ANTIPHASE DOMAIN STRUCTURE OF THE INTERMEDIATE COMPOSITION PLAGIOCLASE FELDSPARS.

C. G. Cinnamon[1] and S. W. Bailey, *Department of Geology and Geophysics, University of Wisconsin Madison. Wisconsin, 53706*

Abstract

By analogy with long-period superlattices in alloys, the split-"b" satellites in intermediate composition plagioclases arise from the extra periodicity of a regular three-dimensional antiphase domain system. Average domain sizes calculated along the X, Y, and Z axes from the separation of split-"b" satellites serve as a basis for discussing the concepts of domain size variation and domain stability. The number of domain boundaries per unit volume is a function of domain size and is one of the factors influencing crystal stability. The breakdown of the intermediate plagioclase domain structure at the peristerite boundary and near An_{75-80} may be related to prohibitively high boundary energies at these compositions. The strength of a plagioclase crystal, as measured by indentation microhardness, is related to domain size and to the nature of the narrow domain walls.

Introduction

The plagioclase feldspars belong to a series in which the composition varies continuously from albite $NaAlSi_3O_8$ to anorthite $CaAl_2Si_2O_8$ by substitution of CaAl for NaSi, but which deviates from the definition of a solid solution series because of several structural complexities. These complexities involve "high," "intermediate," and "low" structural forms that result primarily from differences in cooling rates. The complexities are related ultimately to the degree and nature of ordering of Si and Al. Within the "low" structural series, which is of primary concern in this paper, known complexities include unmixing, symmetry changes, and antiphase domains (Fig. 1).

The crystal structure of the plagioclases of intermediate composition (An_{15} to An_{75-80}) within the "low" series is known only in outline. The existence of two sets of satellite reflections indicates that the true unit cell is composed of numerous quasi-unit cells or subcells. The positions of these satellites in reciprocal space change as a function of composition, which implies that the true unit cell is a periodic unit that changes size with composition.

The origin of the satellites has not been explained successfully, and they have been left out of routine crystal structure analyses. As a result we know only what an "average" unit cell looks like, and have little understanding of the stability relations among various "low" plagioclases from a crystal chemical viewpoint. Megaw (1960) proposed that

[1] Present address: Department of Geological Sciences, University of Maine, Orono, Maine 04473.
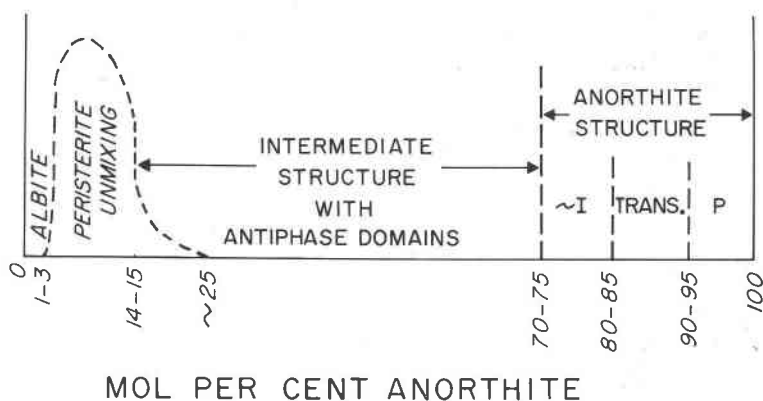
MOL PER CENT ANORTHITE

Fig. 1. Schematic representation of the structural types present in the plagioclase series for the "low" structural state. Lamellate structures observed at An_{45-55} and An_{67-83} have been omitted for the sake of clarity.

the true structural unit is a domain that is antiphase with respect to neighboring domains, and that the crystal structure of the low plagioclases with intermediate composition must be explained on the basis of antiphase domain theory. Recent models of the intermediate plagioclase structure involve domains that are differentiated by incipient chemical unmixing (Korekawa and Jagodzinski, 1967; Smith and Ribbe, 1969; Christie, 1969). Smith and Ribbe (1969) present an extensive review of the available evidence concerning the nature of the plagioclase structure.

In the present paper an analogy is made between the domain structure of the low plagioclases of intermediate composition and a special type of regular antiphase domain system known in alloys—the long-period superlattice. The concept of domain stability is introduced and discussed, and data are presented to show the relationship of micro-hardness to domain size and to domain wall structure. A later paper will study the diffuseness and intensity variations of satellite reflections as a function of composition and their interpretation in terms of domain structure.

SATELLITE REFLECTIONS IN THE PLAGIOCLASES

The diffraction maxima of the plagioclase feldspars can be placed in four classes:

"a"	$h+k$ even,	l even
"b"	$h+k$ odd,	l odd
"c"	$h+k$ even,	l odd
"d"	$h+k$ odd,	l even

In this classification, l refers to the conventional anorthite cell, with $c \cong 14$ Å. In the intermediate composition plagioclase range there are also two groups of satellites that are designated "e" or split-"b", and "f" or split-"a" reflections, which replace or flank the "b" and "a" reflections respectively.

Upon comparison it is obvious that at least a superficial analogy must exist between intermediate composition plagioclases and long-period superlattices in metallic compounds. Both exhibit satellite X-ray reflections around fundamental unit cell (subcell) reflections, which in the plagioclases correspond to those of the 7 Å albite cell. In addition, in both groups the normal superlattice reflections indicative of a multiple repeat distance disappear and are replaced by satellites flanking the vacant superlattice position. In plagioclases the satellites around the fundamental subcell reflections are the split-"a" reflections and the satellites around the vacant superlattice positions are the split-"b" reflections. The splitting vectors of the split-"a" satellites are parallel to, but twice as long as, those of the split-"b" satellites.

