VARIATIONS IN *X*-RAY POWDER DIFFRACTION PATTERNS OF PLAGIOCLASE FELDSPARS

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ABSTRACT

The angular separation between the $(1\bar{3}1)$ and (131) reflections in *x*-ray diffractometer patterns of 66 chemically analyzed natural plagioclases, 11 plagioclases synthesized in the dry way, and 4 plagioclases synthesized hydrothermally has been measured and plotted against composition. By this criterion, plagioclases synthesized in the dry way and natural plagioclases from thick stratiform mafic intrusions constitute two distinctly different series, each of which is closely defined by a single curve. Natural plagioclases from volcanic and hypabyssal rocks and plagioclases synthesized hydrothermally are intermediate between the two series. Other natural plagioclases, some of which have been assumed by others to belong to a "low-temperature" series, do not belong to either of the series mentioned above, and cannot be represented by any single curve. It is concluded that composition determinations cannot be made by the use of the available curves based on the variation of reflection separations, because there is no a priori way of knowing how closely a given plagioclase is represented by a particular curve. However, given the composition of a plagioclase, the curves are useful for making an estimate of its degree of inversion toward some undefined low-temperature state.

INTRODUCTION

Certain parameters in the atomic structures of plagioclase feldspars vary with composition and with thermal history. The variations may be studied by measuring the angular separation of two reflections in *x*-ray powder diffraction patterns, a procedure which avoids some of the errors encountered in measuring the absolute positions of the reflections. Previous workers have published the results of several such studies. Claisse (1950) studied eight samples of plagioclase, disregarding the thermal histories of the samples. Tuttle and Bowen (1950) and Chayes and Robbins (1953) studied samples covering limited parts of the composition range. In a more extensive study, Goodyear and Duffin (1954) presented curves showing the variation of reflection separations with composition in synthetic plagioclases and in natural plagioclases; they also studied the effects of heating natural plagioclases (1955). J. V. Smith (1956) has carried out a detailed study of the variation of lattice parameters and reflection separations in synthetic and natural sodic plagioclases.

The present writers planned to use the *x*-ray methods recommended by Goodyear and Duffin to determine the composition and thermal state of plagioclases synthesized in experimental studies of the system albite–anorthite–water. In order to check the reproducibility of Goodyear and Duffin's results with different equipment, reflection separations in several chemically analyzed samples of natural plagioclases were first measured and compared with Goodyear and Duffin's curves. Significant discrepancies suggested that the 16 samples of natural plagioclases studied by Goodyear and Duffin are not representative of *all* so-called low-temperature plagioclases. In order to investigate more fully the nature of the variations to be expected in *x*-ray parameters of plagioclases, 66 chemically analyzed samples of natural plagioclases, 11 synthetic plagioclases crystallized in the dry way, and 4 synthetic plagioclases crystallized hydrothermally were studied.

EXPERIMENTAL METHODS

The reflections to be measured were chosen on the basis of the following requirements: (*a*) that they be clearly resolved from neighboring reflections, (*b*) that their 2θ values vary sensitively with composition, and (*c*) that two such reflections, between which the angular separation is to be measured, be close together in powder patterns to minimize chart-scale errors. A pair of reflections which satisfies these requirements over the composition ranges An 0 to An 30 and An 60 to An 100* in natural plagioclases is $(1\bar{3}1)$ and (131).† A second pair, (220) and $(1\bar{3}1)$, is equally suitable over the composition ranges An 10 to An 30 and An 60 to An 100, but the (220) reflection crosses the $(2\bar{2}0)$ reflection at An 10 and is masked by strong (040) and (002) reflections from An 0 to An 10.‡ The (220), $(1\bar{3}1)$, and (131) reflections are resolved in plagioclases synthesized in the dry way from An 0 to An 100. Appropriate portions of powder patterns of high-temperature albite and of natural albite, andesine, and anorthite are shown in Fig. 1. The relevant data on the reflections are given in Table 1.

Specimens were prepared by smearing suspensions of powder in lacquer-acetone solution on glass slides. Powder patterns were taken with a

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* Unless otherwise stated, compositions are given as

$$\text{mol }\% \frac{\text{An}}{\text{An} + \text{Ab} + \text{Or}}$$

throughout this report.

† Powder patterns were indexed by comparison with completely indexed patterns of Amelia albite in the natural state and in a high-temperature state (heated at near-melting temperature until no further change took place in the lattice parameters of the quenched product). J. V. Smith (1956) indexed the patterns of Amelia albite, and kindly made the information available to the writers.

‡ This fact is not taken into account by Goodyear and Duffin (1954), who, by measuring the $(2\bar{2}0)-(1\bar{3}1)$ separation for pure albite and including it with measurements of the non-equivalent $(220)-(1\bar{3}1)$ separation for more calcic plagioclases, have introduced considerable error (4 to 7% An) in their *A-B* curve for low-temperature plagioclases in the range An 0 to An 30.

