Relations between the Optic Axial Angle and Triclinicity of Potash Feldspars, and their Significance for the Definition of "Stable" and "Unstable" States of Alkali Feldspars

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With 8 figures in the text

Abstract

Several K-feldspar crystals from the Ticino area, Switzerland, were investigated with respect to the variation of their optic axial angle 2 V and their triclinicity Δ. It was found that sanidine (monoclinic) may have 2 V values as large as 63° within optic axial planes lying perpendicular to (010). This result is discussed in the light of the stability and instability of alkali feldspars as generated in nature during geological times.

Zusammenfassung

Mehrere Kalifeldspat-Kristalle aus dem Tessin, Schweiz, wurden untersucht in bezug auf die Variation ihrer optischen 2 V-Werte und ihrer strukturellen Δ-Werte. Es wurde gefunden, dass Sanidin (monoklin) 2 V-Werte bis zu 63° $\perp$ (010) haben kann. Dieses Resultat wird diskutiert im Hinblick auf das Verhalten stabiler und unstabiler Alkalifeldspat-Zustände, die in der Natur während geologischer Zeiten produziert wurden.

I. INTRODUCTION

In a paper on the lattices of K-feldspars (LAVES 1950) the author was able to show that microcline with an extinction angle of about 18° on (001) and a 2 V angle of about 80° possesses characteristic lattice angles

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showing it to be distinctly triclinic. The term maximum microcline was later introduced for such material. In addition it was found that K-feldspars exist with lattices less triclinic than that of maximum microcline, and that K-feldspars exist with monoclinic lattices. However, some of these „monoclinic" K-feldspars showed X-ray properties that suggested the presence of submicroscopical domains in mutually twinned positions.

As more observations became available, the possibility was discussed (LAVES 1952) of K-feldspar states extending continuously between those of truly monoclinic sanidine and of maximum microcline. Extensive material based on natural feldspars from many localities proving the existence of such structurally intermediate states was presented by GOLDSMITH and LAVES (1945b) and by MACKENZIE (1954). GOLDSMITH and LAVES (1954a) also showed that structurally intermediate states can be produced by heating to about 1000°C. A considerable variation in the optical behaviour of K-feldspars from the Ticino area of Switzerland was reported by PARASKEVOPOULOS (1953) who, in addition, observed qualitative variations in their powder diagrams.

In order to obtain a convenient measure for characterizing the structurally triclinic behaviour of K-feldspars, GOLDSMITH and LAVES (1954a) introduced the term „triclinicity"[1]. This magnitude is defined as $\Delta = 12.5\ (d_{131} - d_{1\bar{3}1})$. The effect on the value of Δ of an Ab-content remains negligible as long as it does not exceed about 20%.

HAFNER and LAVES (1957) reviewed the possible influence on the Δ-value of the infinite number of conceivable three-dimensional Al/Si distributions in triclinic alkali feldspars. They concluded that if structural states be considered that were not formed under equilibrium conditions, no one-to-one relationship between Δ and Al/Si distribution appears to exist. In view of this LAVES (1960) proposed distinguishing between "stable" and "unstable" states. (He defined a "stable" state as one whose Al/Si distribution may be stable under any equilibrium conditions.)

Applying the above definition and as far as we know at present, there exist from a structural point of view only two possible stable series of states that differ with respect to symmetry. If monoclinic the name sanidine can appropriately be applied to the state. If triclinic the state can appropriately be called microcline.

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[1] Many authors objected to this term and now use the word "obliquity". Compare the remarks on this question contained in footnote 1 of the paper by GUBSER and LAVES (1967).

As K-feldspars undoubtedly exist in nature that are "unstable" in the above sense (see BAMBAUER and LAVES 1960), the following comments have to be made with respect to "stable" states:

1. There must be a temperature $T_{S/M}$ above which sanidine is stable (disregarding the influence of pressure and impurities).

