# TWIN AND PSEUDOTWIN INTERGROWTHS IN KAOLINITE

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#### ABSTRACT

Single crystal X-ray study of over 150 flakes and vermicular booklets of sedimentary kaolinite crystals shows that all are pseudotwin intergrowths of three lattice orientations related by 120° spiral rotations about the cleavage normal  $Z^*$ . The three lattice orientations are always present in equal volumes. A low temperature, random selection growth mechanism is proposed in which the vacant octahedral site can be located in any of three possible structural positions (A, B, C) 120° apart. During growth, coherent domains form within the crystal in small regions where cooperative forces influence the vacant site to be in the same position throughout. Each domain has normal triclinic kaolinite geometry, but a domain with site A vacant will have its X and Y axes oriented  $120^{\circ}$  from adjacent domains with sites B or C vacant because of directional distortion around each vacant site. A second type of intergrowth, which is a true twin involving 180° rotation about X and which doubles the number of distinct lattice orientations, is present in most crystals also, especially in the thick vermicular booklets. The frequency of this twinning is variable from crystal to crystal. Both types of intergrowth tend to balance out the directional strain within the distorted, asymmetric kaolinite layer by redirecting the strain on a domain scale.

## INTRODUCTION

Kaolinite occasionally is found in crystals up to a few millimeters in diameter. The two dominant morphologies are as thin, pseudohexagonal (001) platelets and as vermicular booklets for which the thickness may be several times the width. The larger crystals are usually found in porous rocks, such as sandstones and arkoses, but may occur also in massive clay beds. The delicate, fragile shapes of these kaolinite crystals suggest that they have grown *in situ*.

Little information is available in the literature on twinning in kaolinite. The 6th edition (1904) of Dana's System of Mineralogy states that kaolinite sometimes twins according to the mica and penninite laws. Search of the original sources, however, indicates that most of the references are to the varieties dickite and nacrite rather than to kaolinite. Ross and Kerr (1931) in their definitive monograph on the kaolin minerals state "No twinning has been observed in kaolinite crystals." Nevertheless, optical and electron micrographs of kao-

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linite crystals (Conley, 1966; Borst and Keller, 1969) commonly show morphological features suggestive of twinning; these include interpenetrating individuals with reentrant angles on (001), corrugated and striated prismatic surfaces, and the presence within elongate kaolinite stacks of individual packets whose basal surfaces are only quasiparallel.

The present paper reports a single crystal X-ray study of over 150 kaolinite and "anauxite" crystals of different morphologies from eight localities. Intergrowths of three to six lattice orientation in twin and pseudotwin arrangements are universal for these crystals. The intergrowths are considered to be a consequence of growth in a low temperature, sedimentary environment.

EXPERIMENTAL

Samples of macroscopic kaolinite crystals, washed and separated from the enclosing sediments in most cases, were obtained from eight localities in North America, South America, and Europe (Table 1). Two distinct morphologies are represented. The sample from the New Jersey coastal plain, previously described by Lodding (1965) and Isphording and Lodding (1968), consists entirely of thin (001) platelets. All of these flakes exhibit wavy extinction and appear to consist of, many minute but distinct birefringent centers. Thin flakes cleaved from thicker booklets of the Yugoslavia kaolinite were similar to the New Jersey flakes in optical appearance. The other samples contain only vermicular booklets of kaolinite, and even thin cleavages of these proved too opaque for microscopic study in our specimens.

All samples were confirmed to be normal kaolinite by X-ray powder and single crystal study, including the "anauxite" specimens (Bailey and Langston, 1969). A few thin platelets present in the Georgia sample, and similar in appearance to the New Jersey crystals, proved to be muscovite flakes. It is interesting to note that these muscovite flakes are incipiently altered to kaolinite. Weissenberg X-ray photographs show a weak series of 00*l* reflections with a 7 Å repeat distance in parallel orientation with the more intense mica 00*l* reflections.

Kaolinite crystals 0.1 to 0.5 mm in diameter were mounted on glass fibers with (001) parallel to the fiber axis. The crystals were mounted by means of microscope grease in order to avoid any distortion that might be caused by a stronger adhesive. Preliminary Weissenberg examination of several crystals showed that all h0l reflections are either doubled, due to an apparent single twin operation, or are quadrupled, due to two apparent twin operations. Zero and first level Weissenberg photographs were taken about all three pseudohexagonal x axes and about all three y axes for representative examples of singly and doubly "twinned" crystals. A special layer line screen with an adjustable slit was used to insure that the entire length of the elongate reflections could be recorded. Precession photographs of the same representative crystals were taken as a check on the interpretation of the Weissenberg films.

