A NEW LAW OF CRYSTAL MORPHOLOGY
EXTENDING THE LAW OF BRAVAIS

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CONTENTS

THE LAW OF BRAVAIS

This law, proposed by Bravais on speculative grounds, only received general acceptance after Georges Friedel (1904, 1905, 1907) proved it to be substantiated by observation. With certain restrictions the law states that, for any crystalline species, a translation lattice can be found such that: (1) The observed crystal faces are parallel to the net planes with the highest reticular densities (or smallest mesh areas); (2) the greater the reticular density (or the smaller the mesh area) the more important the corresponding form.

The importance of a form is estimated from its size, frequency of occurrence, and presence as a cleavage form. The *reticular density* of a net plane hkl is defined as the number of lattice points (nodes) per unit surface. It is inversely proportional to the *reticular area*, or area S of the smallest mesh in the net, and directly proportional to the *interplanar distance d*, since $S \cdot d = V$, where V, the volume of the smallest unit cell, is a constant for any lattice.

Morphologists of the French School have advocated the acceptance of the lattice indicated by the Law of Bravais, in preference to any other lattice that would also give the observed forms reasonably simple Miller indices. The lattice chosen on this basis has been called the "Haüy-Bravais lattice" or "morphological lattice," in contradistinction to the

"structural lattice" or true translation lattice, determined by x-ray technique. This nomenclature stresses the baffling fact that the two lattices are not always geometrically similar.

Known objections and restrictions to the Law of Bravais include the following:

(1) The forms present on a crystal do not depend only on factors within the crystal, but on external factors as well (conditions of crystallization). A statistical survey of morphological data (locality persistence, in Niggli's sense) usually suffices to detect the influence of external causes, which can then be left out of consideration.

(2) Even in the most favorable instances the law holds for dominant forms only. Minor forms are always irregularly scattered down the list of decreasing theoretical importance.

(3) Frequently the best lattice that can be arrived at leads to an order of importance not wholly satisfactory for the dominant forms and it may even assign small mesh areas to unknown or insignificant forms.

(4) Cleavage forms do not always coincide with the largest and most frequent crystal forms, and conversely.

(5) Complementary merohedral forms have the same mesh area, yet do not always show equal morphological importance.

These objections have given the Law of Bravais the status of a first approximation. G. Friedel, who conceded this point, foresaw an explanation for the encountered anomalies in the "perturbing influence" of the *motif* (the contents of the unit cell).

In this paper we propose to state a law[1] that takes into account one of the important properties of the motif—its symmetry.

SPACE GROUP SYMMETRY

The 230 space groups were derived by combining, in all possible ways, the different symmetry operations: translation, rotation, and reflection (or inversion). This process involved, among others, two composite symmetry elements, which had not been previously recognized (although they are actually implied in some of the 14 Bravais lattices), namely: the screw axis (Sohncke) and the glide plane (Fedorov, Schoenflies, Barlow).

In this paper we shall not concern ourselves with those screw axes and glide planes that are inherent in the translation lattice,[2] since their ef-

[1] A preliminary communication was presented at the January meeting of the *Académie des Sciences* in Paris (1937).

[2] Glide planes and screw axes occur in all the centered modes of all lattices, and also in the hexahedral (primitive) mode of the cubic, tetragonal, and hexagonal lattices (we consider the rhombohedral lattice a mode of the hexagonal). The only lattices devoid of

fect is already taken into account by the Law of Bravais. We shall deal only with screw axes and glide planes that can be combined with a translation lattice, in addition to those it may already possess. The distinction between the two kinds of screw axes or glide planes is made in the Hermann-Mauguin notation: those that exist in the lattice itself are implicitly expressed by the Roman capital indicative of the translation group, the others are explicitly symbolized. The effect of the former is to bring a node into coincidence with another node (which could also be reached by applying a translation to, and which is in parallel orientation with, the first). The effect of the latter is to produce additional points within the lattice, points, however, which are not lattice points (nodes) since they cannot be obtained from any node by translation alone, but only by a translation accompanied by either a rotation or a reflection. Additional points produced by a screw axis are congruent to the true lattice points (nodes), although not in parallel orientation with them; additional points produced by a glide plane are enantiomorphous to lattice nodes. For convenience, in case there are additional points resulting from the action of screw axes and glide planes, we shall term "lattice equipoints" the nodes of the Bravais lattice and "space group equipoints" the additional points.

At this juncture one should recall that a node of a Bravais lattice stands for a whole motif (the contents of a complete unit cell, regardless of how matter may be distributed in it) and that the symmetry of the motif cannot exceed that of the parallelopiped outlined by the cell (holohedry), but may be inferior to it (merohedry). When screw axes and glide planes are present the motif (still represented by the "node") may be thought of as split into several fractions or "submotifs" derived from each other by the available screw rotations and glide reflections; any of these submotifs may be represented by the "lattice equipoint," the others are represented by "space group equipoints."

The Law of Bravais defines reticular density in terms of nodes only. It is our purpose to show that, when a node is replaced by a "lattice equipoint" and one or more "space group equipoints,"the reticular density must be defined in terms of "equipoints" for certain faces: the faces perpendicular to a screw axis and some of the faces perpendicular to a glide plane. Inasmuch as the faces affected in this way are determined by the kind and amount of space group symmetry, they become, con-

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inherent screw axes and glide planes are the triclinic (P), the hexahedral monoclinic (P), and the rectangular hexahedral orthorhombic (P). The figures in the *International Tables for the Determination of Crystal Structures* (Chapter V, Space groups) illustrate the presence of screw axes and glide planes in the lattices mentioned.

versely, criteria indicative of such symmetry. Hence, if the symmetry of the space group is known, its influence on the form development can be predicted and checked; *vice versa* it is possible to ascertain with which space group (or set of space groups) observed morphological features are compatible.

