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Crystal Habit of Synthetic Chalcanthite (Copper Sulfate Pentahydrate) as Related to Position and Orientation in Growth Solution

EDITH CHASEN,

Department of Geology and Geography, Hunter College
of The City University of New York, New York, New York 10021

AND C. W. WOLFE

Department of Geology, Boston University,
Boston, Massachusetts 02215

Abstract

The habit of a crystal of $CuSO_4 \cdot 5H_2O$ can be controlled during growth under refrigeration by varying the depth of the crystal in the host solution, its crystallographic orientation during growth, and the maximum size to which it is allowed to grow.

Crystals were grown in solution 30 mm deep. Those which were grown at the bottom of these solutions rarely produced the form[1] {100}; where that form did occur, it was little more than a line face. Seeds which were set 18 mm up from the bottom yielded crystals with both {100} and {010} forms which were roughly equivalent in size. Those crystals which grew just below the surface produced {100} forms but no {010} forms, the opposite of crystals which developed on the bottom. $\{\bar{1}11\}$ was usually predominant among the terminal forms, but when crystals grew to sizes greater than 0.82 cm^3 in size, at the 2 mm and 18 mm level of the solutions, $\{0\bar{2}1\}$ and $\{\bar{1}21\}$ were almost as large as $\{\bar{1}11\}$. Crystals which were grown to intermediate size at all levels in the solutions developed a new form, $\{\bar{2}11\}$, when either the edge between (010) and $(\bar{1}21)$, or that between (010) and (021) was set uppermost and parallel to the solution surface during growth. This new form generally was equal in size to the $\{\bar{1}21\}$ form. The experimentation demonstrates that as crystals grow, the number of forms diminish in favor of those forms with minimal summation of indices.

Introduction

Wolfe (1960) has suggested that crystal habit is influenced not only by such factors as crystal structure, impurities included during growth, and variations in the nature of the growth medium, but also by crystallographic orientation during growth and by crystal location in the growth medium. To test this premise, crystals of $CuSO_4 \cdot 5H_2O$ were grown under refrigeration conditions, where positive controls on orientation and location in solutions could be maintained. Copper sulfate pentahydrate was chosen for the experimentation partly because crystals of that composition can be grown so readily by the refrigeration technique and partly because the experience of Tutton (1922) as well as our own had indicated that variations in its crystal habit were frequent, notable, and apparently almost random.

Experimental Method

The refrigeration method of crystallization is based on the fact that most salt solutions which are saturated at room temperatures will be supersaturated in a refrigerator. A crystal, *e.g.,* seed, will neither grow nor dissolve if placed in a solution which is saturated for refrigerator conditions and capped to eliminate evaporation. If drops from a room-temperature saturated solution are introduced into the refrigerator solution, precipitation will occur, preferably on any seed crystal already in the refrigerator solution. The seed crystal is usually placed on a small raised platform or pedestal in order to keep other seeds, which might form nearby, from interfering with the perfection of growth of the control seed crystal. The seed crystal is thus being fed at such a rate that it can take on additional ions sufficiently slowly to develop a homogeneous crystal. In order to keep the crystal from growing in only one direction and from growing around the pedestal, the crystal was turned over each

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[1] Form indices are referred to the conventional setting (Palache, Berman, and Frondel, 1951).

day on the pedestal to expose all surfaces to the precipitating ions in the solution.

Preparation of Seeds

In the research described herein, a stock solution was prepared on the basis of the solubility curve of $CuSO_4 \cdot 5H_2O$ in water. The most satisfactory solution proved to be 100 g of water and 16 g of the salt; such a solution is unsaturated at room temperature (26–27°C) but is supersaturated in the refrigerator (6–7°C). To obtain seeds, 30 ml of stock solution in 150 ml beakers, covered with watch glasses, were placed in the refrigerator. Before there was wholesale precipitation, the solutions were decanted into other beakers which had been sitting in the refrigerator. Crystallization proceeded in these beakers, and another decanting took place after many seeds had formed. This process was continued until the host solution came to equilibrium with refrigerator conditions, and no more seeding or crystal growth was taking place.

