THE LAW OF COMPLICATION

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Abstract

The law of complication, advanced by Goldschmidt, rests on an empirical basis. In this paper, the law is investigated rationally. It is shown that if missing terms are neglected, any zone of any crystal must conform to the law of complication for purely geometrical reasons. A transformation to ordinary Miller indices makes it obvious that the law implies the fundamental property of *permutable axes*. Strictly permutable, non-identical axes are only encountered in certain zones of certain classes of isometric, tetragonal and hexagonal crystals, because this condition requires a properly located symmetry element through the dominant node, 1. A re-examination of Goldschmidt's evidence for the reality of the law of complication indicates that this evidence points only to the *statistical* reality of the law, and that individual zones of a given crystal species do not, in general, conform to the law except in the special symmetry cases given above.

Two factors enter into mechanism of controlling the permutability of axes in a zone: the lattice frame, which limits the slopes of crystal faces to definite values but which does not prohibit any crystal from conforming to the law of complication, and the growth environment of the crystal. In any individual instance, the latter factor supplies the mechanism for making the axes of a zone non-permutable. A statistical study of a given crystal species from all environments, however, gives a mass of data from which the specific effect of any individual environment is, at least partly, eliminated. Under these circumstances, the data may fit the crystal species into the law of complication. Junghann's additionrule aspect of the law of complication, although it implicitly includes Goldschmidt's (essentially) reciprocal term law, is a still poorer approximation because it is, without theoretical justification, still more specific.

INTRODUCTION

Peacock¹ has recently refocussed attention upon the law of complication because of its application in the calaverite problem.² This important generalization is due to Victor Goldschmidt,³ and is based primarily upon a statistical study of the frequency of occurrence of crystal faces. Goldschmidt noticed that, referred to the Goldschmidt gnomonic projection coordinates, a zone, $0 \cdots \infty$, contained the face pand its reciprocal 1/p with equal frequency, the simpler values of p oc-

¹ Peacock, M. A., Calaverite and the law of complication: *Am. Mineral.*, vol. **17**, pp. 317–337, 1932.

² Goldschmidt, Victor, Palache, Charles, and Peacock, Martin, Über Calaverit: Neues Jahrb. Min., etc., Bl. Bd., **63-A**, pp. 1–58, 1932.

³ Goldschmidt, V., Über Entwickelung der Krystallformen: Zeit. Krist., vol. 28, 1897. I, pp. 1–35; II, pp. 414–451. curring more often than the complicated ones, of course. This implies that crystal faces occur in zones according to the following plans:

Series					Ce	OORDIN	ATES				
N_0	0										00
N_1	0					1					8
\mathbf{N}_2	0			12		1		2			00
N_3	0		$\frac{1}{3}$	1	23	1	3/22	2	3		8
N_n	0	<u>1</u>		etc.		1		etc.		n	8

The main point of the law of complication is that the *dominant node*, 1, stands symmetrically between reciprocals.

Goldschmidt further showed that in zones not containing the faces 0 and ∞ , the zones could be stretched to do so, so to speak, by a transformation consisting of substituting $p - p_1/p_2 - p$ for each term, p, of the original series, $p_1 \cdots p_2$. The zone so transformed, also conforms to the law of complication. More generally, the transformation may be used rather freely to bring segments of zones into harmony with the law.

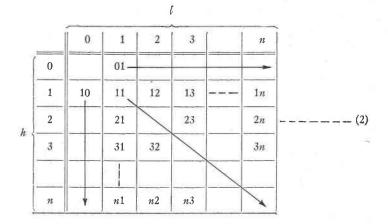
The law of complication is of current interest because of its application by students of surface morphology to the fixing of the axial ratio. As it stands, it is completely empirical. Goldschmidt was inclined to extend the law to cover the fields of music, color, and planetary astronomy, which lent it a distinctly mystical flavor. It is the partial purpose of the present paper to give a rational analysis of the law of complication and to show that it rests on a purely geometrical basis in so far as it concerns crystals.

