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KINETIC CONSIDERATIONS IN THE GENESIS OF GROWTH TWINNING

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ABSTRACT

Invariable growth-twinned overgrowths in some authigenic albite crystals suggest that such twinning results from interfacial kinetic competition during growth. This implies that much growth twinning may be predicted from kinetic considerations and may not be explicable solely in terms of the energy increase resulting from growth accidents.

INTRODUCTION

The genesis of twinning is a subject of continued interest among crystallographers. Twinning is generally considered to result from three more or less distinct phenomena: accidents during growth, mechanical deformation, and transformation from an original to a derivative (usually lower) symmetry. Mechanical and transformation twinning have received a great deal of attention recently, mostly from metallurgists, and the conditions for their formation are reasonably well understood. Growth twinning, however, is only poorly understood, largely because the phenomenon of crystal growth is itself so imperfectly understood.

The current theories on the formation of growth twinning can be summarized as follows: Buerger (1945) believes that growth twinning is the result of a growth accident resulting in the nucleation of a small crystal unit which becomes attached to a host crystal in twinned orientation, and which is neither redissolved nor otherwise rejected. Conditions favoring the formation of growth twins are related to rapidity of growth and to inability of the twin to make an energetic distinction between an untwinned extension and an extension in twinned orientation at the composition plane. Therefore, supersaturation of the solution with respect to the growing crystal and minimal size of the crystal at the time of nucleation of the twinned crystal would be most favorable to twin development. Hartman (1956) explains the relative probability of various growth twins on the basis of energy conditions at the twin boundary—

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stating that the twinning can start on an F-face or, less probably, on an S-face (faces which grow by accretion of layers, or of chains, respectively). Kern (1961) attacks the problem in terms of the angular lattice discordance between the two crystals of the twin. Various angular discordances result in various energy increases (over the energy of the untwinned crystal), and the most favored twin is that which results in the smallest energy increase. All of these theories have one thing in common: they all regard crystal growth as a phenomenon in which the crystal is trying to reach some sort of equilibrium or lowest energy condition, and twinning is an accident during that growth. The energy difference between the twinned and untwinned state is all-important for each of these authors.

Serious objections may be raised to these theories from a purely theoretical viewpoint. The process of crystal growth, with a very few exceptions, is not a reversible process, and there is little reason to believe that crystals grow solely in an attempt to lower the energy of the system. It is perhaps more reasonable to regard crystal growth as a kinetic process, in which the resulting forms reflect more rapid rather than more stable directions of growth.

A second objection to the above theories stems from the growing body of observations on twins which have been made over the past century. For instance, it has long been noted that twins are frequently distinctly larger than are untwinned crystals with which they occur. Secondly, twinning is commonly associated genetically with rapidity of growth, or with high viscosity of the medium. Thirdly, some crystalline species are virtually always twinned (*e.g.*, aragonite). If twinning results in a higher total energy and if attainment of lower energy is the determining factor in the development of a crystal's form, then it is not easy to understand the first of these observations, and the third makes no sense at all. Clearly another explanation must be found.

The chief difficulty in studying growth twins is that it is generally impossible to see them form—we only see them after they have grown. Buerger's hypothesis favoring the formation at a very early stage, when the crystal nucleus is extremely small, cannot be refuted for most observations. However, twinned overgrowths may be observed forming on a nucleus of relatively large size. The present study concerns two examples of this—one being authigenic albite, whose development can be inferred from an examination of its present external and internal morphology, and the second being gypsum, whose growth from sea-water solutions may be observed with a microscope. In both of these examples, it is noted that twinning is an invariable and apparently essential aspect of crystal growth.

AUTHIGENIC ALBITE TWINS

Two excellent examples of authigenic albite twins have been examined. The first of these consists of albite twins (many thousand have been examined) from the Paradox Limestone (Mississippian) of New Mexico, and the second consists of almost identical twins dissolved from the salt of the Tatum salt dome of Mississippi. In neither case is information available as to the size distribution of authigenic crystals (the samples having been sieved before I examined them). Information about coexisting authigenic minerals for the Mississippi example is given by Schlocker (1963). The New Mexico example (from an oil well) is evidently associated with salt deposits, although details of the subsurface lithology are not available.