McConnell and Fleet (1963) have provided important experimental evidence for the existence of a regular domain system in a plagioclase of composition An_{50}. They observed an electron-optical image that consisted of periodically repeating parallel stripes. McConnell and Fleet point out that the electron diffraction intensity distribution in reciprocal space is directly comparable to that observed by Glossop and Pashley (1959) in the long-period superlattice alloy CuAuII. The stripes in both of these studies were observed by isolating the split superlattice satellites in the objective aperture of an electron microscope in the dark field technique. In the case of CuAuII the splitting of the superlattice reflections is definitely due to the existence of an antiphase relationship between domains, and the stripes correspond to the image of the phase lattice that results from the sequence of domains (Sato and Troth, 1965).

Raether (1957) has shown that when a mistake producing an out-of-step occurs between N adjacent point rows of Q atoms each, the phase factor takes the form

$$G_3 = \frac{\sin \pi Q A_3}{\sin \pi A_3} \cdot \frac{\sin \pi N(Q A_3 - \phi)}{\sin \pi(Q A_3 - \phi)} = G_3{}^1 \cdot G_3{}^2$$

The first term $G_3{}^1$ is the known phase factor of a point row of Q atoms and has a maximum at $A_3 = h_3$. The second term $G_3{}^2$ produces maxima symmetric from the normal reflection (h_3) at a distance $\Delta A_3 = \phi/Q$, where ϕ is the mistake phase angle (Fig. 2). The resultant of these two terms for $\phi < 1$ is a zero- or low-intensity maximum at h_3 flanked by two satellites.
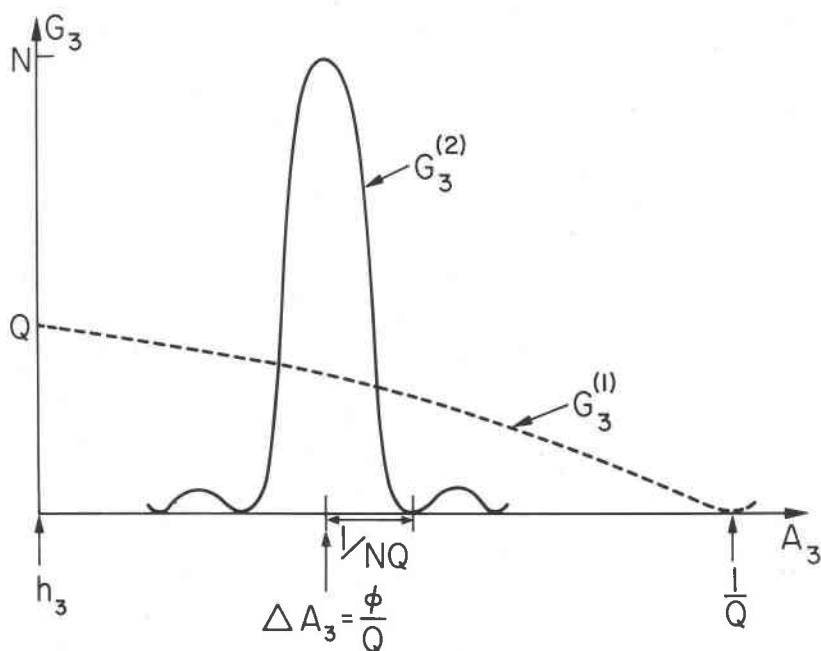
FIG. 2. Variation of the function $G_3 = G_3{}^1 \cdot G_3{}^2$ as a function of the coordinate A_3. Modified from Raether (1957).

A specific solution for this type of equation involves knowledge of the relations between domains. That is, what rules govern the out-of-steps that produce the antiphase relations between domains? In the intermediate composition plagioclases there is little experimental evidence concerning the existence or the nature of out-of-step, and the splitting of the superlattice reflections has not been explained formally. The investigation of McConnell and Fleet, however, provides definite evidence that a phase modulation does occur.

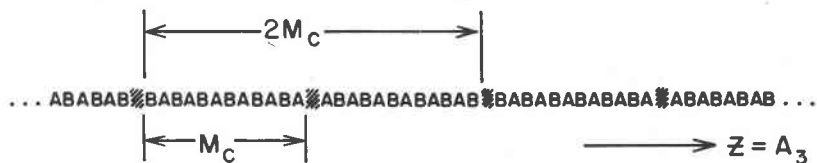Figure 3 shows an example of a simple one-dimensional antiphase

FIG. 3. Schematic representation of a one-dimensional antiphase domain system. The structure repeats after two domains $(2M_c)$ in the Z direction.

structure extending in the Z crystallographic direction. The structure has a c repeat of $2M$, because it contains two non-identical domains of M subcells each that alternate in a regular pattern. By analogy with long-period superlattices that have been studied in alloys, this system can be expected to produce extra reflections directly related to the antiphasing of periodicity $2M$. In metals additional periodic modulations occur with the same period as the antiphase domain size M. These modulations are related to periodic changes of lattice spacing and scattering factors between domains (Watanabe, 1961), and they produce a second set of satellites of periodicity M symmetrical about fundamental unit cell reflections. In the plagioclases the two analogous sets of extra reflections correspond to type "e" (split-"b") and type "f" (split-"a") satellites, respectively.

Based on a three-dimensional model of this sort, the magnitude of the phase modulation in plagioclases can be calculated by setting the observed distance in reciprocal space of a split-"b" satellite from its relevant superlattice position as proportional to $\pm 1/2M$, or the separation between satellite pairs as proportional to $1/M$. The domain size M determined by this calculation will be an average value, because adjacent domains usually will not be of equal sizes.

