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The Lattice Constants and Related Parameters of "Plagioclases (low)"

(Part IV of Laboratory Investigations on Plagioclases)

By H. U. Bambauer (Münster)*), E. Eberhard and K. Viswanathan (Zürich)**)

With 5 figures and 1 table in the text

Zusammenfassung

Von 36 Plagioklasen werden genaue Gitterkonstanten bestimmt und daraus Achsenverhältnis und σ -Winkel des Rhombischen Schnittes berechnet. Die Werte werden graphisch dargestellt und daraus wird versucht, den Verlauf der Grenzkurven für die Reihe "Plagioklas (tief)" anzugeben. Die Kurven zeigen Diskontinuitäten bei An_{1,3}, An₁₆, An₃₃, An₅₀ und An₇₆₋₈₀. Ferner wird ein Diagramm zur Indizierung von Plagioklas-Pulveraufnahmen für den Bereich 2 $\theta = 13^{\circ}$ —54° (Cu K α 1) mitgeteilt.

Abstract

The unit cell dimensions of 36 plagioclases are determined; using these values the axial ratios and the σ -angles of the rhombic section are calculated. The variations in all these parameters have been correlated with the chemical composition of the specimens. An attempt is made to determine the boundary curves, representing the series "plagioclase (low)". Such curves exhibit discontinuities at An_{1.3}, An₁₆, An₃₃, An₅₀ and An₇₆₋₈₀. In addition, a new diagram is presented as an aid to indexing powder patterns of plagioclases in the 2 θ -range 13°—54° (Cu K α 1).

I. INTRODUCTION

The variation of the positions of the lines in the powder patterns of plagioclases, discussed in the accompanying paper (BAMBAUER et al.,

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1967), necessarily depends upon the variation of lattice constants that differ on account of changes in chemical composition and structural state. Therefore, a determination of the six lattice constants should give additional information on the variability of plagioclase-states. The numerous lattice constants to be found in the literature are not used in this investigation for the following reasons: It was felt that at this stage "relative" values are more indicative than "absolute" ones. As different investigation-methods may have different limits of error, especially if chemical analyses are considered, the same X-ray method and the same analytical method were used to determine the properties of all the plagioclases in this investigation. As to the chemical composition of plagioclases described in the literature the following may be said: Frequently the Ancontent was determined indirectly (e.g. optical methods). Further, all the analyses reported in the literature are bulk analyses, which do not take into consideration the inclusions of other minerals or the antiperthitic potash feldspar. In some cases the An-content has been inferred from the bulk Ca-content alone.

Whereas in earlier papers (Brown, 1960; Claise, 1950; Doman et al., 1965; Goodyear and Duffin, 1955; Smith and Yoder, 1956; Smith, 1956, 1958) values e. g. $2\theta_{131}$ — $2\theta_{1\bar{3}1}$ or γ^* already lead to interesting conclusions regarding the behaviour of plagioclases with respect to their chemical composition, a determination of all the cell constants should be more indicative. Therefore, 36 of the feldspars used by Corlett and Ribbe (1967) and by Bambauer et al. (1967) were selected for this investigation. In addition the knowledge of all the cell constants would make it possible to calculate other values of geometrical interest, e. g. σ , the angle of the "rhombic section". The term "plagioclase (low)" is here used to mean the series of plagioclases which extend from albite (low) to antorthite (primitive) and is in a "most ordered" state as far as nature provided it. Both the end members have maximum ordered Al/Si-distributions (Ribbe et al., 1962; Megaw et al., 1962).

II. EXPERIMENTAL

A. Material investigated

The following specimens (36) were selected for the determination of lattice constants from those listed by Corlett and Eberhard (1967):

- a) A representative number (25) of those specimens which were used to draw the boundary curves "plagioclases (low)" in the Δ (θ)-diagrams of Bambauer et al. (1967).
- b) A few other specimens (11) which were intersting because of their higher Or-content or their anomalous Δ (θ)-values. The data for the 36 specimens are summarised in Table 1.

B. The determination of lattice constants

a) X-ray method

Powder photographs were taken using an AEG-Guinier camera and $\operatorname{Cu} \mathbf{K} \alpha_1$ -radiation. Silicon ($a = 5.4305 \,\text{Å}$) was used as an internal standard.

b) Measurement

As far as possible the same films were used from which the Δ (θ)-values (Bambauer et al., 1967) were determined. A few poorly exposed photographs were retaken.

