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Lattice Changes in the Low Plagioclase Series

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Wih 9 figures and 1 table in the text

Abstract

A review of the available lattice constants of low plagioclases, plotted against molar anorthite content, shows that the points fall near linear curve segments. The data are compatible with the assumption that the "kink points" between these segments lie near 25, 37.5, 50 and 87.5 mol % anorthite. At these compositions full Al-Si ordering is possible assuming a cell with $c \sim 14$ Å. The plagioclases between these points are assumed to be composed of two kinds of unit cells with ordered Al-Si and the lattice constants are thus "means" of two cell geometries. The "kink points" at 25 and 87.5 mol % are also indicated in optical data and as maxima of plagioclase composition statistics in metamorphites and plutonites.

INTRODUCTION

The collection and representation of lattice constants in crystalline solid solution series (or of isomorphic compounds with partial substitution) has two main purposes:

1. The determination of chemically intermediate members of the series by comparison of the measured with the known lattice constants.
2. The investigation of "breaks" or "gaps" in the otherwise continuous row of crystal structures, in cases of incomplete isomorphism. These "breaks" are indicated by "kink points" connecting lattice data for various compositions.

For the optical and the physical data in the low plagioclases it has been shown since a long time (e.g. FEDOROW, 1898) that isomorphism in this series is incomplete, and the structural complexity of it made the knowledge of exact lattice constants a prime necessity.

Since about 1960 a large amount of natural (chemically intermediate) plagioclases from various metamorphic and plutonic rocks have been investigated,

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and their anorthite content (determined via optical data or chemically) has been reported together with partial or complete measurements of the six lattice constants which had been obtained either by single crystal methods (COLE et al., 1955, 1960; DOMAN et al., 1965) or from Guinier powder patterns with the help of a least squares refinement computer programm (BURNHAM, 1966). Besides sets of lattice constants for a single substance reported together with structure determinations, the majority of the data obtained with the second method were published in the "Thema Feldspäte" special volume which appeared in 1967 (BAMBAUER et al., 1967; STEWART, 1967; NISSEN et al., 1967). In the same volume, STARKEY (1967) presented the lattice angles α, β and γ as well as the reciprocal lattice angles calculated from the precession data of BROWN (1960), J. V. SMITH (1956) and others.

The lattice data, especially γ^*, reported by BROWN (1960) and DOMAN et al. (1965) had been plotted against anorthite content (in mol %, mainly determined optically), but BROWN also plotted them against the Al/Si ratio, in which diagram the plots showed a linear trend to a first approximation. However, deviations from this linearity were strong, especially in the chemically intermediate members, and a line with several "kink points" fitting the points much more closely, can be drawn. DOMAN et al. (1965) reported that their lattice data, mainly γ^*, when plotted against molar anorthite content, lay near linear segments and a sharp offset existed at the discontinuities which they assumed near 33, 50, possibly 68 and 85 mol %. Obviously, linear curve segments connecting these plots can exist only in one of the two representations, i.e. either when plotted against molar anorthite content or against Al/Si.

BAMBAUER et al. (1967) plotted the lattice constants determined by them only against the proportion Al/Si calculated from anorthite contents. They held that

1. the lattice constants changed along continuous curves, having a number of "kinks";
2. in the plot of lattice constants versus Al/Si the segments between the "kink points" were linear within the error of measurement;
3. the "kink points" were at 17 mol % (if the curves were continued straight through the peristerite exsolution gap), 33%, 50%, 76% and possibly 87 mol %.

Except for the assumption of continuous rather than discontinuous curves these results seemed roughly to correspond to those of DOMAN et al. (1965) except for the calcic (bytownite) region.

The data by STARKEY (1967) calculated from BROWN's and other measurements were only listed in table form and used to calculate σ, the characteristic angle of the rhombic section. This as well as the fact that the data by BAMBAUER et al. (1967) have not yet plotted versus the molar anorthite content,

makes it necessary to plot all data into the same diagrams with molar anorthite contents as abscissa in order to obtain the best possible diagrams for the evaluation of structural breaks and the determination of low plagioclases form their lattice constants. KOREKAWA (1967) has shown that the separation into Ca- and Na-rich domains is the main feature explaining the structural complexity of the low plagioclase and for the present problem this is another reason to prefer the usual molar anorthite scale to the Al/Si scale.

If we plot lattice data from the papers cited above versus molar anorthite content, we must consider two kinds of differences in the methods used to obtain these data:

1. The anorthite content of the specimens has been determined in different ways. Whereas SMITH (1958) and BROWN (1960) determined the anorthite contents mainly with the U-stage (angles of n_α, n_β and n_γ directions with respect to the crystal axes), DOMAN et al. (1965) measured refractive indices of small grains in the "spindle stage" and used plots of refractive indices versus anorthite contents to obtain the latter. Their specimens were in part very inhomogeneous with regard to anorthite content. Therefore the method used by these authors is much preferable to referring all lattice data of one specimen to the anorthite content as determined by wet chemical analysis. It is hoped that microprobe analyses will be executed of the single grains taken for lattice constant and refractive index measurements. The data by BAMBAUER et al. (1967) were determined with the microprobe (CORLETT and RIBBE, 1967).
2. The lattice data have been obtained in different ways. The data by BROWN (1960) and DOMAN et al. (1965) were measured in precession photographs charateristic for a small grain, whereas the data by SMITH (1956) and BAMBAUER et al. (1967) were determined with least squares methods, applied to 2θ-measurements of a powder film representing a mean from a rather large specimen.

In spite of all these differences in methods it was felt that all these data should be plotted together in order to find and resolve discrepancies and analyse them with regard to their value for feldspar determination and the structural meaning of the observed changes. Since Ca/Na exsolution phenomena appear to play an important role in the low plagioclase series, the molar anorthite content was used as the ordinate of all diagrams as is usual also in plots of optical constants (e.g. BURRI et al., 1967).

The lattice constants of exsolved specimens in the peristerite region, the oligoclase region (NISSEN, 1969) and the bytownite region (NISSEN, 1968) have been included in the diagrams, since, in general, these lattice constants fit well into the curves and seem to represent to a high approximation a mean between the two exsolution members. The same seems to be true for labradorites with

lamellae producing the schiller, though in this composition range there is a generally higher scatter of lattice constants.