Korekawa and Jagodzinski (1967), Jamieson, DeFontaine, and Abrahams (1969), and Christie (1969) have suggested more specific models for the plagioclases based on chemical and cell dimension differences between domains, but not including anti-phase relationships. It is true that such models could also account for the observed satellite reflections and striped electron-optical images on a qualitative basis. It is emphasized here, however, that any regular modulation that is not accompanied by narrow, high-energy domain walls is not sufficient to explain the observed physical properties of the intermediate plagioclases that are discussed in the following sections. The present authors favor an antiphase domain model in which adjacent domains also differ chemically and geometrically.

Split-"b" Satellite Separations

The separations of the split-"b" reflections on oscillation photographs about the X, Y, and Z axes have been measured previously by Gay (1956). In the present investigation additional separations were measured only on Z axis oscillation photographs and were added to similar measurements by Doman, Cinnamon, and Bailey (1965). Because of the tendency for diffuseness of the split-"b" reflections the separations were measured from profiles constructed with a microdensitometer. In all investigations the satellite spacings were measured in

terms of 2π times the distance of the satellite from the O-layer line. Gay found that these values of δa, δb, δc vary linearly as a function of composition. Doman *et al.* discovered a discontinuous linear variation of δc. The data of Doman *et al.* have been replotted in the present study to incorporate newly acquired electron probe analyses on their samples, and the discontinuous nature of this curve is ignored in order to simplify the discussion that follows.

Bown and Gay (1958) incorporated the split-"b" separation data into an S vector to give the direction of domain alignment. S is given by

$$S = \delta h \cdot a^* + \delta k \cdot b^* - \delta l \cdot c^*. \tag{2}$$

The reciprocal, $1/|S|$, has been cited as a measure of domain size by Smith and Ribbe (1969). This usage of equation (2), is based on the assumption that the same split-"b" reflections are being viewed in projections along the X, Y, and Z axes, however and reduces the antiphase structure of the plagioclases to a one-dimensional system. This is an unproven assumption that has become accepted as a basic "truth" in the literature concerning the structure of the intermediate composition plagioclases. It seems much more probable from the general nature of the results discussed below that entirely different reflections with different splitting vectors are being viewed along each axis. It is not surprising, therefore, that domain size calculated from the diffraction stripes observed by McConnell and Fleet ($\sim$30 Å in a crystal of composition An_{50}) does not match the domain size calculated from the S vector ($1/|S| = 70$ Å). On the other hand a very good match with the McConnell and Fleet value is obtained for size along the Z crystallographic direction (26.5 Å to 34.5 Å for nine samples An_{49-51}) by the method described below.

Before domain sizes can be calculated, the δa, δb, and δc values representing distances from the O-layer line to the closest satellite-layer line, must be converted to ΔA, ΔB, and ΔC values that represent the separation distances between related satellites. Equations (3) to (5) provide these conversions and relate the separations to average domain sizes M_a, M_b, and M_c along the three crystallographic axes.

$$\Delta A = \frac{\lambda}{a} \cdot \frac{\delta a}{180°} = \frac{\lambda}{a} \cdot \frac{1}{M_a} \quad \text{and} \quad M_a = \frac{180°}{\delta a}$$

$$\Delta B = \frac{\lambda}{b} \cdot \frac{\delta b}{180°} = \frac{\lambda}{b} \cdot \frac{1}{M_b} \quad \text{and} \quad M_b = \frac{180°}{\delta b}$$

$$\Delta C = \frac{\lambda}{c} \cdot \left(1 - \frac{\delta c}{180°}\right) = \frac{\lambda}{c} \cdot \frac{1}{M_c} \quad \text{and} \quad M_c = \frac{180°}{180° - \delta c}$$

These equations are approximations, valid only for first level split-"b" reflections, because they assume a linear variation between the zero and second layer lines of the anorthite cell. According to X-ray diffraction geometry this variation follows a tangential function, but is approximately linear for CuKα radiation between 0 and 11 degrees, which includes the area of measurement used in this study. A small systematic error is introduced by this approximation, but this is probably less than the error involved in the measurement of satellite separations.

Domain sizes in the X, Y, and Z directions were calculated from the available split-"b" separation data in the manner shown below. Let $\delta a = 9°$, as for sample 4 of Gay (1956). Then $M_a = 180°/9° = 20$ albite subcells along X, and $M_a \times a = 20 \times 8.177$ Å $= 163.54$ Å. Values of a, b, and c for conversion of M_a, M_b, and M_c to Ångstrom distances for specific compositions were obtained from smooth curves drawn through the data of Bambauer, Eberhard, and Viswanathan (1967). The results for the conversion of Gay's data are given in Table 1A[1] and for the combined data of Doman *et al.* and of the present paper in Table 1B. The data are plotted in Figure 4 as the number of subcells within a domain along the X, Y, and Z directions, and in Figure 5 as domain sizes in Å units. These plots serve to point out general principles associated with variations of antiphase domain size which are discussed in the following section.

