INTERGRANULAR LATTICE COHERENCY CONTROLS ON LIMESTONE TEXTURE

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ABSTRACT

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ON LIMESTONE TEXTURE

By

Christopher Stevens Plopper

To determine the intergranular lattice relationships of quartz and calcite in a stable recrystallized texture, optic axes orientations for calcite grains surrounding calcite and quartz host grains in the Alton limestone were derived by use of the universal stage. The Alton was chosen to represent textural equilibrium because it is a coarse grained, 720 million year old recrystallized carbonate rock which has experienced no penetrative deformation since its deposition.

Angles between optic axes for combinations of common crystal forms of quartz plus calcite and calcite plus calcite were calculated for orientations in which varying degrees of lattice coherency are maintained at the interface. The angles between "c" axes derived on the universal stage were plotted on orientation-frequency histograms for calcite to calcite and quartz to calcite cases.

When the angular relationships of "c" axes for coherent lattice contacts are compared with the calcite to calcite histogram, it is seen that modes occur adjacent to the positions of maximum coherency. The fact that the positions of maximum coherency are represented as minima on the frequency scale suggests that the energy flux of recrystallization

was locally great enough to destroy those grains which were oriented with maximum lattice coherence at grain contacts, by grain boundary migration. This explains some heretofore unresolved observations in recent literature. The distance of the modes from positions of maximum lattice coherency is a direct measure of the local energy flux experienced by the rock, i.e., the greater the energy flux, the higher the degree of grain contact lattice incoherency that can be overcome by grain boundary migration.

In the quartz to calcite case, the energy needed to mobilize boundaries between these unlike phases is many times greater than that in the like phase contacts. As expected, the orientation frequency histogram for quartz to calcite shows positions of maximum intergranular lattice coherency occupying frequency maxima.

Not only can the evolutionary trend of any carbonate rock be determined by comparison of its orientation-frequency distributions with those of the Alton, but that of any rock in which like and unlike phase contacts are involved.

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Christopher Stevens Plopper

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The National Park Federation was most kind in allowing samples to be taken from Glacier National Park.

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INTRODUCTION

The texture of a recrystallized carbonate rock to a large extent controls its physical and chemical characteristics (e.g., shear strength, porosity, reactivity, etc.). The evolution of a recrystallized texture is in turn strongly dependent upon the nature of the pre-existing texture, the conditions of recrystallization, and the degree of completeness to which recrystallization has proceeded. Given a recrystallized carbonate rock at equilibrium, the orientations of adjacent grains will represent lowest boundary energy conditions. A stable texture may therefore be defined as one in which the grains show a strong affinity for lowest energy contact orientations for the particular conditions of recrystallization. In contrast, an unstable texture would show a random orientation.

At present, no data on crystallographic orientation-frequency distributions of grains in rocks with stable textures exists. The Alton Limestone is a coarse grained, recrystallized carbonate rock which has experienced little or no penetrative deformation since its deposition some 720 million years ago, and due to its age and lack of deformation, it may be assumed a priori that the texture of this rock approaches equilibrium on a grain to grain basis. Another important feature of the Alton is the presence of definite reentrants of one grain boundary into another, showing that this texture has evolved significantly beyond incipient crystal boundary interrelations (Figure 1). Detailed grain to grain orientations in the Alton Limestone may be used as a standard for judging the degree of textural equilibrium in other carbonate rocks.

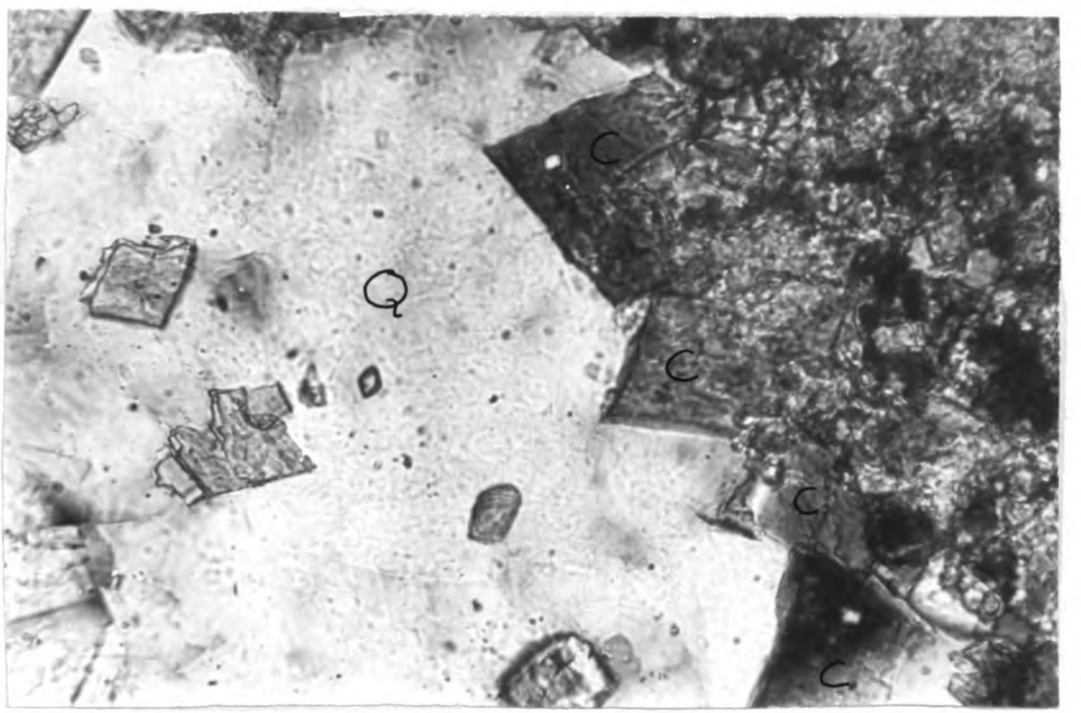

Figure 1. Photomicrograph of calcite reentrants in quartz grain (Crossed Nicols, x 50).

All samples were taken from the Alton formation exposed on the main highway near Many Glacier Hotel (Lat. 48° 02' 54", Long. 113° 08' 42") in Glacier National Park, Montana.