In both examples 100 percent of the albite occurs as twinned crystals arranged as fourlings around a central nucleus, which is rather easily identified as detrital sodic plagioclase of very fine sand to silt size (Fig. 1). All of the twinning is of the albite law, and the individual crystals are oriented so that the opposite quadrants are of the same crystallographic orientation and twinned with respect to the adjacent quadrants. The composition planes are irregular, but tend to approximate (010) between adjacent crystals at the same end of an elongate grain, and a series of inclined, apparently irrational planes closer to (010) than to any other pinacoid between adjacent crystals at opposite ends of the grain. Although it was not possible to make direct observations of the axial angle on the U-stage because of the shape of the grains, a few were measured by the less accurate Dodge method, and these were found to be very close to the low-temperature, ordered condition.

The external morphology of these twins (here called the Paradox type) was investigated with a two-circle goniometer. After several unsuccessful attempts, a twin with reasonably well developed minor faces was measured, and the results are summarized in Table 1. The following forms, in order of their prominence, are present (using the Dana nomenclature): $c\{001\}$, $b\{010\}$, $p\{\bar{1}11\}$, $M\{1\bar{1}0\}$, $z\{1\bar{3}0\}$, $n\{0\bar{2}1\}$. The forms $m\{110\}$ and $f\{130\}$ were found only in renetrants. The twins were not completely euhedral; in the vicinity of the irregular composition planes the facies appeared to be rough, although a more detailed investigation was impossible because the dimensions of what might have been tiny facets in this area were sufficiently close in size to the wave length of light that diffraction effects obscured the images. Similarly, a few of the faces measured (especially *n*) were so narrow that diffraction made their angular measurement difficult. The form of the typical twin is shown in Figures 2 and 3.

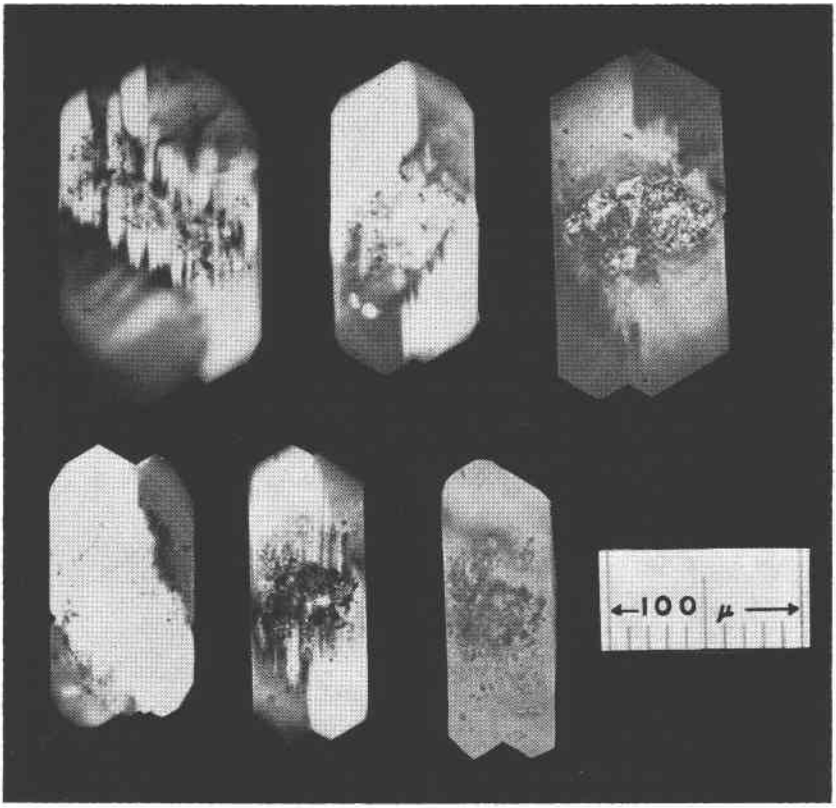

FIG. 1. Twinned authigenic albite from Paradox Limestone, New Mexico (upper photos) and Tatum Salt dome, Mississippi (lower photos). All crossed polarizers except for lower right, which is the same grain as lower center. The outlines of the grains have been retouched. Note the detrital nuclei, one of which (upper left) is itself polysynthetically twinned.