2. It is known rather certainly that sanidines have their optical axial plane parallel to (010) when their Al/Si distribution is rather disordered (high temperature state).
On the contrary, their optical axial plane lies perpendicular to (010) if their degree of order exceeds a certain value (not exactly kown today).

3. In isochemical monoclinic K-feldspars there is a variation in the value of 2 V. This appears to be due to different degrees of Al/Si order. When the plane of the optic axes is parallel to (010) low values of 2 V indicate higher degrees of order. If the plane of the optic axes be perpendicular to (010) high values of 2 V correspond to higher degrees of order.

4. At the temperature $T_{S/M}$ sanidine should have the highest degree of order and the largest 2 V $\perp$ (010) possible under equilibrium conditions.

5. Below the temperature $T_{S/M}$ the plane of the optic axes should move from a position perpendicular to (010) towards a position characteristic of maximum microcline. This shows a deviation of about 20° from the position $\perp$ (010). The higher the degree of order the closer the approach to this position should be. Simultaneously 2 V should increase and approach the value of about 84° characteristic for maximum microcline.

6. According to (4) a value 2 $V_{S/M}$ should exist which would be characteristic for $T_{S/M}$.

In view of points (1) to (6) above, the problem arises as to what the numerical value of 2 $V_{S/M}$ actually is.

This question has in principle already been tackled by MARFUNIN (1961) who investigated a large number of K-feldspars with respect to correlations between optical and structural data. The interpretation of his results indicates that the value of 2 $V_{S/M}$ as defined in (6) above might lie at about 44°. However, data to be presented below indicate that 2 $V_{S/M}$ may actually be as high as 63°. No proof can, however, be

given that this is the highest possible value the axial angle may attain.
Reasons for the difference between MARFUNIN's value of 44° and our
own of 63° will be discussed due account being taken that feldspars
may exist in "unstable" states.

In connection with the problem under discussion two interesting
papers by ANSILEWSKI (1958 and 1959) should be mentioned and taken
into account in future decisions on the terminology of alkali feldspars.

II. EXPERIMENTAL

All the K-feldspars used in this investigation were collected in the
Ticino Canton of Switzerland from pegmatites or from quarz-feldspar-
bearing dikes of pegmatitic appearance. For study of the optical proper-
ties, sections parallel to (001) and approximately perpendicular to the
a-axis were prepared. The optical axial angle 2 V was measured conos-
copically with the universal stage. A special 40 X objective and a special
condensor (Zeiss) were used. The area available for 2 V measurements
was about 60 μ^2. The limits of error are generally $\pm 30'$ but may excep-
tionally amount to $\pm 1°$. All powder photographs were taken with a
Guinier-type camera modified by HOFFMAN and JAGODZINSKI (1935).
The camera is a product of the A.E.G., Germany. Cu-K α_1 radiation was
used. The error in the triclinicity values (Δ) should not exceed 0.03,
though it may reach 0.05 when lines are exceptionally weak. The chemi-
cal analyses were carried out with an electron microprobe (ARL), see
Table 1. The determination of the Or-content should be correct to within
$\pm 1\%$.

Table 1. *Chemical Analysis*[a])

Sp. No.	HOST				GUEST		
	Or	Ab[b])	Cs	An	Ab[b])	An	Or
1758	96.6	2.3	1.1	0.0	—[a])	—	—
1739	94.4	3.9	1.4	0.3	93.1	0.7	6.2
1743	98.7	0.4	0.9	0.0	95.6	0.9	3.5

[a]) The analyses were carried out with the aid of a Microprobe Analyser (ARL). —
The host compositions are considered to be characteristic for solid solution within
the matrix. — In specimen 1758 no perthitic Na feldspar was detected; however,
some thin sections showed appr. 5% exsolved Na feldspar.

[b]) The percentage of albite is determined by subtraction from 100.