THE EFFECT OF A SCREW AXIS

A screw axis, represented by the symbol p_q in the Hermann-Mauguin notation, is a p-fold axis of symmetry, the operation of which is the product of a rotation through $2\pi/p$ and a displacement by a fraction q/p of the shortest translation parallel to the axis. The displacement is known as the *screw component*. The crystallographically possible screw axes are: 2_1; $(3_1, 3_2)$; $(4_1, 4_3)$; 4_2; $(6_1, 6_5)$; $(6_2, 6_4)$; 6_3. Enantiomorphous screw axes, here collected between parenthesis, may be considered as having the same screw component but opposite screw senses. The fraction q/p, therefore, will always be of the form $1/n$, with $n = 2, 3, 4$, or 6.

Consider any two successive lattice planes perpendicular to a screw axis. Let d be their spacing. These two lattice planes will be interleaved by $(n-1)$ other planes brought about by the action of the screw axis, thus resulting in a system of parallel, equidistant planes with the spacing d/n. The lattice planes contain "lattice equipoints" only, the intervening planes "space group equipoints" only. The arrangement of equipoints in any two successive planes of the complete system is the same except for the $2\pi/p$ rotation. All the planes of the system will, therefore, be geometrically equivalent insofar as concerns the crystal form (pinacoid) that parallels them. For such a pinacoid, the faces of which are perpendicular to a screw axis, the "effective" spacing is d/n instead of d. Formally, by virtue of the relations between spacing, reticular density, and mesh area, it is equivalent to consider that the reticular density should be divided by n or the mesh area multiplied by n. Either one of these artifices will adequately express true morphological importance. No other crystal forms, besides the pinacoid just discussed, can parallel equidistant planes containing equipoints of the same sort only; their morphological importance, therefore, cannot be altered. These conclusions are verified by observation.

The reticular area S is given by the formula

$$S_{hkl}^2 = h^2 S_{100}^2 + k^2 S_{010}^2 + l^2 S_{001}^2$$
$$+ 2(hk S_{100} S_{010} \cos \nu + kl S_{010} S_{001} \cos \lambda + lh S_{001} S_{100} \cos \mu),$$

in which $S_{100} = bc \sin \alpha$, $S_{010} = ca \sin \beta$, $S_{001} = ab \sin \gamma$; and $\lambda = (010):(001)$, $\mu = (001):(100)$, $\nu = (100):(010)$. Hence, the value of S will be multiplied by any number n if all three indices are multiplied by n. We propose

generally to adopt this convention and to designate by the symbol $(nh \cdot nk \cdot nl)$ any face (hkl) normal to a screw axis, with screw component d/n. The classical symbol (hkl) merely indicates the face direction. The suggested symbol $(nh \cdot nk \cdot nl)$ acquires a further value—that of a quantitative estimate of the form importance, since the mesh area S is directly obtainable from the "multiple indices" (by introducing them as such in the formula giving S or so-called "quadratic form" S^2). The Miller notation, thus but little modified, assumes greatly enhanced morphological significance.[3]

Example of the Screw Axis Effect: Low Quartz. Hexagonal trapezohedral class $(D_3 = 3\ 2)$. Axial ratio: $c/a = 1.1000$. As demanded by the Law of Bravais and confirmed by x-ray determination,[4] the translation lattice is hexagonal (*not* rhombohedral) and the point group symmetry must be considered holoaxial tetartohedral (*not* holoaxial hemihedral). $a_0 = 4.89$ A.U., $c_0 = 5.38$ A.U. The space group is one of the enantiomorphous pair $D_3^4 = C3_12$ and $D_3^6 = C3_22$.

The 3-fold symmetry axis is a screw axis, whose screw component is $c_0/3$. The basal pinacoid must be written $\{0003\}$ instead of $\{0001\}$. Its mesh area is trebled, or its reticular density divided by 3. In the Bravais sequence of theoretical importance, the base came first $(S_{0001} = 1.13)$. In the modified list, it goes down to the eighth place $(S_{0003} = 3.39)$, after $10\bar{1}0$, $10\bar{1}1$, $11\bar{2}0$, $10\bar{1}2$, $11\bar{2}1$, $20\bar{2}1$, $11\bar{2}2$ (no distinction is made here between merohedral forms).

Thus is explained the extreme rarity of the basal pinacoid in quartz, one of the most striking morphological characters of the species and a flagrant anomaly to the Law of Bravais.

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[3] H. Ungemach (1935), in order to stress the unity of the hexagonal system *sensu vasto*, in the light of recently discovered examples of syntaxy, proposed to use the 4-index Bravais symbols $(hk\bar{\imath}l)$, slightly modified by the omission of the minus sign over the third index, for crystals having a rhombohedral lattice, with the provision that all four indices should be multiplied by 3 when the sum $h + i + l$ is not a multiple of 3. Examples : (0003), (3030). This amounts to considering the rhombohedral lattice a mode of the hexagonal lattice. M. A. Peacock and M. C. Bandy (1936) followed the suggestion in calling the cleavage of their new mineral ungemachite $\{0003\}$ $\{111\}$.

J. D. H. Donnay (1936) proposed to extend the practice to all systems where the lattice may have more than one mode. Examples.—In the body-centered mode of the cubic lattice, multiply all three indices by 2 if their sum is not even: (110), (200), (211), (222), etc. In the face-centered mode, double all three indices if they are not all odd: (111), (200), (220), (311), etc. Such a procedure expresses the Law of Bravais only.

In the present paper we generalize the symbolization so as to include the effect of screw axes and glide planes.

[4] All the structural data quoted in this paper are taken from *Strukturbericht*, where more specific references may be found.