In order to make a statistical study of the frequency of "twin types", oscillation photographs about one of the y axes proved to be quickest and most efficient. After a preliminary Laue photograph taken with the X-ray beam normal to (001), the

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crystal was adjusted so that the nearest y axis was made parallel to the rotation axis. A 15° to 30° oscillation photograph was then taken with (001) normal to the beam at the middle of the oscillation range. The center of the resulting photograph (Fig. 1) shows pseudohexagonal symmetry as a result of the intense 020, 110, 110, and equivalent reflections. Theoretically the 020 and 020 reflections should not occur on oscillation about the y axis. The fact that they do occur on these photos is due to a combination of the mosaic spread of the crystals, the triclinic deviation of the y and  $y^*$  axes, and the presence of several twin and pseudotwin individuals whose  $y^*$  axes do not coincide exactly. The "twinning" is shown best as either a doubling or a quadrupling of the 201, 132, and 132 reflections, which are located 120° apart and along the directions of the three  $x^*$  axes. The doubled reflections, however, show up best on the  $x^*$  axis that lies along the zero layer line. Of the 102 crystals examined in the statistical phase of the study only those crystals too small to give a good pattern could not be identified as to "twin type" by this method.

# INTERPRETATION OF "TWIN" LAWS

All reflections on Weissenberg photographs of kaolinites examined in this study are elongate parallel to the oscillation axis, *i.e.*, along

LOCALITY	DONOR	KAOLINITE DESCRIPTION Thin, colorless (001) platelets, mixed irreg. and hexag. shapes. Wavy extinction, minute bire- fringent centers.	
Woodstown, New Jersey. Alloway Clay Member of Kirkwood Fm.	Prof. T. A. Vogel, Rutgers University		
Huber, Georgia. "soft" clay	Prof. B. F. Buie, Fla. State Univ., & J. M. Huber Corp.	Thick, vermicular hexagonal booklets, white, opaque.	
Simsboro Sandstone in lower Wilcox Fm., central Texas	Prof. E. C. Jonas, Univ. Texas	Thick, vermicular, hexagonal booklets, white, opaque.	
Turin Fm, Restrepo Concession, Colombia, S. A.	Univ. Wis. Coll. #705/43140	Thick, vermicular, hexagonal booklets, tan, opaque.	
Rock Springs, Wyoming. Underclay in Almond Fm. (Asquith, 1968)	Dr. G. B. Asquith, Atlantic Richfield Co.	Thick, vermicular, hexagonal booklets, tan, opaque.	
Kočevje coal mine, Yugoslavia. In sand lens below coal (Krstanović and Radošević, 1961).	Dr. I. Krstanović, University of Beograd, Yugoslavia	Thick, vermicular, hexagonal booklets, white to light greenish gray. Thinner flakes show minute birefringent centers.	
Madera Co., and Amador Co. California. Ione Fm. (Allen, 1928; Langston and Pask, 1968)	Prof. J. A. Pask, Univ. California, Berkeley (through Dr. C. W. Chesterman and Dr. C. S. Ross)	Thick, vermicular, hexagonal booklets in sand and clay matrix with silica scales, white to tan, opaque. Formerly called "anauxite".	
Bilin, Czechoslovakia. Altered basalt (Ross and Foshag, 1928). Smithsonian spec. <b>95535.</b> file 267,168.	Prof. J. A. Pask, Univ. California, Berkeley (through Dr. P. E. Desautels)	Pearly white flakes and booklets in buff clay matrix. Type locality of "anauxite" (Dittler and Hibsch, 1923).	

#### TABLE 1. SAMPLE DESCRIPTIONS

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FIG. 1. 15° oscillation photograph about y axis of Colombia kaolinite with (001) normal to X-ray beam at middle of oscillation range. Note quadrupled reflections (arrows). Fe radiation.

constant  $\theta$ , over an angle varying from 15° to 20°. Aside from this mosaic spread all of the kaolinites are well crystallized. The  $k \neq 3n$ reflections are discrete and, although slightly more diffuse than the k = 3n reflections, do not have any interconnecting streaks due to stacking disorder. It should be pointed out, however, that the degree of crystallinity cannot be determined from precession photographs because the  $k \neq 3n$  "twin" reflections are not well resolved by this technique even with Cu K $\alpha$  radiation.

