

PHYSICAL PROPERTIES OF CALCIC LABRADORITE  
FROM LAKE COUNTY, OREGON<sup>1</sup>

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ABSTRACT

The physical properties of transparent calcic labradorite phenocrysts, USNM 115900, found in basalt in Lake County, Oregon, are described in detail. The phenocrysts occur in a groundmass of labradorite, augite, olivine, magnetite, and the alteration products of these minerals. The cores of the phenocrysts are not zoned and comprise at least 90% of the volume; the rims are complexly zoned toward more sodic compositions. The high-intermediate structural state of the feldspar as deduced by optical and X-ray measurements seems to be typical of the plagioclase in rapidly quenched hypabyssal and volcanic basaltic rocks.

Chemical analysis showed the composition to be (mol per cent) An 67.2 Ab 31.5 Or 1.3. Observed S.G. 2.720, calculated density 2.710 g/cm<sup>3</sup>. Space group  $I\bar{1}$ . Unit-cell parameters:  $a=8.176 \text{ \AA}$ ,  $b=12.865$ ,  $c=14.204$ ,  $\alpha=93^\circ 27.1'$ ,  $\beta=116^\circ 2.9'$ ,  $\gamma=90^\circ 30.6'$ , molar volume=100.84 cm<sup>3</sup>/mol. A powder diffraction pattern indexed on the  $CI$  sub-cell is given. Optically (+),  $2V=85^\circ$ ,  $\alpha=1.5625$ ,  $\beta=1.5668$ ,  $\gamma=1.5718$ .

The cell-edge parameters and linear and volume expansion were determined for 10 temperatures up to 1141° C. An equation is given for the variation of molar volume with absolute temperature; differentiation of the equation yields the thermal expansion, but the values may not be useful at high temperatures because of complications introduced by a possible step on the volume curve that may be caused by a reversible high-order transformation between 650° and 1050° C. It is suggested that this transformation may be the change from a body-centered lattice to a C-centered lattice. It may be possible to establish an equilibrium phase diagram for calcic plagioclases by high temperature X-ray measurements, and to determine the kind and order of the transformations that occur.

INTRODUCTION

Correlation of the physical properties of feldspar with composition has been greatly aided by the availability of samples of uniform high quality and purity, such as the suites described by Spencer (1937) and by Emmons and his coworkers (1953). We here record some of the physical properties and the availability of a sample of calcic labradorite of high quality and a somewhat unusual structural state, intermediate between high and low, but more similar to the high structural state. Many, but not all, of the physical properties of this feldspar, or of material very similar to it, have been recorded earlier (Cole, *et al.* 1951, No. 10, Table 2; Emmons, ed., 1953, No. 19, Table 1; Smith and Yoder, 1956, No. 19(61), Table 2). Remarkably similar crystals also have been obtained from dikes at St. John's Point, near Ardglass, County Down, Ireland (Hutchinson *et al.*, 1912; Barber, 1936; Muir, 1955; Gay, 1956; Joel and Muir, 1964), and from a number of other gabbroic or basaltic rocks mostly of hypabyss-

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177  
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sal or extrusive origin. The composition of our sample is representative of the petrologically important plagioclase from layered mafic intrusives, gabbros and basalts, and the data presented should be widely applicable.

*Availability of Lake County plagioclase.* A number of crystal fragments and several small bottles of cleaned and sized powdered samples have been deposited in the U. S. National Museum under USNM number 115900.

#### GEOGRAPHY AND GEOLOGY

Erosion of highly porphyritic basalt flows in south-central Oregon resulted in lag gravels and poorly developed desert pavement that contain abundant large fragments of exceptionally pure calcic labradorite. The labradorite, locally referred to by mineral collectors as sunstone, is readily obtainable at about 42°43'20" N., 119°51'50" W., in east-central Lake County, 22 miles N. 5° E. of the hamlet of Plush. The locality is in sec. 10 (projected) T. 33 S., R 24 E., Willamette meridian. This may be the same locality briefly mentioned by Aitkens (1931, p. 8). Emmons' sample (1953, No. 19, Table 1) of similar labradorite from Lake County, Oregon, was purchased from a mineral supply house, and the locality was not further specified.

The labradorite described in this paper occurs both as phenocrysts in highly porphyritic basalt flows, mostly about 10 to 20 feet thick, of middle or late Miocene age, and as abundant detrital pebbles and crystal fragments in lag gravels and desert pavement of late Pleistocene (?) age on a gentle dip slope of the basalt. Concentrations of the detrital labradorite are found in pluvial (lake) beach gravels, which are part of an extensive, though thin and discontinuous, veneer of silty and gravelly lacustrine sediments deposited on a weathered and eroded surface on the basalt.

Petrographically, the basalt flows are similar to some plagioclase-rich basalts described from adjacent areas, as for example the Steens Mountain Basalt of Fuller (1931), widely exposed 60 miles to the east, and the Lake Basalt of Powers (1932, p. 286-287), exposed in the Modoc Lava beds about 100 miles to the southwest. Highly porphyritic basalts with nearly identical modes also are exposed on Abert Rim, about 10 to 15 miles to the southwest. Typically these basalts contain about 48 to 50% SiO<sub>2</sub>, 18 to 19% Al<sub>2</sub>O<sub>3</sub>, 10% combined Fe<sub>2</sub>O<sub>3</sub> and FeO, 10% CaO, 5% MgO, 3% Na<sub>2</sub>O, and less than 1% K<sub>2</sub>O (Fuller, Table 8, 1931; Powers, Table 1, 1932; unpublished analyses by U. S. Geological Survey).