III. RESULTS

A preliminary survey of the thin sections showed them to be very inhomogeneous, with extinction angles and optic axial angles varying from place to place (cp. PARASKEVOPOULOS, 1953). In some sections the range of 2 V-variation was less than 5°; in others it exceeded 20°. Measurements were made on specimens from twenty different localities. Optically the material shows widely varying development of twinning, being in some cases distinctly twinned, in others apparently untwinned. The microscope reveals this to be the case even within a given crystal. Sometimes a "patchy" development of twinning is to be observed while in certain crystals (though rather very occasionally in the pegmatites investigated) an excellent development of cross-hatched twinning may be present. There are also some large crystals (in the order of centimeters) which show all degrees of triclinicity but no twinning under the microscope. GOLDSMITH and LAVES (1954) determined the Δ-values of a few similar specimens from the same Ticino area and confirmed the conclusion reached by PARASKEVOPOULOS (1953) that these feldspars belong to "intermediate" varieties.

The three specimens (Nos. 1758, 1739 and 1743) discussed below are from three different areas more than one (but less than fifty) kilometer apart. From each area two or three crystals were selected for this study, and from each crystal two or three successive sections were cut. The observations made were as follows:

1. 2 V distributions may differ widely. Two extreme types of such distributions are shown in the two frequency diagrams Figs. 1 and 2. The measurements were made on two crystals from different areas (approximately 50 km apart). The investigated sections were approximately of the same size (3×3 mm). The intervals between measurements were approximately equal (0.5 mm). Fig. 1 shows the results of 36 measurements of 2 V with a distinct maximum at 64°. This may be considered to be a "good" specimen for the type of diagram to be discussed later. Another specimen (Fig. 2) shows a large variation in 2 V values without a distinct maximum. (Actually this specimen shows values of 2 V varying between 67° and 84° within an interval of but 0.2 mm.)

2. Two different crystals from the same hand specimen may give entirely different 2 V values. In the case of Sp. No. 1743, for example, two crystals were examined, one of which showed a 2 V-variation of 67° to 72° and the other one of 75°—84°. Care must, therefore, be taken

in using individual 2 V-values for interpretations of geological history. Even average values obtained from measurements on one and the same crystal may be misleading.

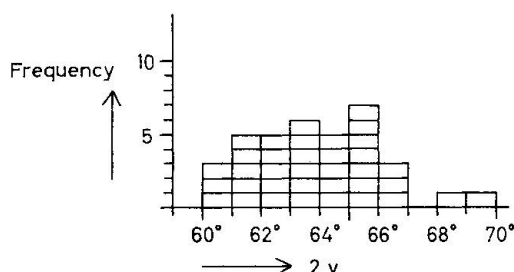Fig. 1. Frequency-distribution diagram of the optic axial angle 2 V. (Specimen No. 1758).
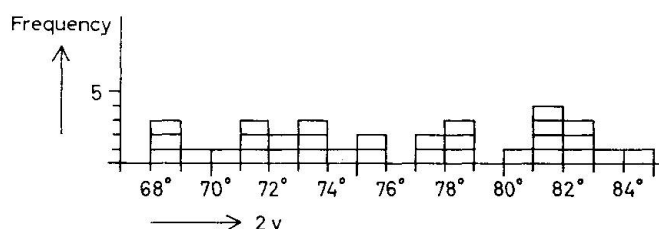

Fig. 2. Frequency-distribution diagram of the optic axial angle 2 V. (Specimen No. 1741.)

3. Though in some cases a continuous increase or decrease of 2 V was to be observed when passing either from one edge of a crystal to the opposite one or from its center to the margins, no general rules exist. For example the 2 V distribution in the specimen depicted in Fig. 2 is most irregular. (An influence of variations in the Na-content on these 2 V variations could not be observed.)

An attempt was made to correlate 2 V with the degree of triclinicity $(\varDelta)$, by measuring 2 V on sections measuring approximately $6 \times 6 \times 0.15$ mm. About 8 to 10 measurements were made on each section which was then used for a powder photograph. In the latter the important reflections $130/1\overline{3}0$ and $131/1\overline{3}1$ were found to be rather broad when the variation of 2 V exceeded $5°$. With a 2 V variation less than $5°$, however, rather sharp lines were observed.