TABLE 1. GONIOMETRIC MEASUREMENT OF TWINNED ALBITE CRYSTAL, MISSISSIPPI

Faces	Measured	Calculated
$(010)\wedge(\bar{1}11)$ $b\wedge p$	60°29′	60°26′
$(0\bar{1}0)\wedge(\underline{\bar{1}11})$ $b\wedge\underline{p}$*	59°44′	60°26′
$(0\bar{1}0)\wedge(0\bar{2}1)$ $b\wedge n$*	46°15′	46°50′
$(1\bar{1}0)\wedge(0\bar{2}1)$ $M\wedge n$*	51°27′	51°35′
$(0\bar{2}1)\wedge(\bar{1}11)$ $n\wedge p$*	87°14′	87°51′
$(\bar{1}11)\wedge(\underline{\bar{1}11})$ $p\wedge\underline{p}$*	59°46′	59°52′
$(0\bar{2}1)\wedge(\underline{\bar{1}11})$ $n\wedge\underline{p}$*	43°01′	46°27′

* Poor measurement.

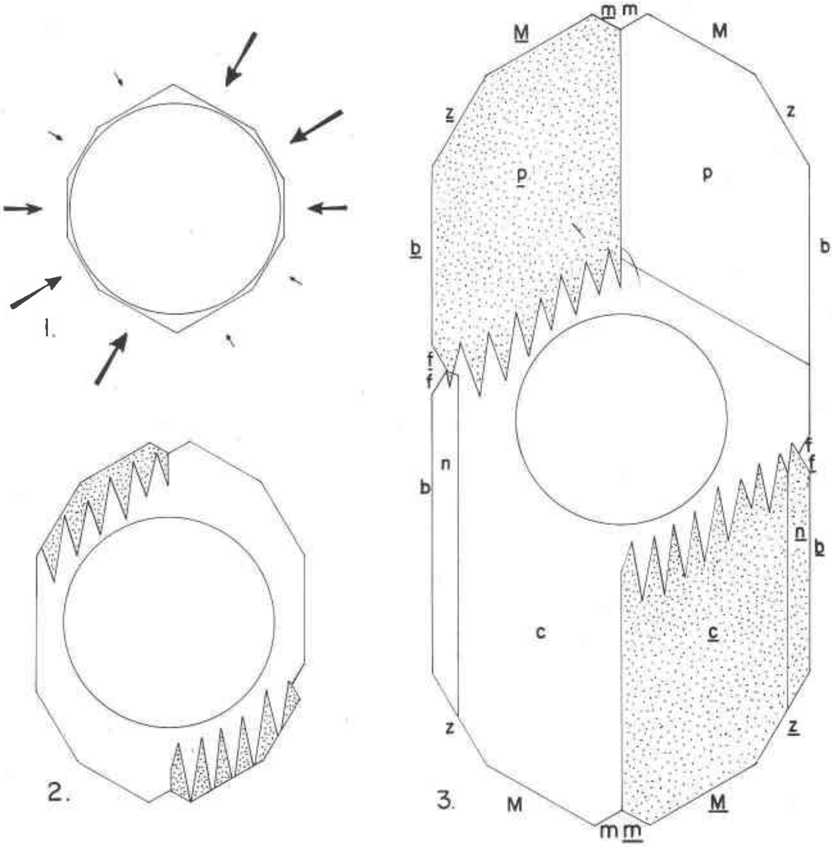

FIG. 2. Three stages in the development of a typical Paradox twin. Twinned portions stippled. Note that some of the facial boundaries are indefinite.

One of the most significant features of these twins is that the twinning is independent of the twinning (if any) of the nuclear grain. Several grains with a twinned nucleus were found; these generally do not have the twinning pattern transmitted to the overgrowth for any appreciable distance away from the nucleus (Fig. 1, upper left). Regardless of the twinned or untwinned condition of the nuclear grain, the same fourling twinning of the overgrowth was developed in all cases.

