Examination of Space-Groupoid Theory

II. OD Structure Theory

Hidewo Takahashi

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Abstract

The meanings of vicinity condition and POs (partial operations) of space-groupoid and OD structure theory are investigated. The vicinity condition does not have any crystallographical meanings, since POs do not transform invariantly the common two-dimensional lattice between layers in an OD structure. By representing POs in correct forms, space-groupoid theory is discussed that there is no room to introduce statistics into it. OD structure theory is concluded not to be able to give any quantative informations for one-dimensionally disordered structures.

1. Introduction

Space-groupoids were introduced by Dornberger-Schiff & Grell-Niemann (1961) in order to form the basis of OD structure theory. If the relation between OD structures and space-groupoids is similar to that between ordinary crystal structures and space groups, diffraction phenomena of OD structures, such as extra-ordinary extinctions or diffuse streaks of X-ray diffraction spectra, should be interpretated by the terms of space-groupoids. We hardly see the papers reasonably discussing these phenomena by space-groupoid theoretical point of view.

The work of Sadanaga & Ohsumi (1979) was one of very few works for the interpretation of the diffraction phenomena by space-groupoid theory. The author (1983) showed that their space-groupoid theoretical interpretation of some structures was wrong and the theory of symmetry of vector sets did not concern with the space-groupoid theory and was wrong. A comment of one of the referees for the paper, which was finally rejected by S.C. Abrahams, Editor of Acta Crystallographica, is shown in Fig. 1. The referee is undoubtedly a space-groupoid theorist. The referee insisted that the structure of Fig. B in Fig. 1 was a space-groupoid structure.

The structure can be modified to an OD structure. The OD structure consists of two layers, the layers are called layer A and layer B in this paper. The layer A consists of small squares of

which centers are at apexes of the large square. The layer B consists of the small square at $\bar{\boldsymbol{b}}$ and its equivalences. The layers A and B have the same two-dimensional translation symmetry.

Recently, Grell (1984) described that the vicinity condition defines OD structures. The

Dr B. T. M. Willis
Joint British Co-Editor
of Acta Crystallographica
Materials Physics Division
AERE Harwell
Oxfordshire OX11 ORA
England

Your Reference No., BW-55

Dear Dr Willis,

Thank you very much for your letter dated 19th June and a copy of Hideo Takahashi's paper, "Examination of Space-Groupoid Theory, I. Theory of Sadanaga and Ohsumi". In reply to your request, I am writing below about the results of my refereeing which will, I hope, meet with your kind approval.

The key to understanding the difference between 'complicated structures' by Takahashi (hereafter abbreviated as T) and 'space-groupoid structures' by Sadanaga and Ohsumi (S-O) will be furnished by Section 3 of T's manuscript (p.6). T states there that X_i is given by $X_i=(B_i,b_i)X_0$, $B_iT=T$ must hold because X_i has the same lattice as X_0, and hence B_i must be an element of the point group of the lattice. These conditions indeed impose a very strong restriction upon B_i and accordingly upon his complicated structures. Let us look at Fig.A attached. In this figure, the entire structure X consists of two substructures X_0 and X_1. X_0 is constructed by distributing a group of atoms with the symmetry of $\bar{4}$ over the lattice points of a tetragonal space lattice T. Then, if B_1 represents a rotation by $\pi/2$ around the tetragonal axis c passing the origin, and b_1 the vector indicating a shift of the origin, both $X_1=(B_1,b_1)X_0$ and $B_1T=T$ hold. The structure shown in Fig.A will thus serve as example of T's complicated structure.

Here, the referee wishes to express his concern about T's 'symmetry operation (B_i,b_i) between a pair of substructures'. Is (B_i,b_i) a symmetry operation of the space group of X (global operation) ? If so, T's complicated structure must be imposed upon by a restriction much severer than that found in Fig.A and will be such as derived by replacing one of the equivalent points of its space group with a configuration of points. Suppose that T's complicated structures are all represented by a structure of this type. Then, if the configuration of points replacing each of the equivalent points of the space group has its own symmetry K_0 which is not isomorphic with 1, the structure in this case can be looked upon as a space-groupoid structure in which K_0 is the kernel and all the space-group operations of X are contained in the hull H. (If $K_0\cong 1$, the structure had better be classified as ordinary space-group one.) On the other hand, if (B_i,b_i) is not a space-group operation, it must be either a local or a partial operation, and what T defines as a complicated structure will be nothing but a space-groupoid structure. Indeed, if one is determined to ignore both local and partial operations, what he is confronted with will become global operations only. However, it is obvious that this determination does not entitle him to blame other people for their taking local or partial operations into consideration.