*Petrography of the basalt.* The basalt that contains the large labradorite phenocrysts typically is holocrystalline (Fig. 1) and exhibits intergranu-

lar and, locally, diktytaxitic textures; ophitic to subophitic clinopyroxene is characteristic. The plagioclase phenocrysts are sporadically distributed and represent 10% of the rock in some places and as much as 55% of the rock in other places. Concentrations of plagioclase phenocrysts are present in the somewhat vesicular upper parts of flows, or, less commonly, near the bottom of flows. In some parts of flows the phenocrysts are preferentially oriented either parallel to the main cooling surfaces or to internal zones of flow-contorted rock. The labradorite phenocrysts occur,

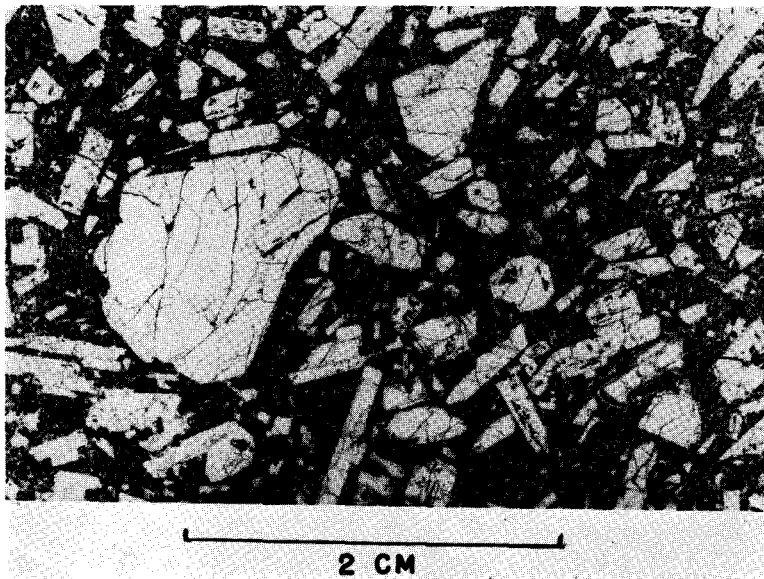


FIG. 1. Photomicrograph of porphyritic basalt showing partly corroded, rounded, and fractured calcic labradorite phenocrysts in basaltic groundmass.

in place, as idiomorphic crystals, partly the result of late overgrowths of somewhat more sodic labradorite, as rounded and embayed crystals or parts of crystals, and, in a few places, as glomeroporphyritic aggregates.

The groundmass is composed of labradorite ( $\approx \text{An}_{60}$ ) microlites (about 50%), augite with a measured (+)  $2V = 46^\circ \pm 3^\circ$  (about 21%), olivine that is partly altered to iddingsite (about 16%), magnetite (8%), and about 2% each of calcite and nontronite(?). The groundmass also contains about 2% of voids that characterize the diktytaxitic texture.

Most of the feldspar microlites are approximately  $\text{An}_{60}$  in composition as determined by extinction angle measurements. The structural state could not be determined directly but is probably intermediate to high by analogy with the structural state of the rims of the phenocrysts (see Fig. 4 and later discussion).

## PHYSICAL PROPERTIES OF THE LABRADORITE PHENOCRYSTS

*Appearance, size, habit.* Many of the plagioclase phenocrysts are tabular plates or large laths ranging greatly in size and shape. The smallest are about 1 mm in greatest dimension whereas the largest crystal observed during the present study is a giant lath 8.3 cm long, 2.6 cm wide and 0.8 cm thick. Some large feldspar pebbles in the lag gravels indicate that thicker and wider phenocrysts are present in some parts of the basalt flows.



FIG. 2 Photomicrograph showing thin peripheral zone and overgrowth on large calcic labradorite phenocryst. Transmitted light; partly crossed polars.

Most of the labradorite is exceptionally clear, translucent, and nearly colorless or pale golden yellow; flecks of hydrated iron oxides on indistinct and poorly developed cleavage impart a faint yellowish-pink hue to some crystals. Both phenocrysts and detrital pebbles generally have subconchoidal fractures. Many of the detrital fragments are partly rounded by erosion and commonly have frosted surfaces.

*Twinning and zoning.* Coarse polysynthetic twins are present in most crystals and are visible in some detrital pebbles as very broad, indistinct lamellae. Some of the broad lamellae may be twinned on the Carlsbad

law but most are albite twins; the latter are found as thin plates in most crystals observed in thin sections.

The labradorite phenocrysts exhibit some evidence of compositional zoning in thin section. In nearly all crystals, however, the zoning is restricted to a very thin peripheral shell; a part of one of the thicker and more continuous zoned shells is shown in Fig. 2, and schematically in Fig. 3. Several measurements on individual medium- to small-sized labradorite crystals indicate that the zoned material in the peripheral shells repre-

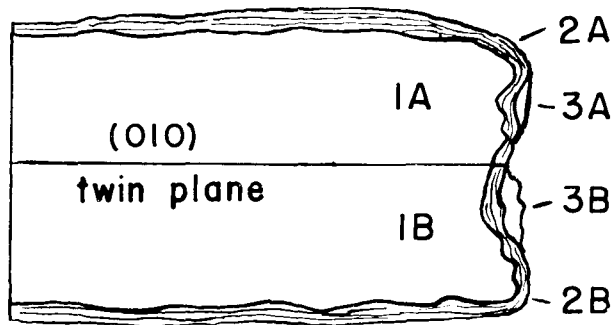


FIG. 3. Sketch of a zoned labradorite crystal. The albite-twinned crystal contains an unzoned core (1A; 1B) of approximately  $An_{68}$  composition and intermediate structural state. The border of the core is marked by resorption and a change in the extinction angle. The core is commonly covered by a rim of plagioclase with intermediate to high structural state and with oscillatory zoning from about  $An_{64}$  to  $An_{60}$ . The outer margin of the rim shows resorption and slight change in the extinction angle, or intergrowth with the groundmass. Additional feldspar (3A, 3B) is locally present on some crystals. This feldspar is strongly zoned normally from approximately  $An_{60}$  to  $An_{36}$ , as estimated from the maximum extinction angle in the zone perpendicular to (010). This material probably has a high structural state.

sents between 3 and 8 volume per cent of the phenocrysts; on the largest labradorite phenocrysts the volume percentage of the zoned material in the outer shells is vanishingly small. A few crystals have central, peripheral, or intermediate zones sparsely speckled with minute inclusions of groundmass material. The peripheral zoned shell has been largely removed by erosion from most of the detrital labradorite pebbles, and in the detrital material that was analyzed in this study no feldspar less calcic than  $An_{60}$  was detected.