All the lines in the approximate 2θ -range 20° — 50° with sufficient intensity were measured (accuracy of measurement 2×10^{-3} mm). Depending on the sharpness and intensity of the photographs, this corresponds to between 40 and 120 lines per photograph. The reproducibility of the positions of the lines in especially sharp photographs is better than 1×10^{-2} mm.

The influence of film shrinkage and camera radius on line positions were corrected by means of the standard substance. During the calculations it was found that no difficulties arose from the relatively long intervals between the powder lines of the standard substance.

c) Calculations

The prerequisite for calculating the lattice constants was a satisfactory indexing of the powder pattern. This was accomplished in the following way: The positions of the powder lines of an albite pattern given by SMITH (1956) were marked on the abscissa of Fig. 1. The positions of the corresponding lines of many plagioclases were marked at the appropriate values on the ordinate, which correspond to the different compositions. The points having the same indices are joined by lines (Fig. 1)¹). The refinement of the lattice constants was carried out with

¹⁾ An-rich plagioclases have a doubled (~14 A) c-dimension. Since it is difficult to determine in powder photographs the boundary of the superstructure region, the superstructure reflexions are omitted.

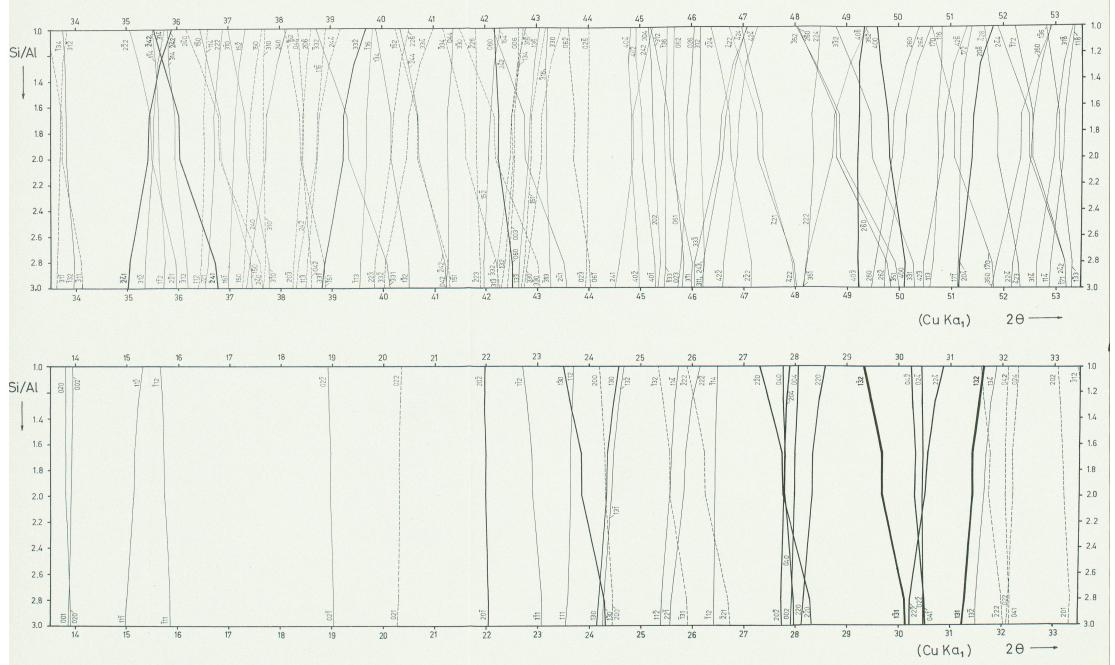
Table 1. Lattice constants and related parameters of 36 plagioclases