THE EFFECT OF A GLIDE PLANE

A glide plane is a plane of symmetry, whose operation is the product of a reflection in the plane and a displacement parallel to the plane. The displacement is known as the *glide component*. The possible glide components are (in vectorial notation): $\frac{1}{2}$ **a**; $\frac{1}{2}$ **b**; $\frac{1}{2}$ **c**; $\frac{1}{2}$ (**a**+**b**), $\frac{1}{2}$ (**b**+**c**), $\frac{1}{2}$ (**c**+**a**); $\frac{1}{4}$ (**a**±**b**), $\frac{1}{4}$ (**b**±**c**), $\frac{1}{4}$ (**c**±**a**). The Hermann-Mauguin notation of a glide plane is obtained by replacing, in the point group symbol, the letter *m* indicative of a reflection plane of symmetry by one of the letters *a, b, c, n, d*, chosen according to the type of glide component.

Consider a glide plane in an othorhombic primitive lattice (hexahedral mode), parallel to any one of the coordinate planes, for instance (100). Let the glide component be parallel to either one of the two coordinate axes in the plane, for example $\frac{1}{2}$ **b**. The effect of the glide plane will be to bring space group equipoints into the centers of the (001) meshes. (See Fig. 1, where the glide plane is parallel to the plane of the figure, the lattice equipoints are represented by white circlets, the space group equipoints by black dots.) The space group equipoints are in enantiomorphous relation to the lattice equipoints. No new points are introduced between successive (001) planes—the spacing of the latter remains unchanged. In the (001) planes the motif comprises both the lattice equipoint and the space group equipoint derived from it by glide reflection—the translation net of (001) remains what it was, its mesh area has not changed, neither has its reticular density. The same observation applies to all planes that contain both space group equipoints and lattice equipoints, *e.g.*, the system of planes (021).

Among the systems of net planes (0*kl*) perpendicular to the glide plane, there are some that contain one kind of points only, either lattice equipoints or space group equipoints, *e.g.*, the system of planes (011). In any such system of planes, true lattice planes (beset with lattice equipoints only) alternate with other planes (occupied by space group equipoints only). Any two successive planes can be considered geometrically equivalent since they contain identical nets with motifs similar in all respects except for the fact that they are mirror images of each other (the motif of true lattice planes being represented by a lattice equipoint, that of intervening planes being figured by its enantiomorphous "space group equipoint"). For any crystal face that parallels such a system of planes, the "effective" interplanar spacing should be taken as one half of the distance between true lattice planes if it is to express morphological importance; one may as well say that the reticular density should be halved or the mesh area S doubled. According to the proposed symbolization the indices should be multiplied by 2, so that (011)

is to be written (022). More generally, all the planes (0kl) that contain points of the two different kinds are those that fulfill the condition $k = 2n$. Those that do not satisfy this condition must have their indices multiplied through by 2. Since $h = 0$ for all planes perpendicular to the glide plane, the condition may be written $h + k = 2n$, which is the criterion of a lattice centered on (001). We may thus express this result by saying that the lattice acts as a C-centered lattice for all faces in the zone [100].

If, in the above example, the glide component had been taken equal to $\frac{1}{2}$ **c**, the lattice would have acted as a B-centered lattice for all faces in zone [100].

When the glide plane is parallel to (010) and the glide component is $\frac{1}{2}$ **c** or $\frac{1}{2}$ **a**, the lattice acts as if A-centered or C-centered, respectively, for all faces in zone [010]. Again, if the glide plane is (001), the effective lattice for the faces of zone [001] will be either B-centered or A-centered according as the glide component is $\frac{1}{2}$ **a** or $\frac{1}{2}$ **b**.

A glide plane n, whose glide component is one half the resultant of the two primitive translations in that plane, e.g., $\frac{1}{2}$ (**a** + **b**), transforms a P lattice (primitive) into an I lattice (body centered) insofar as faces perpendicular to n are concerned.

Similar propositions could be established for all space groups where glide planes occur, invariably leading to the recognition of certain planes (among those perpendicular to the glide plane) whose morphological importance is to be decreased. Such a diminished importance is expressed by a multiplication of the indices so as to comply with the restrictions imposed by the presence of the glide planes. These conditions may be combined with others, resulting from the occurrence of screw axes, or due to the different types of centering of the lattice. It is easily seen that such combined conditions are precisely those that the various orders of x-ray reflections must fulfill in order to escape destructive interference (systematic "extinction," *Auslöschung*). These conditions, worked out by structural crystallographers, are directly available in any table of "space group extinctions."

Our conclusions as to the effect of a glide plane are verified by observation.

Example of the Glide Plane Effect: Orthorhombic sulfur. Rhombic dipyramidal or orthorhombic holohedral class ($D_{2h} = 2/m\ 2/m\ 2/m$). Axial ratios—$a:b:c = 0.8131:1:1.9034$. As demanded by the Law of Bravais and confirmed by x-ray determination, the translation lattice is all-face centered (rhombic octahedral mode). $a_0 = 10.61$ A.U., $b_0 = 12.87$ A.U., $c_0 = 24.56$ A. U. The space group is $D_{2h}^{24} = Fddd$.