*Chemical composition and specific gravity.* Table 1 gives a chemical analysis of our sample together with analyses of Emmons' Oregon sample and the material from St. John's Point, Ireland. The two analyses of Oregon material agree reasonably well, and both require the  $Fe^{2+}$  and  $Mg^{2+}$  ions

TABLE 1. ANALYSES AND OPTICAL PROPERTIES OF CALCIC LABRADORITE OF INTERMEDIATE STRUCTURAL STATE

Oxide	Lake County, Oregon, J. J. Fahey, analyst	Lake County, <sup>1</sup> Oregon, L. C. Peck, analyst	St. John's Point, Ireland, IV. Hutchinson and Campbell Smith (1912)
SiO <sub>2</sub>	51.42	51.08	52.33
TiO <sub>2</sub>	0.04	0.05	not reported
Al <sub>2</sub> O <sub>3</sub>	30.76	31.05	30.22
Fe <sub>2</sub> O <sub>3</sub>	0.24	0.43	0.40
FeO	0.17	0.12	not reported
MnO	not reported	0.01	not reported
MgO	0.05	0.22	not reported
CaO	13.42	13.85	12.52
Na <sub>2</sub> O	3.52	3.38	3.62
K <sub>2</sub> O	0.23	0.12	0.85
ΣH <sub>2</sub> O	0.04	0.06	0.36
Total	99.89	100.52 <sup>2</sup>	100.30
	Numbers of ions on the basis of 32(O)		
Si	9.341	9.293	9.514
Al	6.628	6.659	6.476
Fe <sup>+3</sup>	0.033	0.059	0.054
Ti	0.005	0.007	—
Mg	.013	.060	—
Fe <sup>2+</sup>	.026	.018	—
Mn	—	.002	—
Na	1.248	1.192	1.276
Ca	2.628	2.616	2.439
Sr	—	.015	—
K	0.053	.028	0.197
Ba	—	.001	—
ΣZ <sup>3</sup>	16.01	16.02	16.04
ΣX <sup>3</sup>	3.97	3.93	3.91
Mol % An	67.2	68.9	62.4
Mol % Ab	31.5	30.3	32.6
Mol % Or	1.3	0.7	5.0
α	1.5625 ± .0005	1.5634 (Av. 4 determ.)	1.5630
β	1.5668 ± .0005	1.5682 (Av. 4 determ.)	1.5665
γ	1.5718 ± .0005	1.5725 (Av. 4 determ.)	1.5712
2V <sub>γ</sub>	85° obs. 83.9 calc.	80-90° 85.3 av.	81°48' calc.
S.G.	2.720 ± 0.005	2.713-2.718	2.706

<sup>1</sup> In Emmons (1953), analysis 19, Table 2, p. 18.<sup>2</sup> Includes 0.011 BaO, 0.142 SrO, 0.0013 Li<sub>2</sub>O.<sup>3</sup> Calculated on the basis of 32 oxygens by procedure used in Deer *et al.* (1963, 107-111).

to be in solid solution in the feldspar. The small amount of Or in solid solution despite the relatively high structural state of the Oregon samples suggests that the Or content of the sample from St. John's Point indicated by the old analysis may be too high.

The density calculated for our Oregon sample, using the unit-cell

parameters in Table 3 and Fahey's analysis (Table 1), is  $2.710 \text{ g/cm}^3$ , in good agreement with the pycnometric determination of the specific gravity as  $2.720 \pm 0.005$ .

*Optical properties.* The optical properties of our material are compared in Table 1 with those of the two other samples. Allowance being made for the different methods of computation, our data would agree better with the curves of Chayes (1952, p. 96), Crump and Ketner (in Emmons, 1953, Fig. 6), and J. R. Smith (in Hess, 1960, plate 12), if our material were taken to be approximately 1.5% richer in An. No reason for the discrepancy is apparent.

It is not believed possible to determine the structural state of calcic labradorite by measurements of the refractive indices (Muir, 1955, p. 547; J. R. Smith, 1958). The value of  $2V$  is lower for high plagioclase in the compositional range under discussion, and  $2V$  may be used to estimate the structural state if the composition is known (J. R. Smith, 1958, 1188–1189). So estimated, our material is of intermediate structural state.

The optical method of determining structural state perfected by Uruno (1963) for albite twins was used to determine the structural state of our material (Fig. 4). The results indicate that the unzoned cores are close to  $\text{An}_{70}$  in composition, and that they are of intermediate structural state. The rims on the phenocrysts are more sodic (An 67-56), and are either of higher structural state than the cores, or about the same.

The maximum extinction angle of our sample in the zone perpendicular to (010) is  $-44^\circ$ .

Muir (1955, Table 1, p. 551, Fig. 6A, p. 555) showed that the labradorite from St. John's Point has an intermediate structural state because the (010) pole falls between the curves for (100) of the high and low structural states; heating at  $1000^\circ \text{ C.}$  for 72 hours resulted in the migration of the pole to the curve for the high structural state, and a slight decrease in  $2V$ . Secondary albite twinning developed during the heat treatment.

*X-ray crystallography.* Our colleague, Daniel E. Appleman, examined several single crystals of Lake County plagioclase at room temperature by the precession method. The unit cell was found to be triclinic,  $\bar{1}$ , with  $a = 8.17 \text{ \AA}$ ,  $b = 12.87$ ,  $c = 14.19$ ,  $\alpha = 93^\circ 17.1'$ ,  $\beta = 116^\circ 0.0'$ , and  $\gamma = 90^\circ 46.0'$ . The crystals showed strong type "a" reflections,<sup>1</sup> and weak

<sup>1</sup> For a recent review of the definitions and significance of the various types of reflections shown by plagioclase, see Gay (1962).

but sharp type "b" reflections. No other types of reflections were present. Similar results were obtained by Cole, Sorum, and Taylor (1951, No. 10 of Table 2, p. 22) for Emmons' material from Lake County.

Calcic labradorite from the basalt at Clear Lake, Utah, and from basaltic dikes at St. John's Point, Ireland, also has strong type "a" reflections, and weaker but sharp and single type "b" reflections (Gay,