Orientation of Crystals on Pedestals

In order to test the hypothesis that crystal habit is dependent upon the orientation of the crystal during growth, seeds were selected on the basis of freedom from flaws and on the degree of geometrical perfection. Many more crystals were measured than were subsequently used for growth seeds. Axonometric projections (Wolfe, 1953) of the crystals were made in order to have a rigorous picture of the starting habit of each crystal. Careful two circle goniometric measurements of the *phi* and *rho* angles[2] were made and recorded, and the indices for each face were determined in the Dana setting (Palache, Berman, and Frondel, 1944). With these data available, it was possible to orient the seed in the host solution for growth in any preselected orientation. The following orientations were finally adopted as the most useful and readily obtained:

1. The edge between faces (100) and (110) was set uppermost and parallel to the surface of the solution.
2. The edge between faces (010) and (021) was set as above.
3. The edge between faces (010) and ($\bar{1}21$) was set as above.
4. The *c* axis was oriented perpendicular to the solution surface.
5. The *b* axis was oriented perpendicular to the solution surface.

Depth Placement of Seeds

To achieve the location at various depths small pedestals were contrived. After disappointing trials with red universal wax, we finally settled upon plexiglass pedestals. This material is easily shaped, and slots of various sizes can be cut in it to keep the crystals in any chosen orientation as they are growing. Pedestal heights were cut at 2 mm, 18 mm, and 28 mm.

Growth Procedures

The saturated (at about 6.6°C) host solutions in the refrigerator had a depth of 30 mm in 150 ml beakers. Seeds were placed on plexiglass pedestals at the three depths in the five previously referred to orientations. Several crystals were grown for each of the fifteen conditions listed.

Crystal growth was induced by introducing drops of the room-temperature saturated solution with a medicine dropper three times a day. Care was taken not to overfeed the host solution because the crystals which developed under too rapid feeding were irregular in form, rough, and included some of the mother liquor. The faces on such crystals did not yield good goniometric measurements. Too slow a feeding, on the other hand, produced impractically slow growth. Consideration of these factors and observation of growth rates led to an optimum rate of $\frac{1}{4}$ ml of source solution per feeding.

The crystals were turned over on the pedestals once a day to expose all surfaces uniformly to the source of precipitating ions. Occasionally, additional seeds formed on the growing crystals, although this effect was minimized by use of the pedestals. The randomly oriented and undesired seeds were scraped off manually before becoming embedded when the crystals were being turned.

Final Crystal Sizes and Habits

It was not possible to maintain a steady rate of crystal growth. Therefore, the crystals were simply fed daily until they reached the desired size, at which time they were measured, drawn, and photographed. The ultimate sizes selected were somewhat arbitrary, but they fell into three size ranges. The starting seeds weighed between 0.2 and 0.8 g, with a maximum volume of 1.4 cm^3. Crystals were considered large

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[2] Azimuth and polar distance of a face normal are called *phi* and *rho*, respectively (Wolfe, 1941).

for this work when they weighed between 3.4 and 4.4 g, with a maximum volume of 2 cm^3. The same crystals at various stages of growth and in various orientations were measured on a two circle goniometer and checked for habit development.

A total of 46 crystals were grown on which control measurements were made. Three variables existed in the depth in the solution, five different orientations were used, and three different ultimate sizes were attained. These variables yield 45 possible combinations. Since the same crystals were used to produce different sizes, fifteen crystals would have embraced all of the variables. Thus, the 46 crystals which were grown and measured should be sufficiently great to prove of statistical importance. The experiments extended over a period of 12 months, although single crystals reached maximum growth in about three and a half weeks.

A crystal drawing was prepared for each possible combination of variables. These were consolidated into a few drawings of representative habits which developed for many crystals grown to a particular size, at a certain level, and with a particular orientation. These composite drawings are representative of what could be expected if the experiment were to be duplicated. Small variations from the general habit, of course, are likely. Ten basic habits are shown in Figures 1 and 2. The orientation of all of the drawings, unless otherwise noted, is with the positive end of the *c* axis upward, the normal to face (010) to the right and the normal to face (100) quasiperpendicular to the paper. The face symbols employed on Figures 1 and 2 are those listed in the angle table for chalcanthite in Palache, Berman, and Frondel (1951), an abridged form of which is given here in Table 1 for convenience of reference.