Nr.	194	167	163	110	278	84	171	ÇI	104	29	162	288	166	114	170	193	101	136	10	94	212	237	16,	74	81	84	96	11	45	103	61	36	300	567	011
ь	32.70	32.25	32.03	30.70	22.38	19.77	18.48	16.88	13.60	17.30	17.00	14.30	15.47	7.88	10.53	10.83	9.25	4.25	1.73	-0.87	4.38	5.07	5.27	3.18	2.75	-0.48	-0.53	0.23	-4.42	-5.10	-7.75	-8.83	-11.0	-10.83	-15.15
c: b	0.5599	0.5599	0.5599	0.5592	0.5578	0.5571	0.5564	0.5563	0.5555	0.5562	0.5564	0.5556	0.5560	0.5543	0.5549	0.5550	0.5542	0.5534	0.5535	0.5528	0.5533	0.5531	0.5535	0.5531	0.5527	0.5526	0.5523	0.5526	0.5521	0.5519	0.5516	0.5514	0.5512	0.5513	0.5507
a: b	0.6367	0.6367	0.6366	0.6367	0.6365	0.6361	0.6358	0.6358	0.6354	0.6358	0.6363	0.6358	0.6355	0.6347	0.6353	0.6355	0.6351	0.6346	0.6353	0.6348	0.6353	0.6352	0.6354	0.6358	0.6351	0.6355	0.6356	0.6355	0.6355	0.6355	0.6356	0.6356	0.6356	0.6356	0.6355
Δ	664.5	663.9	664.2	665.6	665.1	665.4	666.5	666.1	666.4	667.2	667.3	667.3	666.7	666.3	667.3	666.7	668.1	6.799	669.2	669.1	668.1	669.4	667.9	668.5	668.4	668.5	670.0	668.8	668.9	669.2	669.5	668.8	668.7	669.1	669.5
*,	90.45	90.40	90.40	90.31	89.70	89.53	89.45	89.33	89.11	86.38	89.35	89.16	89.24	88.73	88.91	88.94	88.85	88.52	88.58	88.24	88.55	88.60	88.61	88.47	88.44	88.23	88.22	88.27	87.93	82.88	87.71	87.63	87.47	87.48	87.18
*	63.50																																		
*8	86.40																																		
*0	0.1565																																		
* q	0.078428																																		
* d	0.1374	47.5	374	372	372	371	370	370	370	369	369	369	370	370	369	369	368	367	366	998	366	365	998	365	365	365	363	364	363	363	362	363	363	363	202
٨	87.69	80.18	87.76	87.85	88.45	88.63	88.74	88.84	89.07	88.80	88.85	89.01	88.94	89.45	89.28	89.26	88.38	89.71	89.68	90.06	89.71	89.66	89.64	89.79	89.81	90.03	90.04	86.68	90.31	90.36	90.55	90.63	90.80	90.79	91.11
β	116.59																																		
8	94.25																																		
ပ	7.159	7.157	7.158	7.156	7.142	7.138	7.137	7.134	7.130	7.137	7.137	7.131	7.134	7.120	7.127	7.124	7.124	7.116	7.119	7.114	7.113	7.116	7.113	7.110	7.108	7.107	7.109	7.107	7.102	7.102	7.099	7.096	$\frac{7.093}{2}$	7.096	080.7
q	12.786	19.789	12.785	12.798	12.804	12.814	12.826	12.824	12.834	12.831	12.827	12.836	12.830	12.847	12.843	12.837	12.853	12.859	12.862	12.869	12.856	12.865	12.851	12.855	12.862	12.861	12.870	12.862	12.865	12.869	12.870	12.869	12.868	178.21	12.875
ಜ	8.141		-	_	$\overline{}$	$\overline{}$	-	-	$\overline{}$	_	-	-	_	-	$\overline{}$	_	-	-	\blacksquare	_	-	\vdash	-	_	$\overline{}$	_	-	_	$\overline{}$	_	-	_	-	-	-
0r	0.3	7.0	9.0	1.4	0.5	0.7	9.0	6.0	0.5	6.0	3.9	6.0	1.2	0.3	1.0	1.0	8.0	0.4	4.2	0.4	0.4	1.3	0.2	1.0	8.0	1.0	1.2	0.3	0.5	0.7	0.6	0.3	0.5	0.0	0.0
An	0.0	0.0	1.1	11.1	13.0	16.5	17.2	17.6	17.9	18.5	20.5	21.0	21.5	23.9	25.3	25.6	28.0	35.3	37.4	41.8	43.2	13.7	47.8	53.00	53.7	54.3	58.1	59.8	58.7	39.0	73.1	0.77	80.0	81.8	93.1
Si/Al	3.0																																		
Nr.	194																																		

Nr.: specimen number given by Corlett und Eberhard; Si/Al: Si/Al-ratio derived from An-content; An: mol-% An from microprobe analyses by Corlett and Ribbe; Or in the matrix; a, b, c: cell edges given in Å-units. The standard deviation is usually ± 0.001 Å; a^* , b^* , c^* : reciprocal cell edges given in Å-1; α , β , γ : cell angles in degrees. The deviation is mostly within 0.01° ; α^* , β^* , γ^* : reciprocal cell angles; V: Volume of the cell, calculated from the lattice constants; σ : angle of the rhombic section.