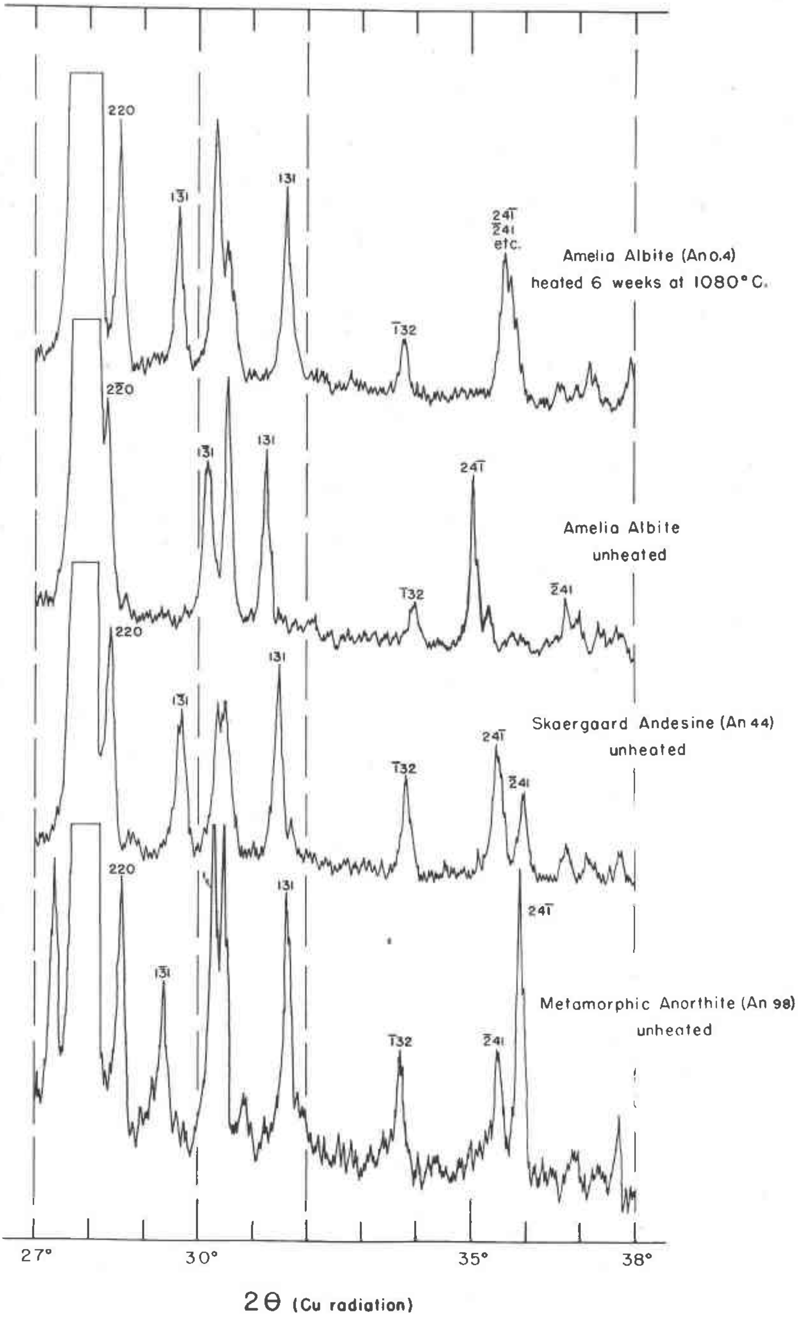

FIG. 1. Portions of powder diffraction patterns of Amelia albite changed by heating to the high-temperature form, and of unheated natural albite, andesine, and anorthite.

TABLE 1. POSITION OF SIGNIFICANT REFLECTIONS

Indices	2θ (CuKα) Low-temp. Ab	2θ (CuKα) High-temp. Ab	2θ (CuKα) An	Remarks
(220)	28.1 (calc.)	28.5	28.6	Crosses $(2\bar{2}0)$ at approximately An 10 in low-temperature plagioclases
$(1\bar{3}1)$	30.1	29.6	29.4	May or may not be resolved in inhomogeneous low-temperature plagioclases in the range An 6 to An 17
(131)	31.2	31.6	31.6	Resolved from An 0 to An 100

Norelco high-angle diffractometer using copper Kα radiation. Divergent and scatter slits were 1°, receiving slit 0.006 inch, scan speed $\frac{1}{4}°$ per minute, and chart scale 2 inches per degree. The appropriate 2θ region was scanned at least three times; the reflection separations were measured to the nearest 0.01° 2θ, and the measurements were averaged arithmetically.

Table 2 is a list of the samples and the measurements obtained from each. In the samples of natural plagioclases, the maximum range of the individual measurements of the reflection separations in one sample is 0.10°, and in most of the samples the range is 0.04° or less. Taking into account the variable quality of the peaks in the powder patterns, the probable accuracy of the average value of a reflection separation for any sample of natural plagioclase is estimated to be $\pm 0.02°$ 2θ or better. The accuracy for the sodic and intermediate synthetic plagioclases is somewhat less than this, owing to the broad character of the peaks. In the plots of the data (Figs. 2, 3, and 4) the size of the symbols has been made 0.04° 2θ, which approximately covers the range of uncertainty of the position of each point in the direction of the reflection separation axis. The uncertainty to attach to the positions of the symbols in the direction of the composition axis cannot be estimated quantitatively on the basis of the chemical analyses themselves, but it is believed to be no greater than ± 2 mol % An for most of the samples. Like most natural plagioclases, the samples used have varying amounts of K_2O (see Table 2). The amounts represent less than 4.5 mol % Or in any sample other than the Larsen samples from volcanic rocks and Howie's sample 2270 from a charnockite, in which samples the maximum content of Or is 10.5 mol %. The effect on *x*-ray parameters of plagioclases of K^+ ions present in the structure is not known. Especially in the samples with high K_2O contents, an undetermined part of the K_2O is probably present in a separate phase, which would contribute small errors to the calculated compositions of the samples.

Discussion of Results

In Fig. 2 the $(1\bar{3}1)-(131)$ separation is plotted against the known compositions of 11 plagioclases synthesized in the dry way (circles) and 11 plagioclases from thick stratiform mafic intrusions (squares). With one exception, the points representing the synthetic plagioclases fall, within the limits of error, on the curve drawn through them. The curve represents plagioclases crystallized at and cooled quickly from temperatures slightly below the solidus for the dry system Ab—An.*

The points representing plagioclases from thick stratiform mafic intrusions—namely, the Skaergaard intrusion, the Great Dyke, the Bushveld igneous complex, and the Stillwater igneous complex—fall on the curve drawn through them within the limits of error. *All these plagioclases probably have similar thermal histories*, inasmuch as they all crystallized at basaltic magma temperatures, and, as a result of the tremendous thickness of the intrusions, cooled very slowly during and after crystallization. In view of their lower temperatures of crystallization and much slower rates of cooling as compared with the plagioclases synthesized in the dry way, it may be assumed that the plagioclases from thick stratiform mafic intrusions are modifications which are either in equilibrium at temperatures lower than the temperatures from which the plagioclases synthesized in the dry way were quenched, or are transitional toward some lower temperature equilibrium state. Since these plagioclases are the most slowly cooled of all magmatic plagioclases in the composition range which they cover (An 36 to An 86), and since they make up a well-defined series by the criterion of reflection separations, they will be called "plagioclases of the Bushveld type" in this report. This is in accordance with Hess's (1952) designation of orthopyroxenes

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* The curve does not represent plagioclases *at* near-solidus temperatures, because MacKenzie (1952) has shown that there are marked differences between reflection separations in synthetic sodic plagioclases at room temperature and at elevated temperatures, and that the reflection separations change on cooling to room temperature, no matter how rapid the cooling rate. Goodyear and Duffin cite discontinuities in the variation of reflection separations measured at room temperature for synthetic plagioclases as evidence of lack of complete solid solution in high-temperature plagioclases, "although the early work of Bowen (1913) on melting phenomena indicated solid solution over the whole range from albite to anorthite" (Goodyear and Duffin, 1954, p. 321). Bowen's data concern equilibrium conditions *at* the solidus temperatures. From MacKenzie's work, it is evident that discontinuities in the variations of x-ray parameters measured at room temperature may have no bearing on whether or not complete solid solution exists at solidus temperatures.