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Possible Indices. Suppose that in any crystal the basal pinacoid and the unit dome are so chosen that the maximum value of h which appears in indices is the same as the maximum value of l. As an elementary instance, suppose the maximum value of h and l does not exceed 3. Omitting the k index number by confining attention to the pinacoidal profile of a zone, the only possible indices which can appear are those formed by taking all combinations of h=0, 1, 2, 3, with l=0, 1, 2, 3. More generally, suppose the maximum value of h and l does not exceed n. Then, the sum total of all indices which can possibly appear on the crystal can be conveniently represented by the square array:

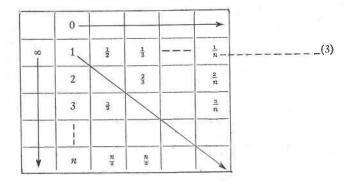
	0	1	2	3		n
0	00	01	02	03		0n
1	10	11	12	13		1 <i>n</i>
2	20	21	22	23		2 <i>n</i>
3	30	31	32	33		3n
1			I	1	\sim	1
n	n0	<i>n</i> 1	n2	<i>n</i> 3		nn

Several of the above combinations are identical. The only distinct ones are the following:



If a gnomonic projection is made of the crystal and the plane of projection chosen normal to the c axis, as established in the first paragraph of this section, the possible Goldschmidt indices, p, corresponding to those given by square array, (2) are obtained by taking each index in (2), reducing it so that the l index number becomes unity, and writing only the other number corresponding to the reduced h index number, thus:

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The order of occurrence of these faces in the zone is as follows:

$$p = \frac{1}{n} \frac{2}{n} \frac{3}{n} \text{ etc.} \quad \frac{1}{3} \frac{1}{2} \frac{2}{3} \quad 1 \quad \frac{3}{2} \quad 2 \quad 3 \quad \text{etc.} \quad \frac{n}{2} \frac{n}{3} \quad n \tag{4}$$

The exact positions of the fractions involving n, with respect to the definite numbers of the sequence, depends upon the value of n. The symbol *etc.* stands for terms involving numbers between 3 and n. Regardless of the value of n, or the terms between 3 and n, however, array (3), from which this series is derived, indicates that any term whatever has a reciprocal an equal distance on the other side of the NW-SE diagonal. In other words, reciprocals are symmetrically placed about the dominant node, 1. This zone sequence, then, is identical with the complication series N_n .

This section may be summarized as follows: with the proper choice of axes (proper choice of basal pinacoid and unit dome, say) the collection of all possible poles of a zone in the gnomonic projection *must* obey the law of complication for purely geometrical reasons. This statement, and the development given above, neglects the possibility of missing and extra terms, which are also well known in actual crystals. These will be discussed subsequently.

Permutable Axes. It appears that the law of complication is really a complicated expression of something more simple and fundamental. If one reverses the reasoning in the last section and assumes the law of complication as empirically demonstrated, then sequence (4) may be taken as a symbolic representation of the law. A rearrangement leads to square array (3), from which a transformation to ordinary indices immediately gives arrays (2) and (1). These arrays indicate that, if a face, (hl), is present on a crystal, a face (lh) is also present. In other words, accepting the law of complication as a postulate, it implies that, so far as crystal surface morphology is concerned, the crystallographic axes may be permuted without affecting the indices of the faces. This conclu-

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sion is not at once obvious in the form of the law as given by Goldschmidt; the reason for this is that the gnomonic projection, used by Goldschmidt for the expression of the law, is a distortion on which all indices are reduced to make l equal unity. It is this distortion which renders the law apparently complex. Indeed, the law of complication might better be renamed, the *law of permutable axes*.

SIGNIFICANCE OF THE LAW

An insight into the possible significance of "permutable axes" may be gained by considering a cubic latticework: say, the two-dimensional analogue of a simple cubic lattice. In a cubic lattice, the a_1 and a_3 axes are symmetrically equivalent and therefore rigorously permutable in any sense (neglecting the modifications introduced by point-group symmetry, and considering only lattice symmetry proper). Hence, any face (hl)must necessarily be accompanied by a face (lh), provided the crystal is grown in an environment field at least as symmetrical as that of the lattice. The two faces, (hl) and (lh), are, of course, symmetrically located on either side of the unit "dome," (11), which is Goldschmidt's dominant node in the law of complication.