The space group criteria include the conditions expressing the all-face

centered mode of the lattice, namely: $h+k=2n$, $k+l=2n$, $l+h=2n$, or in other words "h, k, l either all odd or all even." For planes whose indices include at least one zero this condition becomes "indices all even." In addition to this first criterion the presence of the glide planes d demands, for those planes whose indices contain at least one zero, that the sum of the indices be a multiple of 4, namely: planes $(0kl)$ with $k+l=4n$, planes $(h0l)$ with $l+h=4n$, planes $(hk0)$ with $h+k=4n$. In accordance with our symbolization by means of "multiple indices" the list of crystal forms according to the Law of Bravais will only contain symbols with indices all odd or all even (indices not all odd are made all even by multiplying through by 2). The first two columns of Table 1 (headed "B–F" and "Com") give the Bravais list in such a form. In order to take the glide plane effect into account, form symbols whose indices include at least one zero, and which are already doubled by virtue of the all-face centered lattice (Law of Bravais), should be multiplied by 2 again if necessary to make the sum of their indices a multiple of 4. The mesh area of

TABLE 1. ORTHORHOMBIC SULFUR

Space group: $D_{2h}^{24}=Fddd$

B-F	Com	D-H	B-F	Com	D-H	B-F	Com	D-H
002	—			135			*424	
	111			315			*139	
020	—		*028	—			155	
	—	004	260	—			—	*048
	113			331			*2.0.10	
	022			*242		*406	—	
200	—		*208	—			?1.1.11	
	202			*137			337	
*024	—			—	400		319	
	115			119			?062	
	220		*402	—			*426	
204	—		046	—			*351	
	131			244			*511	
	133			151			*248	
	026			228			*157	
	224			*317		*064	—	
	311		*420	—			*353	
	206			*153			*513	
	—	040		422			442	
	117			335			260	
	313			*0.2.10			—	408
*042	—			*246		*0.4.10	—	

N.B. Crystal forms that have not been observed on sulfur are preceded by an asterisk (*). Doubtful forms are shown by a question mark (?).

such forms is thus quadrupled by the new law instead of simply doubled as it was by the Law of Bravais. The new sequence is shown by the last two columns (headed "Com" and "D–H") of Table 1. The second column in this table contains the forms common to both sequences of decreasing theoretical importance (Bravais-Friedel, on the one hand; Donnay-Harker, on the other).

The example of sulfur was given by G. Friedel (1926, p. 133) to illustrate the finding of the translation lattice by means of the Law of Bravais. Friedel stresses the presence of certain anomalies in the list, namely the fact that the forms {012} and {102} are unknown[5] for the species, in spite of their relatively small mesh areas. The next two gaps, those of {021} and {014}, occurring among observed forms, are just as anomalous. Moreover the order of the forms does not fit the facts satisfactorily: {001} occupying the first rank implies a tabular habit, which has been observed occasionally, but is by no means representative of the majority of sulfur crystals; the other two pinacoids are also placed too high in the sequence with respect to their minor morphological importance.

The modified law (see Table 1), doubling again the mesh area of the forms in the first column, causes them to recede in the list to ranks more in keeping with their observed importance. The pinacoids become {004}, {040}, and {400}. The first four missing forms become {048}, {408}, {084}, and {0.4.16}, and are relegated far down the list. The rhombic dipyramid {111} takes first place, as one would expect of the habit-controlling form. The first unknown form is the 20th in the new list, against the 8th in the Bravais sequence.

Statement and Discussion of the Generalized Law

The relationship that exists between the systematic "extinctions" of certain orders of x-ray reflection in a given space group and the diminished importance of corresponding crystal forms permits the following statement of the law:

The morphological importance of a crystal face is inversely proportional to its reticular area S if the lattice is of the hexahedral mode (no centering) and the space group symmetry does not contain any screw axis or glide plane. The effect of lattice centering, screw axes, and glide planes is corrected for if the face indices are replaced, in the S formula, by the "multiple indices" of the lowest order of x-ray reflection compatible with the space group symmetry.

Instead of considering the mesh area of a plane, it is largely a matter of personal preference to consider its reticular density or its spacing to the

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[5] The form {102}, it is true, has been reported since, but it remains a morphological rarity.

next parallel plane in the system, either one of the last two quantities being inversely proportional to the mesh area.

The law includes the Law of Bravais as a particular case, when, regardless of lattice centering, the space group symmetry is devoid of screw axes or glide planes. It is an extension of the Law of Bravais in the sense that it rests on the same kind of geometrical considerations. The difference is that the reticular constant of a face (its mesh area, reticular density, or interplanar distance) is no longer calculated in *the* translation lattice, but in a variable system of points, changing with the face or zone considered. For instance, in the tetragonal space group $D_{4h}^3 = P \, 4/n \, b \, m$, where the translation lattice actually is primitive tetragonal, the reticular constant is computed as if the lattice were body centered (I) for faces ($hk0$), base centered (C) for faces ($0kl$), simple (P) for faces (hhl), while for faces (hkl)—and this is a general rule—the reticular constant is still to be determined in the true translation lattice.

The new law thus brings out the fundamental importance of zonal development in crystal morphology, a fact which had been anticipated by previous crystallographers (Baumhauer, 1903–1905; Ungemach, 1934 and 1935). It confirms the interest of the investigation of "form series" (Baumhauer's *Zielreihen*, Ungemach's *séries zonaires*, Peacock's *Harmonic-Arithmetic Rule*, 1936).

The law cast in the above statement may, up to a certain point, be said to bridge the gap that existed between structural and morphological crystallographies. It is a further step in the elucidation of the control that the internal pattern of a crystal exercises on its bounding forms. Yet it does not involve any consideration of the special positions that the atoms may occupy, nor does it take into account their nature, their charge, their bonds, or any other physical concept. It only makes use of the "empty frame" of symmetry elements in the cell—it is, in essence, purely geometrical.

Since so many factors are ignored that may well be expected to play a role in the development of crystal forms, the law can claim no other value than that of an improved approximation. Not all the anomalies will be eliminated, as it is easy to foresee that several consequences of the law will not be verified. (1) If space group symmetry were the only influential factor, then all crystalline species belonging to the same space group of the isometric system (where the axial ratios and angles are constant) would have an identical list of forms arranged in the order of decreasing importance, or the same "morphological aspect," as we propose to call such a sequence. (2) Several space groups obey the same general conditions of systematic extinction, thus leading to the same "morpho-

logical aspect" for, say, merohedral as well as holohedral species. (3) The generalized law does not discriminate between complementary merohedral forms in all cases. (4) Anomalies may also result from the fact that no provision is made for differences in the conditions of crystallization (rate of cooling, presence of foreign ions in the solution, and the like) in spite of the attempt to eliminate such external factors by a critical examination of available morphological data.