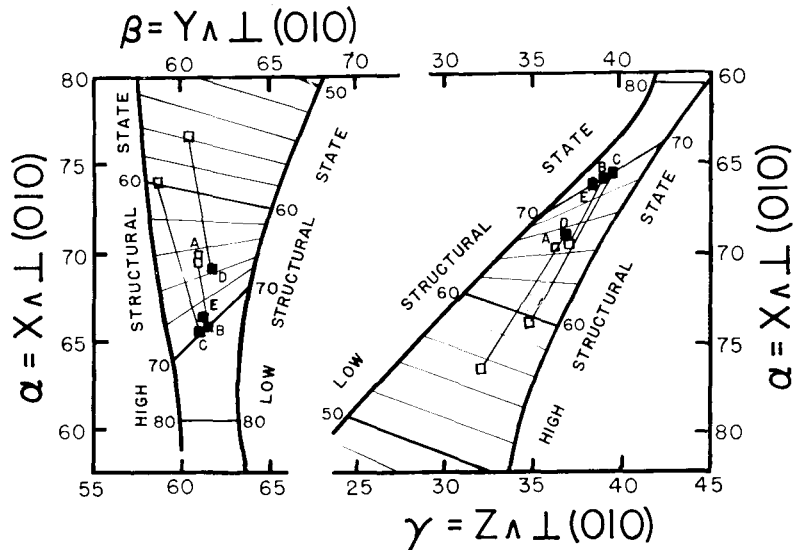


FIG. 4. Orientation of axes of optical indicatrix against (010) for albite twins plotted on the pertinent portions of the curves in orthogonal coordinates of Uruno (1963), modified by taking the angles for each individual of the twins (half of the Kohler angles). The unzoned cores are shown by solid squares and the outer shells (in reality individual zones within sequences of oscillatory zones) are shown by boxes that are connected by lines to the respective cores. The cores are of intermediate structural state, and the more sodic outer shells are of higher structural state than the cores.

1956, p. 32). Gay noted that although associations of type "a" and "b" reflections were common in plagioclase with composition lying between  $An_{70.75}$  and  $An_{80.85}$ , this association was rare in plagioclase less calcic than  $An_{72}$ . It occurs only in specimens from volcanic or hypabyssal environments; all labradorites from plutonic rocks characteristically have doubled, or "split," "b" reflections (now called "e" reflections).

The powder-diffraction lines from Lake County labradorite all correspond to "a" reflections. The strong "a" reflections describe a  $C\bar{1}$  sub-cell with unit-cell parameters like those of the true cell except that the  $c$ -dimension is halved ( $c_{sub} = 7.102 \text{ \AA}$ ). This sub-cell corresponds to



the true cell of sodic plagioclases like albite. The unit-cell parameters of the sub-cell were obtained by least-squares refinement of 20 powder-diffraction lines collected at 26° C. The refinement was accomplished with the fixed index routine of a computer program developed by Evans *et al.* (1963).

We prefer to use the refined sub-cell dimensions to describe our material because the powder-diffraction data are more precise. The sub-cell parameters obtained at 26° C. and the standard errors are given in the first line of Table 3. An indexed powder diffraction pattern is given in Table 2.

Our sample has an intermediate structural state as characterized by the parameters  $\Gamma$  and  $B$  of Smith and Gay (1958). The value of  $\Gamma$  is 1.055, and  $B$  is 0.790. These values are typical of those found in plagioclase of the same general composition from volcanic and hypabyssal rocks. For example, the very similar plagioclase from the dikes at St. John's Point, Ireland, has  $\Gamma=1.08$  and  $B=0.800$ . (*op. cit.*, Table 1).

After the high-temperature measurements described below, the values of  $\Gamma$  and  $B$  were 1.131 and 0.777, respectively, indicating that a higher structural state, close to the highest possible in natural material, had been obtained.

Smith and Yoder (1956, p. 638) reported that the angular separations of the (131) and ( $\bar{1}\bar{3}1$ ) lines and of the ( $\bar{1}\bar{3}1$ ) and (220) lines for Emmons' sample of Lake County labradorite were 2.10° and 1.00°  $2\theta$  CuK $\alpha$ , respectively. Our best values are 2.06°<sub>0</sub> and 1.00°<sub>5</sub>, respectively. Our values plot toward a lower (intermediate) structural state than Yoder and Smith's and are in better agreement with our other data. Their values fall on the curves for the highest structural states yet observed, those of plagioclase synthesized dry from glass.

*Thermal expansion.* The thermal expansions of a series of plagioclases were measured by Kozu and Ueda (1933), but they lacked suitable material between An<sub>43</sub> and An<sub>95</sub>. The composition of our sample is appropriate to help fill this gap, and is representative of the petrologically important plagioclase from layered mafic intrusives, gabbros and basalts.

Data were collected using a heating stage for the x-ray diffractometer (Skinner *et al.*, 1962). The thermocouple was calibrated in the measuring circuit used by reference to the melting points of tin, silver, and gold. Temperature control was within 1% of the value given. The movement of the stage with temperature was corrected by calibration with the position of the (200) line of MgO (CuK $\alpha=42.938^\circ 2\theta$  at 25° C.) using Campbell's values for the linear expansion (Campbell, 1962, Table 10) and MgO from a split of Campbell's interlaboratory standard. Refine-

TABLE 2. INDEXED POWDER-DIFFRACTION PATTERN FOR CALCIC  
 LABRADORITE FROM LAKE COUNTY, OREGON

Triclinic  $\text{Ca}_{0.7}\text{Na}_{0.3}\text{Al}_{1.2}\text{Si}_{2.8}\text{O}_8$ ;  $\bar{1}\bar{1}$ , but indexed on a  $C\bar{1}$  sub-cell with  $a=8.176 \text{ \AA}$ ,  $b=12.865$ ,  $c=7.102$ ,  
 $\alpha=93^\circ 27.1'$ ,  $\beta=116^\circ 2.9'$ ,  $\gamma=90^\circ 30.6'.^1$

<i>hkl</i>	Calculated <sup>2</sup>		Measured <sup>3</sup>	
	<i>d</i> <sub><i>hkl</i></sub> (Å)	$2\theta_{\text{CuK}\alpha}$	$2\theta_{\text{CuK}\alpha}$	$I^\circ/I_{100}$
$\bar{1}10$	6.482	13.660	13.64	1
020	6.416	13.802		
001	6.364	13.914		
110	6.266	14.135		
$\bar{1}\bar{1}1$	5.817	15.231		
$\bar{1}11$	5.645	15.697		
021	4.689	18.926	18.942	5
021	4.365	20.343		
$\bar{2}01$	4.042	21.992	21.987	35
111	3.904	22.775	22.777	15
$\bar{1}30$	3.761	23.659 <sup>1</sup>	23.669	70
111	3.759	23.668		
200	3.670	24.252		
130	3.634	24.497	24.509	12
$\bar{1}\bar{3}1$	3.620	24.593		
$\bar{1}31$	3.497	25.472		
$\bar{1}\bar{1}2$	3.471	25.666	25.667	20
$\bar{2}\bar{2}1$	3.423	26.034	26.007	2
$\bar{2}21$	3.417	26.080		
$\bar{1}12$	3.365	26.490	26.492	30
$\bar{2}20$	3.241	27.520	27.517	40
040	3.208	27.810	27.795	70
$\bar{2}02$	3.201	27.867	27.857	70
002	3.182	28.040	28.047	100
220	3.133	28.491	28.502	35
$\bar{1}\bar{3}1$	3.029	29.492	29.507	25
0 $\bar{4}1$	2.950	30.292	30.282	32
0 $\bar{2}2$	2.935	30.452	30.442	28
$\bar{2}\bar{2}2$	2.908	30.741	30.726	10
131	2.834	31.571	31.567	32
$\bar{2}\bar{2}2$	2.823	31.698		
$\bar{1}\bar{3}2$	2.814	31.799	31.797	15
041	2.786	32.130		
022	2.773	32.281		
201	2.705	33.115		
$\bar{3}\bar{1}1$	2.668	33.589		
$\bar{3}\bar{1}1$	2.654	33.776		
$\bar{1}32$	2.652	33.804	33.818	17
$\bar{2}\bar{2}1$	2.547	35.231	35.237	5
$\bar{1}\bar{1}2$	2.526	35.545		
$\bar{3}\bar{1}2$	2.518	35.659		
$\bar{2}\bar{4}1$	2.515	35.699	35.702	40
$\bar{2}41$	2.510	35.769		
$\bar{3}\bar{1}2$	2.502	35.888		