----: These lines are not readily visible; however they are

marked in the 2 θ -range 13°—45°.



----: The pair $131/1\overline{3}1$, which is important diagnostically.
-----: All other readily visible lines in the 2θ -range 13° — 54° .

Fig.1. Relations between 2θ (Cu K α 1) and chemical composition (given as Si/Al-ratio) in the powder patterns of "plagio-clase (low)".

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the help of the CDC-1604-A-computer, using the lattice constants refinement programm ZH = LCLS²). For every sample approximate lattice constants, inferred from literature data, were used as input. It was usually observed that after the completion of only two cycles the standard deviation did not exceed ± 0.0015 Å for lattice edges and $\pm 0.01^{\circ}$ for lattice angles. In a very few cases the punched cards for some lines had to be removed because of unusually large differences between the calculated and the measured values. These discrepancies may be due to false measurement or false indexing. From Fig. 1 can be seen that such errors in indexing are possible for certain reflexions near the points of intersection of lines. The refined lattice constants and other parameters calculated from these lattice constants are given in Table 1 and the data are presented graphically in Figs. 2, 3 and 5. For the construction of the boundary curves for "plagioclases (low)", shown in the figures, only the 25 points, which satisfied the condition II A a), were considered. Because of the scatter of the plotted data, the drawing of these curves presented some difficulty. Since the behaviour of a few points (e.g. 36 and 84) was always "good" the curves could be drawn using them as reference points. The behaviour of a point is supposed to be "good", if it shows the same deviation (in all the figures) from the general trend (i. e. an average line) indicated by all the other points.

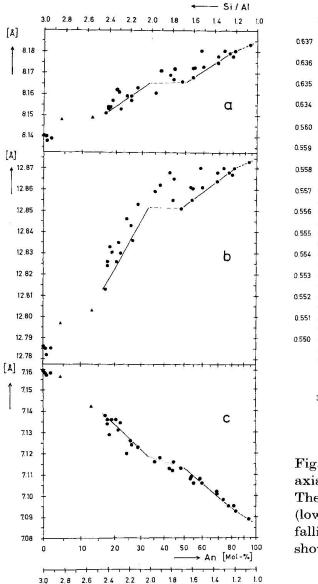
III. RESULTS

According to Brown (1960) a nearly linear relation between the lattice constants and Si/Al-ratios is to be expected: Thus Figs. 2, 3 and 5 have the Si/Al-ratio as abscissa. The following relations are observed in the diagrams:

- 1. In general the lattice constants of "plagioclases (low)" vary with increasing An-content in the following ways:
- a) a and b increase, c decreases; the corresponding a^* , b^* and c^* behave in the opposite way (no diagrams are given for these parameters).
- b) γ increases and β decreases; the corresponding γ^* and β^* vary in the opposite way; α and α^* behave differently: both of them decrease.
- c) The angles γ and γ^* vary much more than the other lattice angles.

²) We thank Dr. Charles W. Burnham who made available to us his programm LSLSQ and Dr. M. Fehlmann for rewriting it to suit the CDC-computer.

- d) a: b varies very little, but c: b shows a distinct decrease. The data available in the literature generally show the same trends (see e.g. Smith, 1956; Brown, 1960).
- 2. There are discontinuities in the lattice-constant/An relations (Figs. 2 and 3) analogous to those of the Δ (θ)/An curves presented by Bambauer et al. (1967). The distinctness of these discontinuities is different for different lattice constants. In Fig. 3 for example, γ^* shows distinct breaks at Si/Al = 2.02 and 1.67, whereas the β^* -curve shows an almost



SI/AL

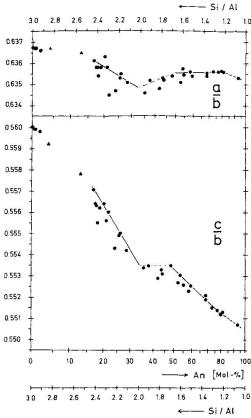


Fig. 2. Relations between cell edges, axial ratios and chemical composition. The boundary curves for "plagioclases (low)" are shown. ▲: plagioclases falling in the peristerite region, but showing a "single phase" powder pattern.