It was decided, therefore, to seek a more precise $2 \text{ V}/\varDelta$-correlation by using even smaller sections (e.g. $3 \times 3 \times 0.15$ mm) with a small 2 V-variation. Even in such small sections 2 V was measured at least at eight places. As most of these sections weighed between 2 and 3 mg, it was necessary to expose their powder to X-rays for about 15—24 hours. As the reflections of $130/1\overline{3}0$ and $131/1\overline{3}1$ are strong, specimens weighing no more than 1.5 mgs could still be used, though the results obtained were, of course, of reduced accuracy. They sufficed, however,

to give an approximate idea of the triclinicity of the specimens in question. For this type of work the Jagodzinski camera seems to be the best. A diffractometer requires at least 10 to 15 mgs of substance for a powder photograph and homogeneous specimens of this size are hard to come by in the material investigated by us. It seems possible that previous workers may have observed broad 130 and 131 reflections, because their specimens were not sufficiently homogeneous. The procedure outlined above led to the following results:

4. In all these specimens the position of the optic-axial plane is approximately perpendicular to (010).

5. Specimens were found showing a 2 V-variation of only 3—5°. Exceptionally specimens were met with showing a variation of 2 V amounting to 67° to 80° within the 2×2 mm area. The material of Fig. 2 was of this character and like all similar cases was not used for the correlation diagrams to be discussed later.

6. The reflections of $131/1\bar{3}1$ and similarly of $130/1\bar{3}0$ were found to be rather sharp when the 2 V variation of the sample remained below 3°. This indicates that the variation in triclinicity is also small.

7. It may be concluded from observations (4) and (5) that the broadening of the 131-reflections, i. e. the variation in triclinicity can be correlated with the variation of 2 V.

8. The $2 V/\Delta$ relations found in three specimens are depicted in the graphs of Figs. 3—5. Following MARFUNIN (1961) 2 V is taken as the ordinate and Δ (the triclinicity) as the abscissa. The range of the 2 V values observed in each sample is shown by lines leading to the triclinicity values as measured by X-rays. These figures indicate that the $2 V/\Delta$-relation is linear in each specimen, though it is evident that the slope of these lines of correlation is different for each of the three specimens.

9. If the $2 V/\Delta$ relations shown in Figs. 3—5 be extrapolated to $\Delta = 0$, i.e. monoclinic symmetry as shown by sanidine (low), 2 V assumes the values of 52°, 60° and 63°, respectively, measured in planes perpendicular to (010).

To obtain the $2 V/\Delta$-relations depicted in Figs. 3—5, several crystals from the same hand specimen had to be used as no single crystal was available showing the entire range of variations of 2 V observable on the hand specimen as a whole. However, since certain parts of different

crystals from the same hand specimen have 2 V/Δ-values which can be connected by "straight" lines, it seems reasonable to suppose that the parts in question have developed under similar conditions. Of course, no variation in the 2 V/Δ ratio should occur among the crystals of any given hand specimen if the latter was formed under equilibrium conditions. However, the rate at which equilibrium conditions are reached may be largely influenced by factors as yet unknown such as impurities (e.g. H_2O) and strain conditions.
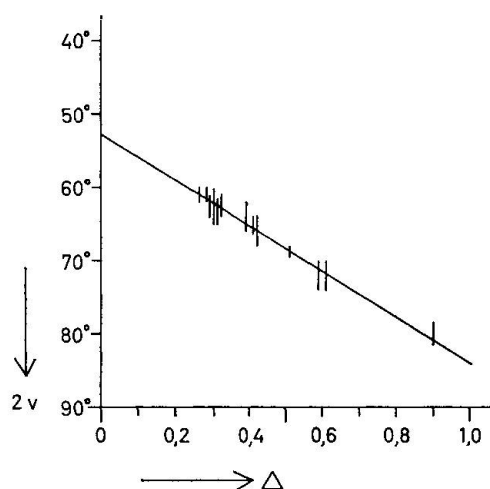

Fig. 3. 2V/triclinicity (Δ) correlation diagram. (Specimen No. 1758.) The ranges of the observed variations in 2V are shown as lines above the corresponding triclinicity values, measured by X-rays on the same samples. The error in triclinicity is ± 0.03.

