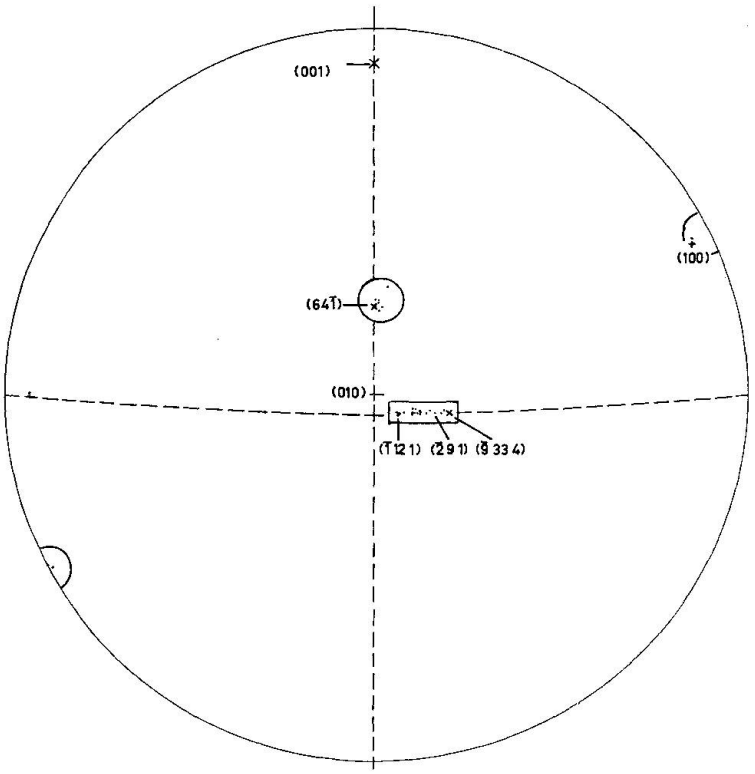


Fig. 6. Position of reflection planes of the schiller according to BØGGILD (1924) Note three different positions.

similar texture [3]). Lamellae similar to "schiller lamellae" were also found by transmission technique in an andesine $An_{43.8}$.

IV. Heating experiments

A large number of specimens with various schiller colours were heated up to 70 days at 1160°C. Many specimens became milky or developed dark patches originating from the inclusions of iron-rich minerals, but the schiller remained equal in colour and visually estimated intensity. The powder data of the material taken out of the oven after 20, 40, 60 and 70 days showed a gradual change to an intermediate structural state, as described by GAY and BOWN (1956).

A labradorite with blue schiller was heated in the heating stage up to ca. 1200°C and no change of visually estimated intensity or colour of the schiller could be observed during heating[4]).

(0kl) precession photographs of a labradorite with schiller were taken at temperatures between 600°C and 1100°C using the equipment described by GUBSER et al. (1963). The "b-split" reflections — probably indicating the presence of antiphase domains (MCCONNEL and FLEET, 1963) — remained essentially unchanged over this entire temperature range.

DISCUSSION OF RESULTS

On the basis of the features described above the intermediate plagioclases may be separated into different groups taking into account the presence of "b-split" reflections, the Or component, the structural state and the presence or absence of lamellae in the 1000 Å order of magnitude:

1. Crystals in the "low" structural state, with < 2 mol-% Or, with "b-split" satellites indicating the presence of antiphase domains. They do not appear to have "schiller lamellae".

2. Crystals in the "low" or "intermediate" structural state, with > 2 mol-% Or, with "b-split" satellites and with "schiller lamellae". Below ca. An_{48} or above ca. An_{58} these lamellae will have too small or too large periodicities to produce visible schiller.

3. ("Intermediate" to) "high" structural state crystals with varying

---

[3]) Further work on these materials is in progress.

[4]) This experiment was executed together with Dr. Korekawa and Mr. Czank.

Or content, lacking "b-split" satellites. No lamellae in the 1000 Å range have as yet been found.

These different groups of crystals normally occur in different petrological surroundings. Their lattice constants show systematic differences and it is therefore principally possible to determine the approximate Or component of a plagioclase with x-ray data alone. — The two phenomena of antiphase domains (indicated by x-ray and electron diffraction satellites) and of lamellar domains in the 1000 Å range (indicated by schiller) do not appear to be directly connected, though it is not impossible that the antiphase fabric is restricted to only one of the two sets of lamellae.

Two extreme models for an explanation of the "schiller lamellae" in the intermediate plagioclases appear possible considering especially the influence of the K content:

1. The chemical Al/Si ratio is different in A and B, as implied by LAVES et al. (1965) (model 1).
2. The Al/Si ratio is not different in A and B (model 2).