RESULTS

The constants (including the reciprocal lattice angles) are represented in Figs. 1–8. In some of these figures data from plagioclases with high structural state have also been plotted for comparison. The synthetic high plagioclases are represented by ×, while the heated natural plagioclases are shown as ○. These plots include measurements published by COLE et al. (1955), SMITH (1956), STEWART (1967) and NAGER et al. (1969). The symbols used for the low plagioclases are as follows:

1. *Dots* are data calculated by STARKEY (1966) after the data of BROWN (1960) and others. STARKEY's anorthite contents have been converted to mol %, whereby they were rounded to 0.5 mol %. In fig. 7 dots represent data from SMITH (1956), triangles those from STEWART (1967).
2. *Crosses* are data from BAMBAUER et al. (1967, table 1).
3. *Triangles pointing up* are data cited in BORG and SMITH (1968) and also unpublished lattice data and microprobe- and optical analyses of three oligoclases kindly supplied by I. Borg.
4. *Triangles pointing down* are data from E. WENK et al. (1968) and H.-R. WENK (1966, 1969).
5. *Squares* are data by NISSEN et al. (1967) from specimens with >2 mol % K-feldspar and data for two specimens from BAMBAUER et al. (1966), also with >2 mol % K-feldspar.
6. *Dots with rings* (⊙) are γ^*-data from DOMAN et al. (1965) (only in fig. 4).

The following details are evident from figs. 1–8.

α (fig. 1). The scatter of all points is smaller than $\pm 0.1°$. From approx. 37.5 mol % to 100 mol % a linear distribution is indicated allowing for the scatter of $\pm 0.1°$. Between 15 and 40 mol % there are in particular many points indicating linear behaviour in the ranges between 0 and 25, and 25 and 37.5 mol %, with a slight kink at 25 mol % and a more pronounced one near 37.5 mol %. The points near 65, 80 and 85 mol % correspond to structurally intermediate plagioclases and are also included in the following figures.

β (fig. 1). If the data from STARKEY (1967) are excluded as has been done in the figure, the diagram shows the best approximation to linearity over the whole anorthite range, with a single "kink point" near 37.5 ± 3 mol %.

This linearity is clearly lost when the same data are plotted versus the Al/Si proportion calculated form the anorthite content. The values for β from

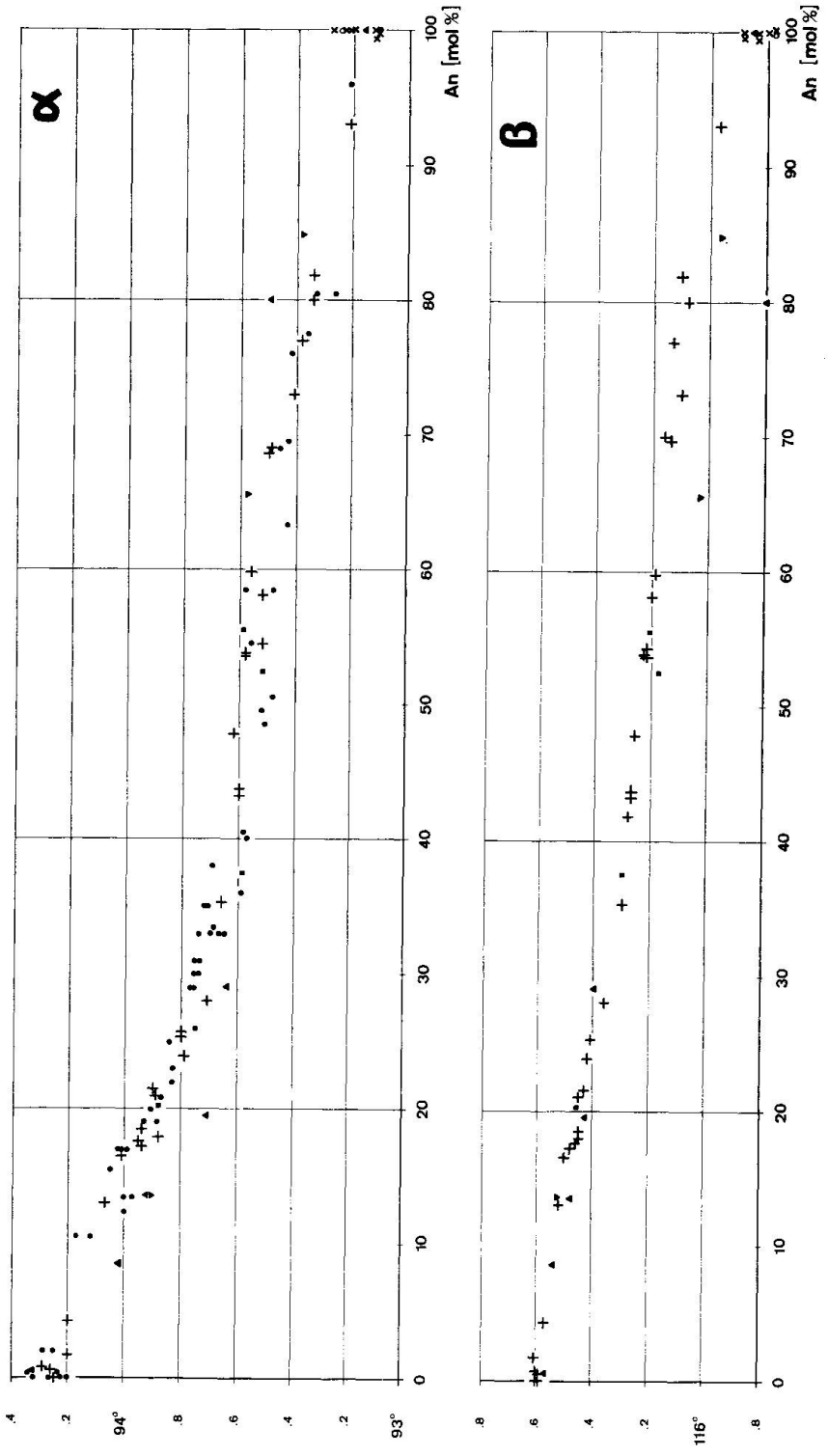

Fig. 1.