Next, let us consider Fig.B to obtain a better understanding to the nature of the space-groupoid structure. This structure X also consists of two substructures X_0 and X_1 in which X_0 is exactly the same as that in Fig.A. The $\bar{4}$-configuration in X_1 is congruent with that in X_0 but shows an angular deviation by θ from that in X_0, where θ is not necessarily one of such rotations around c that bring the lattice to superpose upon itself. Fig.B represents a typical space-groupoid structure. If we assume that X_0 represents its kernel, we may look upon as an element of the hull a composite of a clockwise rotation by θ of each of the $\bar{4}$-configurations in X_0 around an axis parallel to c and passing its lattice point and a partial translation

by b of the entire structure thus derived.

The symmetry of a structure X is a group of all the automorphic transformations of X, namely, such transformations, each of which brings an exact duplicate of X to superpose upon X in its original position and orientation. Therefore, as far as we confine ourselves to the consideration of the symmetry of a structure X, we must certainly presuppose that all the operations we are dealing with are automorphic transformations. However, what S-O deal with in their paper in Acta is not the symmetry of a structure X but the relation in symmetry between X and its vector set V. In their theory of space groupoid, the set of operations in the kernel K_0 of a space groupoid M is indeed a group of automorphic transformations of a geometric configuration representing K_0. On the other hand, the only condition to be imposed upon the elements of the hull H of M is that each of them should bring the geometric configuration representing K_0 to superpose, not necessarily automorphically, upon another configuration whose group of symmetry operations is isomorphic with K_0. It is to be noted here that in the structure in Fig.B, the space group of X_0 is $P\bar{4}$ and is the same as that of X_1.

T points out again on p.9 of his manuscript that the rotational part of the element of the hull must be the element of the point group of the lattice of the structure X. It is indeed true that all the substructures in T's complicated structure have one and the same translation lattice. However, it is T's peculiar assumption that the rotational part of his 'symmetry operations between substructures' brings the lattice to superpose upon itself. By dint of this assumption, T denounces war against the theory of space groupoid by S-O, but he should have looked before he leapt; he should have offered the reason why space-groupoid structures ought to conform to his rule before he so simply insisted so.

From the latter half of p.9 of his manuscript, T starts referring to the structure given in Fig.7 of S-O's paper (p.121) and reproduced in Fig.2 in T's manuscript, and writes on the top of p.10, "It must be noticed that any two squares have common points in this example. According to Loewy (1927), there is no common element between K_0h_i and K_0h_j if $h_i\neq h_j$. Hence, there should be no atom which occupies at the same position in different substructures. This example does not satisfy the postulation of groupoid." The referee thinks that T would have never made such a simple mistake, had he not been so too enthusiastic in fault-finding of S-O's paper ! In Fig.2 of T's manuscript, let us take $\{1,2,3,4\}$ as the geometric representation of the kernel K_0 of the space groupoid. Then, K_0h_i is a set of operations, each of whose elements brings, say, $\{3,8,6,9\}$ to superpose upon $\{1,2,3,4\}$ by h_i and then $\{1,2,3,4\}$ upon itself by an element of K_0. On the other hand, K_0h_j is a set of operations, each of whose elements brings, say, $\{1,5,6,7\}$ upon $\{1,2,3,4\}$ and then $\{1,2,3,4\}$ upon itself, where h_j naturally involves a similarity transformation and the same applies to h_j. Now, there is obviously no element common between K_0h_i and K_0h_j, simply because $\{3,8,6,7\}$ is a configuration entirely different from $\{1,5,6,7\}$. T confuses operations with points.

In the middle of the upper half of p11, T writes that the operations (of the hull of a space groupoid) are not compatible with ordinary space-group operations. Why not ? They are indeed compatible with each other in the space groupoid, as elements of the hull and those of the kernel !