Fig. 4. 2V/triclinicity (Δ) correlation diagram. (Specimen No. 1739.) The ranges of the observed variations in 2V are shown as lines above the corresponding triclinicity values, measured by X-rays on the same samples. The error in triclinicity is ± 0.03.

Fig. 5. 2V/triclinicity (Δ) correlation diagram. (Specimen No. 1743.) The ranges of the observed variations in 2V are shown as lines above the corresponding triclinicity values, measured by X-rays on the same samples. The error in triclinicity is ± 0.03.
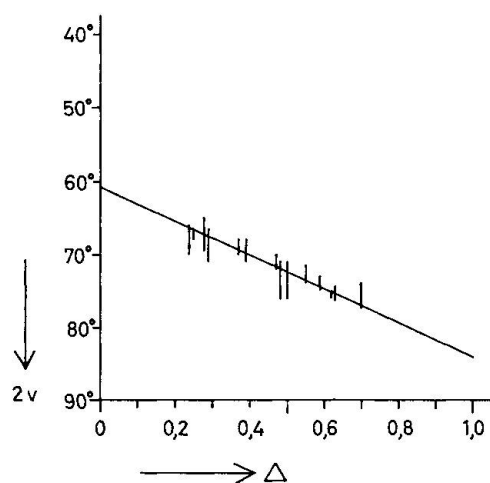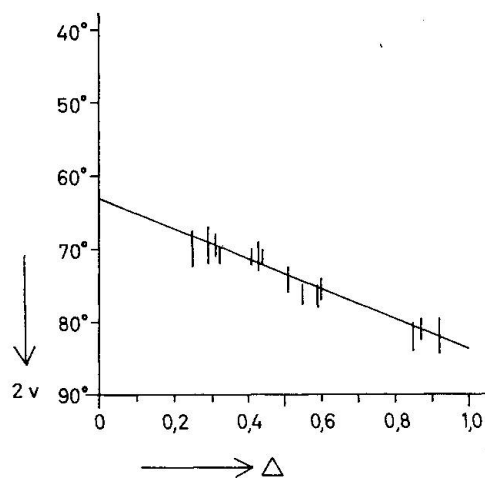
IV. DISCUSSION

The suggestion that the K-feldspars "sanidine" and "microcline" differ in their Al/Si distribution was first made by BARTH (1934). Experimental confirmation of BARTH's hypothesis can be obtained in various ways, a survey of which with references was given by LAVES and HAFNER (1962).

However, the following questions (see LAVES 1960 for references and the discussion in section I) arise and must be answered:

1. *What is the temperature* $T_{S/M}$ above which K-feldspar is monoclinic (sanidine) and below which it is triclinic (microcline) under equilibrium conditions?

2. *What is the maximum degree of Al/Si order* that truly monoclinic material (sanidine low) may have?

Answers to both these questions are difficult to find because of the sluggishness with which Al/Si exchanges take place. Experiments are lacking in which equilibrium states can be considered to have been reached at relatively low temperatures. The answers to the above questions must, therefore, be based on the investigation and interpretation of natural material. Obviously, a truly monoclinic K-feldspar (sanidine) stable at $T_{S/M}$ should have a definite value $2 V_{S/M}$ of its optic axial angle. As stated in section I, this value $2 V_{S/M}$ [$\perp$ (010)] must be the largest one that can be expected in sanidines. Hence of all sanidines for which data are available, the one showing the highest value of $2 V[\perp (010)]$ must be considered to have reached or most closely approached the state of stability at $T_{S/M}$.

Of course, $2 V_{S/M}$ will be dependent on impurities present in the crystal, the most important of which is likely to be the Na-content. As the specimens used for this study were all rather Na-poor, the influence of this factor will be neglected in the following discussion.