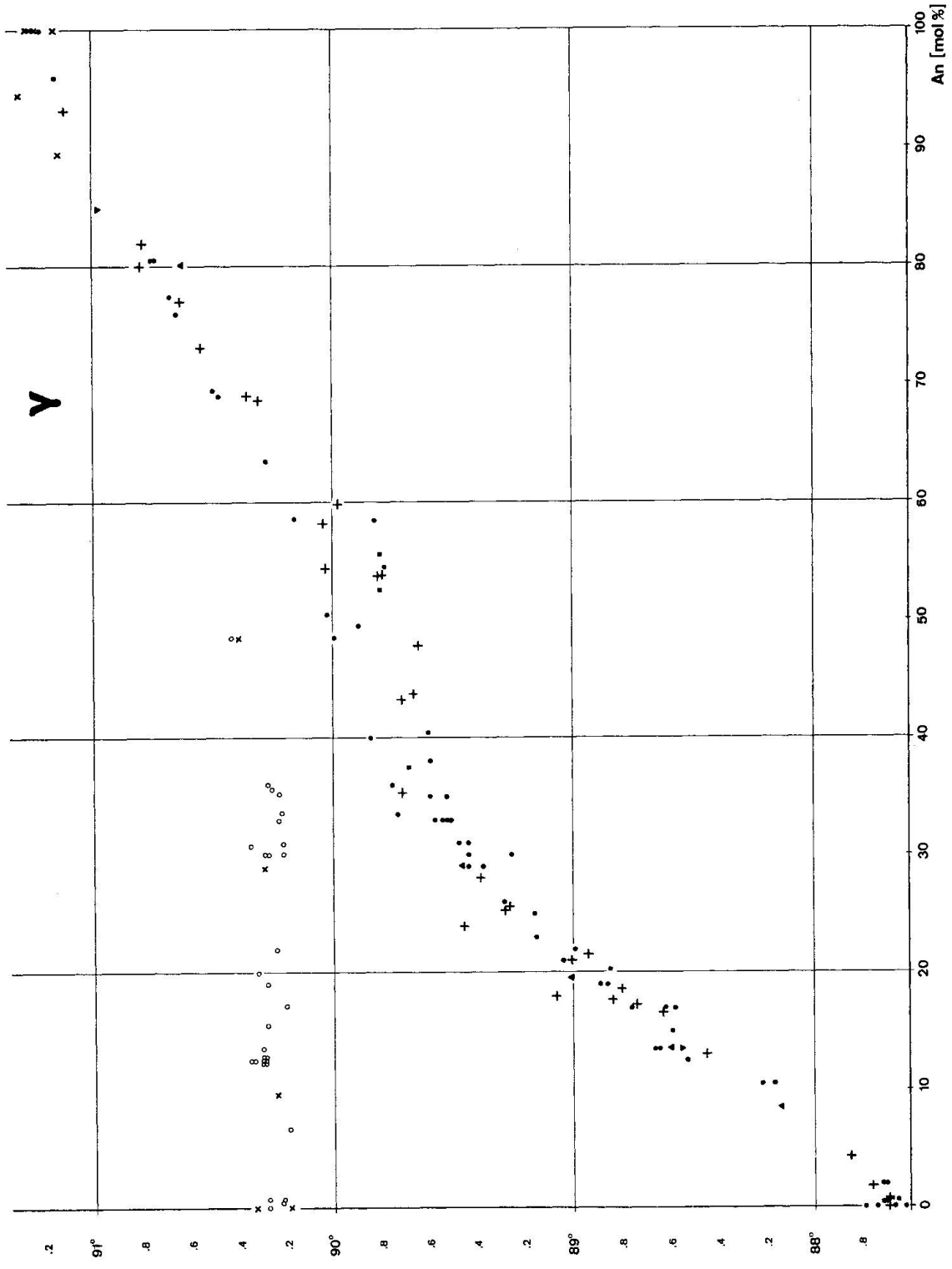

Fig. 2.

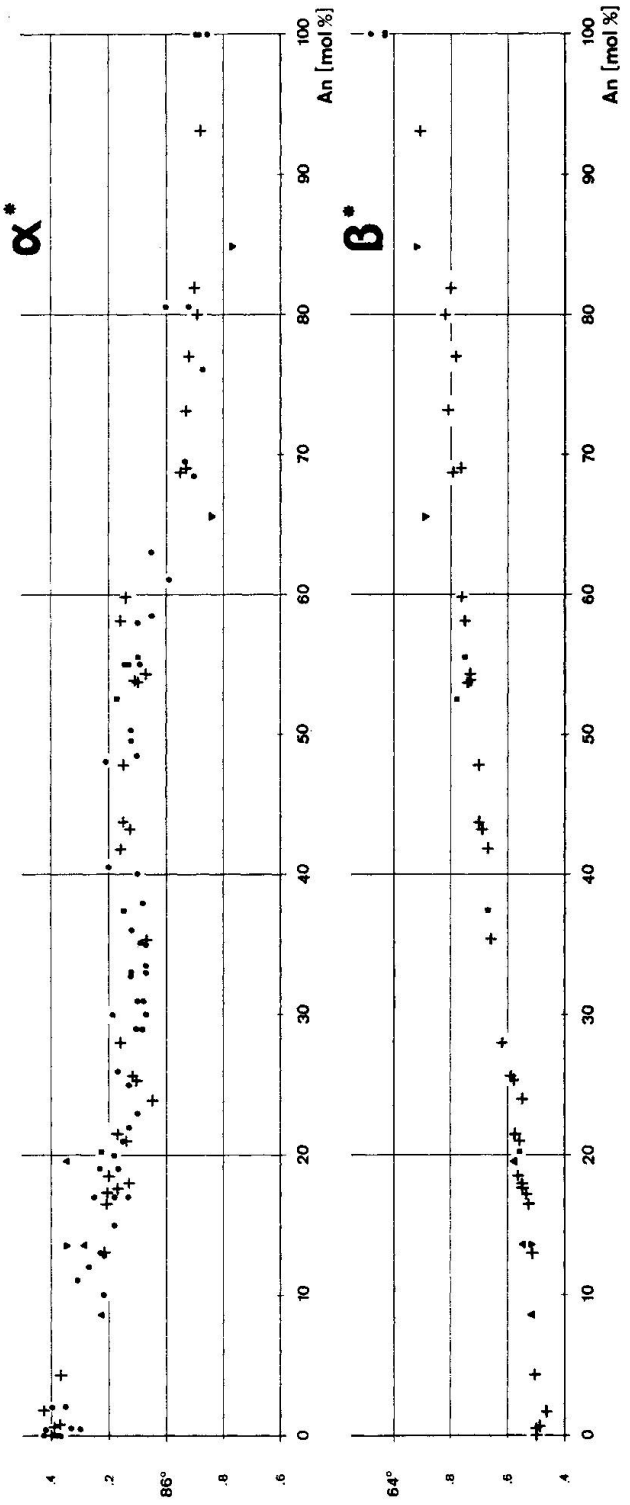

Fig. 3.