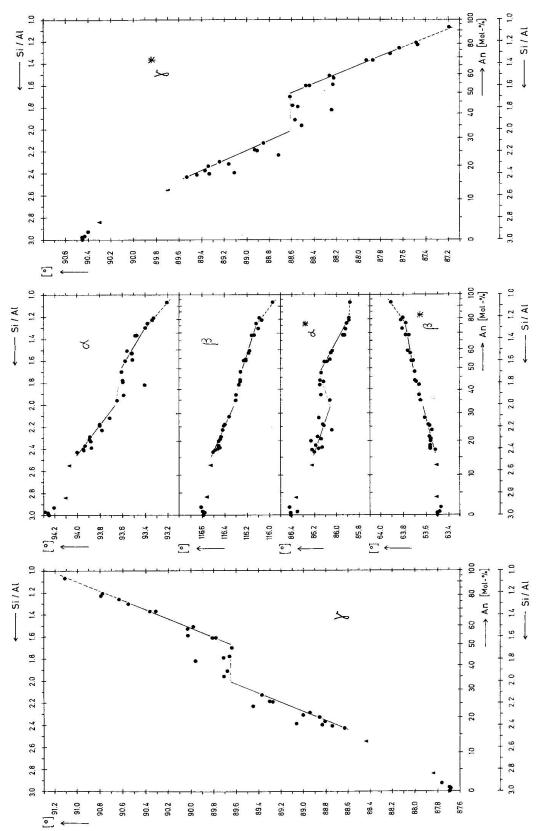


Fig. 3. Relations between lattice angles and chemical composition. The boundary curves are for "plagioclases (low)". ▲: see Fig. 2.

linear behaviour in this range. Considering Figs. 2 and 3 the boundary curves can be subdivided into parts corresponding to:

- a) Si/Al = 3.0 to appr. 2.97 $(An_{0.0-1.3})$: Range of albite (low).
- b) Si/Al = 2.97 2.45 (An_{1.3-16}): Peristerite gap.
- e) Si/Al = 2.45 2.02 (An₁₆₋₃₃): Linear within the limits of experimental error.
- d) Si/Al = 2.02—1.67 (An₃₃₋₅₀): As in the case of Δ (θ)-values uncertainities still exist.
- e) Si/Al = 1.67 1.25 (An₅₀₋₇₆): Linear within the limit of experimental error.
- f) Si/Al = 1.25 1.00 (An₇₆₋₁₀₀): No comments can be made about this region at this stage.
- 3. The Or-content has the greatest effect on a and on a^* . This suggests an analogy to the alkali feldspars. However, due to the insufficient data, no precise information can be given on such a correlation.
- 4. The determination of the lattice constants of both components of peristerites seems to be but rarely possible by the powder method. Among the intense lines which may be indexed with certainity only the paris $131/1\overline{3}1$ and $\overline{2}41/\overline{2}41$ are resolved. Peristerites are, as hitherto, best determined either by single crystal methods (Laves, 1954; Brown, 1960) or by electron microscopy (Ribbe, 1960; Fleet and Ribbe, 1965).

Among the investigated specimens there were occasionally plagioclases whose Δ (θ)-values fell on the interpolated plagioclase (low)-curve in the peristerite-gap. The powder patterns of such specimens could not be differentiated from those of homogeneous phases. The lattice constants were determined for two such specimens (Nrs. 110 and 278). One of them seems to be a peristerite because of its bluish-white schiller. In Figs. 2 and 3 the values of its lattice angles fall rather exactly on to the line interpolated between $An_{1.3}$ and An_{16} , while those of its lattice edges deviate from this line.

- 5. Using the lattice constants derived from Figs. 2 and 3 the following quantities were calculated:
- a) Relations between $2 \theta \operatorname{Cu} K \alpha_1$ and An-content to check Fig. 1. This diagramm was originally intended for indexing the powder patterns of the series "plagioclases (low)". However, plagioclases show the puzzling phenomenon that the lattice constants of a "high" or "intermediate"

plagioclase are approximately the same as those of a "plagioclase (low)" having a different An-content. This means that a powder pattern of any plagioclase, whether "high" or "intermediate" must be almost the same as that of a plagioclase (low); (ignoring the differences in the intensities of the lines). Therefore this diagram can serve to index powder patterns of plagioclases of all compositions and structural states. The best way to index the powder patterns is to measure the value $2 \theta_{131}$ — $2 \theta_{1\bar{3}1}$ in it. Using this value the corresponding Si/Al-ratio given by the boundary curve for "plagioclases (low)" in the diagram (Fig. 5) of BAMBAUER et al. (1967) must be sought. For the Si/Al-value so obtained, Fig. 1 gives the indices along the abscissa in the order of increasing 2θ .