BACKGROUND

A crystal form may be viewed as consisting of faces and edges. A face represents a dense atomic plane of low free energy, which is perpendicular to a slow growth direction in the lattice. An edge is the direction perpendicular to a lattice direction of high atomic density, and is a direction of fast growth. The edge is a direction of high lattice energy (Spry, 1969). In an aggregate of randomly oriented grains, all conceivable lattice orientations at grain contacts are possible, with no regard for surface energy levels. However, in a rock at textural equilibrium, the lowest free energy contacts are preferred.

One way to lower the energy of a grain boundary is for the two grains to maintain a high degree of lattice coherency across the contact (Spry, 1969). The highest degree of coherency occurs when atomic planes in the two grains are parallel at the contact, and the repeat distance is equal so that atoms may be shared. In the ideal situation, like grain lattices are so coherent that an optically continuous crystal is formed and the contact is not visible. This situation occurs when like grains are oriented so that their "c" axes and all other lattice directions are parallel.

A lower degree of coherency occurs when the "c" axes are not parallel but the two lattices are so mutually oriented that equivalent density planes (e.g., probably parallel to a face) are parallel across the boundary. This would correspond in the megascopic case to two crystals sharing a common face. The maximum coherency in this situation

will be produced if the symmetry planes of the two like grains continue across the boundary, ensuring that repeat distances of the corresponding lattices across the contact will coincide.

A third situation is the sharing of different crystallographic lattice planes of high atomic density, comparable to the megascopic sharing of unlike faces between two crystals. Although the repeat distances no longer coincide at the contact, atomic plane and symmetry parallelism can be retained, that is, directions of low and high atomic densities are respectively parallel between the two crystals.

In addition to contacts between like minerals (calcite to calcite), unlike contact relationships (quartz to calcite) are also discussed. The case of lattice coherency in a texturally stable rock still applies, the only difference being that the sharing of a common lattice plane is not possible. However, parallelism of comparable density planes is possible, and some degree of coherency is still maintained by the intersection of a high density direction with a low density direction, if the symmetry planes extend across the boundary.

As stated before, high density lattice planes are considered surfaces of relatively low surface energy whereas the fast growing edges are considered to represent higher inherent energy. Two lattices that are precisely parallel should produce a boundary containing the least surface energy because directions of high lattice energy in one crystal would not intersect those in the other. The high energy directions in the two crystals are "insulated" from one another due to the parallelism of the low energy density planes across the common boundary. A contact formed by two grains whose lattices are oriented such that high energy directions are in direct contact across the boundary, will possess very high surface energy. Boundaries between crystals possessing other lattice orientations should

represent intermediate surface energies.

Since the free energy of a grain contact is inversely related to the degree of lattice coherency at the contact, then a stable texture will contain a high proportion of contact orientations near a coherent position. However, as discussed below, highly coherent contacts have very low activation energies and it would not be surprising if contact relationships at the precise angle of coherency were lost due to grain boundary migration. Under conditions of modest energy flux such as that experienced by the Alton, probably only coherent boundaries between like phases would be activated.

The role of activation energy and its relation to grain boundary migration must be discussed before proceeding. Many grain contacts are stable because a high input energy is required before that boundary will migrate. Once this initial input of activation energy is exceeded, grain boundary migration is rapid, releasing energy as the lattices seek a more stable lower energy orientation (Figure 2). The manifestation of this phenomena in a stable texture will be as follows: 1) like grain "face to face" contacts which are only slightly disoriented from ideal parallelism will have extremely low activation energy levels prior to recrystallization, since very low energy is required to reorient the grains into single crystals. During the first stages of recrystallization the activation energy will first be exceeded for these boundaries and grain boundary migration will result in one continuous crystal with its lattice parallel to one of the interfacing crystals. These contacts should seldom be seen and few should be expected to be recorded. 2) unlike minerals with near parallel "face to face" orientations cannot form single, continuous crystals. These contacts may be expected to be recorded with

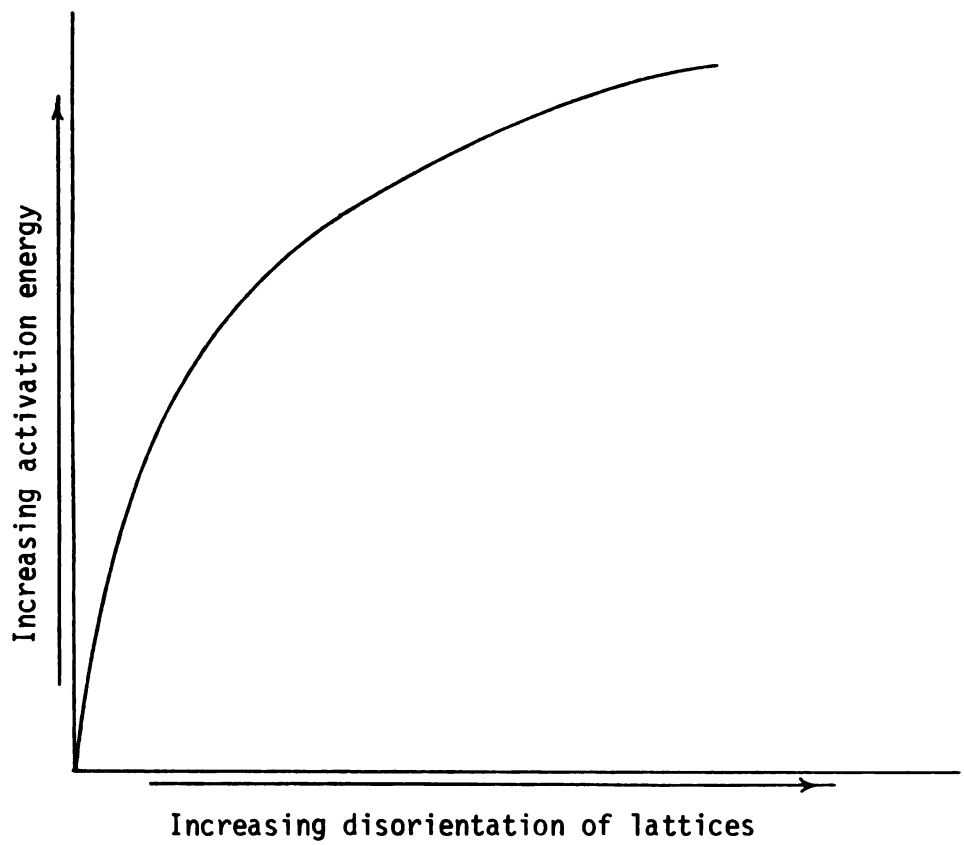

Figure 2. Curve of activation energy as a function of lattice coherency across a contact (After McLean, 1965, p. 109).

a much greater frequency than those of situation 1, and therefore the activation energies are orders of magnitude greater.