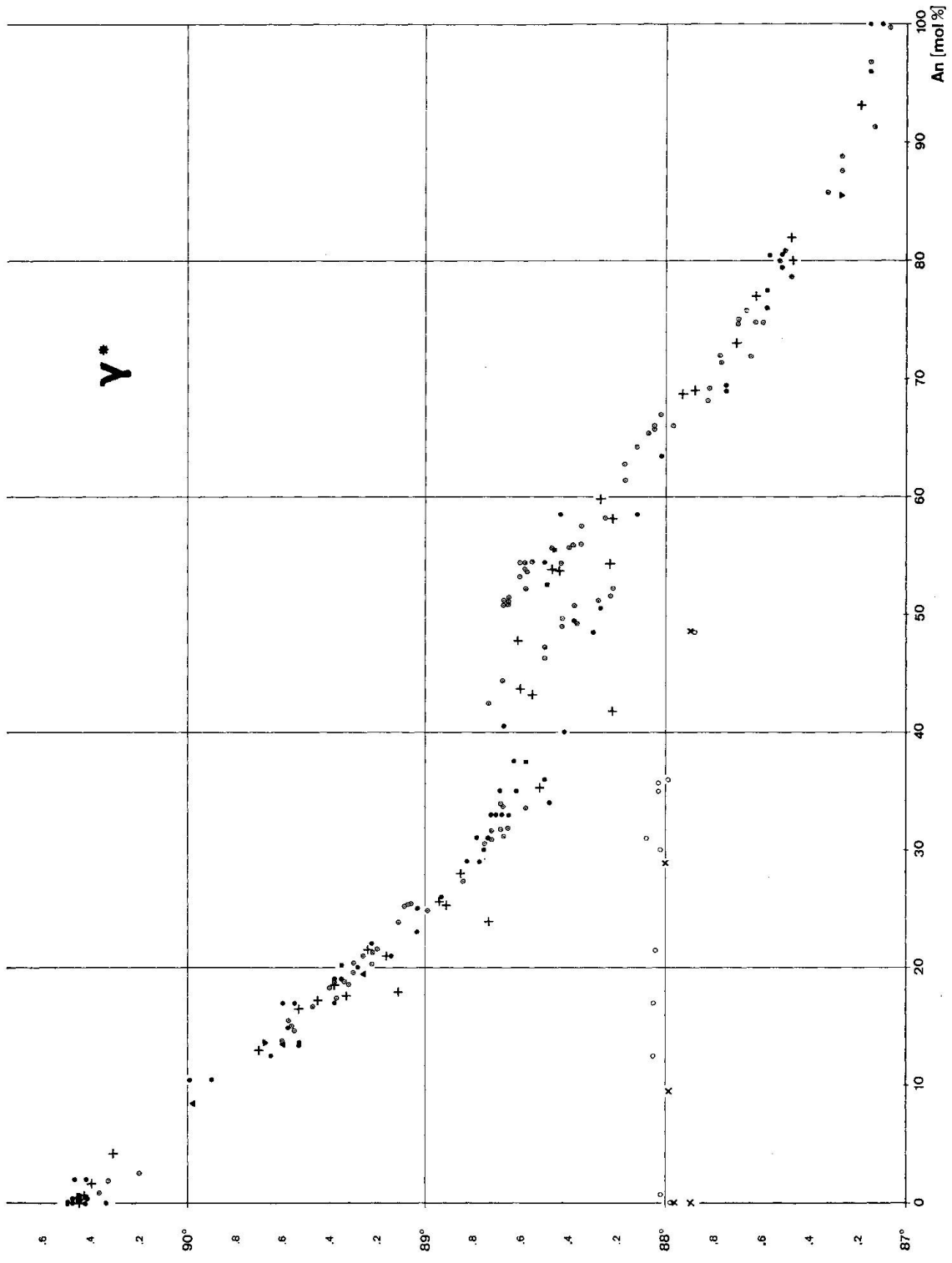

Fig. 4.

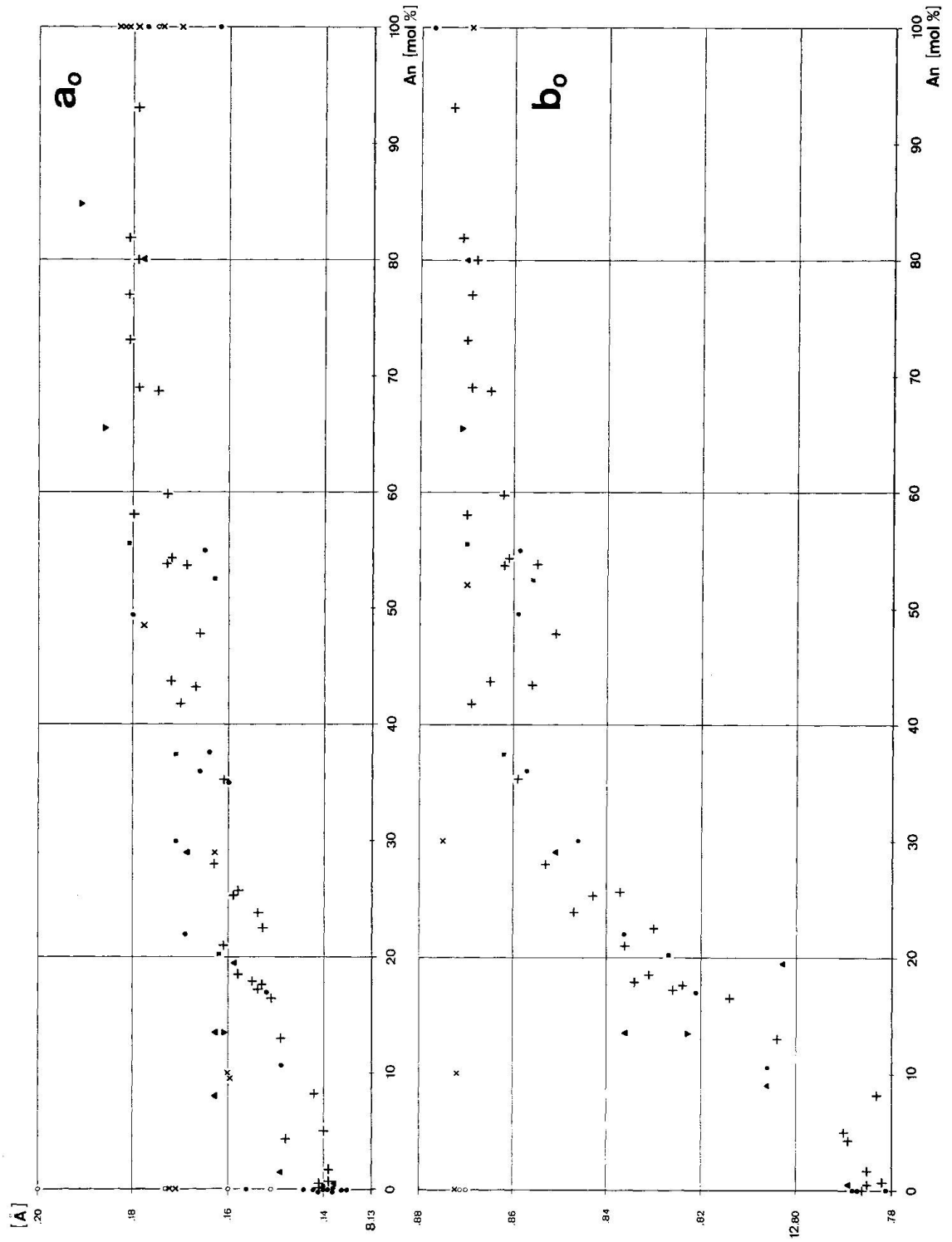

Fig. 5.