TABLE 2. LIST OF SAMPLES AND DATA

Sample No.	Composition Mol o/o An	Ab	Or	Source of Sample	Chemical Analyst (Reference to analysis)*	Occurrence and Locality of Specimen	Measurements of Reflection Separations, with Average in Parentheses $2\theta(1\bar{3}1) - 2\theta(131)$	$2\theta(220) - 2\theta(1\bar{3}1)$
						Plagioclases synthesized in the dry way		
05	4.7	95.3	0	J. F. Schairer	--	"	2.01,2.03,2.02,2.02 (2.02)	1.08,1.07,1.09,1.07 (1.08)
1	9.5	90.5	0	"	--	"	1.98,1.99,2.00,2.00 (1.99)	1.09,1.10,1.09,1.08 (1.09)
2	19.0	81.0	0	"	--	"	1.97,1.95,2.00,1.99,1.95,2.00 (1.98)	1.10,1.13,1.11,1.11,1.10,1.09 (1.11)
3	28.8	71.2	0	"	--	"	1.95,1.95,1.92,1.93 (1.94)	1.11,1.09,1.14,1.12 (1.11)
4	38.6	61.4	0	"	--	"	1.98,1.93,1.95 (1.95)	1.13,1.12,1.11 (1.12)
5	48.5	51.5	0	"	--	"	1.94,1.97,1.93 (1.95)	1.11,1.08,1.08 (1.09)
6	58.6	41.4	0	"	--	"	2.06,2.04,2.05,2.03 (2.05)	1.02,1.05,1.03,1.05 (1.04)
7	68.8	31.2	0	"	--	"	2.10,2.11,2.10,2.09,2.08 (2.10)	0.99,0.99,0.98,0.99,1.00 (0.99)
8	79.0	21.0	0	"	--	"	2.16,2.13,2.16,2.16 (2.15)	0.93,0.94,0.94,0.94 (0.94)
9	89.5	10.5	0	"	--	"	2.25,2.23,2.24,2.23 (2.24)	0.82,0.84,0.85 (0.84)
10	100.0	0	0	"	--	"	2.28,2.28,2.28,2.29 (2.28)	0.78,0.79,0.78,0.77 (0.78)
						Plagioclases synthesized hydrothermally (4 hrs. at 640°C, P_{H_2O} = 10,000 bars)		
S2-1	9.5	90.5	0	Writers	--	Starting material glass	1.86,1.86,1.86,1.84 (1.86)	1.19,1.19,1.18,1.19 (1.19)
S2-2	9.5	90.5	0	"	--	Starting material dry synthetic crystals	1.95,1.97,1.96,1.94 (1.95)	1.11,1.11,1.11,1.11 (1.11)
S2-3	28.8	71.2	0	"	--	Starting material glass	1.87,1.86,1.90,1.90 (1.89)	1.16,1.17,1.17,1.15 (1.16)
S2-4	28.8	71.2	0	"	--	Starting material dry synthetic crystals	1.96,1.98,1.97,1.96,1.96 (1.97)	1.08,1.10,1.09,1.10,1.10 (1.10)
						Plagioclases of the Bushveld type		
4145	36.5	60.7	2.8	L. R. Wager	W. A. Deer (1)	Ferrogabbro, Skaergaard Intrusion, East Greenland	1.76,1.81,1.79,1.74 (1.78)	1.32,1.28,1.30,1.31 (1.30)
2580	40.2	57.0	2.8	"	"	Ferrogabbro, Skaergaard Intrusion, East Greenland	1.79,1.76,1.77,1.77 (1.77)	1.32,1.33,1.31,1.31 (1.32)
1963	44.2	53.7	2.1	"	E. H. Oslund (2)	Ferrogabbro, Skaergaard Intrusion, East Greenland	1.79,1.79,1.77,1.80 (1.79)	1.28,1.30,1.32,1.30 (1.30)
GD29	51.8	46.3	1.9	H. H. Hess	"	Tolerite, Great Dyke, S. Rhodesia	1.82,1.82,1.83,1.81 (1.82)	1.28,1.29,1.28,1.29 (1.28)
BV63	59.2	38.5	2.2	"	"	Gabbro, Bushveld Igneous Complex, South Africa	1.86,1.88,1.88,1.87 (1.87)	1.24,1.23,1.22,1.23 (1.23)
EB41	62.5	36.8	0.7	"	"	Gabbro, Stillwater Igneous Complex, Montana	1.91,1.92,1.91,1.91 (1.91)	1.20,1.19,1.20,1.19 (1.20)
EB42	66.2	33.0	0.8	"	A. H. Phillips (2)	Gabbro, Stillwater Igneous Complex, Montana	1.93,1.95,1.93,1.95 (1.94)	1.17,1.15,1.17,1.15 (1.16)