In his masterly work on the subject, Friedel (1907) proved that the influence of the Bravais lattice is so preponderant that it rarely becomes obscured by that of the motif. Now a painstaking survey of a great many crystalline substances for which the space group is known conclusively shows that, although the symmetry of the space group cannot be the only factor either, it so outweighs all the others in influence that its effect, in the large majority of cases, is not obliterated by the action of the disturbing agents. This result, incidentally, will rehabilitate the symmetry (or pseudo-symmetry) of crystals as one of their most significant properties, a notion which of late had lost more and more credence among structural crystallographers.

Apart from any merit that may be conceded or refused to our speculations on the effect of screw axes and glide planes, the outcome of our statistical survey warrants considering the generalized form of the Bravais principle as a law of observation, valid independently of any theory.

This law leads to the recognition of a great many more "morphological aspects" than did the Law of Bravais. We define a "morphological aspect" as a possible list of decreasingly important forms for any set of axial elements (axial ratios and interaxial angles) in any given crystal system. For instance, in the isometric system, the Law of Bravais led to 3 morphological aspects; the present law leads to 17. These are invariable since cubic axial elements are constant. A "morphological aspect" corresponds to a certain set of general extinction conditions, which characterize one space group or may be common to several space groups. There are, therefore, as many "morphological aspects" possible as there are different sets of systematic space group extinctions. They will be presented, for each crystal system, in later publications.

The next sections will illustrate by examples (chosen among isometric substances) the separation of complementary hemihedral forms and the determination of the space group from morphological data. An anomalous case, that of sodium chloride, will also be discussed.

DISCRIMINATION BETWEEN COMPLEMENTARY MEROHEDRAL FORMS

Since the Law of Bravais is based on the consideration of the lattice only, it cannot make any distinction between complementary merohe-

dral forms. The introduction of space group symmetry enables one to separate (in some cases, at least) complementary hemihedral forms.

Example: Pyrite, FeS_2. Isometric system. Diploidal or parahemihedral class ($T_h = 2/m\ \bar{3}$). $a_0 = 5.40$ A.U. The translation lattice is hexahedral (no centering). The space group is $T_h{}^6 = Pa3$.

None of the three Bravais lists available in the isometric system, corresponding to the three possible lattice modes, is very satisfactory. The agreement with observed form importance is the least bad in the hexahedral mode (simple cubic lattice), which places {012} at the fourth rank. In spite of many anomalies, the correct translation lattice was thus arrived at by the Law of Bravais.

According to the space group criterion the forms {0kl} must satisfy the condition $k = 2n$. If they do not their indices are doubled. This condition separates some complementary pyritohedra; for instance, {021} should have more importance than {012}, which is to be written {024}. The Bravais sequence and the modified list are compared in Table 2. Thefirst column (headed "Bravais-Friedel") gives the Bravais list, which does not discriminate between complementary forms. The second column (headed "Donnay-Harker") gives the new order, in which most of the complementary pyritohedra are separated. The third column (headed "Comparison") presents the data in the same manner as Table 1 for sulfur. In the modified sequence the first three forms are the three most important forms, the first two are the only cleavages (imperfect though they may be). The dominant forms in artificial pyrite crystals are the cube and the octahedron (Groth, 1921, p. 257: "Die nach verschiedenen Methoden erhaltenen Krystalle zeigen meist (100) and (111)").

Two complementary pyritohedra will have equal importance when their symbols contain two odd indices besides the zero; they will be separated if only one of the indices is odd. The data recorded in the standard compendia of mineralogy are insufficient to check the validity of this prediction of the law. Discrimination between positive and negative forms is often uncertain. Hintze (1904) does not attempt to make the distinction. V. Goldschmidt (1920) makes it for a few forms only. Strüver's suggestion that the distinction between positive and negative forms be drawn only if two forms of different sign occur together, in which case the larger and more brilliant one is to be taken as positive, is an arbitrary criterion, at best unsatisfactory. The verification of the law is fortunately made possible by H. Ungemach's valuable observations on form importance in pyrite crystals.[6] It is remarkable that information

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[6] Unpublished data of the great French crystallographer and mineralogist (1880–1936), found in the note books he bequeathed to J. D. H. Donnay. The observations are graphically recorded on a work sheet, a gnomonic projection of pyrite showing the

can thus be obtained on the first *eleven* pairs of pyritohedra that are to be separated by the space group criterion. Here are Ungemach's data: {021} overwhelmingly more important than {012 = 024}; {023} vastly more than {032 = 064}; {041} much more than {014 = 028}; {043} probably more than {034 = 068}; {025} less important than {052 = 0.10.4} (anomaly); {061} known, {016 = 0.2.12} unknown; {045} much more important than {054 = 0.10.8}; {027} unknown, {072 = 0.14.4} known (anomaly); {065} much more important than {056 = 0.10.12}; the next four pyritohedra, {081}, {018 = 0.2.16}, {047}, {074 = 0.14.8} are known, but are minor forms whose relative importance cannot be estimated from Ungemach's projection (they are all represented by small points). The first three pairs of complementary pyritohedra show perfect agreement between facts and theory. Notice that {023} is a negative form, predominant over its positive complementary.

A strange fact is revealed by an inspection of the pyritohedra with two indices odd, which should have equal morphological importance: the two complementary forms coexist in three cases only, {075}, {0.15.11}, {097}; the negative form is found alone once, {0.7.11}; the positive form occurs alone in fourteen cases, {091}, {071}, {0.17.3}, {051}, {0.11.3}, {031}, {073}, {0.11.5}, {0.15.7{, {0.13.7}, {095}, {053}, {0.13.9}, {0.11.9}. These data conflict with the theory. It should be noticed, however, that the pair {013 = 026} {031 = 062} that is encountered first in the sequence of forms occupies the 37th rank! Agreement can hardly be expected to prevail so far down the list. There also remains the looming possibility that forms of uncertain sign may have been labeled positive by Ungemach himself—this would explain the avalanche of positive forms!