Nissen, Eggman, and Laves (1967) have described a lamellate structure with lamellae 1000 Å to 2500 Å in width in structurally low plagioclases of intermediate composition (An$_{45-55}$) that contain more than 2 mol percent Or. Split-"b" satellites indicative of the antiphase domain structure are present whether or not the lamellate structure is observed. This indicates that the phenomena of antiphase domains and of lamellate structure are not directly connected. In addition to this lamellate structure, Nissen (1968) discovered a two-phase structure in the composition range An$_{67-83}$ composed of lamellae of composition An$_{83}$ and a more sodic matrix of composition An$_{65-70}$. The antiphase structure does not appear to be present in the lamellae, whereas it is present in the matrix. The stability of the antiphase domain system may be treated separately from either of the lamellate structures, as is done below, because the two types of structures are on different scales and the existence of the antiphase structure appears to be independent of the lamellae. The ab-

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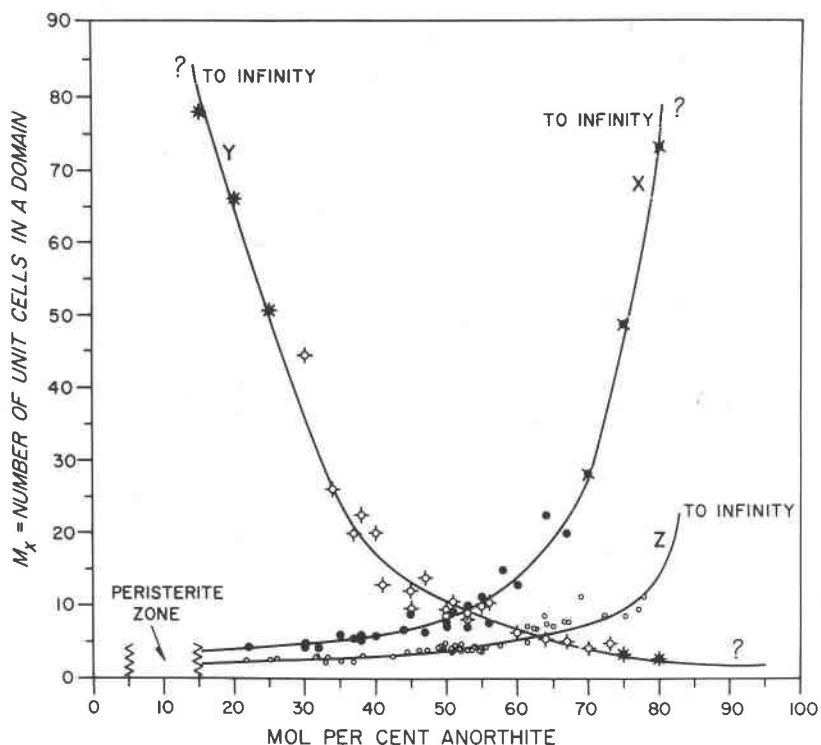
FIG. 4. The calculated variation of domain size (M_x) in terms of the number of subcells in a domain for the crystallographic directions X, Y, and Z. A cross (X) in or on a circle indicates that this is a calculated value derived from the domain volume and the domain size in at least one axial direction. The domain volume was determined from the micro-hardness (H_v) curve (Fig. 10) for these crossed points.

sence of an antiphase domain structure in the lamellae of composition An_{80} may be explained by the fact that the hypothetical domain size for An_{80}, which can be obtained by extrapolation from Figures 4 and 5, would be highly unstable for reasons discussed below.

DOMAIN WALL ENERGY AND STABILITY

Domain walls are regions of high energy, usually treated in a simplified way as a type of surface energy. This energy can arise due to repulsive forces created by the antiphase mistakes. The antiphase boundary energy in the mineral chrysoberyl (var. alexandrite), for example, has been shown to be due to the repulsive force that results when like cations are brought closer together than their equilibrium distance (Phakey, 1969). The less frequently a domain wall is encountered, the smaller is the
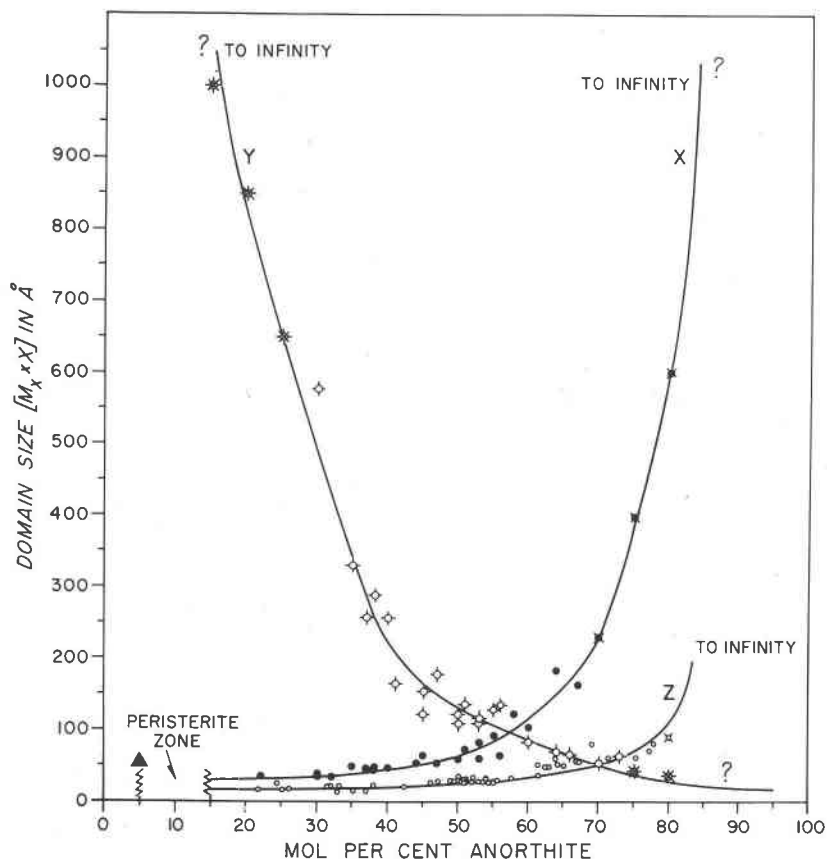
Fɪɢ. 5. Variation of domain size $(M_x \cdot X)$ in Å units, where X is an axial repeat distance, and subcell size in the X, Y, and Z directions for the intermediate composition plagioclases.