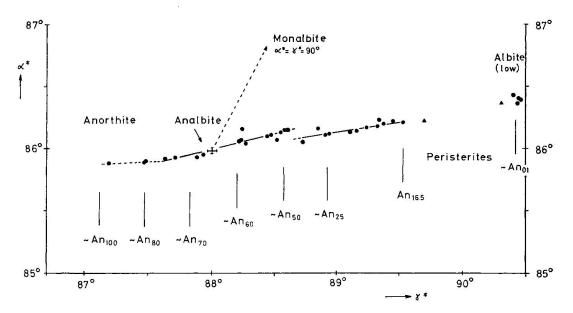


Fig. 4. Relations between α^* and γ^* . The An-calibration is valid only for "plagio-clases (low)". \triangle : see Fig. 2.

b) Relations between Δ (θ)₁, Δ (θ)₂ and An-content. The agreement between the calculated and observed values of Δ (θ)₁ and Δ (θ)₂ appears to be rather good considering the accuracy obtainable by the methods of this investigation. In addition an attempt was made to calculate Δ (θ)-curves for an Or-content of 4 mol-% using the lattice constants of microcline (low), and assuming that Vegard's law is approximately applicable for mixed crystals plagioclase (low)-microcline (low). The agreement between the calculated and observed values is satisfactory in the case of Δ (θ)₁ but poor for Δ (θ)₂, as can be seen from Figs. 5 and 6 of Bambauer et al. (1967).

c) The angle σ of the rhombic section using the relation etg $\sigma = \frac{\cos \alpha^*}{\operatorname{ctg} \gamma}$. Fig. 5 shows the curves for "plagioclases (low)" and "plagioclases (high)". The curve "plagioclase (high)" was calculated from lattice constants, determined for synthetic specimens of Eberhard (1967).

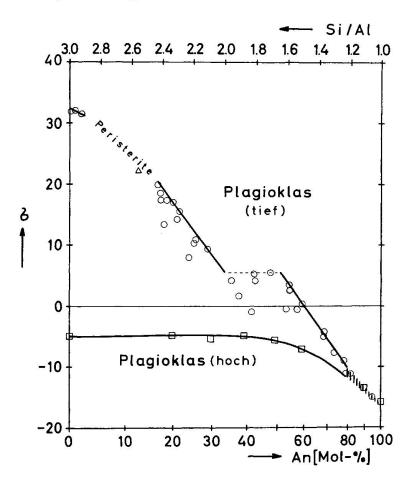


Fig. 5. Relation between the σ -angle of the rhombic section and chemical composition for natural "plagioclases (low)" and synthetic "plagioclases (high)".

IV. DISCUSSION

A. The shape of the curves

The position of the curves cannot be given with certainity in the region Si/Al = 3.0 - 2.97 (Figs. 2-4); therefore they were omitted. However general trends are recognizable.

It is found that the values for "plagioclases (low)" and "plagioclases (high)" nearly coincide in the range An_{76-100} (Fig. 5). Therefore it may

be difficult to distinguish between these two series by powder-diagrams if the An-value is larger than appr. An₇₅. Further additional complications may arise because of the variety of domain-order/disorder states described by Brown et al. (1963). Thus the curves An_{76-100} have not been extended to An_{100} , and are shown only as dotted lines. Future corrections are not expected to affect the main trend of the curves, but amendments in details may be possible.

While the limiting curves for "plagioclases (low)" of the range An_{16-33} and An_{50-76} are considered to be good approximations to straight lines, uncertainties exist for the interval An_{33-50} . Therefore, in Figs. 2, 3 and 5, dotted lines represent an interpolation of our data in this region. Future investigations of additional material are necessary.