The problem of obtaining information regarding $2 V_{S/M}$ (as defined above) has already been tackled by MARFUNIN (1961) in the following way. The 2 V values of about 130 specimens were plotted against the values of the angle (Ng ∧ b*) with which are related the extinction values measured on (001). As these values defining the optical orientation can be correlated rather instructively with the expressions used for defining triclinicity (Δ or γ^*) MARFUNIN's results can be summarized as in Fig. 6. As his $2 V/\Delta$-values lie either *on* the 2 V ordinate or on a line "4" or within the shaded field of Fig. 6, MARFUNIN concluded that the line "4"

divides the $2\,V/\varDelta$-field into two parts: An upper one containing only monoclinic feldspars with $\varDelta = 0$ and a lower one containing feldspars of varying optical behaviour (optically monoclinic and triclinic) and with varying $\varDelta$-values. MARFUNIN considered the line 4 to be characteristic of microclines — a state of equilibrium — and the 2 V-value of 44° (at the intercept of line 4 with the ordinate) to be characteristic of the most ordered condition which a truly monoclinic K-feldspar (sanidine low) can assume.
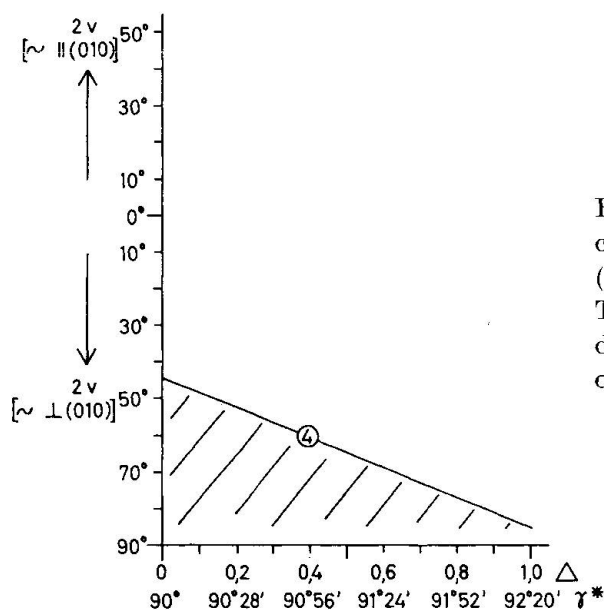

Fig. 6. In this figure a 2V/triclinicity correlation diagram of MARFUNIN (1961) is presented in a modified form. The triclinicity values in his original diagram (Fig. 4, 1961) are based on optical measurements [Ng $\wedge \perp$ (010)].

Fig. 7 is a diagram similar to that in Fig. 6, containing however in addition to MARFUNIN's line "4" further lines "1", "2", "3" derived from Figs. 3, 4, 5 of the present paper. Some $2\,V/\varDelta$ correlations are also included that were measured by BAMBAUER and LAVES (1960) on a specimen of Swiss adularia which is to be considered as rather disordered.

The existence of lines lying below MARFUNIN's line "4" indicates that the latter may not, in fact, represent the equilibrium state of triclinic feldspars. It seems likely that the lowest line found in the present investigation may actually provide a better approach to the $2\,V/\varDelta$ relations of equilibrium microcline material. The expression "better approach" is meant to imply that even lower lines than our line "1" may exist and give correspondingly better values of the $2\,V/\varDelta$-relations of equilibrium material.
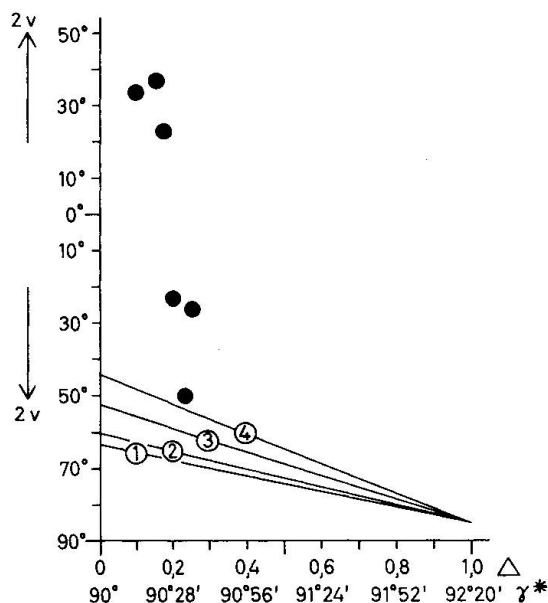
For present purposes and for the sake of discussing the main principles involved, we shall assume our line "1" to be in fact the lowest one and to have the following significance:

1. If only "stable" material be considered and all twinning be disregarded:

A. The line is seen to divide the $2\,V/\Delta$ field into two parts neither of which should contain any "stable" material.

B. The value of $2\,V$ at which the line intersects the ordinate would be the largest one possible for a sanidine (low) with an optic axial plane $\perp$ (010). In our diagram the intersection-value of $2\,V$ is found to be $63°$.

Fig. 7. $2\,V/$triclinicity relations observed in different specimens. Lines 1, 2 and 3 are taken from figures 5, 4 and 3 of this paper. Line 4 is taken from Marfunin's diagram (Fig. 6 in this paper). Solid circles represent $\gamma*/2\,V$ measurements made on different portions of one adularia specimen (BAMBAUER and LAVES, 1960).

2. Assuming submicroscopical twinning to exert an influence on $2\,V$ and Δ it follows that:

A. It is to be expected that such twinning will lead to values of $2\,V$ and Δ that are not characteristic of the Al/Si distributions within the twinned domains.

B. As such twinning becomes finer increasingly low values of $2\,V$ and Δ should be measured.

C. The influence of such twinning ("balanced" or "unbalanced") will be greater on the Δ-values than on $2\,V$.

D. The twinning may have a considerable influence on the position of the plane of the optic axes.

3. If "unstable" states are assumed to be present, conditions become even more complex for the following reasons:

A. BAMBAUER and LAVES (1960) showed that within one and the same natural "adularia" crystal domains exist with considerably

varying properties. If the angle between the optical axial plane and (010) be represented by Ω the following relations hold true:

a) Ω varies continuously between the values of about 0° and 90°.

b) 2 V varies continuously between about 50° in a plane parallel to (010) and about 50° in one nearly perpendicular to (010). A minimum of about 20° lies at $\Omega = 45°$.

c) Δ varies continuously from about 0.00 to 0.25.

B. The facts stated under (A) lead to the conclusion that there may be states which are "unstable" and have properties differing considerably from "stable" ones.

In view of the statements made under (1) to (3) above, no physical property of K-feldspars can be expected to be rigorously indicative of a certain structural state. This is precluded by the fact that the material may be stable or unstable, twinned or untwinned. If twins are present, the twinning may be on such a fine scale that physical measurements represent average values only. Furthermore, the texture of the twinned domains must be taken into account, i.e. consideration has to be given to the size and shape of the domains and to the question of whether these are twinned according to the albite or pericline law or both.

Despite the pessimistic view stated above, available data suggest that 2 V is in fact a rather good indicator of the prevailing structural state, i.e. of the degree of Al/Si order. If 2 V is large and Ω is close to 0° the material may be considered to be rather disordered. Large values of 2 V in connection with Ω values of nearly 90° show the material to be rather
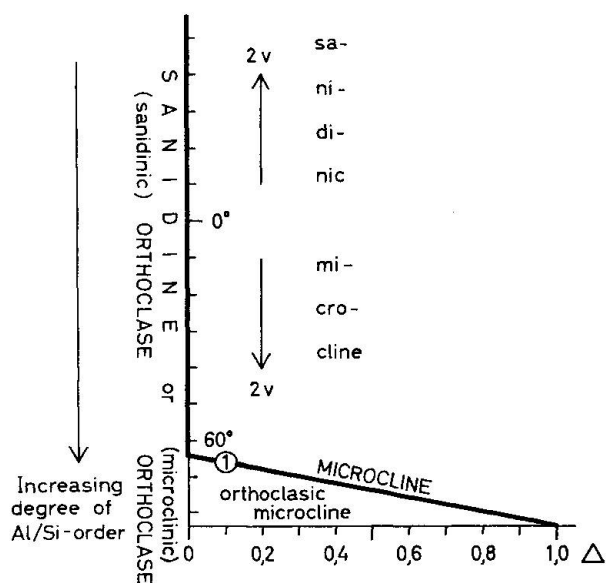