energy per unit volume associated with the domain boundaries, and the greater is the stability of the domain structure. Figure 6 shows the relative domain boundary energy of the plagioclases for the X, Y, and Z directions. At the peristerite boundary the very large domain size in the Y direction has relatively little domain boundary energy associated with it. If domain size here is taken as infinity, there would be no energy or instability associated with the existence of domain walls, because there would be no walls present. The small domain sizes in the X and Z directions have correspondingly high energies associated with them and are unstable. Near An_{75-80} the small domain size along Y likewise has a high domain boundary energy.
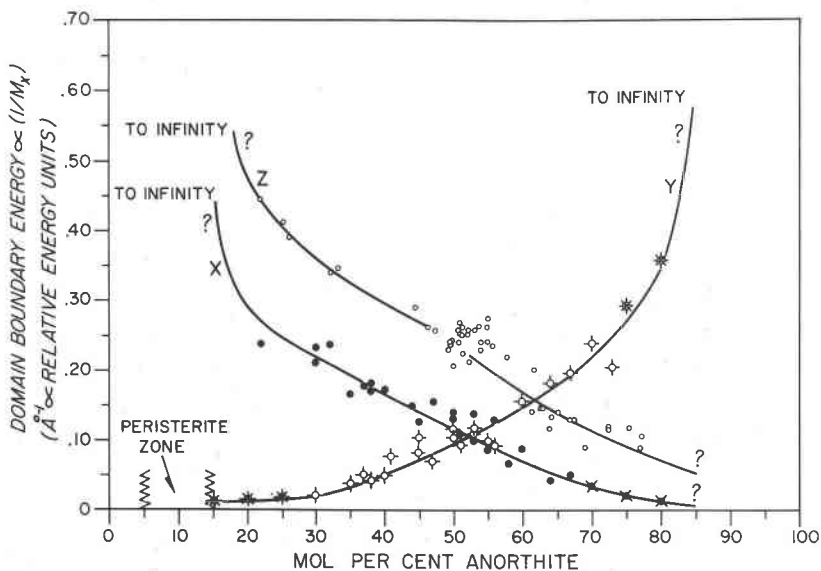
Fig. 6. Relative domain boundary energy of the plagioclase domain system. The scatter of points on the Z curve near An₅₀ is best interpreted in terms of a discontinuous energy curve, but this interpretation is ignored here in order to simplify discussion. Fig. 6 is based in part on the discontinuous δc/An curve given in a paper by Doman *et al.* (1965).

In metal systems it has been found that a domain size of a few sub-cells is the minimum stable size, due to the fact that any domain size smaller than a few subcells gives rise to interactions between domain walls. Because domain walls are areas of high energy of the same sign, the energy term associated with them increases rapidly to infinity when they are close together. This provides an explanation for the behavior of the plagioclase domain size in the Z crystallographic direction, which tends to remain at a two subcell size over a large composition range (Fig. 4). It also explains why the intermediate plagioclase structure breaks down at both the peristerite and transitional-anorthite boundaries. It becomes energetically favorable for the structure to reorganize when one domain dimension becomes unstable and breaks down, rather than to re-form as a two-dimensional or one-dimensional system.

This explanation of domain size variation in the plagioclases requires a different theoretical basis than the presently accepted one. The present explanation of domain size variation is based on minimizing the strain energy associated with domain walls (Megaw, 1960). Based upon this idea, however, it is difficult to see how small domains of only a few sub-cells in size can exist. Due to the repulsive interactions between domain

walls it could be argued that the energy of the domain system would be lowered by maximizing the distance between domain walls and that any tendency toward small domains should be halted at some critical size. This process could even lead to more or less equidimensionally shaped domains.

On the other hand, this difficulty disappears if the stability of domain systems is treated in terms of two domain-related energy terms of different signs that counter-balance each other. On one hand there is the increase in energy due to the existence of domain walls, which is compensated by the reduction in an energy term related to a general property of the lattice. In metal domain systems, the latter is believed to be a reduction in the electronic energy of the crystal (Sato and Troth, 1965). It is suggested here that the existence of a domain system in the plagioclases reduces the general energy term of the lattice due to lattice vibrational energy, so that the domain structure is a stable structure over certain composition ranges. At some point when the balance of energies is dominated by the energy of the domain wall, *i.e.*, at small domain sizes, the domain structure becomes unstable and breaks down. The possibility that the plagioclase domain structure is stable at certain compositions and temperatures should be taken into account in future investigations.

MICROHARDNESS AND DOMAIN SIZE

The study of mechanical properties, such as work hardening, microhardness, and yield strength, has yielded valuable information concerning the nature of specific ordering reactions in different alloys. In the following a comparison of the mechanical properties of long-period superlattices in alloys and in the plagioclases will be presented. This comparison is possible because the energy of domain walls is an important factor in determining physical properties of a crystal, regardless of the stability or origin of a domain system.