The apparent twinning creates groups of closely spaced reflections along h0l and 0kl Weissenberg festoons. The normal and "twin" reflections within the groups are approximately, but not exactly, equally spaced along  $z^*$  so that they simulate a supercell with several layers per unit cell. The 00l reflections appear to be single.

## Single "twins"

This type of apparent twinning is characterized by a doubling of h0l reflections and a tripling of 0kl reflections. Within each pair of h0l reflections one member invariably has twice the intensity of the other. This intensity relationship suggests that three reflections of approximately equal intensities are involved, but that two of them superimpose. Close inspection shows that the more intense reflection is al-

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FIG. 2. Weissenberg photograph about y axis of New Jersey kaolinite showing doubling (arrows) of h0l reflections. Cu radiation.

Reciprocal axis	Measured angle	Calc. angle to Z* = [001]*	Calc. angle to [103]
$X_{T}^{*} = [100]^{*}$	76 <sup>0</sup>	75.2°	76.6 <sup>0</sup>
$X_{2}^{*} = [130]^{*}$		96.1	96.9
$X_{3}^{*} = \left[\overline{130}\right]^{*}$	99	99.0	96.8
$Y_1^* = [010]^*$	89	88,4	90.0
$Y_2^{\pm} = \left[\overline{110}\right] *$	104	103,7	101.7
$Y_3^{*} = \left[1\overline{1}0\right]^{*}$	78	77.8	78,2

TABLE 2. COMPARISON OF CALCULATED AND OBSERVED INTERZONAL ANGLES

ways slightly more elongate than the weaker reflection, so that the superposition making up the more intense reflection is not exact (Fig. 2).

The doubled h0l reflections can be indexed on two separate nets with interaxial angles of approximately 76° and 99°. Fron this geometry it can be deduced that the more intense spot within each pair is a superimposition of 2.0.*l* and  $\overline{1.3.l} + 1$  reflections for the positive *l* quadrant of the first h0l Weissenberg festoon, of 4.0.l and  $\overline{2.6.l} + 1$  for the second festoon, and so forth. The weaker spots along the first festoon are single reflections of index  $\overline{1.3.l} + 1$ . The zones  $[\overline{130}]^*$  and  $[\overline{130}]^*$  are the two pseudo  $+x^*$  axes, which must lie in the true  $z^*x^*$  net plane as a result of the twinning or inter growth.

The Okl Weissenberg photograph of the same crystal shows triplet reflections that can be explained as a result of the three pseudohexagonal  $+y^*$  axes [010]\*, [110]\*, and [110]\* being grouped together so they are nearly coplanar with the z\* axis. In this case there is no overlapping of reflections from the three reciprocal net planes, except for those with the smallest  $\theta$  values within each festoon. The photograph shows an apparent monoclinic symmetry because triplet reflections on one side of  $y^*$  are similar in position and intensity to triplets on the other side, apparently relating 0kl to  $0k\bar{l}$ . Because of the triclinic geometry of kaolinite, however, reflections of similar indices do not have similar  $\theta$  values and are interchanged in position within triplets on opposite sides of  $y^*$ . For example, reflection 062 would be at the top of a triplet on one side of  $u^*$  but  $06\overline{2}$  would be the bottom reflection of the triplet on the other side. The apparent monoclinic relationship results from comparing reflections of differing indices, such as 062 with the 331 "twin" reflection of similar  $\theta$  and intensity value. The "monoclinic kaolinite" described from Yugoslavia by Krstanović and Radošević (1961) and

also examined in this study is, in reality, true kaolinite in a somewhat similar intergrowth of the double "twin" type (see next section).

The theoretical angles between  $z^*$  and the six pseudohexagonal  $x^*$  and  $y^*$  axes have been calculated from the kaolinite unit cell parameters of Newnham, as given by Brindley and Nakahira (1958). The observed values, which cannot be measured very precisely because of the mosaic



FIG. 3 (a) Stereogram of kaolinite Bravais and reciprocal axes.  $y_1^*$  is normal to  $x_1z_1$ , etc. Prime and double prime numbers refer to reciprocal axes after 120° rotations about  $z^*$ . Solid dots represent poles in upper hemisphere, circles those in lower hemisphere. (b) Stereogram of kaolinite reciprocal axes assuming 120° rotations about [103] zone axis.

spread, compare well with the calculated angles (Table 2). The angles from  $z^*$  to the rotated  $x_1^*$  and  $x_2^*$  axes cannot be measured separately because reflections from these two net planes mutually superimpose. The measured value of 76° refers to an axis passing through the superimposed 200 and  $\overline{131}$  reflections, and should be compared with an average of the calculated angles of 75.2° and 75.8° to these individual reflections.