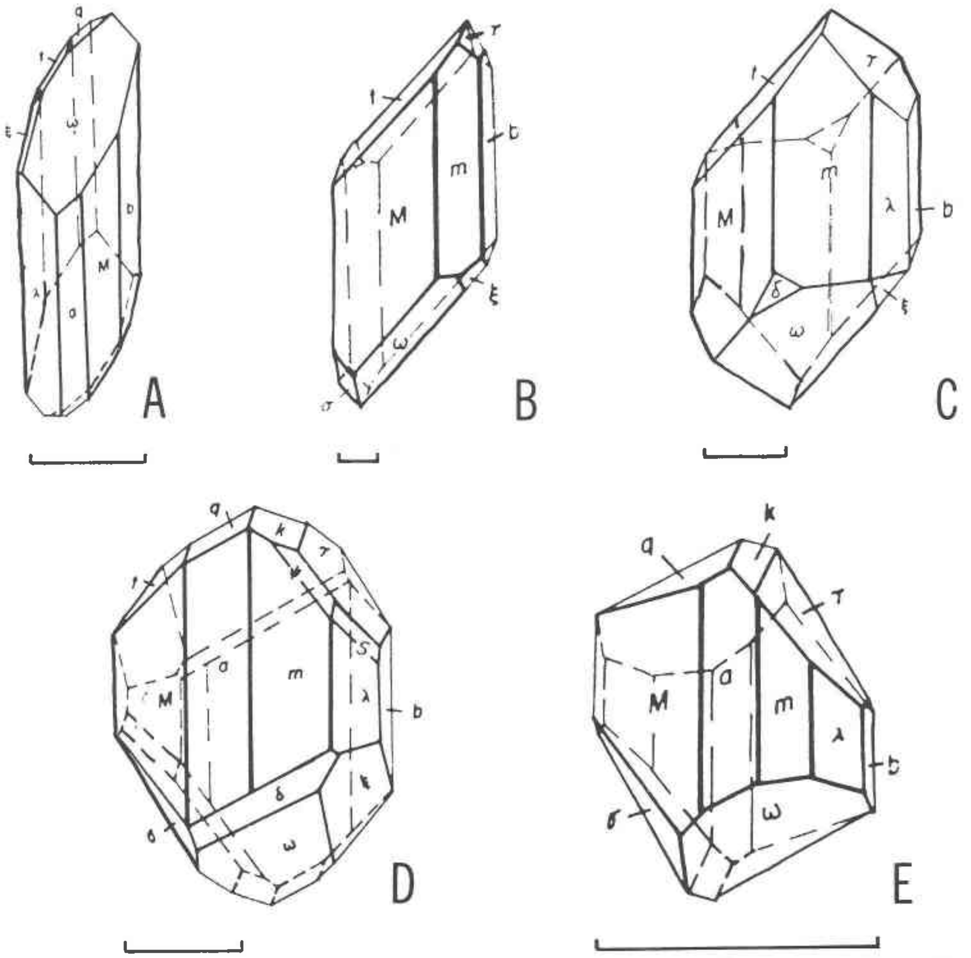

FIG. 1. Crystal drawings of Habits A–E. Habit A is oriented with [00$\bar{1}$] axis up, (0$\bar{1}$0) to the right and (100) out from paper. All bar scales represent 5 mm.