Mookherjee and Sahu (1960) discovered that the Vicker's indentation microhardness (H_v) of the plagioclases has a definite variation with respect to composition. This variation takes the form of a complex curve, shown in Figure 7, which departs markedly from linearity in the intermediate composition range. Mookherjee and Sahu suggest that this curve is related to the submicroscopic crystal structure. In the present investigation the essential correctness of the microhardness curve in Figure 7 has been verified (Table 2A). Mookherjee and Sahu used the highest value obtained with a particular load on randomly oriented, polished grains mounted in bakelite molds to represent the maximum microhardness value of the mineral for that load. The microhardness
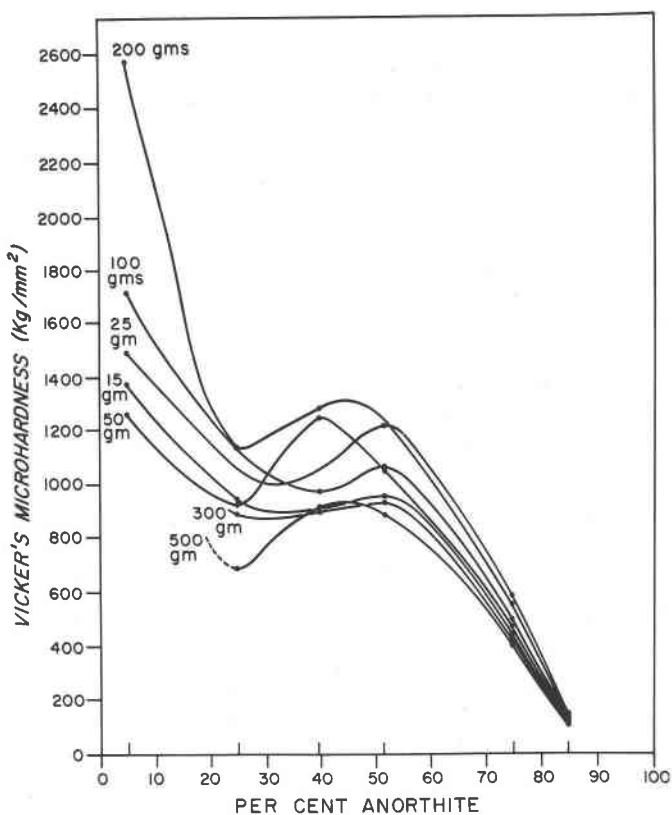
FIG. 7. Variation of Vicker's indentation microhardness for the plagioclases under various loads. After Mookherjee and Sahu (1960).

values in the present study were obtained on cleavage surfaces of large, randomly oriented grains. Numerous indentation tests also were performed on small polished mounts identical to those used for studying grains in the electron-microprobe analyser, but these grains yielded higher hardness values than unpolished cleavage flakes, suggesting that work-hardening of the grain surfaces had occurred due to prolonged polishing. The results quoted by Mookherjee and Sahu indicate that the polishing technique employed in their study induced only limited work-hardening of the grain surface.

According to the theory developed below, the orientation in which indentation hardness is measured should be only a secondary factor in determining hardness, because directional bond strength should be secondary in importance to the average domain size. This will be true
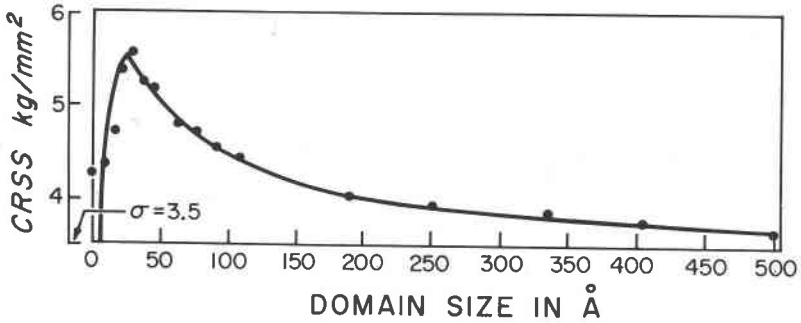
FIG. 8. The effect of domain size upon crystal strength. The circles are data points obtained for the alloy Cu_3Au annealed at different temperatures. The full line is a plot of equation (6) given in the text. After Ardley (1955).

as long as all the domain dimensions are small compared to the tip of the pyramid-shaped diamond used to measure hardness. This restriction appears to hold true for the intermediate composition range. There is a large margin of error (± 100 kg/mm²) in the measured hardness values, however, that appears to be due in part to the brittleness of the sample and the consequent development of fracture at the test pit corners. This indicates that plagioclases at room temperature are on the boundary between brittle and plastic deformation modes of deformation. The large range in microhardness for plagioclases is striking because a range of this magnitude (~ 1100 kg/mm² for the 50 gm load) is not present in most metal systems. This difference in magnitude of the microhardness range as well as the slightly brittle deformation behavior of the plagioclases are probably related to the difference in bond type between the rather weak metallic bonding systems and the relatively stronger ionic-covalent bonding of the plagioclases.

Ardley (1955) demonstrated that a long-period superlattice type of structure has characteristic mechanical properties. In brief, the critical resolved shear strength (CRSS) for slip of Cu_3Au is a function of both local and distant order, and is directly dependent upon the size of an average domain diameter (Fig. 8). The CRSS is the component of the applied force necessary to initiate slip along the slip plane. The points on the graph of Figure 8 represent Ardley's experimental observations while the full curve represents the equation

$$\sigma_{APB} = \frac{\gamma_s}{\epsilon} \left(\frac{\epsilon - l}{\epsilon} \right)^3 \tag{6}$$