If the cubic lattice is slightly distorted both with regard to angle and lengths, it becomes a general two-dimensional lattice. Suppose, now, that one assumes that with each lattice point is associated a sort of discrete, self-contained, packet of atoms, both in the cubic and general cases. Then, whether the packets pile together in cubic or general array in any particular case will have very little control on which faces are left exposed on the surface, for the significance of crystal system in this case is mainly the type of packing required by the shapes of the packets themselves. In other words, with these simple assumptions, one might expect that non-cubic crystals would show a sort of quasi-equivalence of (hl)and (lh) faces corresponding to those rigorously equivalent in the cubic case.

If one modifies the simple assumptions just mentioned, and endows the lattice-point packets with directional bonds, then practical equivalence of (hl) and (lh) no longer need hold for non-isometric crystals. This is, indeed, borne out in the distribution of faces in actual crystals. The very table used by Goldschmidt to demonstrate the statistical reality of the law of complication, speaks plainly on this point. Three isometric examples are cited, two with an analysis of the zone p0. Both of these show ideal examples of the law of permutable axes, as they necessarily must, for a_1 and a_3 are rigorously permutable for the two examples, garnet and halite. The third example contains an analysis of the zone p1in fluorite. The zone p1 implies reference axes, [011] and [100], which are not equivalent. The dominant node, 1, is flanked on one side by 2 and 3 and on the other with its reciprocals, but this is the limit of the application of the law of complication, for there are, in addition, three terms on one side of the dominant not having equivalents on the other, and *vica versa*; this is to be expected in view of the non-equivalence of the reference axes [011] and [100].

The effect of non-equivalence can be extended to the analysis of zones of non-isometric crystals, for which Goldschmidt gives a list of 18 examples. Of these 18, used to prove the law of complication statistically, only two are in rigorous accordance with the law individually: diopside and copper vitriol. Every other example shows one or more terms without corresponding reciprocals. Strictly speaking, then, the law of complication is only a statistical truth. The reason for this is that, in a collection of examples used for statistical study, a certain number of faces, of a given particular type (hl) will be present. Whether this index is written (*hl*) or (*lh*), giving Goldschmidt indices, h/l or l/h respectively, will depend on the convention of choosing the first or second axis as the c axis. This is purely arbitrary in the general case, the triclinic system, and the convention bears no relation to essentials in any other system. In any statistical collection of crystals, therefore, there tends to be an equal number of faces of Goldschmidt index p and 1/p. This is exactly what Goldschmidt found, and it is on this basis that he suggested the law of complication. It should be clearly understood that this is of no rigorous application to any individual non-isometric crystal, or to an individual zone referrable to non-equivalent axes in even an isometric crystal, or, in general, to the case of an individual zone in which the symmetry of the crystal does not require a symmetrical distribution of faces about the node 1. In general, non-identical axes, can be rigorously permutable only in certain classes of the isometric, tetragonal, and hexagonal system.

As a particular example of this feature, the 0q zone of a pyrite crystal of cubo-pyritohedral habit may be cited. The zone sequence is:

indices:	001		3	021	010	
q =	0	×	×	2	∞	(5)

Not only is there no arrangement of reciprocals about the dominant node, 1, in this case, but the dominant itself is absent. Furthermore, the law of complication is not necessarily followed by the crystal even when this simple cubo-pyritohedral habit becomes more ornate by development of additional faces. The truth of this can be instantly grasped by a glance at the figures of pyrite collected by Goldschmidt.⁴ The reason why

⁴ Goldschmidt, Victor, Atlas der Kristallformen, 6, 1920, Tafel 116, figures in the region of 247-250, for example.

pyrite does not adhere to the law of complication is that its point-group does not contribute a properly located symmetry element through the dominant node, 1, and therefore there is no strict requirement for equivalent faces on each side of this node.

This non-equivalence of axes gives rise to missing or extra terms in the complication series. Thus, in series (5) above, the term $\frac{1}{2}$ is missing, or the term 2 may be regarded as extra, depending on the viewpoint. In many actual examples, the number of missing or extra terms is not large and the correspondence of important simple terms with those required by the ideal law of complication is good. Thus in Goldschmidt's statistical table used to demonstrate the law of complication, the term 1 appears in each of the 21 examples, as do also the terms 0 and ∞ ; the term 2 and $\frac{1}{2}$ find their reciprocals missing nine times, the terms $\frac{3}{2}$ and $\frac{2}{3}$ find their reciprocals missing twelve times; but the more complicated terms do not give the law of complication much backing.