The common crystal forms for both calcite and quartz are well known (Figure 3). These faces and the edges formed at the face intersections are megascopic manifestations of the relative orientations of high density atomic lattice planes. In the light of the preceeding discussion, angular relationships between the common faces and the "c" axis of quartz and calcite forms were taken from Dana (1951) and plotted on stereographic projections in order to determine the angle between edges and the "c" axis (Figure 4). With this information, resulting angles between optic axes for all possible coherent contact orientations of quartz to calcite and calcite to calcite forms were calculated and are presented in Table 1. The sharing of a density plane (e.g., face direction) does not uniquely define such angular relationships, since this situation can be preserved through a 360° rotation of the adjacent forms about the pole perpendicular to the common density plane. The positions which will have maximum lattice coherency are those which occur when the plane containing the "c" axes of both forms is parallel to a symmetry plane in both forms. Then the atoms near the interface in one lattice possess a symmetrical relationship to those near the interface in the adoining crystal. Because the symmetry planes in the trigonal system intersect the "c" axis, the plane containing the "c" axes of the two adjoining crystals can include a symmetry plane in at most three positions for each form, i.e., one per face and one per edge. It is the resulting angle between "c" axes of the two adjacent forms in these special positions which are tabulated in Table 1. The face and edge directions utilized in various combinations are those designated as "very common" and "very very common" in Dana (1951).

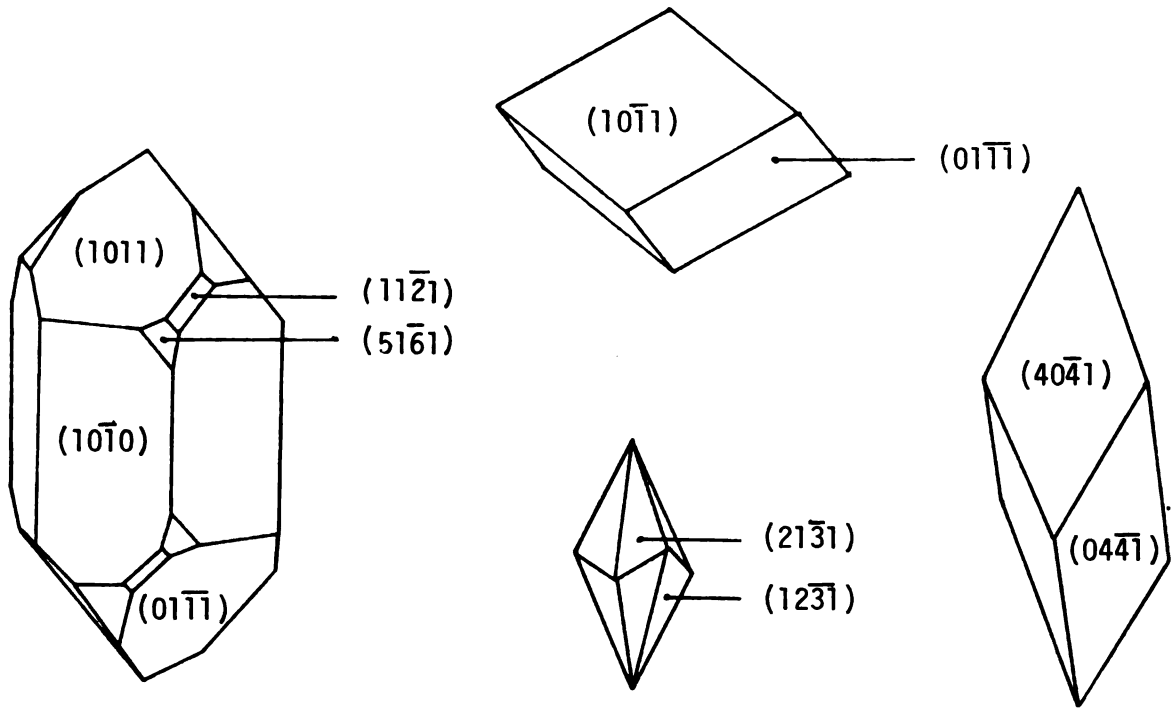

Figure 3. Common forms of quartz and calcite.