Weissenberg photographs taken 120° apart by oscillation about each of the three pseudohexagonal x axes of the same kaolinite crystal are identical, as are photographs taken 120° apart about each of the three y axes. As a result, the apparent twin operation must involve 120° rotations about  $z^*$ . In a single crystal of kaolinite the six pseudohexagonal  $x^*$  and  $y^*$  axes are coplanar in a net inclined 15° to the (001) cleavage. Rotations of 120° about the cleavage normal  $z^*$  keeps the (001) morphology intact and groups the reciprocal axes as shown in the stereogram of Figure 3a. Because of the triclinic geometry  $+x_2^*$  is not coincident with  $+x_3^*$  after rotation, but deviates by 3°.

 $Z^*$  cannot be a true twin axis according to the present definition of twinning because it does not coincide exactly with any row line of the direct lattice for kaolinite. In layer silicates for which the layer shift is exactly a/3,  $z^*$  will coincide with the [103] direct zone. For kaolinite, however,  $z^*$  deviates from [103] by 2°03'. The stereogram in Figure 3b shows the relation of axes that would result by true 120° twin rotations about [103] for kaolinite. This model predicts interaxial angles of 76.6° from [103] to  $x_1^*$  and of 96.9° to  $x_2^* + x_3^*$  (Table 2). The observed angles of 76° and 99° can be measured accurately enough to prove the model in Figure 3a. Measurements to the  $y^*$  axes (Table 2) also agree best with the model of Figure 3a, in which  $z^*$  is the "twin" axis. Perhaps the most convincing evidence in favor of  $z^*$  as the "twin" axis is that the alternative model in Figure 3b predicts that the rotated  $x_2^*$  and  $x_3^*$  axes should overlap and, from reconstruction of the three integrown reciprocal lattices, that reflections of index  $\overline{1.3.l} + 1$  and  $\overline{1.3.l} + 1$  from these two nets should coincide in position with reflections of index 2.0.1 from the  $x_1^*z^*$  net. Thus there would be no doubling of hol reflections, contrary to observation (Fig. 2).

The intergrowths in this paper will be described as pseudotwins rather than true twins. It should be noted, however, that the geometry involved is equivalent to that of one of the forms of the mica twin law in a centrosymmetric 2:1 type layer structure, where the spiral 120° rotations about  $z^* = [001]^*$  can be described alternatively as 180° rotations about each of the direct lateral axes  $y_2 = [\overline{310}]$  and  $y_3 = [\overline{310}]$ . Either (001), (110), (110), or an irregular surface can be the composition plane. Some micas and chlorites twinned by the mica law exhibit crystal faces that indicate the lateral axis description is correct for these crystals, because the layer has been inverted by the twinning. For example, see the muscovite from Abühl, illustrated in Figure 18 by Tschermak (1877). For most crystals twinned by the mica law the crystal morphology is inconclusive, however, and X-ray studies have not been attempted. The frequency of intergrowths in kaolinite, where X-ray study shows the acentric 1:1 type layer is not inverted, is at least suggestive that similar intergrowths could occur in other layer silicates without layer inversion. Rotation about the normal to a net plane should then be accepted as a true twinning operation, even if the twin axis is not a row line in the direct lattice.

The pseudotwin intergrowth involving  $[001]^*_{120^\circ}$  rotations has been observed for every sedimentary kaolinite and "anauxite" crystal examined in the present study. The equal intensities observed for equivalent "twin" reflections indicate that equal volumes of the three individuals must be present in all crystals. Clearly, some universal growth mechanism is required by way of explanation.

### Double "twins"

The 0kl Weissenberg photographs of crystals showing two types of apparent twinning are not distinguishable from those for the single pseudotwin intergrowths. Differentiation must be made by means of h0l Weissenberg photographs, on which there are closely spaced groups of four reflections (Fig. 4) in contrast to the pair of reflections found for the single "twins" (Fig. 2). Within each set of four h0l reflections two are identical in position and intensity to the pairs created by the three pseudotwin orientations previously discussed. The additional two reflections arise from a second set of three orientations that result from a second twin operation.