MODIFYING CONDITIONS

Cause of Non-permutable axes. It is evident that, for a particular crystal having axes rigorously non-permutable in the zone in question, the law of complication is, at best, an approximation. It is now desirable to inquire into the nature of certain factors rendering the axes non-permutable in an effort to ascertain to what extent the law of complication may be used.

The entire law of complication as usually recognized, rests on the appropriate appearance of external crystal faces. The appearance of a crystal face is conditioned by two factors:

(a) A crystal face can only appear parallel to a lattice plane. The lattice is the framework of crystal faces, and it provides the most fundamental control on their appearance by restricting slopes to certain permitted values. If it were the only factor in the appearance of faces, i.e., if every lattice plane appeared as a face on the crystal, then every crystal would rigorously conform to the law of complications in its major aspect of providing reciprocal terms symmetrically arranged about the dominant node, 1. Furthermore, it would conform to the law regardless of the choice of direction of crystallographic axes. This follows from the fact that permuting any two crystallographic axes only has the effect of permuting the index numbers corresponding to them. The more formal proof follows the lines indicated in the section entitled *Possible Indices*.

If, instead of studying a crystal by means of the reflection of visible light from external planes, it is studied by means of light of x-ray wavelength reflected from internal planes, then the additional variables regulating the appearance of external faces are of no importance, and the

collection of planes so determined conforms rigorously to the law of complication in its reciprocating aspect. This statement must be qualified by noting that the complication series so determined contains missing terms due to systematic extinctions caused by space group and atom parameter requirements.

(b) The second factor which limits the appearance of external faces is the accident of environment in which the crystal made its appearance and continued growth. This set of variables is very complex, but the general nature of its control is known. Generally speaking, the faces which appear on the surface of a crystal are those whose growths are too slow to permit them to be eliminated at the expense of other faces during the growth process. This may mean either that the face in question would not have been eliminated, or it may mean that it was on its way toward elimination, but that the process was arrested by interruption of the growth of the crystal. The last point is important, for it implies that the forms observed on crystals are not necessarily equilibrium ones; the elimination of a face of this sort depends on the size of the crystal under observation, small crystals having a richer variety of faces which are less representative of conditions of growth.

The relative growth velocities of crystal faces depend on a number of conceivable factors, among which position in coordination energy series,⁵ temperature, supersaturation and especially impurity content of solution are important.⁶ The last general factor has been the theme of the researches of Buckley,⁷ who has been able to show that an impurity is able to impede the growth of a crystal face provided the impurity ion (etc.) has a configuration and nature comparable with units exposed on the crystal face. This means that in all but the simplest and purest possible artificial solutions, the habit of the crystal is controlled by variables of a very complex nature.

⁵ The following papers give an introduction to this subject:

Stranski, I. N., Wachstum und Auflösen der Kristalle vom NaCl-Typ: Zeit. physik. Chemie, vol. 17B, pp. 127–154, 1932.

Anderson, Paul A., The molecular process of crystal growth in hexagonal metals. Deposition upon monocrystalline hemispheres of zinc: *Physical Review*, vol. **40**, pp. 596–606, 1932.

⁶ Many authors emphasize "recticular density" as a factor. The very term itself, however, has lost its significance since it has become apparent that crystals generally are not built of molecular units, and that the units are not generally bonded by central forces. In place of "recticular density," a coordination energy series is important.

⁷ Buckley, H. E., The crystallization of potash-alum and the effect of certain added impurities on its habit: *Zeit. Krist.*, vol. **73**, pp. 443–464, 1930. This is the first of a series of papers by this author. Further contributions have appeared in the same place from time to time up to the present year.

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APPROXIMATION FOR A SINGLE SPECIES

As mentioned previously, the first requirement, (a), puts very little restriction on the development of crystal faces. The second, (b), however, is the one which supplies the mechanism for actually making the axes of a zone non-permutable, for the structural configurations of faces related by the permuted indices are not the same. For any given impurity in the solution, a particular crystal will, therefore, develop a certain set of faces, which will not, in general, be in accord with the law of complication. There are two alleviating circumstances, however: Small crystals may not have grown sufficiently to eliminate fast-growing faces, and a small crystal, may, therefore, be more nearly in accord with the law of complication. Secondly, if the study of a crystal species is based, not on a single individual, or on a single crop, or on the representatives of a single mineral locality, but rather if the study is based on numerous crystals grown under a wide range of varying conditions, or on minerals from a large number of localities, then the study takes on a statistical aspect. In this case, there is a chance that differing solutions and other conditions bring out different sets of faces, and that, therefore, the sum total of all the faces represents a large number of the possible simple lattice planes. This tends to eliminate the low symmetry effect of an individual growth environment.