EB43	74.3	25.1	0.6	"	"	Gabbro, Stillwater Igneous Complex, Montana	2.05,2.01,2.04,2.02 (2.03)	1.04,1.07,1.06,1.05 (1.06)
26(109)	77.0	21.5	1.5	R. C. Emmons	L. C. Peck (3) S. S. Goldich (4)	Norite, Bushveld Igneous Complex, Rustenberg, Transvaal, South Africa	2.09,2.11,2.10,2.10 (2.10)	1.00,1.00,0.99,0.98 (0.99)
EB38	80.0	19.5	0.5	H. H. Hess	A. H. Phillips (2)	Norite, Stillwater Igneous Complex, Montana	2.14,2.12,2.13,2.12,2.11 (2.12)	0.96,0.94,0.95,0.97,0.98 (0.96)
EB18	85.8	13.9	0.3	"	"	Norite, Stillwater Igneous Complex, Montana	2.19,2.20,2.21 (2.20)	0.90,0.90,0.88 (0.89)
						Plagioclases from pegmatites and granites		
1(87)	0.1	98.7	1.2	R. C. Emmons	L. C. Peck (3)	Pegmatite, Peerless Mine, Keystone, S. D.	1.06,1.07,1.08,1.09 (1.08)	(220) not measurable
D-761-1	0.2	98.7	1.1	C. S. Ross	R. E. Stevens (3)	Pegmatite, Ramona, Calif.	1.06,1.05,1.04,1.11,1.10,1.11 (1.08)	"
D-761-2	0.4	98.2	1.4	"	"	Pegmatite, Pala, Calif.	1.09,1.08,1.06,1.07 (1.08)	"
D-761-3	0.4	98.2	1.4	"	"	Pegmatite, Amelia, Va.	1.09,1.08,1.09,1.08 (1.09)	"
KN13	0.6	98.6	0.7	F. C. Kracek	E. G. Zies (5)	Pegmatite, Varuträsk, Sweden	1.07,1.08,1.08,1.07,1.06,1.07 (1.07)	"
2(10)	11.0	87.4	1.6	R. C. Emmons	L. C. Peck (3)	Granite, Peekskill, N. Y.	Unmeasurable	"
3(73)	15.4	81.8	2.8	"	"	Granite, Parishville, N. Y.	1.42,1.41,1.41,1.45,1.46,1.42,1.47 (1.43)	1.71,1.73,1.70,1.72,1.69,1.70,1.58 (1.70
4(156)	15.9	82.5	1.6	"	"	Granite, Petrick Quarry, Llano Co., Texas	1.40,1.40,1.39,1.37 (1.39)	1.73,1.72,1.74,1.73 (1.73)
KN8	16.8	82.0	1.2	F. C. Kracek	O. von Knorring (5)	Pegmatite, S. C.	1.42,1.43,1.41,1.44,1.41 (1.42)	1.71,1.68,1.70 (1.70)
5(94)	18.7	77.1	4.2	R. C. Emmons	L. C. Peck (3)	Granite, Tigerton, Wis.	1.48,1.50,1.46,1.52,1.52 (1.50)	1.64,1.64,1.67,1.63,1.60 (1.64)
D-761-4	19.5	76.7	3.8	C. S. Ross	R. E. Stevens (3)	Pegmatite, Hawk Mine, Bakersville, N. C.	1.49,1.51,1.50,1.50,1.49,1.50,1.50 (1.50)	1.62,1.60,1.61,1.60,1.60,1.57 (1.60)
KN9	21.1	75.5	3.4	F. C. Kracek	F. A. Gonyer (5)	Pegmatite, Hawk Mine, Bakersville, N. C.	1.51,1.53,1.52 (1.52)	1.60,1.58,1.58 (1.59)
KN10	21.6	74.5	3.9	"	E. Chadbourn (5)	Pegmatite, Hawk Mine, Bakersville, N. C.	1.53,1.52,1.52 (1.52)	1.57,1.58,1.58 (1.58)
KN11	28.7	67.8	3.5	"	"	Pegmatite, Little Rock Creek, Mitchell Co., N. C.	1.59,1.60,1.59 (1.59)	1.50,1.50,1.51 (1.50)
D-638	29.8	69.7	0.5	C. S. Ross	R. E. Stevens (3)	Pegmatitic vein in dunite, Corundum Hill, Macon Co., N. C.	1.74,1.73,1.74,1.74,1.74,1.74 (1.74)	1.35,1.34,1.34,1.35,1.35,1.36 (1.35)
6(152)	34.6	64.0	1.4	R. C. Emmons	L. C. Peck (3)	Gneissoid granodiorite, Spanish Peak, Calif.	1.72,1.71,1.73,1.74 (1.73)	1.37,1.35,1.36,1.37 (1.37)
8(144)	36.6	61.5	1.9	"	"	Granodiorite, Crestmore, Calif.	1.76,1.79,1.76,1.76,1.74,1.77,1.77,1.76,1.75 (1.76)	1.34,1.31,1.32,1.32,1.31,1.32,1.33,1.37,1.35 (1.33)