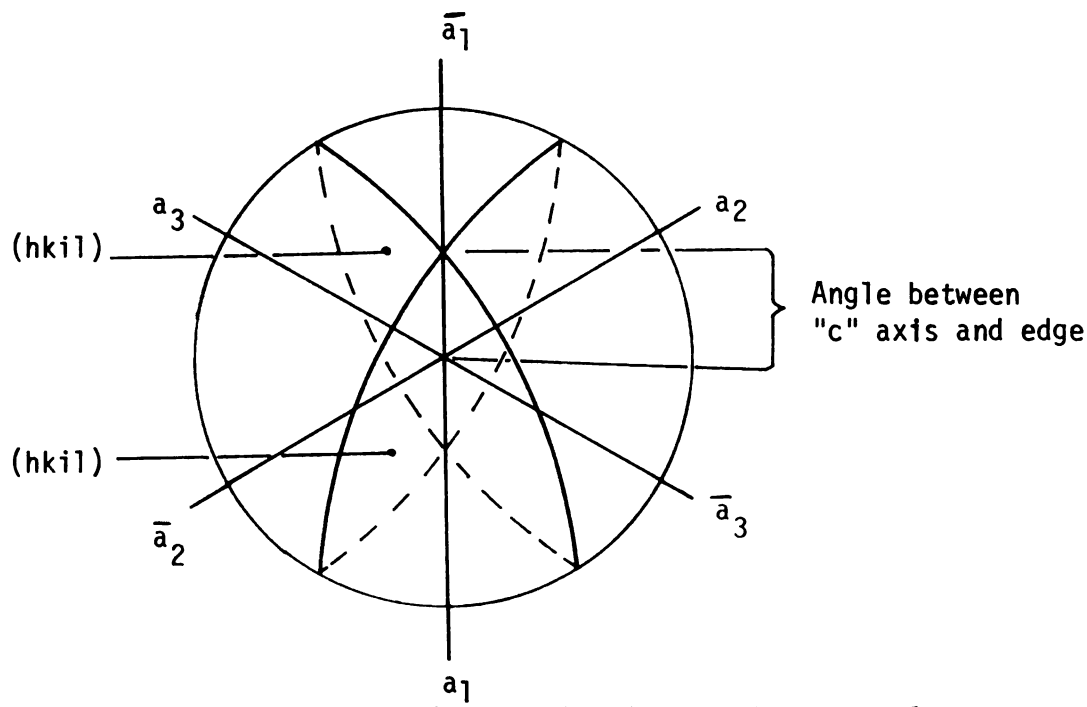

Figure 4. Sample stereographic projection for determining crystallographic edge to "c" axis angles.

Table 1. Angular relationships of "c" axes of adjacent forms with coherent lattice orientations.

Calcite to Calcite

Orientation	Angle between "c" axes (°)
$(21\bar{3}1)+(21\bar{3}1)$	41.6
$(21\bar{3}1)+(12\bar{3}\bar{1})$	0
Edge $A_{(21\bar{3}1)}$+Edge $A_{(21\bar{3}1)}$	62
Edge $A_{(21\bar{3}1)}$+Edge $B_{(21\bar{3}1)}$	53
Edge $A_{(21\bar{3}1)}$+Edge $A_{(12\bar{3}\bar{1})}$	0
Edge $A_{(21\bar{3}1)}$+Edge $B_{(12\bar{3}\bar{1})}$	9
Edge $B_{(21\bar{3}1)}$+Edge $B_{(21\bar{3}1)}$	44
Edge $B_{(21\bar{3}1)}$+Edge $B_{(12\bar{3}\bar{1})}$	0
$(10\bar{1}1)+(10\bar{1}1)$	89
$(10\bar{1}1)+(01\bar{1}\bar{1})$	0
$(10\bar{1}1)+(21\bar{3}1)$	66.3
$(10\bar{1}1)+(12\bar{3}1)$	24.7
$Edge_{(10\bar{1}1)}$ +$Edge_{(10\bar{1}1)}$	54
$Edge_{(10\bar{1}1)}$ +$Edge_{(01\bar{1}\bar{1})}$	0
$Edge_{(10\bar{1}1)}$ +Edge $A_{(21\bar{3}1)}$	86
$Edge_{(10\bar{1}1)}$ +Edge $B_{(21\bar{3}1)}$	47.5
$Edge_{(10\bar{1}1)}$ +Edge $A_{(12\bar{3}\bar{1})}$	32
$Edge_{(10\bar{1}1)}$ +Edge $B_{(12\bar{3}\bar{1})}$	23.5
$(40\bar{4}1)+(40\bar{4}1)$	28.4
$(40\bar{4}1)+(04\bar{4}\bar{1})$	0
$(40\bar{4}1)+(10\bar{1}1)$	59.7
$(40\bar{4}1)+(01\bar{1}\bar{1})$	31.3
$(40\bar{4}1)+(21\bar{3}1)$	35

Table 1 (cont'd)

$(40\bar{4}1)+(12\bar{3}1)$	6.6
$Edge_{(40\bar{4}1)}$ $+Edge_{(40\bar{4}1)}$	54
$Edge_{(40\bar{4}1)}$ $+Edge_{(04\bar{4}1)}$	0
$Edge_{(40\bar{4}1)}$ $+Edge\ A_{(21\bar{3}1)}$	58
$Edge_{(40\bar{4}1)}$ $+Edge\ B_{(21\bar{3}1)}$	49
$Edge_{(40\bar{4}1)}$ $+Edge\ A_{(12\bar{3}1)}$	4
$Edge_{(40\bar{4}1)}$ $+Edge\ B_{(12\bar{3}1)}$	5
$Edge_{(40\bar{4}1)}$ $+Edge_{(10\bar{1}1)}$	90
$Edge_{(40\bar{4}1)}$ $+Edge_{(01\bar{1}\bar{1})}$	36

Quartz to Calcite

Orientation	Angle between "c" axes (°)
Quartz + Calcite	
$(10\bar{1}1)+(21\bar{3}1)$	59.2
$(10\bar{1}1)+(12\bar{3}1)$	17.6
$(10\bar{1}1)+(10\bar{1}1)$	83.9
$(10\bar{1}1)+(01\bar{1}\bar{1})$	7.1
$Edge_{(10\bar{1}1)}$ $+Edge\ A_{(21\bar{3}1)}$	69.4
$Edge_{(10\bar{1}1)}$ $+Edge\ A_{(12\bar{3}1)}$	7.4
$Edge_{(10\bar{1}1)}$ $+Edge\ B_{(21\bar{3}1)}$	60.4
$Edge_{(10\bar{1}1)}$ $+Edge\ B_{(12\bar{3}1)}$	16.4
$Edge_{(10\bar{1}1)}$ $+Edge_{(10\bar{1}1)}$	75
$Edge_{(10\bar{1}1)}$ $+Edge_{(01\bar{1}\bar{1})}$	21
$Edge_{(10\bar{1}1)}$ $+Edge_{(40\bar{4}1)}$	69
$Edge_{(10\bar{1}1)}$ $+Edge_{(04\bar{4}1)}$	15

Table 1 (cont'd)