It is possible therefore, to utilize the law of complication if one does not expect perfect agreement with it, provided the comparison with the ideal law is made with the accumulation of all known crystal faces of the species in question, and provided that a rather large number of observations has been made on crystals from many environments. As a matter of fact, it is a statistical mass of data of this sort to which Goldschmidt has applied his law of complication.

It should be pointed out that this means of fixing the lattice of a crystal actually consumes months of time for the accumulation of the necessary data, not to mention the fact that one is never sure whether the mass of data is really sufficient for the purpose. Against this one should weigh the *unique* method of fixing the lattice within a few days' time which is possible by x-ray means, utilizing the Weissenberg⁸ method.

It should also be pointed out that x-ray methods are unique simply because they do not depend on the development of *surface* planes but rather upon internal planes. X-ray methods fix the lattice uniquely because they study the lattice directly; ordinary optical reflection methods

⁸ Turnell, George, Determination of the space-lattice of a triclinic mineral by means of the Weissenberg *x*-ray goniometer: *Am. Mineral.*, vol. **18**, pp. 181–186, 1933.

are non-unique because they study this fundamental structure as modified by other complex variables,—because they thus study a secondary property.

Special Cases. There is one group of special cases in which individual actual crystals with non-permutable axes may nearly conform to the law of complication, namely: the appropriate zone of a crystal which is based upon a high symmetry isometric frame, or more generally, the appropriate zone of a crystal whose axes are semi-permutable because they correspond to really permutable axes in a framework of proper symmetry. The simplest example of this is a crystal whose essential structural plan is simple, but which has a superstructure which lowers the symmetry. The compound AuCu is a representative of this relation. Both Au and Cu alone have cubic close-packed structures; the compound AuCu also has a cubic close-packed structure if one does not distinguish between Au and Cu atoms. The mutual arrangement of these two sorts of atoms, however, gives the space pattern of the compound tetragonal symmetry. PtCu, similarly, has a rhombohedral superstructure modification of cubic close-packing. β -brass, CuZn, must also belong to this class of crystals. Its essential arrangement is body-centered cubic (usually spoken of by metallographers, however, as cesium chloride type); it is thought to have a cubic superstructure, but actually the crystals are anisotropic by reflected, polarized light, so the symmetry is no greater than tetragonal. A slightly different aspect of this relation is given by martensite, which is α -iron with up to 6% carbon. This has an essentially body-centered cubic structure with a few carbon atoms placed interstitially in such a way as to degrade the symmetry of the structure to tetragonal. Unfortunately, crystals of these compounds are not known with well-enough developed faces to test their approximate conformation to the law of complication.

THE ADDITION RULE

Finally, it should be noted that the appearance of crystal faces in the law of complication is an aspect of the addition rule.⁹ This is easy to see in a simple series, say N₃. Combinations of all possible index numbers up to 3 give:

indices = 01	13	12	23	11	32	21	31	10	(6)
$N_3 = 0$									

It will be observed that the addition of any two indices separated by an

⁹ Rogers, Austin F., The addition and subtraction rule in geometrical crystallography: Am. Mineral., vol. 11, pp. 303-315, 1926. odd number of indices, gives the index half way between the two. This property can be easily demonstrated in simpler cases down to the limiting series, N_1 . This suggests that it can also be generalized to higher series. If so, the next higher series, N_4 , can be produced by adding adjacent indices to obtain the new ones appearing between them. Applying this to (6) gives,

01 14 13 25 12 35 23 34 11 43 32 53 21 52 31 41 10 (8) 0 $\frac{1}{4}$ $\frac{1}{3}$ $\frac{2}{5}$ $\frac{1}{2}$ $\frac{3}{5}$ $\frac{2}{3}$ $\frac{3}{4}$ 1 $\frac{4}{3}$ $\frac{3}{2}$ $\frac{5}{5}$ 2 $\frac{5}{2}$ 3 4 ∞ (9)