<sup>1</sup> From least-squares refinement of data measured.

<sup>2</sup> All calculated spacings are shown for  $d \geq 2.100 \text{ \AA}$ —calculated spacings less than  $2.100 \text{ \AA}$  are listed only when they correspond to an indexed observed reflection.

<sup>3</sup> Average of three observations with annealed  $\text{CaF}_2$  as internal standard,  $a=5.4622 \text{ \AA}$  at  $25^\circ\text{C}$ . Ni-filtered  $\text{CuK}\alpha$  radiation ( $\lambda=1.54180 \text{ \AA}$ ). Lower limit of  $2\theta$  measured =  $13^\circ \text{ CuK}\alpha$  ( $6.809 \text{ \AA}$ ). Pattern obtained at  $26^\circ\text{C}$ .

TABLE 2—(continued)

Triclinic  $\text{Ca}_{0.7}\text{Na}_{0.3}\text{Al}_{1.7}\text{Si}_{2.3}\text{O}_8$ ;  $I\bar{1}$ , but indexed on a  $C\bar{1}$  sub-cell with  $a=8.176 \text{ \AA}$ ,  $b=12.865$ ,  $c=7.102$ ,  
 $\alpha=93^\circ 27.1'$ ,  $\beta=116^\circ 2.9'$ ,  $\gamma=90^\circ 30.6'$

<i>hkl</i>	Calculated <sup>2</sup>		Measured <sup>3</sup>	
	<i>d</i> <sub><i>hkl</i></sub> (Å)	$2\theta_{\text{CuK}\alpha}$	$2\theta_{\text{CuK}\alpha}$	$I^\circ/I_{100}$
240	2.464	36.466	36.454	2
112	2.459	36.548		
150	2.453	36.636	36.677	3
221	2.441	36.816	36.847	3
310	2.421	37.136	37.178	4
151	2.414	37.254		
150	2.393	37.581		
310	2.386	37.699		
240	2.369	37.983		
151	2.352	38.268		
042	2.344	38.395		
203	2.336	38.543		
113	2.320	38.821		
242	2.310	38.995		
331	2.309	39.005	39.026	3
331	2.281	39.498	39.486	2
113	2.267	39.769		
132	2.255	39.985		
223	2.234	40.381		
151	2.230	40.441	40.479	3
242	2.225	40.547		
332	2.212	40.791	40.838	1
042	2.183	41.366		
332	2.181	41.403		
330	2.161	41.805		
223	2.158	41.859	41.842	2
152	2.141	42.207		
060	2.139	42.258	42.235	15
241	2.131	42.410	42.387	35
003	2.121	42.616	42.532	20
132	2.119	42.666		
313	2.118	42.684		
133	2.105	42.960		
151	2.099	43.084	43.060	12
402	2.021	44.851	44.898	2
422	1.929	47.123		
422	1.926	47.178	47.157	3
333	1.882	48.366		
260	1.880	48.409	48.372	5
222	1.8795	48.429		
351	1.8791	48.440	48.437	2
403	1.848	49.316	49.272	2
243	1.842	49.483	49.432	1
400	1.835	49.684	49.684	10
113	1.834	49.714	49.719	1
260	1.817	50.213	50.165	1
113	1.798	50.791	50.762	10
204	1.772	51.569	51.579	12
242	1.754	52.136	52.115	1
043	1.714	53.453	53.397	3
442	1.711	53.548	53.547	3
441	1.691	54.253	54.167	1
242	1.652	55.623	55.577	5

TABLE 3. UNIT-CELL PARAMETERS OF THE SUB-CELL OF CALCIC LABRADORITE  
REFINED BY FIXED INDEXING

Measurement Sequence	T, °C.	a, Å	b, Å	c, 2 Å	$\alpha$	$\beta$	$\gamma$	$\frac{1}{2}$ Volume Å <sup>3</sup>	Volume cm <sup>3</sup> /mol	No. of lines used	Standard Error % <sup>29</sup>
(1)	26	8.176	12.865	7.102	93 27.1	116 2.9	90 30.6	669.5	100.84	20	.0146
	±	.003	.003	.002	1.4	1.5	1.3	.4	.06		
(11)	26*	8.173	12.871	7.101	93 23.9	115 59.8	90 35.3	669.7	100.87	21	.0236
	±	.005	.005	.003	2.3	2.4	.4	.4	.06		
(2)	190	8.176	12.882	7.106	93 21.8	116 2.0	90 29.4	670.8	101.03	34	.0247
	±	.003	.003	.002	1.7	1.6	1.7	.5	.06		
(4)	330	8.186	12.887	7.108	93 18.1	116 0.8	90 24.8	672.3	101.26	30	.0152
	±	.002	.002	.001	1.3	1.1	1.2	.4	.06		
(8)	429	8.193	12.888	7.111	93 13.8	116 0.5	90 26.1	673.3	101.41	22	.0235
	±	.004	.004	.003	2.2	2.3	2.1	.4	.06		
(3)	508	8.198	12.900	7.111	93 12.7	116 2.2	90 21.2	674.2	101.55	28	.0234
	±	.003	.004	.002	1.9	1.7	1.8	.4	.07		
(5)	654	8.214	12.904	7.117	93 10.9	116 1.8	90 17.3	676.4	101.88	21	.0234
	±	.004	.005	.003	2.5	2.3	2.2	.4	.06		
(6)	813	8.220	12.907	7.118	93 1.8	115 58.8	90 17.7	677.6	102.06	22	.0238
	±	.005	.004	.003	2.2	2.5	2.1	.4	.06		
(7)	961	8.222	12.914	7.123	92 58.4	115 58.8	90 10.1	678.7	102.22	24	.0290
	±	.005	.006	.003	2.6	2.6	2.5	.5	.08		
(9)	1051	8.221	12.923	7.118	92 53.3	115 51.1	90 10.9	679.4	102.33	24	.0298
	±	.005	.005	.003	2.7	2.8	2.6	.6	.09		
(10)	1141	8.233	12.927	7.120	92 48.7	115 49.4	90 7.6	681.0	102.57	24	.0178
	±	.003	.003	.002	1.6	1.6	1.6	.4	.06		
Linear and volume expansion in per cent											
	26°	0.000	0.000	0.000				0.000			
	26*	-.037	.046	-.015				.029			
	190	0.000	.132	.056				.194			
	330	.122	.171	.084				.418			
	429	.208	.178	.126				.567			
	508	.269	.272	.126				.702			
	654	.464	.303	.211				1.030			
	813	.538	.326	.225				1.209			
	961	.562	.380	.295				1.374			
	1051	.550	.450	.225				1.478			
	1141	.697	.481	.253				1.717			