The modified list of forms does not make any distinction between complementary diploids, {hkl} and {hlk}, the effect of the glide plane being restricted to faces perpendicular to it (one index equal to zero).

In conclusion, one should be well satisfied with the verification obtained, which is vastly better than any agreement could be if due to chance alone. A detailed morphological study of pyrite is, nevertheless, badly needed to confirm or ascertain the sign of the hemihedral forms.

The case of pyrite is a first example of space group determination, without the aid of x-rays, by means of the scantest morphological information, namely: isometric system (whatever the class); {001}, {111}, {012} dominant (regardless of relative importance). Among the 17 possible "morphological aspects" of the isometric system, only one gives

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relative importance of the forms by the size of the corresponding poles (the latter vary from tiny pencil points to circlets five millimeters in diameter).

TABLE 2. PYRITE

Space group $T_h^6 = Pa3$

Bravais-Friedel Lattice P	Donnay-Harker Aspect $Pa3$	Comparison		
		B-F	Common	D-H
001	111	001	—	
011	002	011	—	
111	— 021		111	
012=021	112		—	002
112	022	012 —	— 021	
122	122		112	
*013=031	113		—	022
113	023 —		122	
023=032	123=132	*013=031	—	
123=132	— 041 ⎫		113	
014=041 ⎫	223 ⎬	— 032	023 —	
223 ⎬	114		123=132	
114	133	014 —	— 041 ⎫	
133	024 —		223 ⎬	
124=142	124=142		114	
233	233		133	
034=043	— 043		—	024 —
*015=051 ⎫	134=143		124=142	
134=143 ⎭	115		233	
115	025 — ⎫	034 —	— 043	
025=052 ⎫	234=243 ⎭	*015=051	— ⎫	
234=243 ⎭	*125=152		134=143 ⎭	
*125=152	144 ⎫		115	
144 ⎫	225 ⎭	— 052	025 — ⎫	
225 ⎭	334		234=243 ⎭	
*035=053 ⎫	*135=153		*125=152	
334 ⎭	— 061		144 ⎫	
*135=153	116 ⎫	*035=053	225 ⎭	
*016=061	235=253 ⎭		— ⎫	
116 ⎫	*026=062		334 ⎭	
235=253 ⎭	*126=162 ⎫	*016 —	*135=153	
*126=162 ⎫	045 — ⎬		— 061	
045=054 ⎬	344 ⎭		116 ⎫	
344 ⎭	*145=154		235=253 ⎭	
*145=154	335		—	*026=062
335	*245=254		*126=162 ⎫	
*245=254	136=163	— 054	045 — ⎬	
136=163	?236=263		344 ⎭	
?236=263	345=354		*145=154	
*017=071 ⎫	117 ⎫		335	
345=354 ⎭	* 155 ⎭		*245=254	
117 ⎫	— 064		136=163	
*155 ⎭	*027		?236=263	

TABLE 2. (Concluded)

Bravais-Friedel Lattice P	Donnay-Harker Aspect Pa3	Comparison		
		B-F	Common	D-H
*027=072 ⎫	*146=164	*017=071	— ⎫	
*146=164 ⎬	*127=172 ⎫		345=354 ⎭	
*127=172 ⎬	255 ⎭		117 ⎫	
255 ⎭	227 ⎫		*155 ⎭	
	445 ⎭		—	— 064

N.B.—Unknown forms marked by asterisk (*), doubtful forms by query (?).

top ranks to the three dominant forms; to that aspect the space group Pa3 corresponds uniquely.

SPACE GROUP DETERMINATION BY MORPHOLOGICAL CRITERIA

A "morphological aspect" corresponds to one or more space groups in a crystal system. Within a crystal class, however, a morphological aspect corresponds to one space group only (or, at the most, to two enantiomorphous space groups). Knowledge of the relative importance of the crystal forms usually suffices to limit the possible space groups to a very few, especially if the crystal class (point group) can be determined. Even if the crystal class is not known it is still possible, in certain favorable cases, to determine the space group uniquely. Pyrite is one example. Garnet is another. It will be given here as an illustration of the fact that the dominant form, in an isometric crystal, need not be the cube, the octahedron, or the rhombic dodecahedron, as demanded by the Law of Bravais and implied by the French nomenclature: hexahedral mode, octahedral mode, dodecahedral mode. Derivation of the 17 isometric "aspects" has shown that either one of these three forms can be dominant in all three lattice modes, except that the octahedron can never be dominant in the dodecahedral mode, where, however, the leucitohedron {112} can take first place.

Examples: Garnet, $Al_2Ca_3(SiO_4)_3$. Isomettic system. Hexoctahedral or holohedral class. ($O_h=4/m\ \bar{3}\ 2/m$). $a_0=11.8$ A.U. Translation lattice, body centered (dodecahedral mode). Space group $O_h^{10}=Ia3d$. Here are the most salient morphological data, as given in Dana: the dodecahedron {011} and the trapezohedron {112} are the most common simple forms; the hexoctahedron {123} comes next; the cube {001} and the octahedron {111} are rare.

In order to account for the great importance of the leucitohedron {112} the Law of Bravais can only select the body centered lattice. This

choice is confirmed by x-rays. Among the 17 isometric aspects only one gives top ranks to the three most important forms; this aspect uniquely determines the space group $Ia3d$.