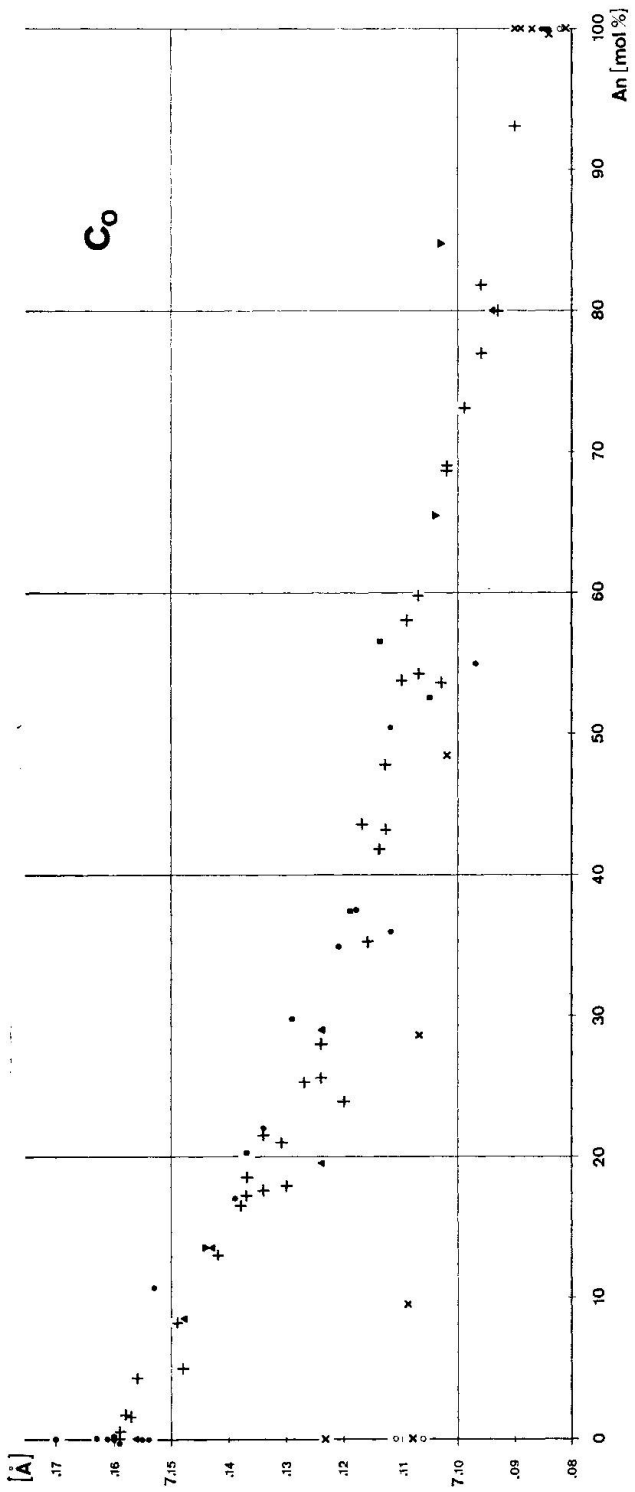
c_o
[Å]
.17
.16
7.15
.14
.13
.12
.11
7.10
.09
.08
0
10
20
30
40
50
60
70
80
90
100
An [mol %]

Fig. 6.

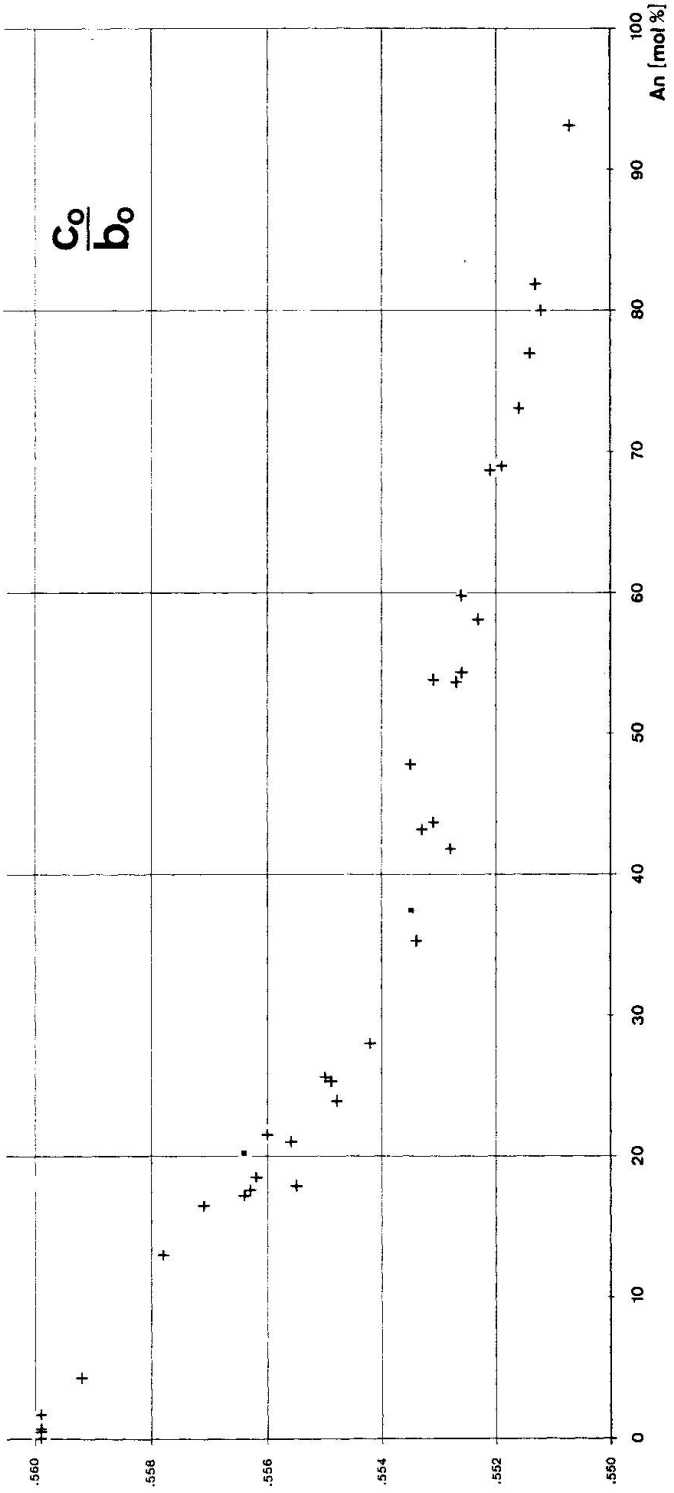
$\frac{c_o}{b_o}$
.560
.558
.556
.554
.552
.550
0
10
20
30
40
50
60
70
80
90
100
An [mol %]

Fig. 7.