\* After heating to 1141° C

ment of the data was accomplished with a digital computer by alternately using the fixed and variable indexing options of the crystallographic indexing program developed by Evans *et al.* (1963). Feldspar lines that

were interfered with by mullite lines from the sample holder or by platinum lines from sublimed platinum from the furnace windings were omitted from consideration. Data were collected in the sequence shown in Table 3 so that any irreversible changes that might have occurred could be detected from the refined results.

At least 20 uniquely indexed lines within the range  $22^\circ$  to  $56^\circ 2\theta$  (CuK $\alpha$ ) were used in the final cycle of fixed indexing. The number of lines used, the unit-cell parameters, the unit cell and molar volumes, and the standard error associated with the measurements for each temperature are indicated in Table 3,<sup>1</sup> together with the per cent linear axial and volume expansions for each measurement. It is interesting that the three non-orthogonal linear expansions sum to less than the true volume expansion (compare Yoder and Weir, 1951, p. 684).

The sample remained triclinic throughout the heating interval, and the change of the interaxial angles was small. The per cent linear axial expansion is greatest along  $a$ , and least along  $c$ .

The change of molar volume with increase of absolute temperature was fitted by the method of least squares with a digital computer to the equation (Skinner *et al.*, 1961, p. 665)

$$V = \frac{A}{T} + B + CT + DT^2$$

The value at room temperature was given three times unit-weight; all others were given unit-weight. The value of  $A$  is  $160.99 \text{ cm}^3 \text{ }^\circ\text{K/mol}$ ,  $B = 99.528 \text{ cm}^3/\text{mol}$ ,  $C = 2.7331 \times 10^{-3} \text{ cm}^3/\text{mol}^\circ\text{K}$ , and  $D = -5.0064 \times 10^{-7} \text{ cm}^3/\text{mol} (\text{ }^\circ\text{K})^2$ . The standard deviation of the array is  $0.0523 \text{ cm}^3/\text{mol}$ . The observed molar volumes and the least-squares curve of molar volume are shown on Fig. 5. The molar volume calculated from the least-squares equation, the two expressions of the thermal expansion ( $(dV/dt)_p$  and  $\alpha_V$ ), and the calculated per cent volume expansion given in Table 4 must be regarded with suspicion for high temperatures because the equation for the curve of the molar volume takes no account of the possible transformation to be described below. Comparison of the calculated per cent volume expansion ( $\Delta V$ , Table 4) with the results of Koza and Ueda (1933, Fig. 2) for plagioclase shows good agreement up to  $800^\circ$ ; at higher temperatures our observed and calculated values are

<sup>1</sup> A document listing the computer input (fixed Miller index and associated observed  $2\theta$ ) and the final cycle of refinement for each temperature studied has been deposited as Document No. 8673 with the American Documentation Institute, Auxiliary Publications Project, Photoduplication Service, Library of Congress, Washington, D. C. Copies may be received by citing the document number, and remitting \$2.50 for photoprints or \$1.75 for 35 mm. microfilm. Advance payment is required.

both lower than predicted by their figure, and the high-temperature volume thermal expansion of labradorite appears to be almost the same as that of anorthite.

*Changes in twinning relations on heating.* J. V. Smith (1958) summarized both old and new studies on the effects of temperature, structural state,

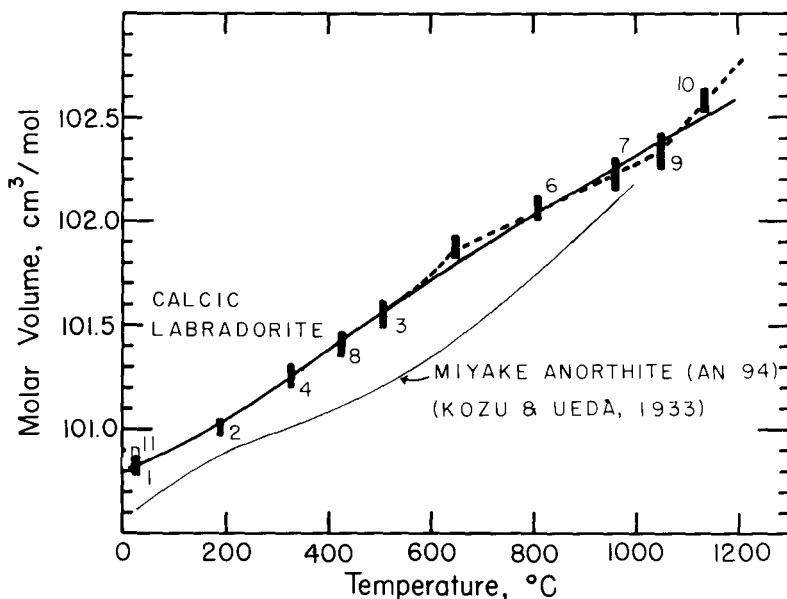


FIG. 5. Observed molar volume of calcic labradorite at various temperatures, and the least-squares curve derived for these observations (heavy line). The dashed curve indicates the volume through a possible high order transformation. The size of the spot is proportional to the standard error; numbers indicate the sequence of measurement in data collection. The curve for Miyake anorthite is approximate and is displaced by  $-0.2 \text{ cm}^3/\text{mol}$  in order to separate it from the curves for calcic labradorite; it is discussed in the text.

and composition on the obliquity of albite twins,  $\phi$ , at room temperature. It is pertinent to inquire what changes occur on heating to the temperature range where feldspars commonly are formed, because as the obliquity decreases the likelihood that twinning will occur increases. The data in Table 3 have been used to calculate  $\phi$  for our sample as a function of temperature (Table 5). The obliquity decreases appreciably as a nearly linear function of temperature. Twinning ought to be very common in crystals formed at high temperature; the relative scarcity of twinning in our material is not easily interpreted. It should also be recalled that

the heating and cooling of calcic plagioclase resulted in the formation of secondary albite and pericline twinning (Muir, 1955, p. 563-566).