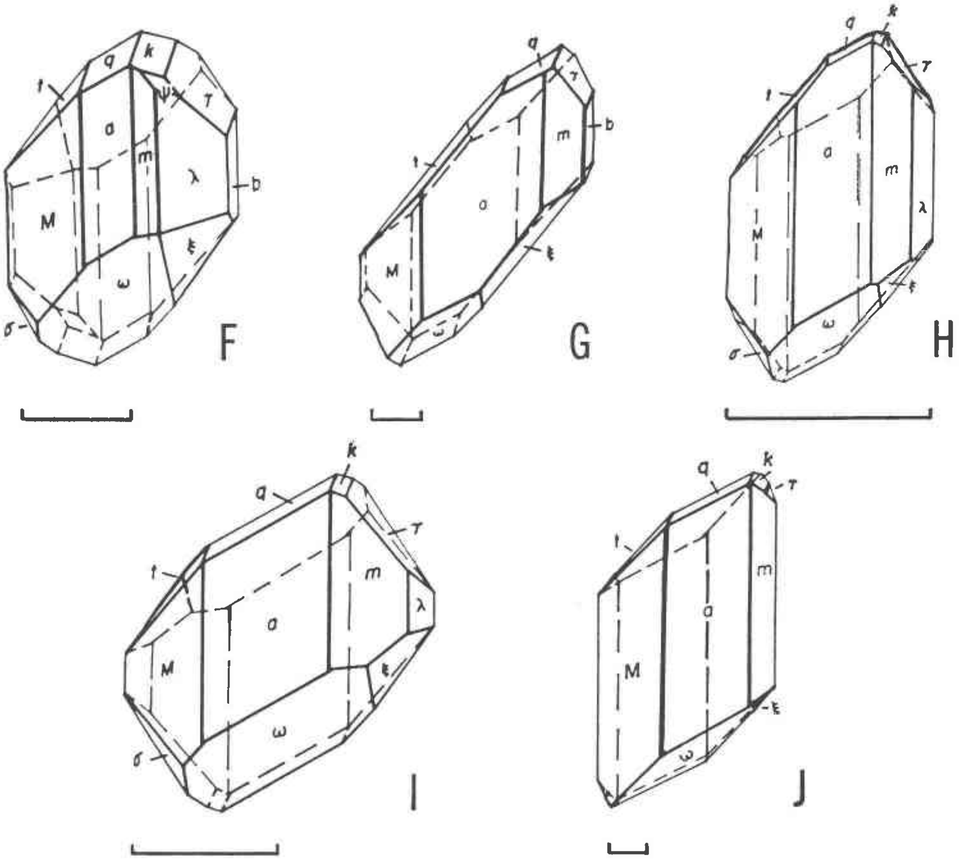

FIG. 2. Crystal drawings of Habits F–J. All bar scales represent 5 mm.

Results

In addition to those forms listed in the Dana System angle table, one new form {$\overline{21}1$} is included here. This form occurred on all of our intermediate sized crystals at all levels in the solution when the edge between (010) and (021) or that between (010) and ($\bar{1}21$) was at the top of the crystal and oriented parallel to the surface of the solution. The new form was usually equal in size to {$\overline{12}1$} (see Figure 1, habits C and D). All of the forms listed in the angle table were observed in this study. The ten distinctive habits that were obtained by varying seed orientation, depth in the solution, and size to which the crystals grew are tabulated in Tables 2 and 3 and are shown in Figures 1 and 2.

TABLE 1. Angle Table of Forms Obtained*

Forms Letters	Forms Indices	ϕ	ρ	A	B	C
b	010	0°00	90°00	100°54.5	--------	85°45.5
a	100	100 54.5	90 00	--------	100°54.5	73 46.5
π	130**	33 54	90 00	67 01	33 54	76 49
λ	120	47 08.5	90 00	53 46.5	47 08.5	74 22.5
m	110	69 49	90 00	31 05.5	69 49	72 11
M	$\bar{1}10$	126 59	90 00	26 04.5	126 59	78 48.5
k	011	25 45	35 51	81 22.5	58 10	27 35.5
τ	021	14 23	51 38	87 16.5	40 35	45 10.5
q	$0\bar{1}1$	147 38	30 23	69 45.5	115 17.5	29 32
t	$0\bar{2}1$	163 37.5	48 04	70 03.5	135 32.5	49 47.5
ω	$\overline{11}1$	-112 49	37 33	120 27.5	103 40	55 19
ψ	131	39 56.5	64 20.5	64 03.5	46 17	50 30
S	141	31 36	68 35.5	70 48	37 32.5	57 01.5
r	$1\bar{2}1$	133 26	61 29	42 11.5	127 10	53 07
ξ	$\overline{12}1$	-140 52.5	48 26.5	110 43	125 29	63 26.5
ζ	$\overline{13}1$**	-153 52	58 07.5	102 53	139 40.5	70 28.5
δ	$\overline{21}1$***	-93 19	59 56	147 05	87 08	77 51

* *Chalcanthite, $CuSO_4 \cdot 5H_2O$, is triclinic, pinacoidal $\bar{1}$, with a:b:c = 0.5715:1:0.5575; α = 97°44', β = 107°26', γ = 77°20'; $p_o:q_o:r_o$ = 0.9907:0.5452:1; λ = 85°45.5'; μ = 76°46.5'; ν = 100°54.5'; p_o'1.0413, q_o'0.5730, x_o'0.3139, y_o'0.0778. Elements from Palache, Berman, and Frondel (1944).*