$(10\bar{1}0)+(21\bar{3}1)$	20.8
$(10\bar{1}0)+(10\bar{1}1)$	45.5
$(10\bar{1}0)+(40\bar{4}1)$	14.2
$Edge_{(10\bar{1}0)}$ +Edge $A_{(21\bar{3}1)}$	31
$Edge_{(10\bar{1}0)}$ +Edge $B_{(21\bar{3}1)}$	22
$Edge_{(10\bar{1}0)}$ +$Edge_{(10\bar{1}1)}$	63
$Edge_{(10\bar{1}0)}$ +$Edge_{(40\bar{4}1)}$	27
$(51\bar{6}1)+(21\bar{3}\bar{1})$	28.9
$(51\bar{6}1)+(12\bar{3}\bar{1})$	12.7
$(51\bar{6}1)+(10\bar{1}1)$	53.5
$(51\bar{6}1)+(01\bar{1}\bar{1})$	37.4
$(51\bar{6}1)+(40\bar{4}1)$	22.3
$(51\bar{6}1)+(04\bar{4}\bar{1})$	6.1
$(11\bar{2}1)+(21\bar{3}1)$	45.3
$(11\bar{2}1)+(12\bar{3}\bar{1})$	3.7
$(11\bar{2}1)+(10\bar{1}1)$	70
$(11\bar{2}1)+(01\bar{1}\bar{1})$	21
$(11\bar{2}1)+(40\bar{4}1)$	38.7
$(11\bar{2}1)+(04\bar{4}\bar{1})$	10.3

METHODS

To determine the exact nature of the non-random texture, optic axis orientations of twenty-one quartz grains and six calcite neighbors per grain were derived by use of the four axis universal stage. Likewise, the calcite to calcite optic axis orientations were derived from twenty calcite grains and five or six neighbors per grain. In all, eight thin sections were examined. The planar angles between optic axes of host and neighbor grains were calculated and plotted on orientation-frequency histograms (Figures 5 and 6).

The measurement error was estimated to be at the most ± 1.5°. For this reason histograms relating grain frequency to angle between "c" axes of adjacent grains were plotted with a class interval of 3°.

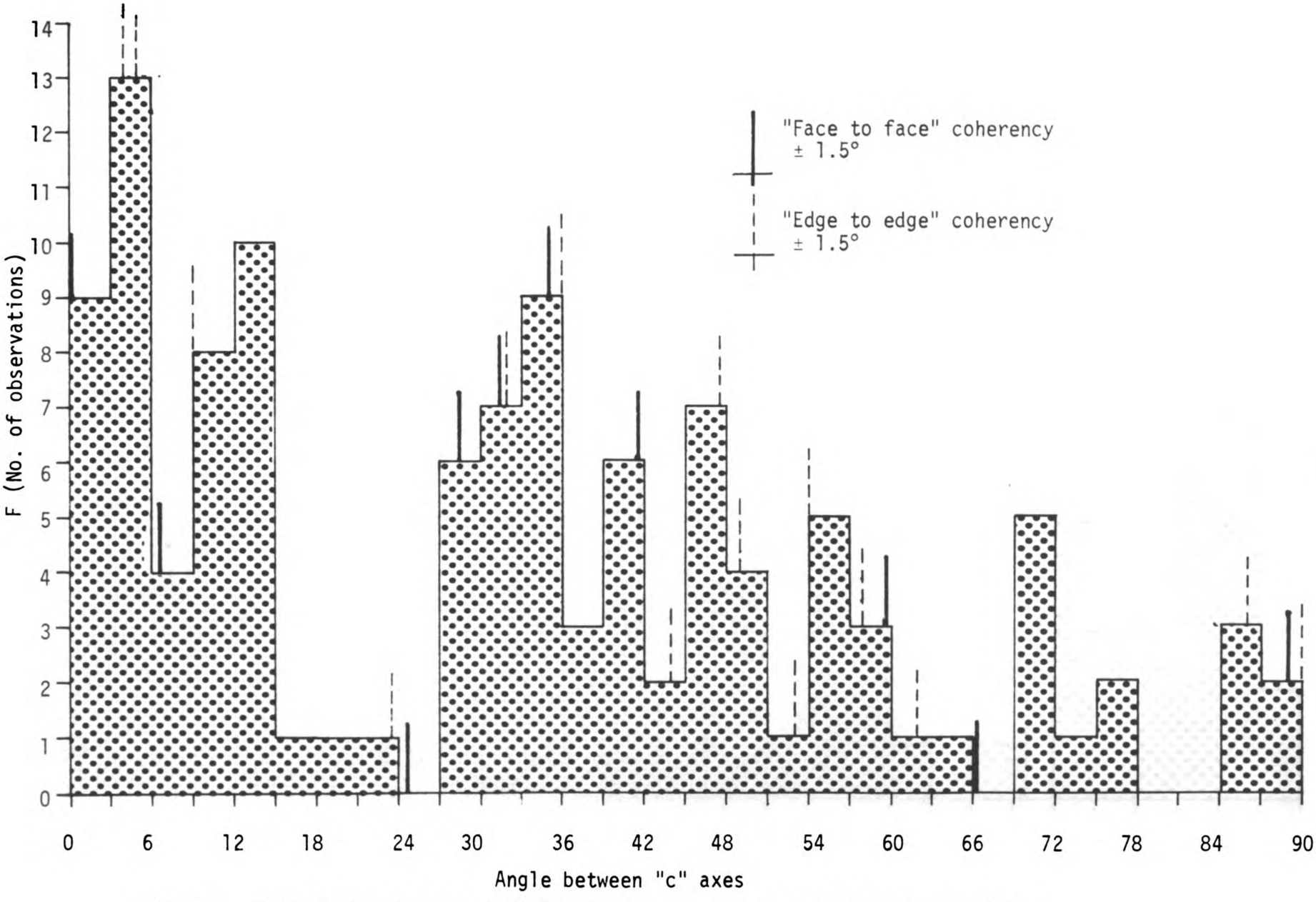

Figure 5. Orientation-frequency histogram for calcite to calcite grain contacts.