Hypothetical formation of the fourling twin. The fact that the twinning formed during overgrowth around a nucleus of relatively large size enables us to draw reasonably clear inferences concerning the stages of development of the twin and to speculate on the factors favoring twinning. Very probably the initial overgrowth shell was formed in crystallographic continuity with the nucleus, and simply represented a development of euhedral form (including possibly several minor forms) approximating the original detrital shape. Continued growth eliminated more minor forms, and at a very early stage of overgrowth the grain probably resembled drawing 1 of Figure 2. The wedge-shaped interface between the untwinned extension of the nucleus and the twinned quadrants (which are shown by stippling in drawings 2 and 3 of Fig. 2) suggests that shortly after the initiation of overgrowth several nuclei formed in twinned orientation on the faces of the crystal. Only those nuclei that were oriented according to the albite-twin law and developed on the original *m* and *f* faces have survived. We can only speculate that nuclei with other twin orientations, or with albite-twin orientations forming on other faces, were either redissolved or never attained visible size before they were enveloped by the surrounding crystal. A more significant inference can be drawn from the wedge-shaped interface between the crystals. Once formed, those twinned nuclei, that exposed *M* and *z* faces on a portion of the host crystal with *m* and *f* faces, were apparently assured of success. That is, the velocity of growth of these nuclei was greater than that of the host. The host crystal was displaced entirely by the twinned nuclei, which coalesced and formed the twinned quadrants of the grain.

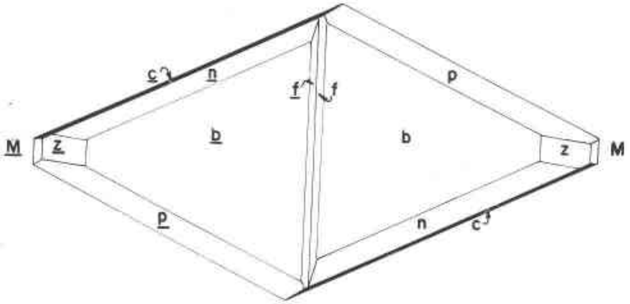

FIG. 3. Appearance of Paradox twin viewed perpendicular to {010}.

The reasons for this twinning must lie in the different growth characteristics of the faces involved. In a triclinic, centrosymmetric crystal, the growth characteristics of faces will be related only by the center of symmetry. We may suppose that *M* and *m*, as well as *f* and *z*, faces will attract ions[1] at different rates, and the rate of increase in their size will not be the same. Regardless of the precise nature of the growth mechanism, the diffusion of materials inwards along concentration gradients in

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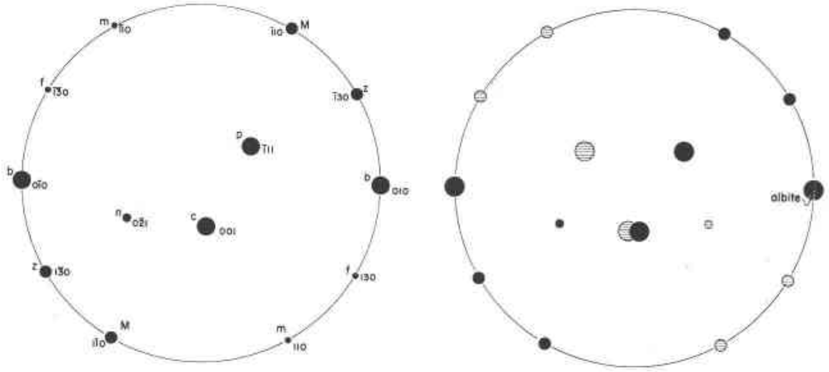
[1] The word ion here does not exclude the possibility that the growth process is one of accretion of molecular groups.