In order to define uniquely the twin operation involved it is necessary to determine the positive and negative ends of the crystallographic axes. These follow directly from the preceding descriptions of the h0l and 0kl Weissenberg photographs, and can be checked further on higher level photographs.

The observation that the two different pairs of reflections do not superimpose on h0l photographs but lie along the same Weissenberg festoon is a result of the matching of  $+\beta^*$  and pseudo  $+\beta^*$  of three lattice orientations with  $-\beta^*$  and pseudo  $-\beta^*$  of the second set of three lattice orientations. This is only possible if the sense (+ or -) of the rotation axis y is different for the two sets. It also requires that the



FIG. 4. Weissenberg photograph about y axis of Colombia kaolinite showing quadrupling (arrows) of h0l reflections. Cu radiation.

sense of either  $x^*$  or  $z^*$ , but not both, must be different for the two sets. Exactly as for the single pseudotwin examples, one spot within a given reflection pair is always twice the intensity of the other spot as a result of superposition of reflections from equal volumes of two individual crystal orientations. The weaker spots from each reflection pair do not overlap, and pseudotwin reflections of index 13*l* or  $\overline{13l}$  from one orientation lie immediately adjacent to twin reflections of index  $\overline{1.3.l} + 1$  or 1.3.l - 1, respectively, from a second orientation. This is true both on hol and 1kl photographs.

On 0kl Weissenberg photographs all twin reflections from the second set of three orientations exactly superimpose on reflections from the first set, e.g., 0kl on  $0\bar{k}\bar{l}$  or  $0\bar{k}l$  on  $0k\bar{l}$ , and it is possible to measure the  $\alpha^*$ angle of 88.4°. Because it is known from h0l photographs that the sense of the y axis must be different for the two sets of orientations, the superposition observed on  $0k\bar{l}$  photographs requires that the sense of the  $z^*$  axes must be different also.

The geometrical relationship that is thus defined is a true twin involving a  $180^{\circ}$  rotation about the x axis. It can be described either as a rotation of the first set of three lattice orientations about a single x axis or as a rotation of single individuals about all three pseudohexagonal x axes. There is no consistent intensity relationship between reflections arising from the two different sets of lattice orientations, so that different volumes of the two sets must be involved in different crystals. This is contrary to the pseudotwin intergrowths involving 120° rotations about  $z^*$  within each set, which do have equal volumes of three individuals.

#### STATISTICAL STUDY OF "TWIN" TYPES

In order to determine the relative frequency of the two "twin" types, a statistical study was undertaken on the samples from New Jersey, Georgia, and Colombia. A total of 100 crystals was selected at random from the three samples and their "twin" types determined by the X-ray oscillation method described previously. The results are as follows:

	New Jersey	Georgia	Colombia
Number of crystals	52	23	25
Number of usuable photographs	50	20	21
Percent singly "twinned"	44	20	5
Percent doubly "twinned"	56	80	95
Deviation at confidence level 0.95	$\pm 2.0$	$\pm 2.0$	$\pm 2.1$

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An additional 61 crystals were examined in the course of the study from the other five samples. These crystals are not included in the statistics above because they were selected as a result of their perfection of morphology rather than at random. It is noteworthy, however, that all 61 crystals were doubly "twinned".

It is evident that the great majority of macroscopic kaolinite crystals are doubly "twinned" and consist of six individual lattice orientations. There is an especially noteworthy correlation of double "twinning" with thick, vermicular, hexagonal crystals. This is true even when the vermicular booklets are cleaved into as thin (001) platelets as possible on a macro-scale. The New Jersey sample is unique in consisting entirely of thin (001) platelets, rather than thick, vermicular booklets. It differs also in showing approximately equal numbers of single and double "twin" types.

## ORIGIN OF INTERGROWTHS

The so-called single pseudotwin has been shown to be an intergrowth of three lattice orientations related by  $120^{\circ}$  spiral rotations about  $z^*$ . It has also been noted that the observed intensities of the X-ray reflections from the three units involved are always equal and that this must require equal volumes of the three lattice orientations. This suggests origin by a random selection growth process in which there are equal probabilities of adopting three orientations  $120^{\circ}$  apart.