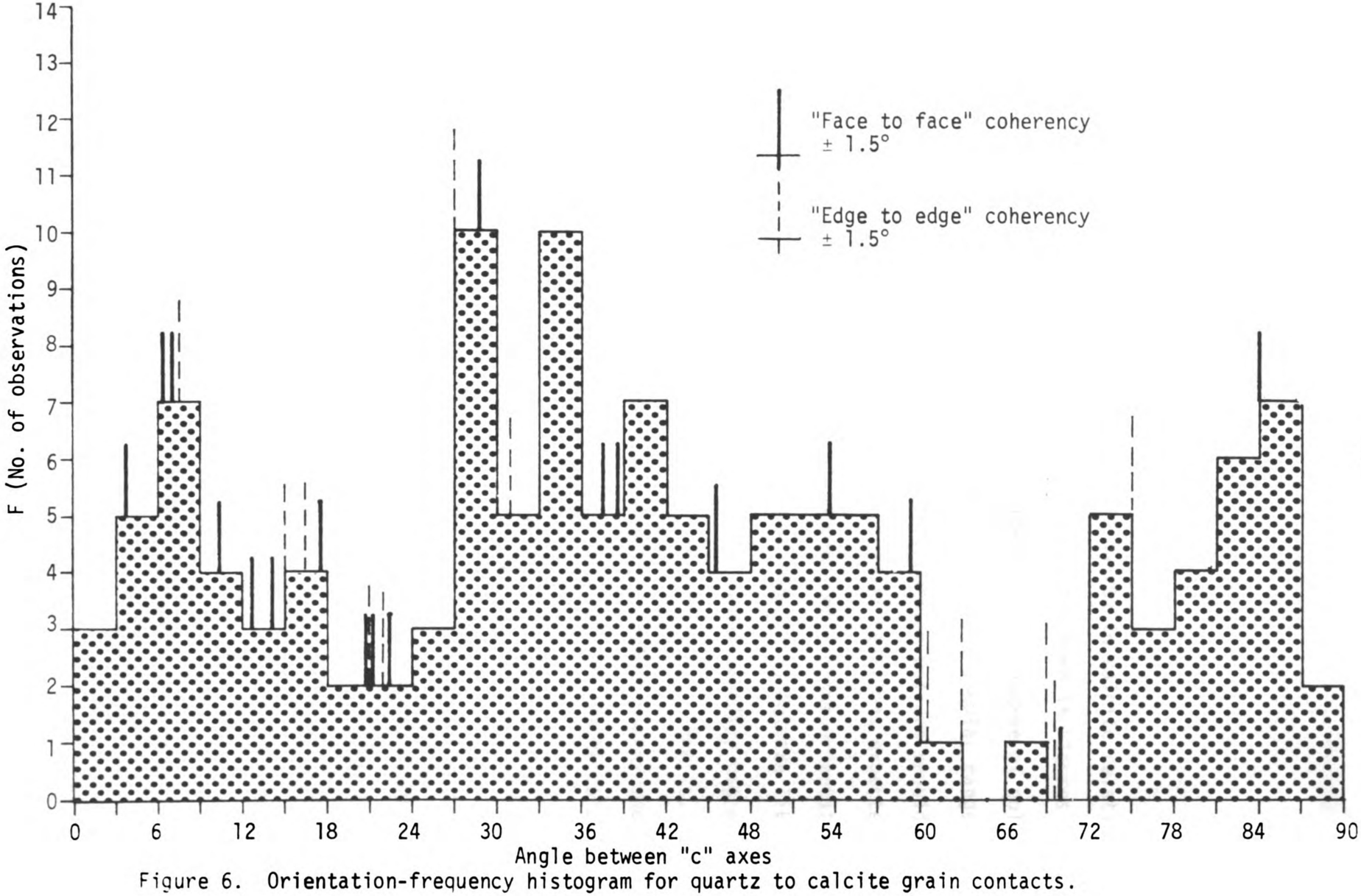

Figure 6. Orientation-frequency histogram for quartz to calcite grain contacts.

RESULTS

Histograms for calcite to calcite and quartz to calcite relationships between frequency and angles between "c" axes are shown in Figures 5 and 6. Simple inspection of these histograms suggests a high degree of polymodality with maxima and minima occupying both large and small angular ranges. According to the preceeding discussion, this complex pattern should be largely explained with reference to those angular relations expected to occur at high frequencies with respect to coherent lattice orientations.

The calcite to calcite case, concerning interfaces between like phases, should involve a higher probability of grain boundary migration for coherent positions, than the quartz to calcite case. That is, the activation energy for the most coherent boundary between quartz and calcite should be many orders of magnitude greater than for calcite to calcite.

Calcite to Calcite

Figure 5 illustrates the histogram relating frequency of occurrance to the angle between grains which share an interface. In addition, locations of particularly high and low coherence are designated. As might be expected from theory, positions of very high coherence are represented by frequency minima, but are accompanied a few degrees away by frequency maxima. For instance, when grains interface in a precisely parallel position (0° between "c" axes), the boundary is the most coherent. Figure 5 displays a minimum in the interval 0°-3° with a maximum in the interval 3°-6°. The actual relationships over this region might be more extreme if

the histogram were constructed on a 1/2° class interval rather than 3°, inasmuch as the activation energy increases quite rapidly with departure from the parallel position (Figure 2). This deficiency of grains at the most coherent position is interpreted to be the result of a reduction in the number of grains in this position due to grain boundary migration. Inspection of Figure 5 reveals that every major mode is bordered by minima at coherent positions.

Orientations where lattices are in an "edge to edge" position should not undergo grain boundary migration because of the high activation energy involved. Indeed, crystals once nucleated in that position should undergo little growth at all. Therefore, during recrystallization these orientations are destroyed. In Figure 5, these orientations are also generally low frequency position. When a mode occurs at this position, it is also closely situated to a coherent "face to face" position. Very precise investigation might resolve an interesting fine structure in the histogram in this region.

The distance of a mode from a coherent position should be a function of the maximum energy flux experienced by the rock. The mode adjacent to the most coherent position at 0° occurs in the region 3°-6°. Thus, the activation energy for grains whose lattice discordance exceeds this value was greater than the energy flux experienced by the rock. Greater energy fluxes would presumably "erode" the low angle side of the mode.