where σ_{APB} is the resolved shear strength related to the anti-phase

domain system, ϵ is the average diameter of an antiphase domain, γ_s is the energy associated with the creation of an antiphase boundary along the slip plane, and t is the thickness of the grown-in antiphase boundary, which is the transitional surface (including the antiphase out-of-step) separating one antiphase domain and its neighbor. Relationship (6) was derived by equating the energy of the domain boundaries created during slip by paired dislocations in the superstructure to the work done by the applied stress. Dislocations tend to move as pairs in an ordered material, because the second dislocation of a pair restores the order destroyed by the first member of the pair, so that little energy is required to move the pair across the slip plane. When the paired dislocations intersect an antiphase boundary, however, additional antiphase boundary is created as shown in Figure 9. Figure 9a shows an ordered structure containing domain boundaries, before slip, and Figure 9b shows the same crystal after slip. After slip, the amount of antiphase boundary has increased, and the energy to cause slip has to be supplied by the externally
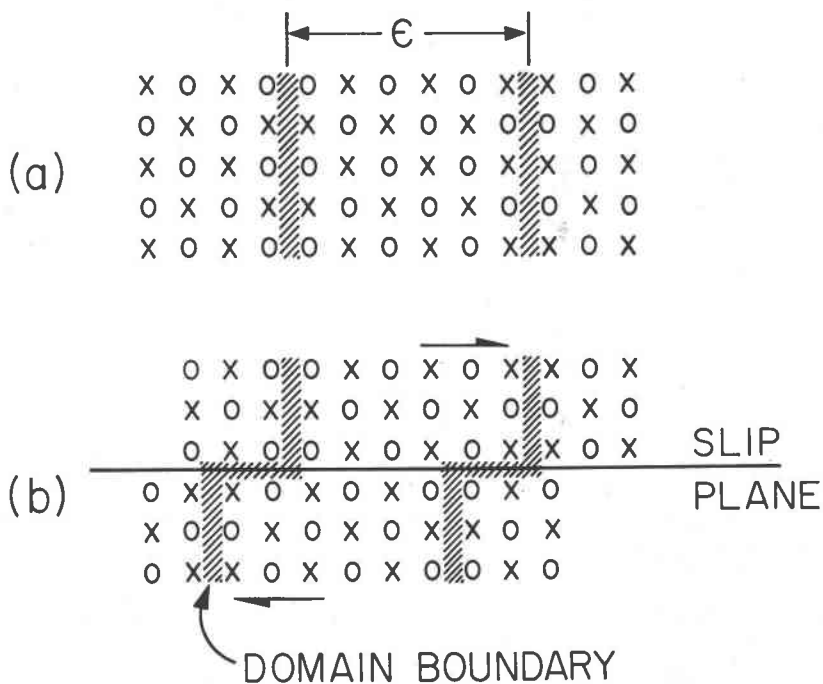

FIG. 9. Schematic diagram of an antiphase domain system with an average domain size of ϵ. After Ardley (1955). a) The crystal before slip. b) The crystal after slip and showing the creation of an additional antiphase boundary.
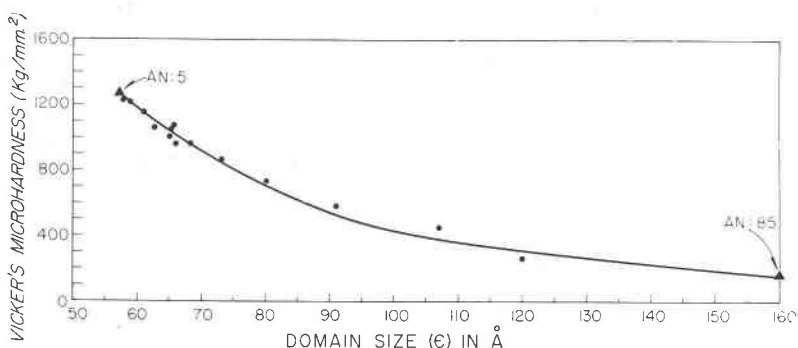
FIG. 10. Average domain size (ϵ) and its relations to Vicker's indentation microhardness. The filled circles represent the cube root of domain volume as calculated for a triclinic unit cell at intervals of 5 mol % An. The triangles are values of domain size at An_5 and An_{85} obtained indirectly from experimentally determined microhardness values. The curve is fitted to this data by trial.

applied stress. It follows that the smaller the domain size, hence the more domain boundaries that exist, the larger the stress will have to be in order to cause slip.

The fact that the observed microhardness in plagioclase is greatest at small domain sizes, and least at large domain sizes indicates that an inverse relationship between size and microhardness is valid (Fig. 10). In measuring the mechanical strength of a polycrystalline ceramic material, however, it is often the strength necessary to initiate brittle deformation by crack formation and propagation throughout the crystal that is being measured, rather than the strength necessary to initiate plastic deformation by slip along well defined planes. This is true so long as the grain size is reasonably large. Under these conditions the relationship between grain size (G) and crystal strength is

$$\sigma = kG^{-a} \tag{7}$$

where (k) and (a) are constants (Davidge, 1969). At small grain sizes (≤ 200 Å), however, the stresses near the crack tip may be high enough for localized plastic deformation processes involving slip to become important.

In the plagioclase feldspars the antiphase domains comprise a "polycrystalline" texture of internal boundaries, somewhat similar to grain boundaries separated by less than 200 Å, and it may be expected that the indentation diamond produces enough stress concentration at its tip to produce local slip. This suggests that crystal strength as measured by indentation microhardness is controlled primarily by the resistance of

the domain walls to slip and only secondarily, if at all, by the resistance of the domain wall to crack propagation throughout the crystal. At elevated temperatures plastic deformation processes are operative without doubt, as indicated by the experimental production of mechanical twins and slip bands in the intermediate composition plagioclases (Borg and Heard, 1969). Whichever mechanism controls crystal strength in the plagioclases at room temperature, the main conclusion of this discussion is that the measured crystal strength is governed primarily by average domain size in an inverse relationship. The analysis of slip outlined above indicates that the twinning behavior of the plagioclases is directly related to their domain structure. This relationship is now being investigated by one of us (C.G.C.).