(continued)

TABLE 2. LIST OF SAMPLES AND DATA—(*Continued*)

Sample No.	Composition Mol °/o An	Ab	Or	Source of Sample	Chemical Analyst (Reference to analysis)*	Occurrence and Locality of Specimen	Measurements of Reflection Separations, with Average in Parentheses $2\theta(1\bar{3}1) - 2\theta(131)$	$2\theta(220) - 2\theta(1\bar{3}1)$
						Plagioclases from metamorphic rocks		
C41	50	50	0	H. H. Hess	E. H. Oslund (2)	Metamorphosed gabbro, Salem District, India	1.87,1.89,1.88,1.88 (1.88)	1.22,1.20,1.22,1.21 (1.21)
12(97)	54.5	43.3	2.2	R. C. Emmons	L. C. Peck (3)	Greenstone recrystallized to anorthositic rock, Eland, Wis.	1.82,1.82,1.82,1.83 (1.82)	1.30,1.27,1.27,1.26 (1.27)
13(92)	54.9	42.8	2.3	"	"	Greenstone recrystallized to anorthositic rock, Tigerton, Wis.	1.82,1.80,1.80,1.81 (1.81)	1.26,1.30,1.32,1.31 (1.30)
Howie 2941	56.7	40.0	3.3	R. A. Howie	R. A. Howie (7)	Metamorphosed norite, Salem District, India	1.86,1.86,1.86 (1.86)	1.23,1.24,1.23 (1.23)
HGIF-An-53	88.7	8.8	2.5	A. H. Nicol	E. H. Oslund (8)	Vein-like masses in schist, Olricksfjord Area, N. W. Greenland	2.22,2.23,2.23 (2.23)	0.86,0.84,0.84 (0.85)
354	93.3	6.1	0.6	F. S. Miller	F. A. Gonyer (9)	"Hornblende-olivine norite," San Luis Rey Quad., Calif.	2.25,2.24,2.25,2.23,2.26 (2.25)	0.81,0.81,0.81,0.81,0.80 (0.81)
C50	98.0	2.0	0	A. P. Subramaniam	E. H. Oslund (10)	Metamorphosed anorthosite, Salem District, India	2.28,2.26,2.27,2.28 (2.27)	0.78,0.78,0.78,0.77 (0.78)
						Plagioclases from volcanic rocks		
Larsen 9	11.1	82.1	6.8	E. S. Larsen, Jr.	F. A. Gonyer (6)	Phenocrysts from obsidian, No Agua, N. M.	1.70,1.71,1.71,1.70 (1.71)	1.36,1.36,1.35,1.36 (1.36)
Larsen 8	28.2	64.6	7.2	"	"	Phenocrysts from rhyolite, Uncompahgre Quad., N. M.	1.77,1.77,1.77 (1.77)	1.27,1.27,1.26 (1.27)
Larsen 4	30.2	62.5	7.3	"	"	Phenocrysts from quartz latite, Uncompahgre Quad., N. M.	1.76,1.77,1.72 (1.75)	1.29,1.28,1.31 (1.29)
7(64)	34.8	62.2	3.0	R. C. Emmons	L. C. Peck (3)	Dacite, San Luis Obispo Co., Calif.	1.85,1.85,1.84,1.84,1.84,1.87,1.83 (1.85)	1.21,1.23,1.21,1.21,1.23,1.21,1.22 (1.22)
Larsen 7	35.4	54.1	10.5	E. S. Larsen, Jr.	F. A. Gonyer (6)	Phenocrysts from quartz latite, San Cristobal Quad., N. M.	1.83,1.83,1.82 (1.83)	1.22,1.21,1.21 (1.21)
Larsen 5	41.0	53.9	5.1	"	"	Phenocrysts from latite-andesite, Conejos Quad., N. M.	1.85,1.84,1.84 (1.84)	1.20,1.19,1.20 (1.20)
Larsen 3	45.8	48.1	6.1	"	"	Phenocrysts from andesite, Summitville Quad., N. M.	1.84,1.85,1.86,1.84 (1.85)	1.22,1.20,1.20,1.22 (1.21)
Larsen 2	60.5	37.5	2.0	"	"	Groundmass of basalt, Servilleta Plaza, N. M.	2.01,2.01,2.01 (2.01)	1.07,1.09,1.08 (1.08)
KN7	63.8	35.0	1.2	F. C. Kracek	O. von Knorring (5)	Phenocrysts from basalt, Clear Lake, Utah	2.07,2.06,2.06 (2.06)	1.02,1.02,1.03 (1.02)
Larsen 1	65.5	33.3	1.2	E. S. Larsen, Jr.	F. A. Gonyer (6)	Groundmass of basalt, Buffalo Butte, N. M.	2.05,2.06,2.05 (2.05)	1.05,1.04,1.06 (1.05)
19(61)	68.8	30.4	0.8	R. C. Emmons	L. C. Peck (3)	Basalt porphyry, Lake Co., Ore.	2.12,2.10,2.10,2.10 (2.10)	0.98,1.00,1.00,1.03 (1.00)
						Plagioclases from anorthositic masses near tops of gabbroic sills, Northern Minnesota		
15(53)	58.6	38.6	2.8	R. C. Emmons	L. C. Peck (3)	East of Duluth, Minn.	1.96,1.96,1.97,1.98 (1.97)	1.12,1.12,1.12,1.13 (1.12)
17(52)	63.2	34.0	2.8	"	"	Grand Marais, Minn.	1.94,1.95,1.95,1.95 (1.95)	1.14,1.13,1.14,1.13 (1.14)
21(51)	69.2	29.3	1.5	"	"	Crystal Bay, Minn.	2.05,2.07,2.07,2.06 (2.06)	1.02,1.03,1.03,1.03 (1.03)
22(54)	69.5	29.8	0.7	"	"	Grand Marais, Minn.	2.06,2.08,2.08,2.10 (2.08)	1.00,0.98,0.99,0.98 (0.99)
KN5	69.7	30.3	0.0	F. C. Kracek	O. von Knorring (5)	Crystal Bay, Minn.	2.13,2.11,2.12 (2.12)	0.96,0.97,0.97 (0.97)
24(1B)	75.7	23.4	0.9	R. C. Emmons	L. C. Peck (3)	Split Rock Point, Minn.	2.14,2.15,2.17,2.18 (2.16)	0.93,0.93,0.91,0.91 (0.92)
25(1A)	76.2	23.1	0.7	"	"	Split Rock Point, Minn.	2.15,2.16,2.18,2.17 (2.16)	0.92,0.95,0.94,0.90 (0.93)
						Plagioclases from Adirondack-type anorthosite massifs		
9(24)	47.7	48.8	3.5	R. C. Emmons	L. C. Peck (3)	Anorthosite, Essex Co., N. Y.	1.78,1.79,1.80,1.80 (1.79)	1.29,1.30,1.29,1.30 (1.29)
KN6	51.9	45.8	2.3	F. C. Kracek	O. von Knorring (5)	Anorthosite, Isle of Paul, Labrador	1.75,1.72,1.75 (1.74)	1.35,1.35,1.35 (1.35)
						Plagioclases from plutonic and hypabyssal rocks, details of occurrence not known		
Howie 8-347	30.0	68.0	2.0	R. A. Howie	R. A. Howie (7)	Charnockitic rock, Mt. Wati, Uganda	1.72,1.73,1.72 (1.72)	1.34,1.33,1.32 (1.33)
Howie 2270	31.8	61.8	6.4	"	"	Charnockitic rock, Salem District, India	1.71,1.68,1.69 (1.69)	1.40,1.39,1.40 (1.40)
Howie 6436	32.1	66.0	1.9	"	"	Charnockitic rock, Meenambakam, Madras, India	1.73,1.70,1.70 (1.71)	1.36,1.39,1.39 (1.38)
10(132)	50.2	48.8	1.0	R. C. Emmons	L. C. Peck (3)	Hornblende gabbro, Shelby, N. C.	1.85,1.85,1.83,1.85,1.84 (1.84)	1.24,1.24,1.25,1.23,1.24 (1.24)
11(151)	50.8	46.7	2.5	"	"	Diorite, Fresno Co., Calif.	1.82,1.84,1.83,1.84 (1.83)	1.27,1.26,1.23,1.22 (1.25)
14(157)	58.3	40.2	1.5	"	"	Gabbro, Merrill, Wis.	1.80,1.83,1.82,1.82 (1.82)	1.29,1.25,1.27,1.28 (1.27)
16(132)	59.0	40.0	1.0	"	"	Hornblende gabbro, Shelby, N. C.	1.89,1.90,1.90,1.89 (1.89)	1.18,1.18,1.19,1.19 (1.18)
18(134)	65.0	32.9	2.1	"	"	Diabase, Chester Co., Penn.	2.00,2.02,2.01,2.01 (2.01)	1.12,1.10,1.09,1.08 (1.10)
20(22)	69.2	29.8	1.0	"	"	Orthoclase quartz gabbro, Wichita Mts. Okla.	1.97,1.99,1.97,1.97 (1.97)	1.12,1.13,1.15,1.12 (1.13)
23(165)	72.7	26.3	1.0	"	"	Gabbro, Lincoln Co., Wis.	2.03,2.05,2.03,2.06 (2.04)	1.05,1.07,1.06,1.05 (1.06)
27(156)	80.6	17.5	1.9	"	"	Troctolite, Merrill, Wis.	2.16,2.16,2.14,2.15 (2.15)	0.92,0.91,0.93,0.91 (0.92)

*References to chemical analyses are at the end of the paper.