** *Indicates this form not present on Figures 1 and 2 due to poor quality or rare occurrence.*

*** *New form.*

Tables 2 and 3 and the figures reveal some major variations in habits as functions of depth of immersion, orientation, and size to which the crystals were allowed to grow. Habits A, E, and H are those of small crystals having, respectively, eight, ten, and eleven forms. Habits C, D, and F are intermediate sized crystals with nine, fifteen, and thirteen forms. Habits B, G, I, and J are for the largest crystals grown in these experiments and have eight, nine, ten, and nine forms. The average number of forms for crystals less than one gram in size was just under ten; for those crystals which were between 1.9 and

3.2 g the average number of forms was twelve, while for crystals between 3.4 and 4.4 g, the average number of forms was nine. These empirical results are in close agreement with natural crystals of different mineral species. Very small crystals and very large crystals usually show few forms, but intermediate crystals usually have a multiplicity of forms. It is interesting to note that the new form $\{\bar{2}11\}$ occurred only on these crystals of intermediate size.

The dominant form among the $\{hk0\}$ forms is usually M $\{\bar{1}\bar{1}0\}$, but the importance of {100}, {010}, and {120} varies greatly. {100} does not occur on two of the three habits obtained at 2 mm depth; {010} does not occur on habits obtained at 28 mm depth; {110} does not occur on habit A, and {120} does not occur on habits B, G, and J.

Among the terminal forms $\{\bar{1}11\}$ is usually the dominant form, although in habits B, C, and F, $\{0\bar{2}1\}$ or $\{\bar{1}21\}$ are practically equal to it in size; and in habit G, $\{\bar{1}21\}$ is larger than $\{\bar{1}11\}$. The size importance of other forms varies from habit to habit, as can be noted from Figures 1 and 2 and from Tables 2 and 3.

Of particular interest is the occurrence of $\{\bar{2}11\}$, a form not reported in the literature before. Its angles were in close conformity with its calculated theoretical position. The form was developed on crystals grown at all depths where the orientation during growth was with either the edge (010)–(021) or the edge (010)–($\bar{1}$21) up and parallel to the surface of the solution. The striking conformity between observed and calculated angles and the frequency of this form is remarkable, since the form had never before been recognized. The best explanation for such lack of observation in the past must be that during previous experimentation the crystals of synthetic chalcanthite were never positioned in the attitude which would encourage the development of this form; or, perhaps, they were never examined upon reaching the intermediate size.

TABLE 2. Variations in Growth with Orientation, Depth in Solution, and Size to Which Crystal Was Grown

Habit symbol	Relationships* of face sizes in zone [001]	Relationships* of face sizes for terminal faces	Orientation of crystals on pedestals	Depth of crystal in solution 30mm deep	Crystal size in grams
A	$b > a$ no m $\lambda = M$	large $\xi > t > q$	Edge (100)-(110) up and parallel to solution surface	2 mm above bottom	0.2-0.8
B	no a $m < M$ no λ	$\sigma = \tau = \xi$, all small but equal in size $\omega = t$	Same as for A	2 mm above bottom	3.4-4.4
C	no a $m = M$ $\lambda < m$	t and ω large and larger than $\delta = \xi$	Edge between (010) and ($0\bar{2}1$) or ($1\bar{2}1$) up and parallel to solution surface	2 mm above bottom	1.9-3.2
D	$a > b$ $m = M$ $\lambda < m$	ω large $q = t$, both $> k$ $\tau > q$ and t $\xi = \delta$	Same as for C	18 mm above bottom	1.9-3.2
E	$a = b = m = \lambda$ M much greater than other {hk0}	ω large $\sigma = \tau$ $\tau > q > k$	Same as for A	18 mm above bottom	0.2-0.8
F	$a > b$ $m < M$ $\lambda > m$	ω almost equal to ξ $\sigma = \psi$ $q = k$, $t = \tau$	+ c up	18 mm above bottom	1.9-3.2
G	$a > b$ $m < M$ no λ	$\omega < \xi$ $\xi = t$ $q = \tau$	+ b up	18 mm above bottom	3.4-4.4
H	no b $m < M$ $\lambda < m$	ω large $t > \tau > q > k$ $\sigma = \xi$	+ b up	28 mm above bottom	0.2-0.8
I	no b $m = M$ $\lambda < m$	ω large $t = q$ $\sigma = \xi$ $\tau > t$	+ c up	28 mm above bottom	3.4-4.4
J	no b $m < M$ no λ	ω large $t = q$ $\xi = \tau$ $\tau < k$	Same as for A	28 mm above bottom	3.4-4.4

* Roman and Greek letters are face symbols given in Table 1 and Figures 1 and 2.