Fig. 8. 2 V/Δ relations of natural K-feldspars. The heavy lines correspond to "stable" states (sanidine or microcline) or to "unstable" states (orthoclase), see text. — As to the fields above and below the "microcline line" see section IV: Discussion.

ordered. If Ω is different from $0°$ and smaller than about $70°$, the presence of an "unstable" state is indicated.

Fig. 8 is based on the facts mentioned above and is intended to show the $2\,V/\varDelta$ correlations. The figure calls for the following remarks:

A. The heavy lines represent the $2\,V/\varDelta$-relations, indicated by the results of the present investigation, for stable material undisturbed by submicroscopical twinning.

B. Of the branches shown in the diagram the one with $\varDelta = 0$ corresponds to (stable) *sanidines* while the one with $\varDelta$ varying from 0 to 1 applies to (stable) *microclines*.

C. When $\varDelta$ is different from zero the microcline branch divides the $2\,V/\varDelta$-field into two parts. Material falling into the uppermost of these is to be considered as unstable with regard to its Al/Si distribution. As such material is triclinic[2]) but more disordered than stable microcline it might be called *"sanidinic microcline"*. Material falling into the lower of the two fields is to be considered as consisting of unbalanced submicroscopical twins with domains having a higher degree of order than indicated by their $\varDelta$-value. For such material the term "orthoclasic microcline" might be appropriate.

D. When $\varDelta = 0$ two sub-cases must be distinguished from the point of view of structure.

   a) When $2\,V$ is smaller than about $65°$ the material *may* be truly monoclinic, i.e. *sanidine*. On the other hand, it *may* show an apparently monoclinic optical behaviour due to a (balanced) submicroscopical twinning of triclinic domains having an unstable Al/Si distribution as in "sanidine microcline". In this case (sanidinic) orthoclase would appear to be the proper designation.

   b) When $2\,V$ is greater than about $65°$ the material *must necessarily* consist of balanced, submicroscopically twinned domains that are more ordered than the sanidine stable at $T_{S/M}$. The name (microclinic) *orthoclase* appears suitable for such material.

There has been much discussion in the literature on the meaning of the word orthoclase. Some authors call all K-feldspars that are optically monoclinic "orthoclase" and consider sanidine and adularia to be subdivisions of orthoclase in this sense. Others make a distinction between orthoclase and sanidine using a rather arbitrary value of $2\,V$ to dis-

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[2]) Either single crystalline or as a result of unbalanced submicroscopic twinning.

criminate between the two. The reasons for such diverging definitions are those given above and Fig. 8 shows that, in principle, the arbitrary use of terms can be avoided if *structural* properties are taken into consideration. Of course, the name orthoclase still retains its practical value for the classification of K-feldspars according to *optical* properties and *geological* modes of occurrence.

The name "adularia" has been avoided in this discussion as its meaning is predominently a *mineralogical* one based on morpological appearance and geological occurrence. From a *structural* point of view adularia may be sanidine, microcline or orthoclase (this term being considered to mean a "pseudostable" state in the sense of footnote 2 on page 549 of a paper by LAVES, 1952).

As a rule, adularia occurs in states intermediate between those capable of being exactly defined as sanidine, microcline or orthoclase. Some properties of adularia are discussed in an accompanying paper by GUBSER and LAVES (1967).

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