TABLE 3. CHEMICAL ANALYSIS OF BYTOWNITE HGIF-AN-53

	Wt. %	Mol numbers
SiO_2	44.87	747
Al_2O_3	34.93	342
TiO_2	0.01	
Fe_2O_3 FeO	0.09	
MgO	0.00	
SrO	0.70	7
CaO	17.41	311
BaO	0.00	
Na_2O	0.98	16
K_2O	0.42	4.5
Rb_2O	0.00	
H_2O+	0.59	
H_2O-	0.04	
	100.04	

Mol % An*

$$= \frac{311+7}{(311+7)+2(16+4.5)} \times 100$$

$$= 88.7$$

Analyst: Eileen H. Oslund.

Collector: Allan H. Nicol.

Occurrence and locality: Large single crystal in vein-like mass in hornblende-actinolite schist, Olricksfjord area, N. W. Greenland.

Alteration: Slight alteration to unidentified material along cracks.

* With SrO calculated as CaO and K_2O presumed to be present as potash feldspar in solid solution.

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(8) TABLE 3 of this paper.

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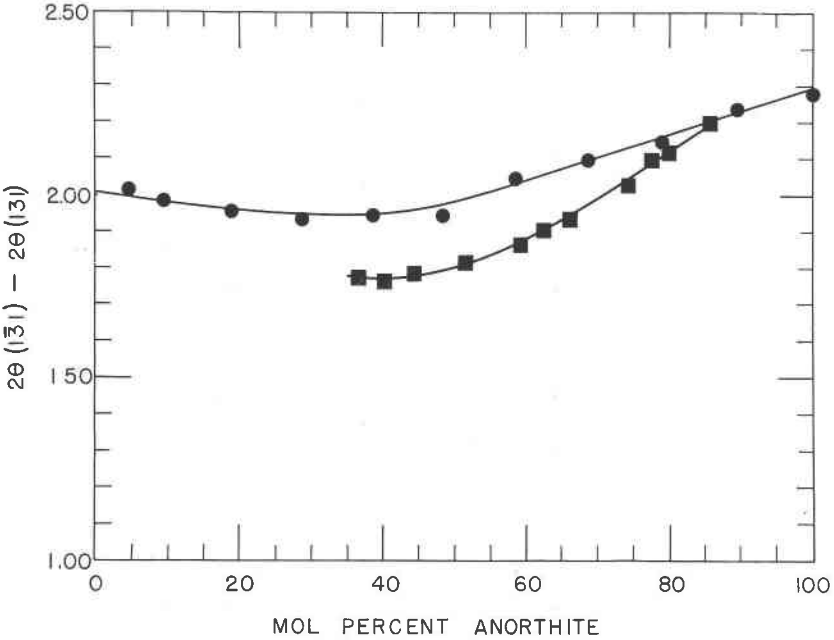

FIG. 2. Variation of 2θ $(1\bar{3}1)-2\theta$ (131) with composition in plagioclases synthesized in the dry way (circles) and in plagioclases from thick stratiform mafic intrusions (squares).

from some of the same intrusions and avoids the use of the term "low-temperature plagioclases," which has yet to be defined.

The curves for the plagioclases synthesized in the dry way and for the plagioclases of the Bushveld type will now be used as a basis for comparison with the other plagioclases which were studied. The $(1\bar{3}1)-(131)$ separations for all the other plagioclases are plotted in Fig. 3, in which the curves of Fig. 2 are reproduced as full lines. The dashed line is representative of the studied samples of sodic plagioclases from pegmatites and granites (denoted by ×'s). The curve must be regarded as tentative, because there is some doubt as to the existence of homogeneous plagioclases of composition An 6± to An 17± in rocks formed at low temperatures or slowly cooled from higher temperatures.* If there is no

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* Laves (1954) has shown that in the composition range An 6 to An 17 all the plagioclases which he studied from pegmatites and granites consist of two submicroscopically discrete phases. J. V. Smith (1956) states that the phases may or may not be resolved in powder diffraction patterns; if the phases are resolved, reflections representing only one phase may be mistakenly measured as reflections representing plagioclase of the bulk composition of the sample; if the phases are not resolved, peaks in powder patterns may be too broad to be measurable, as in the case of our sample number 2(10)(An 11.0). Two of the samples represented in Fig. 3, numbers 3(73)(An 15.4) and 4(156)(An 15.7), were

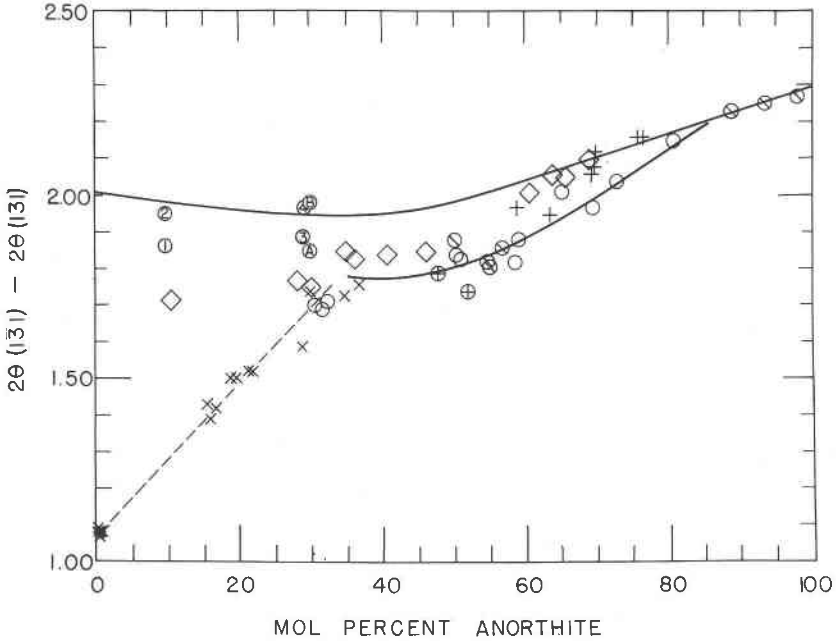

FIG. 3. Curves in full lines are the curves of Fig. 2. Symbols represent $2\theta\,(1\bar{3}1)-2\theta\,(131)$ versus composition for plagioclases of the following origins:

×	from pegmatites and granites
⦸	from metamorphic rocks
◇	from volcanic rocks
+	from anorthositic masses near the tops of gabbroic sills, northern Minnesota
⊕	from Adirondack-type anorthosite massifs
○	from miscellaneous rocks, details of occurrence not known
① and ③	synthesized from glass at 640° C. and 10,000 bars water pressure
② and ④	synthesized from crystalline material at 640° C. and 10,000 bars water pressure
Ⓐ	natural plagioclase from pegmatite heated for 9 hours at 1122° C.
Ⓑ	natural plagioclase from pegmatite heated for 24 hours at 1140° C.

real curve in this composition range, considerable uncertainty must be attached to the portions of the curve from An 0 to An 6 and from An 17 to An 35, because the slope of these portions depends to a large extent on the assumed validity of the projection of the curve through the An 6 to An 17 region.