In model 1, K may promote plagioclase "unmixing", while, in model 2, K may tend to develop a state which is a precursor towards an antiperthitic exsolution. In model 2, the Al/Si ordering state must be different in A and B because of the persistence of schiller on heating (p. 299). Both reactions assumed in these two models may in fact play a role in the actual formation of schiller lamellae.

Further chemical and electron-microscope work is planned to find supporting evidence in favour of one of the two models. At present, the authors consider the second one more probable.

The "schiller lamellae" are not submicroscopic twins: several fragments of labradorite with schiller could be found showing no trace of twinning in long-exposed precession photographs. Neither can they be "normal" exsolution lamellae, as in moonstones and peristerites, since their x-ray as well as electron diffractograms show only one set of diffraction spots.

Since submission of this paper, the authors obtained the paper "On the origin of the colour of labradorite" by BOLTON, BURSILL, MCLAREN and TURNER (phys. stat. sol. *18*, p. 221, 1966). Their (mainly optical) considerations are consistent with the present publication.

Acknowledgements

The microprobe analyses were made by Dr. M. Corlett and most kindly put to the authors' disposition, for which assistance they are very grateful. Miss Corlett also made valuable suggestions and read the manuscript. Many thanks are due to Mr. E. Schärli for help in preparation, to Dr. W. Hoffmann for collaboration in the high temperature precession study, to Mr. G. Brunner and Dr. W. Bollmann for helpful discussions and to Mr. R. Gubser for various experimental help. Mr. W. Villiger, Basle, kindly made some additional replica electron micrographs. — A gift of specimens by Dr. E. Gübelin and the Mineralogisches Institut der Universität Bern is gratefully acknowledged. — The work was made possible through the generosity of Professor K. Mühlethaler, Inst. für Allg. Botanik, who provided electron microscope facilities during the first stage of the work.

LITERATURE

Baier, E. and Pense, J. (1957): Elektronenmikroskopische Untersuchungen an Labradoren. Naturwiss. *44*, 110.

Bøggild, O. B. (1924): On the labradorisation of the feldspars. Kgl. Danske Vidensk. Selsk. Math.-fys. Medd. *6*, 1—79.

Brown, W. L. (1960): Lattice changes in heat-treated plagioclases. The existence of monalbite at room temperature. Z. Krist. *113*, 297—329.

Fleet, S. G. and Ribbe, P. H. (1962): An electron microscope investigation of a moonstone. Phil. Mag. *8*, 1179—1177.

Fleet, S. G. and Ribbe, P. H. (1964): An electron microscope study of peristerite plagioclases. Min. Mag. *35*, 165—176.

Gay, P. and Bown, M. G. (1956): The structures of the plagioclase feldspars: VII. The heat treatment of intermediate plagioclases. Min. Mag. *31*, 306—313.

Gubser, R., Hoffmann, W. und Nissen, H.-U. (1963): Röntgenaufnahmen mit der Buergerschen Präzessionskamera bei Temperaturen zwischen 1000° C und 2000° C. Z. Krist. *119*, 264—272.

Jäger, E. und Huttenlocher, H. (1955): Beobachtungen an basischen Plagioklasen der Ivrea-Zone. Schweiz. Min. Petr. Mitt. *35*, 199—207.

Jagodzinski, H. und Korekawa, M. (1965): Supersatelliten im Beugungsbild des Labradorits (Ca, Na) (Si, Al)$_2$O$_8$. Naturwiss. *52*, 640.

Kôzu, S. and Endô, Y. (1921): X-ray studies of adularia and moonstone, and the influence of temperature on the atomic arrangement of these minerals. Tôhoku Imp. Univ. Sci. Repts., 3. ser., *1*, 1—17.

Laves, F. (1952): Phase relations of the alkalifeldspars. II. J. Geol. *60*. 549—574.

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

Laves, F., Nissen, H.-U. and Bollmann, W. (1965): On schiller and submicroscopical lamellae of labradorite (Na, Ca) (Si, Al)$_4$O$_8$. Naturwiss. *52*, 427—428.

McConnel, J. D. C. and Fleet, S. G. (1963): Direct electron-optical resolution of antiphase domains in a silicate. Nature *199*, 586.

Mellis, O. und Nilsson, C. A. (1965): Der Labradorit von Ylämaa in Finnland. Manuscript „Kurzreferate für die 43. Jahrestagung der Deutschen Mineral. Ges.", p. 39—40.

Nissen, H.-U. and Bollmann, W. (1966): Submicroscopic fabrics in feldspars. VI. Int. Congr. Electron Microscopy, Kyoto, vol. I, p. 591—592.

Schröder, A. (1964): Die Schillerfläche am Labradorit. Naturwiss. *51*, 507.

Tröger, W. E. (1952): Tabellen zur optischen Bestimmung der gesteinsbildenden Minerale. 145 p., Stuttgart.

Manuscript received November 14th, 1966.