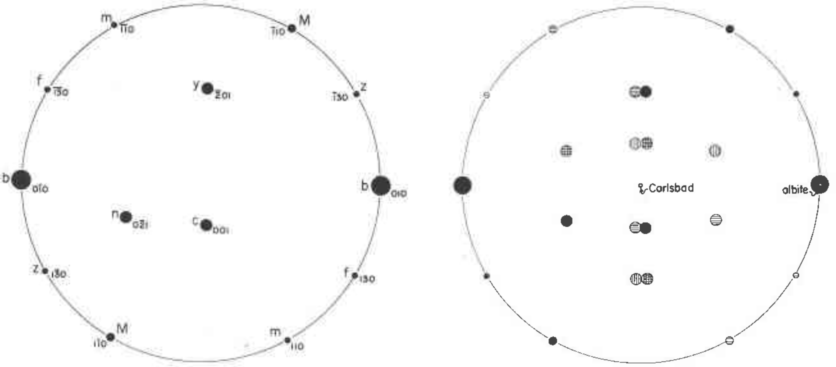
response to potentials (not necessarily electrostatic) on the growing face is a well established theory. The present hypothesis simply states that where the supply of ions or groups is limited with respect to the rate at which ions or groups can be incorporated on the faces, then more rapidly growing faces (faces with higher potential) will attract the majority of inwardly diffusing materials, and hence will be more successful in their growth.

Several situations may be envisaged as creating conditions under which kinetically favored growth forms will prevail. A very low solubility of the crystal in the solvent will require ions to diffuse a relatively long distance. Viscosity of the medium (if liquid) or rate of solid or boundary diffusion (if the matrix is solid) may similarly retard the supply of materials. A high degree of supersaturation or a rapidly changing physical environment (such as rapid cooling or rapid release of pressure) may lead to the same favoring of kinetic considerations over energy consideration. In no case here is the growing grain acting only to bring its internal energy to a minimal value; in fact, the invariable formation of these twins shows that the minimization of internal energy is a secondary consideration. Instead the grain may be thought of as growing so as to expose the most successful faces to the environment in a spatial array that presents these faces in the widest distribution of directions. Offsetting this effect, to a certain extent, is the increased internal energy of the twin, and we might conclude that the most common twin laws (representing perhaps the minimal increases of internal energy) might be favored, unless a less common twin law resulted in an obviously more favorable array of rapidly growing faces. The illustration of this principle is seen in Figure 4 (top), in which the relative size of faces on the Paradox type of twin is shown by size of circles arranged on a stereographic projection (upper half only). In addition to the [001] zone faces, the faces *c*, *p*, and, to a lesser extent, *n*, are dominant. The operation of the albite-twin law shifts the *p* and *n* faces to regions in which there is no other rapidly growing face present, and so this twin law results in a more favorable angular facial array. The Carlsbad law in the present case would give little advantage either in addition to the albite law or by itself; the *p* and *n* faces of the Carlsbad-twinned crystals would be oriented nearly parallel to the *n* and *p* faces, respectively, of the original crystal, and the *c* face would be rotated to a position directly between the *p* face of the original crystal and the *p* face of the albite-twinned crystal.

The present arguments should not be construed as suggesting that internal energy considerations are unimportant in determining the form of a twinned crystal grown under these conditions. On the contrary, the increased internal energy becomes a locally important consideration at



Paradox– untwinned Paradox– twinned



Roc Tourné– untwinned Roc Tourné– twinned

FIG. 4. Stereographic projections (upper hemisphere only) showing spatial arrangement of faces of Paradox (upper) and Roc-Tourné (lower) twins, with size of circular symbols showing their relative prominence. Original orientation, full circles; albite-twinned faces, horizontal lines; Carlsbad-twinned faces, vertical lines. Twin axes shown.

the outcropping of the twin plane on the twin surface. In all cases where this could be observed, this outcrop had the form of a re-entrant (with *m* and *f* faces, which are otherwise absent) or a roughened, depressed area. We might conclude that the increased energy of the twin plane is expressed during growth as a locally decreased potential for the diffusion of ions and that these twin-boundary outcrops represent locally starved zones.

The final form of the twinned crystal is more equant than its untwinned counterpart would be, if we assume that the relative importance of the faces would be the same. The twinned crystal, then, represents the result of a more efficient ionic transport to the faces, in that, if the original distribution of ions were homogeneous, the mean distance traveled by an ion to the twin was less than would have been the case for the untwinned counterpart.