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studied by Laves (1954) and are known to consist of two phases. The two phases were not resolved in our powder patterns, and, since the values for the samples fall close to the dashed line in Fig. 3, the separation of the unresolved composite peaks is apparently representative of plagioclase of the bulk composition of the samples.

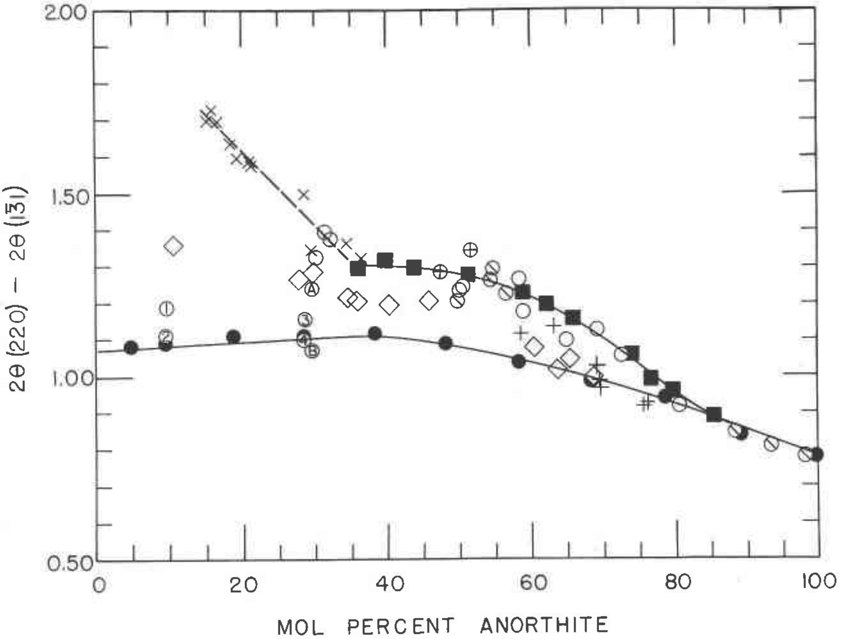

FIG. 4. Variation of 2θ (220) − 2θ (1$\bar{3}$1) with composition for plagioclases synthesized in the dry way (closed circles) and plagioclases from thick stratiform mafic intrusions (closed squares). Other symbols are the same as in Fig. 3.

In the composition range An 30 to An 80, the large scatter of the points representing natural plagioclases is to be noted. Most of the scatter is due to the fact that the values for plagioclases from volcanic rocks and from anorthositic masses in thin gabbroic sills are intermediate between the curve for the plagioclases synthesized in the dry way and the curve for the Bushveld-type plagioclases; they approach the curve for the plagioclases synthesized in the dry way in the composition range An 65 to An 75. This presumably means that these plagioclases are "frozen" in somewhat higher temperature states than those represented by the Bushveld-type plagioclases, probably as a result of the more rapid cooling of the small extrusive and hypabyssal bodies in which they occur. However, the values obtained for some other samples are not readily explainable in terms of cooling rates. For instance, sample number *C*41 (An 50) is from a highly metamorphosed gabbro in a gneiss terrane in India (Subramaniam, 1956); the (1$\bar{3}$1) − (131) separation is significantly greater (i.e., tending toward "higher temperature" values) than that for Bushveld-type plagioclase of the same composition, which is not to be expected if the rate of cooling is the chief factor controlling the variation. Again, sample number *KN*6 (An 51.9) is from an Adirondack-type anorthosite massif at Isle of Paul, Labrador (Kracek and Neuvonen, 1952); the $(1\bar{3}1)-(131)$ separation is significantly smaller than that for Bushveld-type plagioclase of the same composition, suggesting that under certain (unspecified) conditions plagioclases may attain states of inversion which are apparently representative of lower temperatures than are the states attained by the Bushveld-type plagioclases. These isolated observations serve to point out the fact that the evidence of the state of inversion of a plagioclase afforded by *x*-ray parameters may be difficult to explain in terms of the geological evidence; the obvious conclusion is that more must be learned of the *x*-ray parameters of plagioclases from various geological environments.

In the composition range An 85 to An 100, there is no measureable difference between the $(1\bar{3}1)-(131)$ separations for the plagioclases synthesized in the dry way and for the natural plagioclases which were studied, namely one Bushveld-type plagioclase and three from metamorphic rocks.

In order to discover whether similar relations would have resulted from measurements of other reflection separations, the (220)—$(1\bar{3}1)$, $(\bar{1}32)$—$(24\bar{1})$, and $(\bar{1}32)$—$(\bar{2}41)$ separations were also measured. Figure 4 is a plot of all measurements made of the (220)—$(1\bar{3}1)$ separation. A comparison with Figs. 2 and 3 will show that the relations are almost identical (although the relative positions of the curves are inverted). Closely similar results were also obtained from the $(\bar{1}32)$—$(24\bar{1})$ and $(\bar{1}32)$—$(\bar{2}41)$ separations.

The variation with composition of the *F–G* separation of Goodyear and Duffin (1954) appears to be fundamentally different from the variation of the $(1\bar{3}1)$—(131) separation. In attempting to measure the *F–G* separation in Geiger counter powder patterns, the writers found that both reflections are weak, and that the *F* reflection is not resolved from neighboring reflections. J. V. Smith (1956, p. 66) makes the following statement: "The *F* and *G* reflections of Goodyear and Duffin have the following indices: *G*, $(20\bar{4})$; *F*, probably mainly $(04\bar{3})$ but includes other reflections as well. Diffractometer records of reflection *F* often show a complex structure and it is doubtful whether it is suitable for measurement." Since the *F–G* separation could not be measured with an accuracy comparable to the other separations, its variation was not studied by the writers.