The corresponding series of Goldschmidt indices (9) has the new terms $\frac{1}{4}, \frac{2}{5}, \frac{3}{5}, \frac{3}{4}$, and their reciprocals. These new terms are just the ones which Goldschmidt's statistical study indicates to actually occur next in order of importance, and series (9) is indeed complication series N₄. Further tests of the addition rule in predicting actual complication series cannot be completely carried out, for lack of sufficient data on the more complicated faces. It can be said, however, that the terms which Goldschmidt's statistical study indicates to occur next most abundantly are among those to be obtained by an extension of the addition rule. Goldschmidt arrived at an identical ideal complication series by what he believed to be the rules of combination of crystal forces to give crystal faces.¹⁰

The occurrence of faces according to the addition law was first recognized in anorthite by Junghann.¹¹ Subsequent writers have investigated this property empirically for other crystals, as well as theoretically. A summary of the development of this subject is given by its latest contributor, Baumhauer.¹²

Goldschmidt's statement of the law of complication may be said to be included in Junghann's empirical addition law. Thus, given any two pinacoids, the indices are taken as 01 and 10. Adding these gives 11, which gives the first Goldschmidt series N_1 , etc. Junghann's law is more restrictive than Goldschmidt's, however, for it definitely predicts the required new indices for any given complication. Actually, Goldschmidt's law does the same, but in his case, more emphasis has been laid on the symmetrical arrangement of reciprocal terms on each side of the dominant, which gives it a more general aspect. It should be pointed out

¹⁰ Reference 3.

¹¹ Junghann, Gustav, Ein einfaches Gesetz für die Entwickelung und die Gruppierung der Krystallzonen: *Pogg. Ann.*, vol. **152**, pp. 68-95, 1874.

¹² Baumhauer, H., Untersuchungen über die Entwickelung der Krystallflächen im Zonenverbande: Zeit. Krist., vol. 38, pp. 628-655, 1904.

that up to the series N3, there is no difference between Junghann's rule and Goldschmidt's (essentially reciprocal) law, because N3 uses every possible combination of h=0, 1, 2, 3 with l=0, 1, 2, 3. In the next sequence, N4, all combinations with 4, and some, but not all, combinations with 5, enter (see sequences (6), (7), (8), and (9)). Higher series are still less regular, according to the strict interpretation of the addition rule. In spite of Goldschmidt's statistical evidence, however, it is the reciprocating aspect which individual crystals most nearly obey, not necessarily the addition principle. Thus, anorthite, which Junghann¹¹ used to demonstrate the addition rule, conforms to that rule without missing terms only to N1, beyond which it is abnormal. Goldschmidt's law implies permutable axes in all crystals, while the addition rule, strictly applied not only implies this but much more with regard to the appearance of particular faces. Needless to say, crystals are found to conform more nearly to the less restrictive Goldschmidt reciprocal term approximation than to the highly restrictive addition rule approximation.

SUMMARY

1. A rational derivation of the reciprocal term aspect of the law of complication is given, and it is shown that the significance of the law is more easily understood divorced from the gnomonic projection. It is then evident that the law of complication might better be renamed the *law* of *permutable axes*.

2. The law of complication is, at best, a poor approximation, both theoretically and as shown by actual crystals. It implies permutable axes in the zone in question, which are actually only realized in certain zones of certain classes of the isometric, tetragonal and hexagonal systems. A rigorous application of the law of complications can only be made for a crystal having an appropriately located symmetry element through the dominant node, 1.

3. The lattice proper does not restrict the application of the law of complication, but the growth of external faces supplies the mechanism for the non-permutable character of axes. However, if the crystal species is studied statistically, i.e., if the comparison with the law is made on the basis of crystals from many growth environments, then the effect of any particular environment may be, in part, eliminated. Only in this way can the law of complication be compared with the actual occurrence of crystal faces.

4. Certain crystals based on cubic, tetragonal or hexagonal patterns, but with a superstructure, may conform to the law of complication.

5. The strict addition rule is a still worse approximation to the actual occurrence of crystal faces than Goldschmidt's law of complication in its reciprocal term aspect, because the addition rule is still more restrictive, with no better theoretical basis.