Accepting plastic deformation as the most important process for controlling deformation in the plagioclases at room temperature, an important conclusion can be reached. Under these conditions a proportionality exists between microhardness (H_v) and ($CRSS$). It is also clear that average domain size (ϵ) and domain wall thickness (t) play an important role in determining the physical properties of plagioclases with a domain structure. In the intermediate composition plagioclases the energy associated with the creation of an antiphase boundary along the slip plane (γ_s) should depend on ordering patterns of Si/Al. Because these ordering patterns are unknown, only the ratio (γ_s/ϵ) may be obtained from available experimental data. With these restrictions the relation between domain size and crystal strength may be written

$$H_v \propto k\sigma = 100\left(\frac{\gamma_s}{\epsilon}\right)\left(\frac{\epsilon - t}{\epsilon}\right)^3 \qquad (8)$$

where k is an arbitrarily determined constant given a value of 100. In Figure 10 the measured microhardness data have been plotted to show the variation of microhardness as a function of average domain size rather than of composition. Average domain sizes for sample points were determined by finding the cube root of the triclinic domain volume, obtained from the domain size curves along X, Y, and Z (Fig. 5) and values of lattice angles listed by Doman *et al.* and Bambauer *et al.* (1967).

The triangular points labeled An_5 and An_{85} in Figure 10 are indirect estimates of domain size based on experimental microhardness values. At An_5 there is no direct diffraction evidence for an antiphase domain system, and the large hardness value for this composition is probably related largely, if not completely, to the high stress required to move the leading dislocation of a pair through the unique ordering pattern of the albite structure rather than through closely spaced antiphase domain

walls. This argument is similar to that proposed by Laves (1952) to explain the difficulty of twin formation in ordered albite.

At An_{85} the "b" reflections are not split. This indicates that the regular domain structure of the intermediate composition plagioclases does not exist. The existence of a *disordered* antiphase domain system with domains of unspecified size, however, has been suggested by Ribbe and Colville (1969), on the basis of diffuse "c" reflections, for plagioclases of this composition.

If specific values of domain wall thickness (t) are assumed, relative
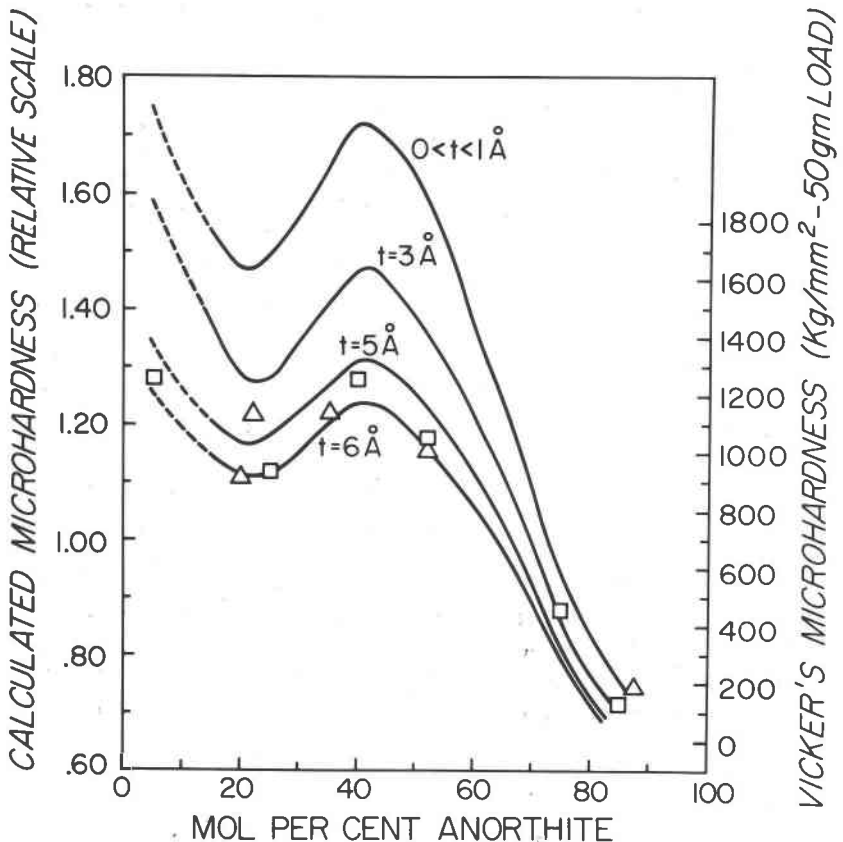

Fig. 11. Calculated microhardness of the plagioclases compared to experimental points obtained for a load of 50 grams. The squares are data obtained by Mookherjee and Sahu (1960) and the triangles are data obtained in this investigation. The solid lines represent calculated values of microhardness for sample points of the average domain size (ϵ) and for different assumed values of domain wall thickness (t). The value of $0 < t < 1$Å is for an infinitely small domain wall thickness.

H_v values can be calculated (Table 2B) from (8) and these can be compared to the experimental data, as shown in Figure 11. The dashed line segments are extrapolated from the solid lines calculated from (8) for different values of t. When t is approximately 5 Å to 6 Å a good fit between the calculated curve and the experimental points is obtained. Due to the large error in measuring microhardness, however, domain wall sizes obtained from Figure 11 should not be considered definitive, but may be taken as a qualitative argument that domain boundaries are on the order of one unit cell in thickness. This result is in keeping with predictions made by previous researchers. Megaw (1962) indicated that it might be possible for boundary stresses in the plagioclase feldspars to be confined to a layer that is a fraction of a unit cell. Ardley (1955) and Logie (1957) found that domain boundaries in Cu_3Au are from one to two atomic diameters in thickness, and Flinn (1960) has argued that a narrow domain wall would be a region of lower energy than a wide disordered domain wall.

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