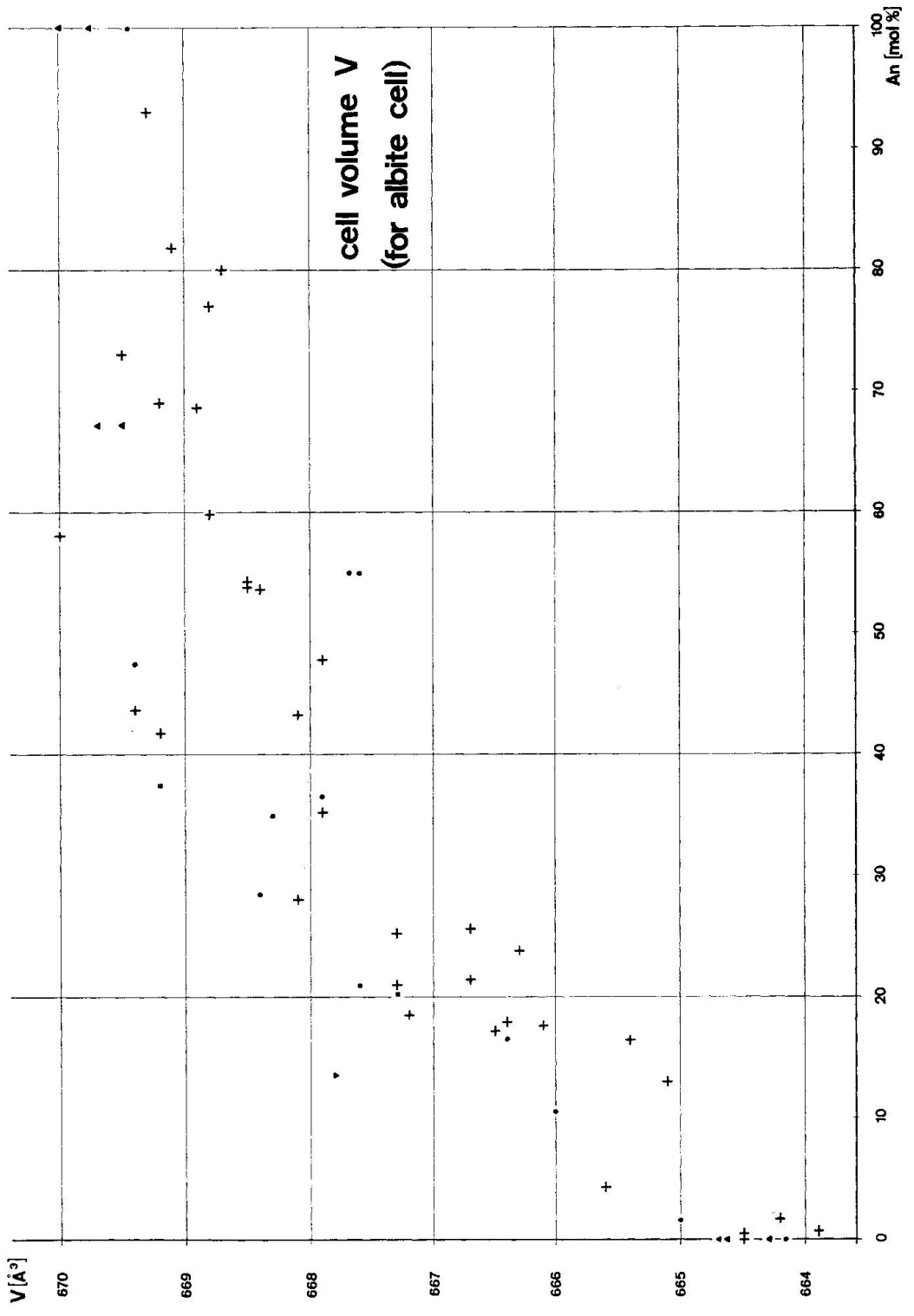
cell volume V
(for albite cell)
V [Å³]
670
669
668
667
666
665
664
An [mol %]
0
10
20
30
40
50
60
70
80
90
100

Fig. 8.

STARKEY (1965) were mainly calculated from the measurements of BROWN (1960) using the expression

$$\cos\beta = \frac{\cos\alpha^* \cos\gamma^* - \cos\beta^*}{\sin\alpha^* \sin\gamma^*},$$

where β^* was already calculated from the original measurements of α^*, γ^*, and β using the expression

$$\cos\beta^* = \left(\frac{\cos\alpha^* \cos\gamma^*}{\cos\gamma^* \sin\alpha^*} - \cos\beta\right) \sin\alpha^* \sin\gamma^*.$$

Obviously, the error becomes too large during this double calculation. STARKEY apparently made this calculation to replace Brown's measurements of β made directly on the dial of the precession camera. However, both sets of data have a much higher scatter than the β data obtained from least squares refinements of powder data and were therefore not included in fig. 1.

γ (fig. 2). If the diagram of γ-values is inverted and the angular scale shifted, the diagram of γ can be superimposed with high accuracy on to that of γ^* which will be described below.

Since many more measurements of γ^* than of γ are available, the γ-diagram should be compared with that of γ^* for the evaluation of "kink points". The diagrams of γ and γ^* show the strongest changes (measured in degrees). γ is linear (less than $\pm 0.1°$ scatter) between 0 und approx. 25 mol % and between 25 and 36–40 mol %. From approx. 40–60 mol % the scatter is very large but a straight line can be drawn approx. from 50 to 87.5 mol %. Between 87.5 and 100 mol % the curve may be flatter again, but data in this range are badly lacking.

α^* (fig. 3). This diagram shows more clearly than others the kink point near 25 mol %. Otherwise the scatter is too large to be very indicative. Another "kink point" must be between 60 and 70 mol % but is difficult to fix more precisely.

β^* (fig. 3). Like β, this diagram (with STARKEY's data excluded for the reasons given above) shows very good linearity, with a very weak "kink" possibly near 50 mol % and another strong "kink" near 87.5 mol % (i.e. between 83 and 95 mol %).

γ^* (fig. 4). Similar to the γ-diagram (fig. 2), this figure shows a high degree of linearity (less than $\pm$ 0.1°) except between 35 and 60 mol % where the scatter is very high and the anorthite determinations of some specimens are a matter of discussion. The Na-rich and the Ca-rich parts of the curve both show a "kink", the change is only slight near 25 mol % anorthite but rather pronounced near 87.5 mol %. In the range between 35 and 60 mol % the data with microprobe analyses of anorthite content must be given preference. These indicate "kink points" near 37.5 and 50 mol % anorthite. The measurements made by

DOMAN et al. (1965) on specimen No. GD-1805 (Ferrogabbro from Shepherd Mountain, Missouri) have been left out of the diagram since they appear improbable in the light of the other data. A verification is awaited. A new determination of the anorthite contents of specimens No. 10 and 11 might also change these points which now appear from the γ^* diagram as structurally intermediate specimens. The same seems to be the case for specimens No. 104, 114, 94 and 48 of BAMBAUER et al. (1967) and for specimens No. 88, 91 and 92 of STARKEY (1967) (viz. No. 29 and 30 of BROWN, 1960).

a_0 (fig. 5). The scatter of the a_0-values is too big to indicate definite "kink points", except between 82 and 92 mol % (possibly at 87.5 mol %).

b_0 (fig. 5). The curve of the b_0 values shows a definite kink point near 37.5 ± 4 mol %. For lower anorthite values, b_0 rises rapidly with rising anorthite content and for higher anorthite values it remains nearly constant (approx. 12.87 Å).

c_0 (fig. 6). The diagram indicated a "kink point" near approx. 25 mol %; but the scatter is rather high, so that other possible "kink points" near 50 and between 85 and 90 mol % are only weakly indicated.

c_0/b_0 (fig. 7). This diagram drawn to supplement diagrams of b_0 and c_0 and including only the data of BAMBAUER et al. (1967) shows a linear behaviour with good approximation in the ranges 0 to approx. 25 mol % and approx. 40 to 100 mol %. Calculation of c_0/b_0 from further lattice constants in the andesine range may show whether two "kink points" occur in this range corresponding possibly to those found for the b_0 and c_0 diagrams (i.e. approx. 25 and 37.5 mol %) and in the lattice angles.