Tunell (1952) and MacKenzie (1956) discussed  $\sigma$ , the angle between the  $a$ -axis and the trace of the rhombic section on  $\{010\}$ , which is of interest because the rhombic section is the composition plane of pericline twins. We have used our data to calculate  $\sigma$  for the temperature range we investigated. A large scatter is obvious because  $\sigma$  is very sensitive to slight errors in the determinations of the lattice angles, but a marked

TABLE 4. CALCULATED MOLAR VOLUMES AND THERMAL EXPANSION OF CALCIC LABRADORITE AT HIGH TEMPERATURES

T° C.	V cm <sup>3</sup> /mol	( $\partial V/\partial T$ ) <sub>p</sub> cm <sup>3</sup> /mol °C. × 10 <sup>-3</sup>	$\alpha_V$ °C <sup>-1</sup> × 10 <sup>-5</sup>	$\Delta V$ % of V <sub>26</sub>
0	100.827	0.300	0.35	-0.01
26	100.839	0.633	0.72	0.00
100	100.909	1.201	1.19	0.07
200	101.049	1.540	1.52	0.21
300	101.211	1.669	1.64	0.37
400	101.380	1.704	1.68	0.54
500	101.550	1.689	1.66	0.71
600	101.717	1.648	1.61	0.87
700	101.879	1.589	1.55	1.03
800	102.034	1.519	1.48	1.19
900	102.182	1.442	1.41	1.33
1000	102.322	1.359	1.32	1.47
1100	102.454	1.273	1.24	1.60
1200	102.577	1.184	1.15	1.72

decrease of  $\sigma$  with increasing temperature is clear. These observations may be useful in optical studies of the formation of secondary twins on heating.

*Possible transformation in Lake County labradorite at high temperatures.*

The least-squares curve falls outside the interval of standard error for the observation at 654° C., and the observation at 1141° C. is colinear with the three observations at lower temperatures. Further work to confirm the reality of this irregularity is needed. A possible explanation of the irregularity and the fact that the rate of change of volume with temperature ( $\partial V/\partial T$ )<sub>p</sub>, Table 4, decreases at high temperature is that a transformation takes place above 654° C. and causes a step on the volume curve as shown by the dashes on Fig. 5. The sequence of measure-

ments was such that the changes that occurred at high temperatures are known to be reversible; observation 8 falls smoothly between observations 4 and 3, and 9 falls on a smooth extension of 5, 6 and 7. The apparent step, if real, may result because antiphase domain boundaries (Megaw, 1962) or the Al:Si order are changing in a continuous and reversible way in the interval between 650° and 1050° C. This proposal could be evaluated by an x-ray study of the "b" reflections from single crystals at high temperatures, but we are not equipped to do this. The "b" reflections are too weak to be recorded on our diffractometer patterns. Megaw recognizes and emphasizes the importance of geometrical arrangements of formula units in plagioclase that are to a first approxi-

TABLE 5. EFFECT OF HEATING CALCIC LABRADORITE ON THE OBLIQUITY,  $\phi$ , AND THE ANGLE BETWEEN THE  $a$ -AXIS AND THE TRACE OF THE RHOMBIC SECTION ON  $\{010\}$ ,  $\sigma$

T° C.	26	26 <sup>1</sup>	190	330	429	508	654
$\phi$	-4° 7.3'	-4° 6.8'	-4° 1.0'	-3° 53.7'	-3° 49.7'	-3° 46.1'	-3° 41.4'
$\sigma$	-7° 6.6'	-8° 14.2'	-7° 1.5'	-6° 5.6'	-6° 31.2'	-5° 23.5'	-4° 29.2'
T° C.	813	961	1051	1141			
$\phi$	-3° 31.5'	-3° 23.8'	-3° 18.5'	-3° 11.2'			
$\sigma$	-4° 48.3'	-2° 50.8'	-3° 9.3'	-2° 16.7'			

<sup>1</sup> After heating to 1141° C.

mation independent of the degree of ordering of silicon and aluminum atoms, or even of the Al:Si ratio. At high temperatures increased thermal vibrations may lead to a greater variation in bond angles within the feldspar structure and a transition from a "puckered" structural arrangement to a straightened arrangement with an effective increase of symmetry even though the space group may remain unaltered.

The cell volume appears to increase continuously with rising temperature over a long temperature interval, so that the possible phase transformation is at least second order, analogous to the symmetry change in anorthoclase (MacKenzie, 1952; Donnay and Donnay, 1952).

*High temperature transformations of other plagioclase samples.* It is clear that because nonquenchable transformations occur in plagioclase feldspar measurements of the physical properties of feldspar must be made at high temperatures. Such measurements can lead to an equilibrium phase diagram for the plagioclase series, and aid in the interpretation of thermal expansion measurements. Diagrams constructed from the available data



collected at room temperature such as that of Megaw (1962, p. 120) are vital to the formulation of significant experiments, but are already in need of revision to accommodate our results and those of Brown *et al.* (1963).