Figure 5 illustrates a plan view of a 1:1 type layer, in which the three possible locations for octahedral Al cations are labeled A, B, C. One of these three locations must be vacant in dioctahedral kaolinite. Adoption of the same vacant site in each successive layer, and distortion of the structure around the vacant site causes kaolinite to be triclinic (Brindley and Nakahira, 1958; Bailey, 1963).

It is proposed here that there is in fact an equal probability of each of the three possible octahedral sites being vacant and that during the initial phase of crystal growth A, B, and C are adopted equally as the vacant site, but in different small regions (domains) of the growing seed crystal. Cooperative bond forces and the tendency to attain as low a structural energy as possible will work toward enlarging each domain in a coherent fashion, both laterally and vertically, so that the same site is vacant throughout the domain as the crystal grows. Domain enlargement is limited, however, by the opposing tendency for random establishment of adjacent domains having the vacant site in a different location. As a result the macro-crystal will tend to be made up of a large number of small domains that are not structurally coherent with their neighbors. The sharpness of the diffraction spots

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suggests a lower limit to the domain size of a few hundred Ångstroms. Within each of the three types of domains that are created by having sites A, B, or C vacant, repulsion between adjacent  $Al^{3+}$  cations and shortening of shared octahedral edges will enlarge the vacant octahedral site relative to the occupied sites and will distort each domain to triclinic geometry. The three types of domains will be distorted in three different directions, however, as a result of having a different octahedral site vacant. The direction of distortion in each domain can be predicted according to the rules summarized by Bailey



FIG. 5. Schematic (001) diagram of kaolinite structure to illustrate domains oriented 120° apart as a result of adoption of sites A, B, or C as the vacant octahedral site. Each domain is distorted differently as a result of the different vacant site, so that the conventional triclinic unit cells must be related by 120° rotations about  $z^*$ . Upper plane of OH atoms is not shown.

(1966). Figure 5 illustrates the resulting lattice orientations, 120° apart, within the three domain types relative to the conventional orientation of the triclinic axes.

All of the specimens examined in this study are believed to be of sedimentary origin. Their fragile shapes and hexagonal morphology suggest they have grown *in situ* at essentially earth temperatures. A random-choice growth process of the sort proposed here should be especially favored at low temperature. Cations arriving at the growth surface will not have sufficient kinetic energy to migrate to those positions that are most favorable energetically and that would produce a true single crystal. Rapid growth, in which arriving cations are incorporated into the crystal rapidly, should also favor a randomchoice process. If true single crystals of kaolinite exist, therefore, they should be sought in higher temperature occurrences under conditions of relatively slow, undisturbed growth.

The second "twin" type is a true twin involving  $180^{\circ}$  rotation about the crystallographic x-axis, but undoubtedly originates during the growth process also. It may or may not be present in a given crystal. When present, the proportions of twin components are quite variable from crystal to crystal. Rotation about x does not change the position of the vacant octahedral site, but does turn the layer upside down. This may explain why the probability of twinning of this type increases in the thicker booklets and why electron micrographs of the edges of kaolinite booklets appear corrugated (Conley, 1966, Fig. 6). Reversal of the structure by rotation about x necessarily reverses the direction of stagger of successive layers and would create a prismatic reentrant angle. Repeated twinning would create a corrugated prismatic surface.

Most dioctahedral layer silicates have 2-layer periodicity in which the location of the vacant octahedral site alternates regularly from layer to layer. This tends to balance the distribution of strain from layer to layer. In kaolinite the 1:1 layer type and the adoption of the same vacant octahedral site in each layer combine to produce a very asymmetric and strained structure. Kaolinite probably owes its customary small grain size to difficulty in extending its distorted structure coherently over many unit cells. The two types of intergrowths observed in macro-crystals of kaolinite can be viewed as two methods of alleviating the strain by redirecting the distortions on a domain scale.

#### ACKNOWLEDGMENTS

This study has been supported in part by a grant from the Georgia Kaolin Company and in part by grants GA-1681 from the National Science Foundation

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and grants 1176-A2 and 4899-AC2 from the Petroleum Research Fund, administered by the American Chemical Society. Samples were provided by T. A. Vogel, B. F. Buie, E. C. Jonas, G. B. Asquith, I. Krstanović, and J. A. Pask. All of this support is acknowledged gratefully. Valued advice on twinning was received from J. D. H. Donnay.

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Manuscript received, August 30, 1971; accepted for publication, October 27, 1971.