HEATED NATURAL PLAGIOCLASES AND PLAGIOCLASES SYNTHESIZED HYDROTHERMALLY

Tuttle and Bowen (1950) found that the values of *x*-ray parameters of natural plagioclases are changed to values similar to those of synthetic plagioclases by prolonged heating at temperatures near the melting points, and that in some cases intermediate values can be achieved. Goodyear and Duffin (1956) and J. V. Smith (1956) have confirmed and enlarged upon these results. In the present study, heating of pegmatite plagioclase *D*638 (An 29.5) for nine hours at 1122° C. caused the reflection separations to change to values about halfway between the original values and those of plagioclase of the same composition synthesized in the dry way. Heating for 24 hours at 1140° C. caused a change to values similar to those of plagioclase synthesized in the dry way (see Figs. 3 and 4). These and the previously mentioned experiments demonstrate that the state of inversion of a plagioclase is in some way a function of temperature, but the inability to reverse the reactions accompanying changing temperature has to date hindered experimental studies of the equilibrium relations.

Preliminary experiments to investigate the nature of plagioclases synthesized hydrothermally have been carried out. Plagioclases of composition An 10 (wt. %) and An 30 (wt. %) were synthesized at 10,000 bars of water pressure and 640° C., a temperature which is slightly below the extrapolated solidus temperatures at this water pressure (Yoder, unpublished data on Ab–An–H_2O). For each composition, two different starting materials were used, one glass and the other crystalline plagioclase synthesized in the dry way. In each case, the plagioclase produced from the crystalline starting material has reflection separations nearly the same as the starting material, whereas the plagioclase produced from the glass starting material has reflection separations significantly closer to the reflection separations of natural plagioclase of the same composition from pegmatite (see Fig. 3). It is doubtful whether either product represents the modification of plagioclase which is in equilibrium at 640° C., but the results suggest the possibility that experiments involving long periods of crystallization in the presence of water at low temperatures may ultimately lead to an understanding of the equilibrium relations.

LACK OF CONTINUITY OF THE CURVES

As may be seen in Figs. 3 and 4, there is a marked break in slope between the curve for plagioclases from granites and pegmatites and the curve for Bushveld-type plagioclases around An 35; there is a less marked discontinuity between the curves for Bushveld-type plagioclases and metamorphic plagioclases at An 85–90. At about the same compositions, one of the writers (J. R. Smith, 1954) has found discontinuities in the optical property curves for plagioclases from the same environments. Single-crystal *x*-ray studies to date have not disclosed any discontinuous change in the crystal structure of plagioclases near An 35, but rather a gradual disappearance in the An 20 to An 30 region of certain subsidiary reflections characteristic of more calcic plagioclases from low-temperature environments (Gay and Smith, 1955). Likewise, in the range An 80–90, there appears to be a gradual change from a body-centered to a primitive unit cell in plagioclases from low-temperature environments (Gay, 1954). Inasmuch as the relations in these apparently critical regions might be different for suites of plagioclase samples from different geological environments, no interpretation of the discontinuities can be given on the basis of the present restricted sampling.

USE OF REFLECTION SEPARATIONS FOR DETERMINATIVE PURPOSES

As previously stated, other workers have recently published several studies of the variation of reflection separations with composition and with thermal history in the plagioclase feldspars. *The applicability and the accuracy of determinative curves based on the variations have been overestimated by some of the workers*, largely because the plagioclases studied were too few to adequately sample the variations existing in natural plagioclases from various source rocks. The curves may be valid for the particular samples studied, but had different samples been studied, different curves would have resulted. For instance, Claisse (1950), after studying reflection separations in eight natural plagioclases of various origins, concludes that a plagioclase composition can be determined to an accuracy of 1% An with the use of his curves, regardless of the origin of the sample. Goodyear and Duffin (1954) subsequently measured the same reflection separations measured by Claisse, using different samples; their data indicate that Claisse's curves are incorrect, and should not be used for determinative purposes. To a lesser degree, Goodyear and Duffin's curves for plagioclases "of presumably low-temperature origin" (Goodyear and Duffin, 1954, p. 306) are in turn subject to limitations, mostly because of the small number of samples used, but partly because three of the samples presumed by them to be of low-temperature origin are transitional between Bushveld-type plagioclase and plagioclase synthesized in the dry way (Goodyear and Duffin's samples numbered 7, 8, and 9; the same samples as our numbers 17(52), 18(134), and *KN*5). From the present study of a large number of natural plagioclases of various origins, the following conclusions regarding the use of reflection separations for determinative purposes may be drawn:

(*a*) The distribution of the points plotted in Fig. 3 demonstrates that natural plagioclases do not fall into two unique series. Volcanic plagioclases do not appear to constitute a unique high-temperature series, but rather to be in various states of inversion between a "maximum" high-temperature state and some undefined lower temperature state. Similarly, plagioclases which have been hitherto presumed to be of "low-temperature" origin do not all fall into a unique low-temperature series. This being the case, the composition of a plagioclase cannot be determined by comparing its reflection separations with the curves of the present study or with curves previously published by others, because there is no a priori way of knowing how closely a particular plagioclase is represented by any of the available curves.

(*b*) Reflection separations for Bushveld-type plagioclases fall on a well-defined curve (Figs. 2 and 4). This suggests the possibility that a reflection separation versus composition curve could be determined for any suite of plagioclases of closely similar origins and thermal histories; such a curve could then be used for determinations of composition of other plagioclases from the same suite in the composition ranges where the slope of the curve is great enough to attain the desired accuracy. Curves so determined should be used with discretion. For instance, it is possible that plagioclases in the border zones of an intrusive body would be represented by a curve different from that representing plagioclases from the central parts of the body.

(*c*) If the composition of a plagioclase is known, and if it is in the range An 0 to about An 75, an estimate of its state of inversion may be made by measuring the $(\bar{1}31)$—(131) separation and comparing its value with Fig. 3. Little quantitative value can be placed on the estimate, because different reflection separations give somewhat different estimates of the degree of inversion, as may be seen by comparing the positions of the points representing volcanic plagioclases in Fig. 3 with the positions of the same points in Fig. 4. The interpretation of the state of inversion of a plagioclase is subject to uncertainty because of the lack of knowledge concerning the relative importance of the various factors which might influence it.

ACKNOWLEDGMENTS

Most of the chemically analyzed samples studied were available at the Geophysical Laboratory, having been previously used for other purposes. The writers are indebted to the collectors and contributors of the samples, whose names are listed in Table 2. J. F. Schairer of this Laboratory generously made his samples of synthetic plagioclases available for study. Much valuable information was gained from the manuscript of J. V. Smith's paper on "The powder patterns and lattice parameters of plagioclase felspars," to which reference has been made throughout the text.

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