Although there have been several investigations in which plagioclases have been examined before and after heating (summarized by Brown, 1959), no one has reported on enough data collected at high enough temperatures to be directly comparable to ours.<sup>1</sup> However, the heating experiments of Gay and Bown (1956, Table 1b, nos. 5 and 7, and p. 310) on the phenocrysts ( $An_{65}$ ) from basaltic flows at Clear Lake, Utah, and the phenocrysts ( $An_{63}$ ) from basaltic dikes at St. John's Point, Ireland, are significant, because these materials are both comparable to ours and show the anomalous optical and x-ray properties of the material under discussion. For their samples, no changes in optical or x-ray properties toward higher structural states could be detected at room temperature after heating at up to 950° C. for 3 days, but changes were detected after heating to 1000° C. and higher for 3 days. The "b" reflections, as observed after cooling to room temperature, on heating above 1000° C. become weaker with increasing heat treatment and finally disappear, so that the lattice symmetry becomes *C* face-centered (*CI*), which is characteristic of sodic plagioclases. J. R. Smith (1958, p. 1190) found that irreversible changes toward higher structural states that were measurable at room temperature could be induced by hydrothermal experiments in plagioclases of  $An_{30}$ – $An_{80}$  composition only at temperatures above 990° C. The heating time for any particular temperature was less than 4 hours in our experiments, so the higher temperature (>1051° C.) at which irreversible effects become apparent in our investigation is quite compatible with the earlier results. The temperature after which "b" reflections will be absent at room temperature is probably different from the temperature at which "b" reflections disappear during heating. The latter temperature is presumably closer to the equilibrium temperature for the transition, and thus it is probable that the equilibrium diagram for calcic plagioclase can only be determined by measurements of physical parameters made at high temperatures.

The importance of combining thermal expansion measurements with

<sup>1</sup> Gay (1961, p. 161) mentions that Bown has investigated the rate of disappearance of split "b" reflections at 1050° C. in plagioclase,  $An_{63}$ , from the Stillwater intrusive, but no further details are given. Gubser, Hoffman, and Nissen (1963) have determined the variation of  $\alpha^*$  of anorthite with temperature up to 1400° C., and have also measured the relative intensities of several "a" reflections as a function of temperature. We are also aware that thermal expansion measurements have been made at the University of Chicago by A. A. Kayode.

high temperature studies of single crystals is well demonstrated by a comparison of the results of Koza and Ueda (1933) on the thermal expansion of sodic anorthite with the single crystal *x*-ray studies of anorthite by Brown, *et al.* (1963) at elevated temperatures. The molar volume of anorthite (taken as 100.827 cm<sup>3</sup> from the data of Cole *et al.*, 1951) was multiplied by the per cent volume expansion given by Koza and Ueda for An<sub>95</sub> from Miyake Jima. The resulting molar volumes, less 0.2 cm<sup>3</sup>/mol to separate them from the results for Lake County labradorite, are plotted in Figure 5. The volume curve of Miyake anorthite has a step in the interval between 200 and 500° C.<sup>1</sup> Laves and Goldsmith (1954a, p. 407; 1954b, p. 469-471) showed that Miyake anorthite has diffuse "c" reflections. Brown *et al.* showed that "c" reflections of sodic anorthite weaken and disappear on heating from 25° to 350° C., and it is probable that the step on the volume curve and the disappearance of the "c" reflections are related because both occur in the same temperature interval. Furthermore, Bloss (1964) found reversible changes in the orientation of the optical indicatrix corresponding to the same temperature interval, and located an additional displacive transformation at about 800°. Kohler and Weiden (1954) gave DTA curves for two anorthite samples that show peaks in these same two temperature ranges. While it is not possible yet to state structurally exactly what both of the transformations are that take place, it is interesting to observe that the temperature of the continuous and reversible loss of "c" reflections in sodic anorthite deduced from samples quenched from 1100° C. and studied by Laves and Goldsmith (1954a) at room temperature and by the measurements of Brown *et al.* at high temperatures differ by more than 750° C.!

Kohler and Weiden (1954) also gave a DTA curve for sodic labradorite from Labrador. An exothermic peak centered at ~350° C. was attributed by them to admixed hematite or ilmenite, and an endothermic peak at ~790° C. was attributed to a transformation to a high-temperature form. Our results cannot be correlated simply with these thermal effects. No effect was discernible in our more calcic material at 350° C. The peak at 790° C. falls within the temperature interval of our supposed transformation, but does not appear to cover as much of the temperature range as we would have expected. There is, furthermore, no correlation between the differential thermal effects they reported in albite and the cell parameters of low albite at high temperatures as determined by Stewart and von Limbach (1964).

<sup>1</sup> A somewhat less pronounced step at somewhat higher temperatures was observed by Koza and Ueda (1933) in the per cent volume expansion curve for sodic anorthite from Mitaki, Japan.

## DISCUSSION

*History of crystallization of feldspar.* The petrographic observations described earlier can now be interpreted in view of the physical measurements that have been made on the calcic labradorite. The lack of zoning and mineral inclusions in the large cores of the phenocrysts indicate that these formed by primary crystallization in a magma chamber at depth under relatively uniform conditions. The temperature was probably greater than 1100° C., because split "b" reflections and other changes in physical properties occur in calcic labradorite that has been exposed for long periods of time to lower temperatures (Gay and Bown, 1956; Laves and Goldsmith, 1955; Gay and Muir, 1962). The release of pressure accompanying eruption initiated the resorption observed on the surface of the core (zone, 1 Fig. 3), and the rapid fluctuations of temperature and pressure during eruption and extrusion caused the oscillatory zoning and resorptions observed in the rims of the phenocrysts. The temperature changes involved were probably small, because the difference in composition and structural state of the core and the outer shells is also small. The outer shells,  $\approx 0$  to 8% of the volume of the phenocrysts, represent crystallization that took place during eruption to the surface, and the intermediate structural state of the core and rim feldspar is evidence that rapid cooling and low vapor pressure of volatiles followed this process, which would be expected in flows only a few tens of feet thick. The mechanical fracturing of the cores and outer shells (Figs. 1, 2) and the presence of some bent twin lamellae testify to rapid and violent flow movements during extrusion, though it is also possible that cataclasis may have occurred in the magma chamber in crystal-liquid mush. Microlites of the same composition as found in the rims of the phenocrysts were formed in part during the interval of extrusion and flow. After the rock had become sufficiently rigid to resist deformation, the outermost shell of the phenocrysts formed by crystallization *in situ* with strong fractionation of the residual liquid. The higher structural state of the feldspar of the outermost shell and groundmass is not a consequence of a higher temperature of crystallization, but rather reflects a compositional effect on the rate of attaining a given structural state during cooling (Wright, 1963; Gay and Muir, 1962; Uruno, 1963).

It is probable that much of the calcic plagioclase in extrusive rocks and hypabyssal rocks has undergone a thermal quench on emplacement. Such material can be expected to have physical properties like those described above.

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