Discussion

The appearance of the new form is a strong argument in favor of the conclusion that habit is a function of crystal orientation, during growth, relative to

TABLE 3. Habits Developed by Crystals with Different Orientations, Depths, and Sizes*

mm	Edge (100)-(110) up and parallel to surface			Edge (010)-(021) up and parallel to surface			Edge (010)-($\bar{1}$21) up and parallel to surface			c axis vertical			b axis vertical		
	Height in mm above bottom														
28	E	J	J	E	D	J	D	C	C	F	F	I	H	I	I
18	E	J,E	J	D	D	C	D	D	C	F	F	I	H	H	G
2	A	E	B	D	C	C	D	C	C	H	F	I	H	H	I
	Weight in grams														
	0.2	1.9	3.4	0.2	1.9	3.4	0.2	1.9	3.4	0.2	1.9	3.4	0.2	1.9	3.4
	0.8	3.2	4.4	0.8	3.2	4.4	0.8	3.2	4.4	0.8	3.2	4.4	0.8	3.2	4.4

* See Figures 1 and 2.

TABLE 4. Reciprocal Lattice Vectors of Chalcanthite Forms

Theoretical Importance	Form	Reciprocal Lattice Vectors	Theoretical Importance	Form	Reciprocal Lattice Vectors
1	010	.5452	11	$\bar{1}\bar{1}1$	1.3598
2	100	.9907	51	112	2.5984
3	001	1.0000	43	$1\bar{1}2$	2.4536
8	110	1.2179	28	$\bar{1}12$	2.0318
4	$1\bar{1}0$	1.0364	30	$\bar{1}\bar{1}2$	2.0529
17	120	1.6060	32	121	2.0722
10	$1\bar{2}0$	1.3271	21	$1\bar{2}1$	1.7759
31	130	2.0663	14	$\bar{1}21$	1.5391
19	$1\bar{3}0$	1.7444	18	$\bar{1}\bar{2}1$	1.6925
49	140	2.5602	58	212	3.3178
36	210	2.1523	57	$2\bar{1}2$	3.1412
54	230	2.7978	41	$\bar{2}12$	2.4002
6	011	1.1738	47	$\bar{2}\bar{1}2$	2.5013
5	$0\bar{1}1$	1.1029	55	122	2.8300
13	021	1.5329	48	$1\bar{2}2$	2.5585
12	$0\bar{2}1$	1.4239	37	$\bar{1}22$	2.2308
34	012	2.1115	38	$\bar{1}\bar{2}2$	2.2693
29	$0\bar{1}2$	2.0337	45	131	2.4628
27	031	1.9790	23	$\bar{1}31$	1.9316
22	$0\bar{3}1$	1.8528	35	$\bar{1}\bar{3}1$	2.1153
39	$0\bar{4}1$	2.3308	56	141	2.9036
15	101	1.5922	50	$\bar{1}41$	2.5843
7	$\bar{1}01$	1.1949	52	211	2.6115
44	201	2.4563	42	$2\bar{1}1$	2.4168
25	$\bar{2}01$	1.9542	24	$\bar{2}11$	1.9462
46	102	2.4675	33	$\bar{2}\bar{1}1$	2.1082
26	$\bar{1}02$	1.9683	40	$\bar{2}\bar{2}1$	2.3800
20	111	1.7656	53	$\bar{2}\bar{3}1$	2.7347
16	$1\bar{1}1$	1.5959	--	---	------
9	$\bar{1}11$	1.2654	--	---	------

the major source of ions. One of the well known observations of crystal synthesis, which has been strikingly confirmed in this work, is that as crystals grow larger, small faces disappear in favor of larger ones. As ions are deposited on any crystal face, that face is extended further from the crystal center; and if the starting size is maintained for that face, it becomes relatively smaller in comparison with the other faces on which no ions precipitate. In practice, continued precipitation of ions on small crystal faces ultimately results in the disappearance of the small face from the growing crystal. Small faces receive more ions because the energy of crystallization can be released most readily on faces which approach a point in dimensions; that is, small faces are better heat sinks than larger ones simply because of their geometry, and precipitation of ions on small faces grows them out of existence.