Roc Tourné vs. *Paradox twinning.* A similar type of twin in certain authigenic albites from Europe has been known for a century and a half. These were best described by Rose (1865) and have come to be known, from the location of Rose's most distinctive material, as Roc Tourné twins. Fuchtbauer (1956) has added some further notes on this type of twinning, showing, for example, that the crystals flattened on $\{010\}$ are twinned on a combination of the albite and Carlsbad law. Fuchtbauer notes, and the various American examples recently studied substantiate, that crystals flattened on $\{010\}$ are generally much more scarce than are crystals flattened on $\{001\}$ and $\{\bar{1}11\}$. Because the Roc Tourné twins (referring to those flattened on $\{010\}$, with the combination of albite and Carlsbad twinning) show a different association of twin laws than the more common Paradox type, it might be profitable to examine them more closely. Rose's drawings show that *b* is the dominant face, and that *c*, *y* $\{\bar{2}01\}$, and *n* are all of nearly the same, secondary importance. *M* and *z* are of secondary or tertiary importance; the distinction is of no interest here. Apparently the face *p* is absent, or nearly so. Rose labels one small face $o\{\bar{1}11\}$, but he gives its angle with *b* as 57°37′. The calculated value for the angle between *b* and *p* is 57°49′ and between *b* and *o* 55°53′, so his identification of this face, and the arrangement of this part of his drawing, must be considered doubtful. However, it is only important to note here the dominance of *b*, the equivalence of the secondary faces *c*, *y*, and *n*, and the apparent absence of *p*. Twinning *c*, *y*, and *n* on the albite law alone leaves a large portion of the angular space around the crystal unoccupied by normals to these rapidly growing faces. Additional twinning on the Carlsbad law, however, fills many of these gaps nicely and results in a more equant distribution of rapidly growing faces, and, hence, a more equant, evidently more efficiently grown crystal (Fig. 4, lower). We might conclude, then, that regardless of what specific growth conditions were responsible for the different development of faces, this different facial development resulted inevitably in the formation of the two twin laws, rather than the albite law by itself.

Observations on Gypsum

A simple experiment that requires no more than some seawater and a polarizing microscope is the observation of gypsum growth during evaporation. The crystals precipitate at early stages of evaporation, forming the familiar swallowtail twins. Most of these twins evidently form at very small crystal sizes (Fig. 5, left); at any rate it is difficult to observe an untwinned crystal in the act of beginning its twinning. The hourglass configuration of the two crystals comprising the twin, and the orientation of the boundary between them, suggests that twinning began after the original crystal had reached a small size, and that the crystals in twinned orientation with respect to the host crystal initially grew with a velocity greater than that of the host. Eventually a stage was reached at which the velocities of the two crystals (it is now useless to talk of the original untwinned grain and the twinned portion of the overgrowth) were equal, with minor fluctuations, and the boundaries between adjacent crystals approximate the directions parallel to the *c*-axis and perpendicular to it.

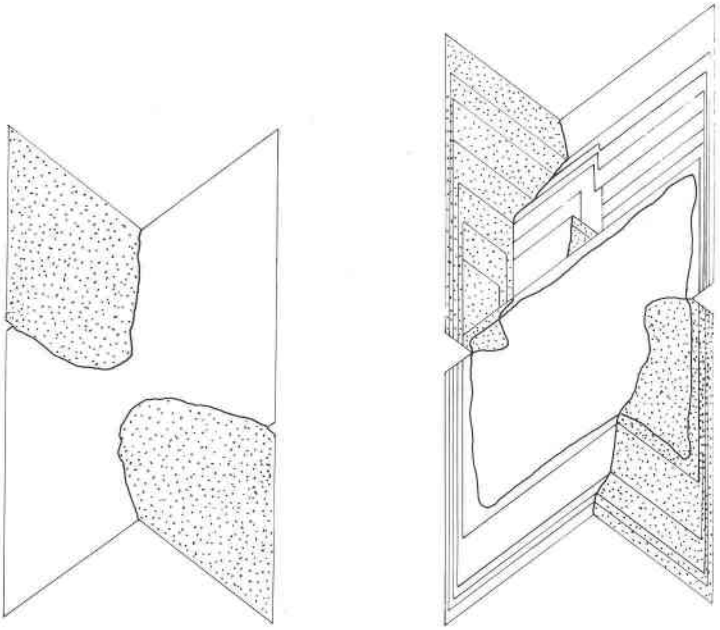


FIG. 5. Two gypsum crystals (omitting the very small prism and dome faces) precipitated from sea water. The crystal on the right shows, by successive outline, overgrowth on a partially dissolved nucleus (heavy, irregular outline).