Many discrepancies between the facts and the Bravais sequence are corrected by the new list (see Table 3, arranged like Table 1 for sulfur). The Bravais list expresses the condition that the sum of the indices must be even, which is imposed by the mode of the lattice. The new list takes into account the additional restrictions of the space group: $\{0kl\}$ with k and l even, $\{hhl\}$ with $2h+l=4n$. The cube, which came second, is now fourth. The octahedron, which was fifth, goes down to the fourteenth place. The leucitohedron comes first, followed immediately by the dodecahedron and the $\{123\}$ hexoctahedron.

Analcime, $NaAlSi_2O_6 \cdot H_2O$. This mineral is selected as an example of a pseudo-symmetric species. The dominant "form" is, by far, the trapezohedron $\{112\}$, which often occurs alone. Other "forms" that have been observed: cube, dodecahedron, octahedron, tetrahexahedron $\{012\}$, and

TABLE 3. GARNET

Space group: $O_h^{10} = Ia3d$

B-F	Com	D-H	B-F	Com	D-H	B-F	Com	D-H
011	—			233			145	
002	—		*015	—		226	—	
	112			134			*136	
	—	022		*125			—	444
013	—		035	—		*017	—	
222	—		334	—			*345	
	123		244	—			046	
	—	004		*116			*127	
*114	—			*235			*255	
	024			—	026	*037	—	

N.B.—Crystal forms that have not been observed on garnet are preceded by an asterisk (*).

trisoctahedron $\{233\}$. Of these the octahedron is certainly the rarest. It is hard to assign relative importances to the remaining forms. The only combination given by Hintze that shows five forms (apparently the richest observed) may serve as an indication; its forms are, in order: 112.011.001.012.233. It was found at Friedensdorf, in Hessen.

Incomplete though the data may be, they can be accounted for by one of the morphological aspects only, the same as for garnet, namely: 112.022.123.004.024.233. $\cdots$ (octahedron coming 14th in the list). The fact that $\{123\}$ has not been reported is an anomaly. It does not obscure the otherwise perfect agreement.

The pseudo-isometric character of the species being recognized, simple data on habit and form development permit one to predict that the symmetry of the structure will closely simulate that of the space group $O_h{}^{10} = Ia3d$. The results of the x-ray investigation have led to the recognition of a "pseudo-structure" ($S6_1$ type of *Strukturbericht*) based on a cubic cell ($a_0 = 13.7$ A.U.) and space group $O_h{}^{10} = Ia3d$. The confirmation is striking.

Spinel, $MgAl_2O_4$. The most typical feature of this species is the predominance of the octahedron. The dodecahedron and the trapezohedron {113} come next in importance. The cube never is a "habit form."

Out of the 17 isometric aspects four give {111} the first place. One is the aspect of pyrite, corresponding to the space group $T_h{}^6 = Pa3$, with simple cubic lattice. This aspect is immediately eliminated as it gives the cube second rank, the dodecahedron fifth rank, and {113} seventh rank; not to mention its characteristic hemihedry. It would also demand the presence of {123}, which is unknown, as the first general form, whereas {135}, typical of the F lattice, is observed instead. The other three aspects are: (1) the Bravais list for an all-face centered lattice; (2) the same, slightly modified by symbolizing the cube {004} instead of {002} and making it retreat from second to fourth place; (3) the same list again, but further altered by the "extinction"[7] of all {0kl} forms for which the sum of the indices is not a multiple of 4.

The Bravais list is not as good as either one of its two modifications, on account of the rarity of the cube. The third list fits the facts better that the second, since neither {024} nor {046} are known, whereas {026} is observed. Comparison between the three aspects is shown in Table 4. The first aspect (headed "F" because it is the expression of the all-face centered lattice) corresponds to five space groups, one in each point group, namely: $T^2 = F23$, $T_h{}^3 = Fm3$, $T_d{}^2 = F\overline{4}3m$, $O^3 = F43$, $O_h{}^5 = Fm3m$. The second aspect uniquely defines the space group $O^l = F4_13$. The third aspect is compatible with two space groups: $T_h{}^4 = Fd3$ and $O_h{}^7 = Fd3m$. We have seen that the form development points towards the last two space groups, even if the point group is unknown. Since the diploidal class can be ruled out, $Fd3m$ is the only possibility.

The x-ray results confirm the morphological conclusions. The röntgenographically possible space groups are given as $Fd3m$, $Fd3$, and $F4_13$, in *Strukturbericht*.

[7] This use of the word "extinction," that recalls the correlated phenomenon in x-ray diffraction, may be found convenient. In the first and second lists, {012}, extinguished as such by the all-face centered lattice, appears as {024}. It is extinguished as {024} and occurs as {048} in the third list, owing to additional restrictions of the space group.

TABLE 4. SPINEL, $MgAl_2O_4$

F	O^4	T_h^5, O_h^7
111	111	111
002	—	—
022	022	022
113	113	113
—	004	004
133	133	133
*024	*024	—
224	—	224
*115	—	*115
135	—	135
244	same	244
026	as	026
*335	list	*335
*117⎫	F	*117⎫
*155⎭	—	*155⎭
*046	—	—
*246	—	*246

N.B.—Unknown forms are marked by an asterisk (*).

PERSISTING ANOMALIES

Inasmuch as the law presented in this paper is of a strictly geometrical nature only, it cannot be expected to hold in all cases. Some anomalies do persist.

Example: Halite, NaCl. The space group is known to be $O_h^5 = Fm3m$. The translation lattice is all-face centered, the motif is composed of a sodium ion and a chlorine ion, one half of the cube edge apart (which explains why some authors describe the structure as composed of two "interpenetrating lattices"). In this case the Law of Bravais and the generalized law coincide (no screw axis, no glide plane). The new law is, therefore, just as powerless as the Law of Bravais. The anomalies shriek.