The Law of Haüy, which states, in effect, that the importance of a form is directly related to the sum of its indices, does not differentiate between various possible forms with the same sum of indices. Bravais suggested and G. Friedel demonstrated that the importance of a form is directly related to its reticular density (or its interplanar spacing). In the triclinic system we do not have to be concerned with the effects of space group symmetries on the spacings of planes (Donnay and Harker, 1937). In the triclinic system, in particular, it is easier to consider the spacing of planes in terms of their reciprocal lattice vectors.

If the importance of a form, $\{hkl\}$, is directly proportional to its interplanar spacing [usually indicated as $d(hkl)$], it is inversely proportional to the length of the reciprocal lattice vector $\mathbf{L}^*[hkl]$. A test of the validity of this approach to the importance of forms in terms of size and frequency of occurrence can well be made here in this triclinic substance.

We shall use as the three basic vectors in the reciprocal lattice the values for p_0, q_0, and r_0, which are listed in Table 1. The three basic reciprocal lattice angles are λ, μ, ν, also listed in Table 1. Since the approach here is morphological, it suffices to use the reciprocal (polar) lattice constants derived from goniometric measurements, rather than the X-ray structural ones. The general formula for the square of the length of the reciprocal lattice vector for any form $\{hkl\}$ is:

$$|\mathbf{L}^*(hkl)|^2 = h^2 p_0{}^2 + k^2 q_0{}^2 + l^2 r_0{}^2$$
$$+ 2q_0 r_0 kl \cos\lambda + 2r_0 p_0 lh \cos\mu + 2p_0 q_0 hk \cos\nu .$$

The fact that $r_0 = 1$ simplifies the calculation. The values for the calculated lattice vectors for various permutations of indices between 0 and 2 (with a few more complicated ones added) are given in Table 4. The computed theoretical importance of each form, based on the magnitude of its reciprocal lattice vector, is also given for the 58 forms listed. The importance of a form, from a geometrical point of view, should be inversely proportional to its reciprocal lattice vector.

Table 5 indicates the observed importance of the forms which developed on the ten habits shown in Figures 1 and 2. Under each of the habits (*Habits* A-J columns, Table 5), the *size importance* from 1,

TABLE 5. Size and Frequency Rating of Forms

Size Rating	Form	A	B	C	D	E	F	G	H	I	J	Frequency Rating	Importance Rating $\frac{\text{Size Rating}}{\text{Frequency Rating}}$
5.4	b 010	1	4	6	4	8	7	6	-	-	-	0.6	9.0
2.25	a 100	5	-	-	3	2	3	1	1	1	2	0.8	2.8
3.2	m 100	-	2	2	2	5	5	3	3	4	3	0.8	4.0
1.7	M $1\bar{1}0$	4	1	1	1	1	1	2	2	3	1	1.0	1.7
5.0	λ 120	3	-	4	7	7	4	-	5	-	-	0.5	10.0
7.75	q $0\bar{1}1$	6	-	-	11	6	9	9	10	6	5	0.8	9.7
10.3	k 011	-	-	-	13	10	11	-	11	10	7	0.6	17.2
7.1	t $0\bar{2}1$	7	5	5	12	-	10	4	6	9	6	0.9	7.9
7.0	τ 021	-	8	7	8	4	8	8	7	5	8	0.9	7.8
3.5	ω $\bar{1}\bar{1}1$	2	3	3	5	3	2	7	4	2	4	1.0	3.5
7.2	ξ $\bar{1}\bar{2}1$	8	7	8	6	-	6	5	9	7	9	0.9	8.0
9.0	σ $\bar{1}21$	-	6	-	10	9	13	-	8	8	-	0.6	15.0
9.0	δ $\bar{2}\bar{1}1$	-	-	9	9	-	-	-	-	-	-	0.2	45.0
13.0	ψ 131	-	-	-	14	-	12	-	-	-	-	0.2	65.0
15.0	S 141	-	-	-	15	-	-	-	-	-	-	0.1	150.0

largest, to 15, smallest, is indicated for each form in relationship to the other forms occurring on that particular habit. The size importance is simply an arithmetical rating based on the size of the form as compared with the others on a particular habit. The largest face is rated 1, second largest 2, and so on. The column labelled *Size Rating* is determined by the summation of the size importances for each form in the various habits, divided by the number of habits on which the form appears. On this basis, the size rating of the lowest magnitude represents the largest form occurring on the crystals with habits that exhibit that form. The *Frequency Rating* simply indicates the ratio of the number of occurrences of the form as seen on the ten habits. Thus, a form which is observed eight times out of ten habits has a frequency rating of 0.8. The *Importance Rating* of a form is inversely proportional to its Size Rating relative to its Frequency Rating, as shown in the last column of Table 5.