B. Comparison with data in the literature

It is assumed that the material, which was selected for this investigation from about 300 specimens is representative for the most ordered state of "plagioclase (low)" produced by nature. Some of the published values of the lattice constants for plagioclases show very good agreement if plotted in Figs. 2 and 3; other values deviate considerably to both sides of the curves presented. An explanation of this fact cannot be given at this stage. Perhaps the scatter may be due to inhomogeneities of the material investigated. The bulk composition of a "crystal" may not be representative of the homogeneous "host"-phase which is responsible for the position of the powder lines measured. As is now well known the relation of the powder data of the "plagioclases (low)" to the An-content exhibits discontinuities. Such discontinuities became obvious with varying distinctness in the data published by Claisse (1950) at An₃₀₋₄₀; by Goodyear and Duffin (1954, 1955) at An_{30} and An_{70-80} ; by Smith and Yoder (1956) at An_{30-40} and by Smith and Gay (1958) at An_{35-40} and An_{50} .

A comparison between literature data and those presented in this investigation is possible in certain cases. Goodyear and Duffin (1955) show breaks in the γ^* -curve at about An_{30} and An_{70} . The data of Smith (1956) refer mainly to the region An_{16-33} . The data of Brown (1960) reveal some interesting features. However, since the precession method and the bulk An-content (without Or) were used in this work, the resolution of his data with respect to boundary curves was less than that of the data presented in this investigation. If the smooth curves drawn by

Brown (relating γ^* and $(010) \wedge (\overline{1}01)$ with chemical composition) are reexamined, there are strong indications of discontinuities at An₃₀ and An₅₀₋₆₀. His α^*/γ^* diagram is very similar to Fig. 4 in this investigation.

Until now the most detailed information on structural discontinuities in the series "plagioclases (low)" is from Doman et al. (1965). Relating γ^* (determined from single-crystal photographs) with An-content (determined mainly by refractive indices), these authors found that $\gamma^*/\text{An-values}$ group themselves along more or less parallel, mutually displaced lines. Their diagrams indicate discontinuities at about An₃, An₁₆, An₃₃, An₅₀ and An_{92–96}. Neglecting indications found during this investigation³) that An_{1,3} is a maximum value for the An-content in albite (low) and that the upper limit of the peristerite gap occurs at An₁₆, there is a relatively good agreement between the present values and those of Doman et al. However, whereas Doman et al. observed mutually displaced "segments" in the pertinent curves, we only observed "breaks" in them. Further work appears to be necessary for clarifying the situation. In addition, there are the following differences between the results of Doman et al. and those of this investigation:

- a) Doman et al. find a discontinuity at appr. An₉₄ whereas our data indicate such a discontinuity at appr. An₇₅ (it should be noted that within the An-rich region the $\gamma^*/\text{An-diagram}$ of Doman et al. is hardly indicative).
- b) There is no agreement in the region An_{33-50} . In this range our γ^* -values are always $\geq 88.61^\circ$ and the γ^* -values found in the literature are always $\geq 88.69^\circ$. Doman et al. however show an isolated line in this region, which runs more or less parallel to the general trend of the γ^*/An -relations from $\gamma^* = 88.22^\circ$ (An_{52}) to $\gamma^* = 89.08^\circ$ (An_{34}). It should be taken into consideration that all the points on this curve with $\gamma^* > 88.75^\circ$ are determined by the use of one, perhaps inhomogeneous, plagioclas specimen (GD 1805) from a ferrogabbro. This specimen appears to be of considerable interest, as no similar values of γ^* have been obtained by us for plagioclases of this range. (As according to our experience, plagioclases from such rocks are likely to have a high Or-content which cannot be determined optically, it would be of interest to examine the specimen GD 1805 of Doman et al. with a microprobe analyser for any "impurities" that may be present in the matrix in solid solution.)

³) This value is based on an analysis of the Ab-rich phase of a peristerite using the electron microprobe analyser.

C. Significance of the discontinuities

The reasons for the discontinuities, found and depicted in Figs. 1—5, are still unknown. Structure determinations (as far as Al/Si-distributions are concerned) of chemically intermediate "plagioclases (low)" are very desirable. In addition, electron microscopy (Laves et al., 1965) and very high resolution X-ray techniques developed by Jagodzinski and Korekawa (1965) may lead to a better understanding of the variety of feldspar structures. This variation appears to be due to their Al/Si distributions, the significance of which was first envisaged by Barth (1934). A mathematical treatment of the possibilities of Al/Si order under the assumption that no Al-tetrahedra have common oxygen corners is given by Niggli (1967) in an accompanying paper.

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