Cell volume V (fig. 8). The cell volume shows a scatter up to ± 1 Å within a small anorthite range. It increases until approx. 25–40 mol % anorthite and for higher anorthite contents it remains constant within the scatter range. The possible "kink point" may indicate the change from the albite cell ($c \sim 7$ Å) to the anorthite cell ($c_0 \sim 14$ Å). The calculation of more cell volumes and also density data is in preparation. These data are needed before a structural evaluation is possible. The high scatter of the data may be due to summation of errors inherent in the measurement of the single lattice constants used to calculate the volume, i.e. incomplete or inaccurate powder diffraction measurements, and also – perhaps less important – to differences in structural state and isomorphic K-content. The cell volumes determined for structurally intermediate labradorite and bytownite by WENK (1966) and WENK et al. (1968) are unusually high (671 and 672.5 Å^3) and are not plotted in fig. 8.

Diagram of $2\theta_{(131)} - 2\theta_{(1\bar{3}1)}$ (fig. 9) for specimens with < 2 mol % K-feldspar. The points for the curves reported by BAMBAUER et al. (1966) are here plotted against the molar anorthite content. Specimens reported by CORLETT and RIBBE (1967) to be particularly poor in isomorphic K content and homogeneous with regard to the main non-tetrahedral cations Ca, K and Na are shown as rings.

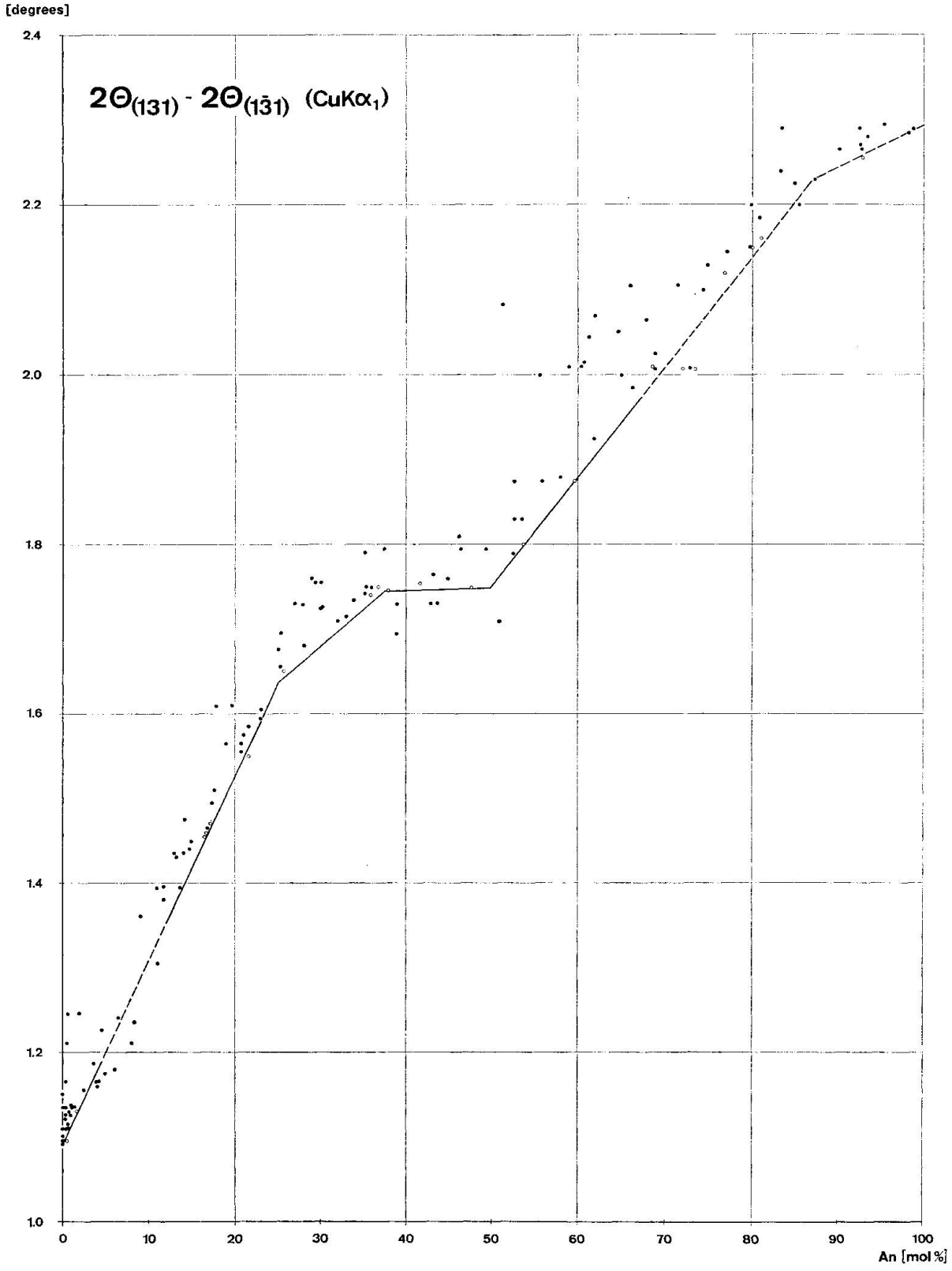
[degrees]
$2\Theta_{(131)} - 2\Theta_{(1\bar{3}1)}$ (CuKα_1)
2.4
2.2
2.0
1.8
1.6
1.4
1.2
1.0
0
10
20
30
40
50
60
70
80
90
100
An [mol %]

Fig. 9.

These points can be connected by linear segments with "kink points" near 25, 37.5, 50 and 87.5 mol %. Only some of the specimens marked as rings in the bytownite range deviate significantly from this curve; these have been found as well as many others in the same anorthite range to be unmixed (NISSEN, 1968). It is interesting to note that if the very numerous measurements of MORSE (1961) and ROMEY (1969) are plotted into the diagram of fig. 9, a large percentage falls below the curve drawn as lowest curve for homogeneous K-free specimens. This does not seem to be entirely due to high K contents of these (anorthositic) materials because homogeneous K-contents in samples with more than approximately 60 mol % anorthite are usually relatively low (< 2 mol % K-feldspar).