Table 6 gives the sequential importance rating of the forms noted on the various habits, with the lowest importance rating (for $\{1\bar{1}0\}$) from Table 5 set equal to one. These ratings are compared with the expected importance based on reciprocal lattice vectors, from Table 4.

No general agreement exists. Of particular interest is the omission from the form list of $\{001\}$, theoretical importance 3, and $\{\bar{1}01\}$, theoretical importance 7. Otherwise, it is rather impressive that of the forms which do occur among the first twelve, all are found within the first eighteen on the theoretical list. Since secondary faces usually develop on the edges between dominant faces, the absence of $\{001\}$ as a primary form (third most expectable form, theoretically) makes the lack of development of $\{\bar{1}01\}$ (seventh most expectable form, theoretically) somewhat understandable. One of the factors which is impossible to control in the experiments is the rather fortuitous nature of the form development of the initial seed which was employed. If the seeds could have formed in an isotropic environment in midsolution, rather than on the bottoms of the beakers, a closer approximation to the theoretical importance of forms might have been obtained in practice.

It is to be expected that the larger the crystals grow, the closer would their form development approach the theoretical importance based on reciprocal lattice vectors, since the influence of the initial seed habit would be in part vitiated. From Tables 2 and 3 we recognize that habits B, G, I, and J were those of crystals which were grown to largest size in these experiments. On these habits, not only is the number of faces relatively reduced, but there is a somewhat better agreement between theoretical and observed importance of forms. The reduced size or absence of $\{010\}$ is not expectable, however.

The data, at present, do not warrant a more detailed analysis of the discrepancies between observed and theoretical importance of forms; these discrepancies probably are attributable to crystal

TABLE 6. Comparison of Observed Importance Rating to Theoretical Reciprocal-Lattice-Vector Rating

Importance Rating $\{hkl\}$ / Importance Rating $\{1\bar{1}0\}$		Form	Predicted importance based on reciprocal vectors (From Table 4)
1	1	$1\bar{1}0$	4
2	1.6	100	2
3	2.1	$\bar{1}11$	11
4	2.4	110	8
5	4.6	021	13
6	4.7	$0\bar{2}1$	12
7	4.8	$\bar{1}21$	18
8	5.3	010	1
9	5.7	$0\bar{1}1$	5
10	5.9	120	17
11	8.8	$\bar{1}21$	14
12	10.1	011	6
13	26	$\bar{2}11$	33
14	38	131	45
15	88	141	56

structural factors rather than to the simple morphological approach. Donnay (1972) suggested that in some cases it might be well to use a different basic morphological cell from that of the structure. As an anonymous referee put it: "The morphological lattice (instead of being recalculated as a new primitive one) may be described as *the X-ray lattice centered on the C face*. . . . the fact that the Cu atoms lie on this *C* lattice, which is thus both the rigorous morphological lattice and a pseudo-lattice for the structure" produces a multiple cell which, if used for the calculation of reciprocal lattice vectors, would produce even closer agreement between theoretical and actual importance of the various forms. In general, the external geometry can be expected to reveal the internal relationships, and the present work indicates a special structural relationship which controls form development.

It should be mentioned that the new form observed in this study, {$\overline{21}1$}, is 33rd in expectability out of the 58 forms listed in Table 4, which means that its actual occurrence is consistent with its theoretical probability.

Conclusions and Implications

Crystal habit is at least partially determined by attitude of the seed during growth and to proximity of source ions for growth. Habit, also in part, can be predicted on the basis of lattice considerations; the frequency and size of a form (form importance) is inversely proportional to the reciprocal lattice vectors implicit in the coordinates indicated by form indices. Structural factors connected with bonding strengths in differing directions in the lattice will control, in large measure, the final habit attained. The extent to which habits vary from predicted development is a direct indication of the deviation of the geometry of a lattice from coincidence with bonding strengths in differing directions. Other controls on habits, which were not considered in this paper, would, of course, be variables in the physical-chemical environment, such as pH, foreign ions, and rate of feeding.

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