If we take one of these twins and partially dissolve or otherwise break it up with a pin, we can obtain an untwinned, irregular nucleus, whose shape is considerable different from the equilibrium, or favored, shape. The evaporation of successive portions of seawater will cause overgrowths on these irregular nuclei. These overgrowths will commonly be crystallographically continuous with the nucleus, but at various places on the perimeter of the nucleus, especially at places where the local facial outline is less favorably oriented with respect to the receipt of ions than might be some other facial orientation, portions of the overgrowth in twinned orientation will develop. As the overgrowth continues to grow, a competition between the two facial orientations can be observed—in most cases the crystal with the more favorable orientation with respect to the local ion supply will grow faster. As the twin slowly changes its shape, the relative velocities of these portions of the overgrowth change also, and a small, twinned part of the overgrowth that was originally favored may later find itself at a disadvantage with respect to another, untwinned, portion of the overgrowth, which will then go on to envelop it and seal it off. The ultimate arrangement of crystals in an overgrown, irregular nucleus will be very nearly the same as for a twin which grew in one stage from a very small nucleus (Fig. 5, right-hand drawing).

The original overgrowths on this nucleus included one crystal in twinned orientation (small stippled triangle) whose original orientation advantage was removed when the growth of the adjacent, untwinned portion of the grain altered the local supply of ions. This grain also shows, by the isochronous outlines, that certain adjacent portions of the grain, twinned with respect to each other, (upper left portion) grew at different rates, again according to their orientation with respect to the supply of ions.

CONCLUSIONS

The most obvious conclusion to be drawn from these observations is that there are definitely instances in which growth twinning is not accidental. The important point is that the twinned configuration may be more favorable for growth in a system in which the rate of supply of ions to the crystal is relatively slow in comparison to the rate at which these ions will be incorporated in the growing crystal.

That facial orientation is the sole cause of growth twinning (or the sole reason for the success of a twin, once initiated by accident) is not implied. Very possibly certain twins might be favored by totally different kinetic mechanisms; such as high rate of generation of screw disolcations on the composition plane as the result of the torsion imposed on the two crystals across this plane. Some growth twins, on the other hand, might not be especially kinetically favored at all, and their occurrence might be simply a matter of chance. The experiments of Löffler (1934) on NaCl crystals are a case in point. She found that the incidence of twins could be raised to approximately half by the addition of an optimum amount of $MnCl_2$ to the solution. Her explanation was that the twin (a typical fluorite-type twin with cube faces) resulted from a stacking fault in the close-packed planes parallel to (111). If the stacking sequence became

1 2 3 1 2 3 1 2 3 2 1 3 2 1 3 2 1, a twin would occur. Two, or any even number of stacking faults formed early in the crystal's growth would cancel each other out, and the twin would disappear. Löffler found no situation in which all or even the majority of the crystals would twin, and it appears that twinning of these crystals offered no competitive advantage to their growth.

The previous theories relating growth twins solely to accidents miss the point in many cases. Observations that certain species of crystals are virtually invariably growth twinned, that certain physical conditions of formation favor growth twinning in other cases, and that the twins are commonly larger than untwinned crystals with which they occur suggest some sort of advantage imparted to the growing crystal by the act of twinning. In a few cases, this twinning can be shown to be invariable and can be made to form as overgrowths on essentially infinitely large nuclei; the inferred success of the twin is here explained by the spatial relationship between the faces which it produces. We should stop asking the question, "How can twins possibly form?", and ask the question, "Under what conditions should (or even *must*) twins form?"

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