CONCLUSIONS

From the evidence shown in figs. 1–9 the following conclusions can be drawn:

1. In spite of many and in part apparently reliable measurements available to date, the lattice changes of the low plagioclase series are yet very insufficiently known. More determinations on materials analysed with the microprobe are needed, especially for the ranges 40–60 mol % and for 80–100 mol % anorthite (where structurally high plagioclases are difficult to differentiate from structurally low specimens).
2. The lattice constants of low plagioclases show such small scatter for given anorthite contents that the mean of these measurements (after the exclusion of structurally intermediate specimens) appears to indicate the data typical for the low structural state and should be used to construct curves of lattice constants rather than the lowest measured values.
3. The lattice constants of low plagioclases plotted against anorthite contents change along curve segments which are *linear* within the scatter inherent

Table 1. *Plagioclases which may have a fully ordered Al, Si configuration, assuming the anorthite cell ($c_0 \sim 14$ Å) and, partly, absence of centrosymmetry.*

anorthite fraction	anorthite [mol %]	Formula for 8 (Na+Ca)	Al/Si	Si/Al
0	0.0	$Na_8 \quad Al_8 Si_{24}O_{64}$	8 : 24	3.00
1/8	12.5	$Na_7 Ca \quad Al_9 Si_{23}O_{64}$	9 : 23	2.55
2/8	25.0	$Na_6 Ca_2 \quad Al_{10}Si_{22}O_{64}$	10 : 22	2.20
3/8	37.5	$Na_5 Ca_3 \quad Al_{11}Si_{21}O_{64}$	11 : 21	1.95
4/8	50.0	$Na_4 Ca_4 \quad Al_{12}Si_{20}O_{64}$	12 : 20	$1.66\overline{6}$
5/8	62.5	$Na_3 Ca_5 \quad Al_{13}Si_{19}O_{64}$	13 : 19	1.46
6/8	75.0	$Na_2 Ca_6 \quad Al_{14}Si_{18}O_{64}$	14 : 18	1.29
7/8	87.5	$Na \quad Ca_7 \quad Al_{15}Si_{17}O_{64}$	15 : 17	$1.1\overline{3}$
8/8	100.0	$Ca_8 \quad Al_{16}Si_{16}O_{64}$	16 : 16	1.00

in the measurements. No lines have been drawn into the diagrams in figs. 1–8 in order not to bias the evaluation of the data.

4. The available measurements are consistent with the assumption that "kink points" in the low plagioclase series occur at n/8 mol % anorthite, where n = 2, 2, 4, 7, i.e. at 25, 37.5, 50 and 87.5 mol % anorthite. For the 32 (Al+Si) atoms and the 8(Ca+Na) atoms per unit cell (with $c \sim 14$ Å) these are possibilities for even-numbered partitions of both Al and Si as well as Ca and Na, i.e. values for which complete ordering of Al and Si is possible (table 1; cf. A. NIGGLI, 1967) assuming, for a part of these cases, lack of centrosymmetry.

DISCUSSION

For purposes of determination and structural interpretation the exact position of the curves may be useful to construct. This can be done by visual estimation or by statistical methods, which may be even applied to estimate the most likely position of the "kink points". When microprobe analyses are available for all sets of lattice constants (so that all points have equal weight in the statistical treatment), it is hoped that such methods can successfully be applied to construct curves which have been deliberately omitted in figs. 1–8.

The linearity of lattice changes between 0 and 25 mol % can be explained on the basis of the domain structure of oligoclases (KOREKAWA, 1969; NISSEN, 1969). Domains composed of two kinds of unit cells with Al/Si configurations corresponding to low albite and oligoclase (with approx. 25 mol % anorthite) have been suggested in these materials. The lattice constants of members intermediate in composition between the two kinds of cells are therefore a mean and correspond to the relative amount of the two kinds of unit cells present. A similar explanation of the lattice changes in the intermediate and Ca-rich plagioclases can be envisaged. However, in the intermediate plagioclases, from approx. 25 to 75 mol %, the domain textures and hence the X-ray satellites are very similar. The "kink points" near 37.5 and 50 mol % therefore must have another structural meaning than those at approx. 25 and 87.5 mol %. It is contended that at the latter two compositions only one kind of unit cell is present, the crystal is submicroscopically homogeneous, highly ordered with respect to Al/Si and possibly a stable phase. This is in good agreement with statistical data on the frequency of plagioclase compositions in plutonic and metamorphic rocks (HUNAHASHI et al., 1969; WENK, 1962; WENK and KELLER, 1969) supposed to represent equilibrium compositions. These statistics show compositional maxima at 25 and 87.5 mol %.

It is interesting to compare the suggested "kink points" of low plagioclases with the "kinks" in the changes of optical data. For this comparison three kinds of data may be taken from BURRI, PARKER and WENK (1967):

1. The diagram of 2 $V_{(x)}$ (Pl. XII).
2. The position of the optical directions n_α, n_β and n_γ with regard to crystal axes, expressed as stereographic projections (Pl. V–X) or as Eulerian angles (Pl. I–III).
3. (Dependent on 2.) The intersections of mean optical planes for different anorthite contents (fig. 3.6, p. 228 of WENK in BURRI et al., 1967).

2 $V_{(x)}$ shows a maximum at exactly 25 mol % for low plagioclases, a minimum at 50 mol % and a kink between 80 and 90 mol % anorthite.

The curves of n_α, n_β and n_γ in the stereographic projection show a kink (and thus a deviation from the regular position on small circles) between 25 and 30 mol %, between approx. 70 and 75 mol % (this is assumed here to be due to the bytownite unmixing gap; cf. NISSEN, 1968) and between 80 and 85 mol % anorthite. The same is indicated in the plots of Eulerian angles I, II, and III.

The optical planes, drawn from ten to ten mol % for low plagioclases, show a common intersection for 0, 10 and 20 mol % and another common intersection to a high degree of precision for low plagioclases with more than 30 mol % anorthite, i.e. the "break" is between 20 and 30 mol %.

We see that the two most important points in the plagioclase series, at 25 and 87.5 mol %, one separating oligoclases (incl. peristerites) from the "intermediate structure" with its typical satellites, the other probably representing the exact case of the "body-centered" bytownite structure, are well marked in the optical data, while the "kink points" at 37.5 and 50 mol % falling within the range of the intermediate structure are not indicated. The latter seem to represent points delimiting compositional ranges with minor differences within the range of the "intermediate structure".

It is hoped that the diagrams given here can be renewed soon, when more and possibly more accurate data have been gathered.

Acknowledgement

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LITERATURE

SMPM = Schweiz. Mineralogische und Petrographische Mitteilungen.

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