The most dominant crystallographic relations across grain boundaries for the calcite to calcite case therefore possess neither highest nor lowest surface energy, but instead possess the lowest surface energies whose concomitant activation energies barely exceeded the local energy flux.

Quartz to Calcite

In common with the calcite to calcite relationships, the most coherent boundaries between quartz and calcite possess the lowest activation energy for grain boundary migration. However, because this case concerns unlike phases, that activation energy is many orders of magnitude higher than that for the situation between like phases. A rock such as the Alton quite likely has not encountered a high enough energy flux to mobilize such boundaries.

Accordingly, inspection of Figure 6 suggests that major modes occur at the most coherent positions, and that the frequency minima occur near the most incoherent positions. The histogram is dominated by a central region containing a large number of "face to face" lattice positions, bounded by regions with large numbers of "edge to edge" relations. Low and high degree maxima are also related to "face to face" orientations.

In the calcite to calcite case, about one third of all pairs measured were segregated in the region 0°-16°. In the case of quartz to calcite, the data are more uniformly distributed. This is a direct manifestation of the fact that whereas the 0° position is the most coherent for like phases, it need not possess that attribute for unlike phases.

DISCUSSION AND CONCLUSIONS

The results discussed above for the like calcite to calcite and unlike quartz to calcite cases are applicable to like and unlike grain relationships in general. For example, Ransom (1971) displays orientation-frequency histograms for recrystallized quartz grains in contact with one another (Figure 7). All such histograms display a pronounced minimum at the most coherent parallel orientation. Gordon and Vandermeer (1966) also reported a paucity of grains in coherent positions where like phases (zone-refined lead) share a common boundary (Figure 8).

Because the Alton was subjected to a relatively low energy flux, only the orientations with lowest activation energies are mobilized. Byerly and Vogel (in press), examining the textural evolution of a granodiorite pluton under increasing levels of thermal metamorphism, observed that at the highest grades (e.g., upper amphibolite) most grain boundaries except those with highest activation energy were mobilized. In that case concomitant segregation of impurities to highly incoherent boundaries stabilized them, resulting in a texture composed principally of boundaries possessing high degrees of incoherency as well as high activation energy.

The frequency-orientation histograms such as Figures 5 and 6 are inversely proportional to a functional relationship between surface energy and orientation, with the exception of the absence of a high frequency mode at very coherent positions. Assuming the existence of such modes, schematic surface energy-orientation relationships similar

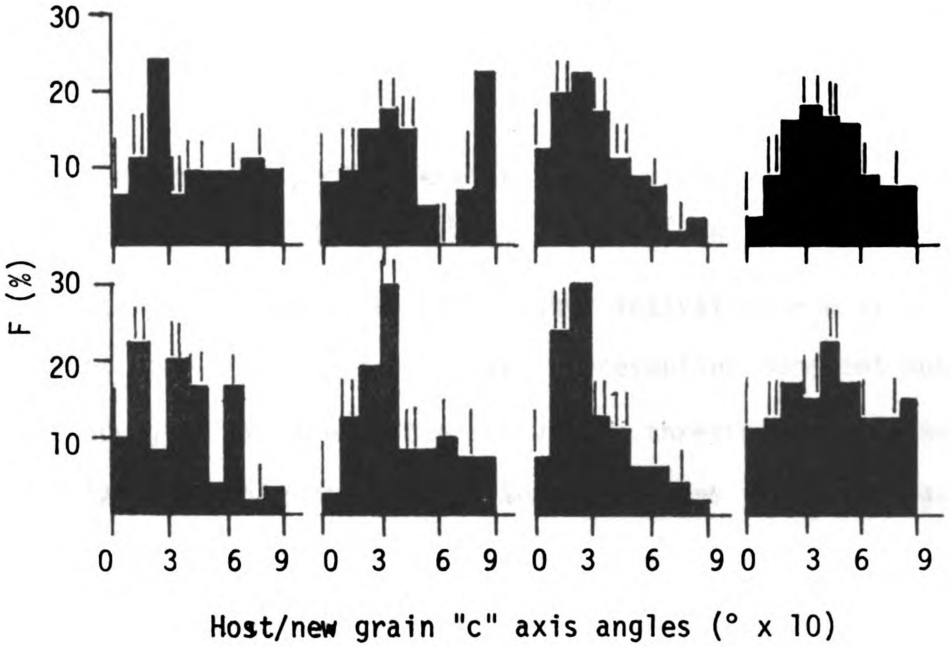

Figure 7. Orientation-frequency histograms for quartz to quartz "c" axis angles (From Ransom, 1971, p. 84).

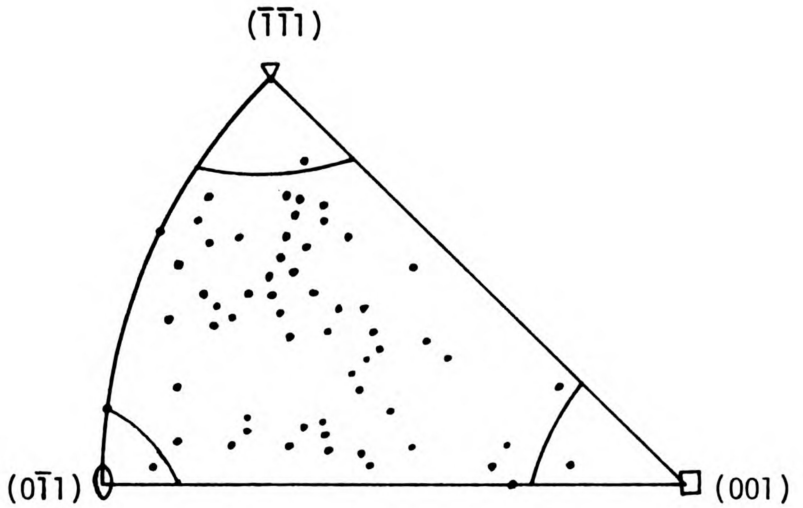

Figure 8. Stereographic plot of new grains to matrix grain in zone-refined lead (From Gordon and Vandermeer, 1966, p. 213).

to that of Spry (1969, Figure 8, p. 33) can be constructed (Figures 9 and 10).