As a natural mineral, NaCl usually occurs in cubes. The other forms known are {011}, {111}, {012}, {123}. Artificially produced crystals show various habits according to the conditions of crystallization (the addition of urea to the solution produces octahedra, as is well known) and several new forms, the principal ones being {112} and {122}; the others, {014}, {034}, {035}, {045}, {233}, {345}. The cleavage, which is perfect, parallels the dominant habit form {001}. Only three of the 17 isometric aspects will give the cube the leading rank (see Table 5, first three columns): (1) the Bravais sequence for a simple cubic lattice (Table 5, first column, headed "*P*"); (2) the aspect that

corresponds to the space groups $T_d{}^5$ and $O_h{}^6$, with an all-face centered lattice; (3) the aspect that corresponds to the space group $T_h{}^7$, with a body centered lattice. None of these is the one demanded by the law (fourth column, headed "F"). The best agreement is found in the "P" list. G. Friedel (1926, p. 144) had already stressed the anomaly. He pointed out that, if one did not discriminate between Na ions and Cl ions, the smallest cell would become simple cubic.

It is a curious fact that cadmium oxide CdO, which belongs to the same space group as NaCl and has the same structural type, does obey the law. The data given in Groth permit listing the forms as follows: {111}, dominant; {001} next; {011} third; and last a trapezohedron, which has not been measured, tentatively given in Groth (1906, p. 176) as {112}?. This list is in perfect agreement with the "F" sequence (see Table 5), even to the last form if the trapezohedron is {113}, which it may well be. Geometrical considerations are obviously insufficient to account for such cases. The reason why cadmium oxide conforms to the law whereas sodium chloride does not is probably to be sought in the less marked ionic character of the cadmium oxide structure.

TABLE 5. HALITE, NaCl

P	$T_d{}^5, O_h{}^6$	$T_h{}^7$	F
001	002	002	111
011	022	112	002
111	222	022	022
012	024	222	*113
112	*135	123	*133
—	224	*114	024
122	244	024	224
*013	*026	*233	*115
*113	*226	*134	*135
*023	*046	*125	244
123	246	*334	*026
—	*137	244	*335
014}	028}	*116}	*117}
*223}	*446}	*235}	*155}

N.B.—Unknown forms are marked by an asterisk (*).

It has been shown above that, when the motif is composed of two "submotifs" enantiomorphous to each other or congruent but rotated with respect to each other, the reticular constant (reticular density, mesh area, or interplanar spacing) should be defined in terms of "equipoints" (representing "submotifs") instead of lattice nodes (standing for motifs) for all planes containing one kind of equipoints only (either

lattice equipoints or space group equipoints). In the case of NaCl, on account of the ionic character of the structure, the motif may perhaps be thought of as composed of two "submotifs," one Na^+, the other Cl^-. Inasmuch as "submotifs" need not be in parallel orientation, it may perhaps be justified to enlarge the tolerance by supposing that ions of different nature can play the rôle of "submotifs." In the case of the sodium chloride structure, this hypothesis is particularly tempting; if the Na ions are imagined to lie at the nodes of the translation lattice, they can be considered as "lattice equipoints" while the Cl ions will become "space group equipoints." In such an hypothesis all the planes with indices all odd will have their spacing halved (or their mesh area doubled) and hence their indices multiplied by two. All other planes have their indices doubled already by virtue of the all-face centering of the lattice. The final result is thus to have all planes modified in the same manner, namely by doubling the indices, which means that the sequence of forms according to decreasing importance will be identical with that of a simple cubic lattice (the edge of the unit cube halved). All anomalies would then disappear.

We have shown that *the two-dimensional periodicity of a plane is the effective factor determining its morphological importance, regardless of what the motif of the mesh may be;* it need not be the same as the three-dimensional motif of the lattice, but may be a submotif thereof, *i.e.,* a part of the motif obtainable from the other parts by screw rotation or glide reflection. The working hypothesis introduced in interpreting the NaCl morphology is that the submotifs need not even be chemically identical.

Summary

Independently of its theoretical justification the following law can be considered a law of observation, including the Law of Bravais as a particular case:

The morphological importance of a crystal face is inversely proportional to its reticular area S if the lattice is of the hexahedral mode (no centering) and the space group is devoid of screw axes and glide planes. The effect of lattice centering, screw axes, and glide planes is corrected for if the face indices are replaced, in the S formula, by the "multiple indices" of the lowest order of x-ray reflection compatible with the space group symmetry.

Many anomalies of the Law of Bravais are accounted for; some persist, since the law is purely geometrical, involving no physical consideration. The influence of space group symmetry is so preponderant, however, that it seldom becomes obscured by that of other factors. The law introduces a distinction between certain complementary merohedral

forms. It leads to the recognition of the various possible "morphological aspects" in any crystal system. The law determines the relative importance of the forms for a given space group; conversely the space group can be predicted from morphological features alone (without the help of x-ray methods).
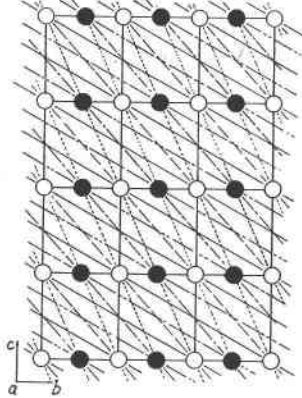

FIG. 1.

Orthorhombic Primitive Lattice.
Glide plane (100) parallel to plane of figure.
Systems of Points:—

      Lattice equipoints: Open circles O

      Space group equipoints: Solid circles ●

      The space group equipoints are mirror images of the lattice equipoints.

Systems of Planes:—

      (021) – – – – – – – – – – – – – –

      (011) Through lattice equipoints —— —— —— —— —— ——

      (011) Through space group equipoints —– —– – —– —– – —– —– – –

      (012) Through lattice equipoints ————————————

      (012) Through space group equipoints — – – —— – —— – – – – — – –

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