One would expect, therefore, that with increasing energy flux, the texture of the Alton would evolve through the activation of calcite to calcite grain boundaries with higher activation energy thresholds, thus moving modes further from angles representing coherent positions. When the energy flux exceeds the activation threshold involving coherent quartz to calcite boundaries, then the modes in these angular positions would be destroyed. It is likely that most calcite to calcite boundaries would be activated before the energy level needed to activate the quartz to calcite boundaries is reached. In fact, that activation energy might exceed that energy wherein calcite and quartz would react chemically to form wollastonite.

These results indicate that rock textures, through their intergranular relationships are sensitive to and respond to energy fluxes over a wide range. This response is of a continuous nature. Relative and absolute energy fluxes could thus be directly determined from such orientation-frequency diagrams. In this light, such investigations would be of importance in fully understanding the recrystallization of rocks.

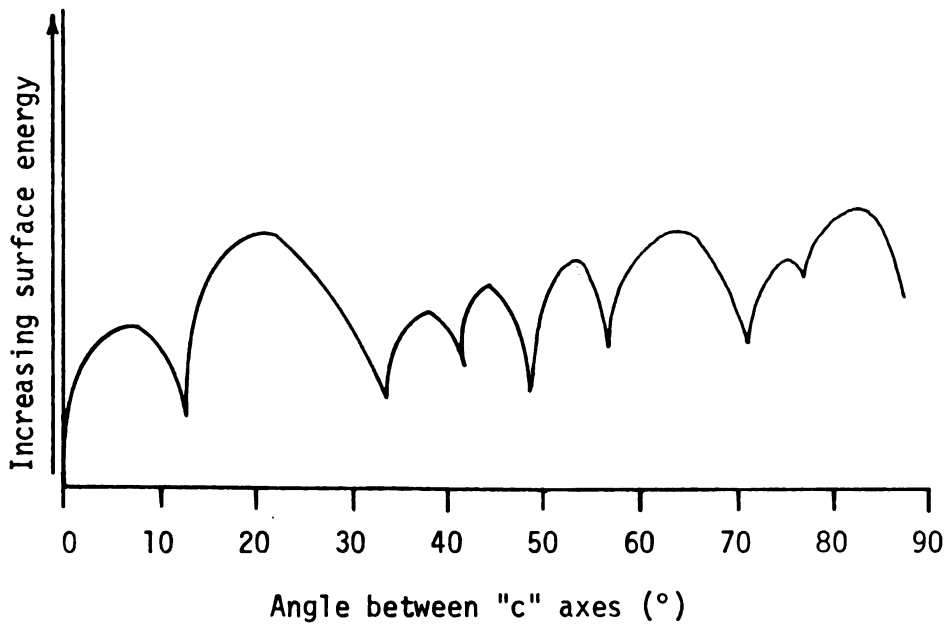

Figure 9. Orientation-surface energy curve for calcite to calcite (After Spry, 1969, p. 33).

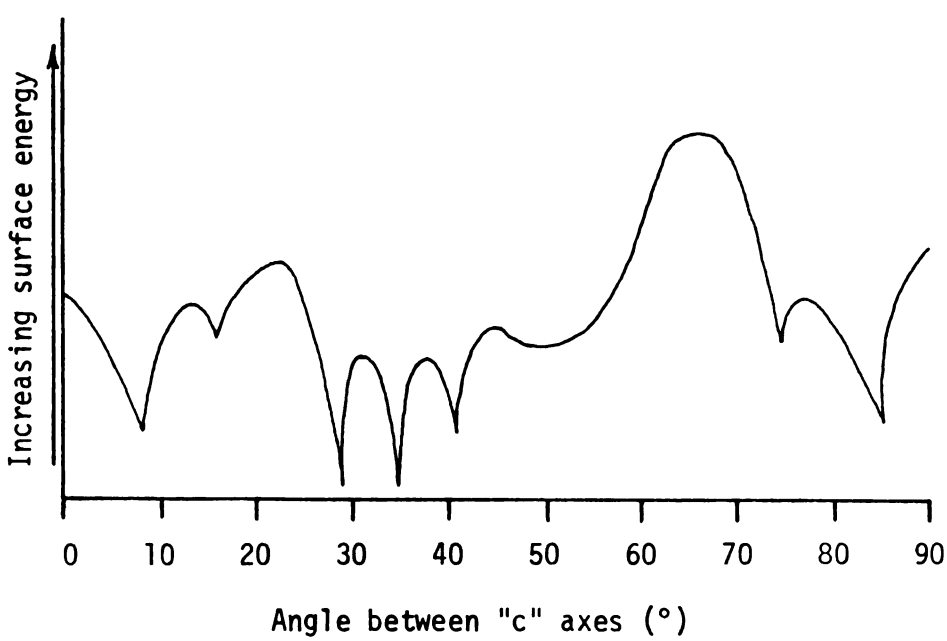

Figure 10. Orientation-surface energy curve for quartz to calcite (After Spry, 1969, p. 33).

LIST OF REFERENCES

LIST OF REFERENCES

Byerly, G. R., and Vogel, T. A., In Press, Grain boundary phenomena in metamorphic plagioclase feldspars: Lithos.

Dana, J. D., 1951, The system of mineralogy, Rewritten by, Palache, C., Berman, H., and Frondel, C.: John Wiley and Sons, Inc., New York.

Gordon, P., and Vandermeer, R. A., 1966, Grain boundary migration, in Recrystallization, grain growth, and textures: American Society for Metals, Metals Park, Ohio, 617 p.

McLean, D., 1965, The science of metamorphism in metals, in Pitcher, W. S., and Flinn, G. W., Editors, Controls of metamorphism: Oliver and Boyd, London, 368 p.

Ransom, D. M., 1971, Host control of recrystallized quartz grains: Mineralogical Magazine, v 38, p. 83-88.

Spry, A., 1969, Metamorphic texture: Pergamon Press, New York, 350 p.