The referee cannot be bothered with pointing out each of the minute misunderstandings scattered here and there in Section 5 of T's manuscript and wishes to proceed to Section 6 on p.13. Here, T's argument is very much complicated, if not confused. So, the referee will try to explain Section 4 of S-O's paper in plain terms. Let $\vec{a}$ be an inter-atomic vector arbitrarily chosen in a structure X consisting of atoms of one

kind. If there is found in X another inter-atomic vector $\bar{a}'$ which satisfies $|\bar{a}'| = |\bar{a}|$ and whose direction makes with the direction of $\bar{a}$ a certain angle θ around a certain line N, X is specified by S-O as self-homometric. The vector set V of X in this case will be brought to superposition upon itself by a rotation by θ around the direction parallel to N. The case in which X is symmetric by the rotation by θ around N is included in the above category of self-homometry. Three kinds of cause will then be conceived of the given vector symmetry of the structure X: it may be due to the global symmetry of X, or a local symmetry in X, or such a self-homometry of X that is neither the global symmetry nor a local symmetry of X. The vector-symmetric structure due to this last cause is called properly self-homometric. S-O show in 5(A) that it is indeed possible to construct a model of properly self-homometric structure. If a structure which is not properly self-homometric gives rise to diffraction enhancement of symmetry, a local symmetry is responsible for the enhancement.

The referee believes that the present paper is not acceptable for publication in Acta, because the most part of the author's argument is unfortunately based upon his misunderstanding of the nature of space groupoid.

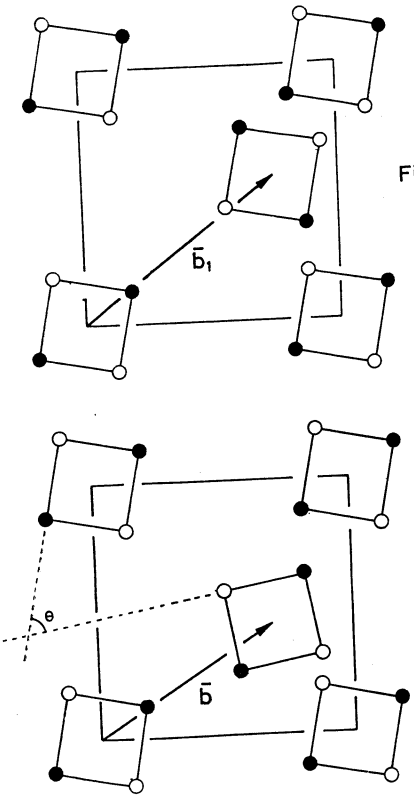


Fig. 1, The comment of the referee of Acta Crystallographica.

vicinity condition requires every layers of OD structures to be of the same two-dimensional translation symmery. The vicinity condition is satisfied for the structure. However, OD structures should have POs transforming a layer to another layer. We can not bring the layer A to the layer B by an operation. The small square in the layer B can be obtained by rotating the small square in the layer A by $\pi/2-\theta$. But the rotation does not bring the large square in the layer A to that of the layer B. That is to say, the rotation does not transform invariantly the two-dimensional lattice.

If we postulate that POs should satisfy the vicinity condition the logical consequence is unique and the comment of the referee becomes nonsense. In this paper, the author discusses shortly the relation between the vicinity condition described by Grell (1984) and POs defined by Dornberger-Schiff & Grell-Niemann (1961).

2. Vicinity condition and partial operations

According to Grell (1984):

"The vicinity condition (VC) is said to hold for a structure (a set of structures) if and only if

(α) it (they) may be considered as consisting of disjunct parts periodic in two dimensions (layers) which are either of the same or of a small number of kinds;

(β) all layers of a crystal have a translational group $m\boldsymbol{a} + n\boldsymbol{b}$ in common ($\boldsymbol{a}$, $\boldsymbol{b}$ non-collinear);

(γ) equivalent sides of equivalent layers are in any and all considered structures faced by adjacent layers in such a way that the layer pairs thus formed are equivalent."

Dornberger-Schiff & Grell-Niemann (1961) described:

"As all layers of the structures discussed are equivalent, there must exist partial operations which transform any one of these layers either into itself or into any other layer. Such partial operations will be called POs for short.

We shall not consider any partial operations referring to parts of a layer (or of layers) only. Thus any repeating operation under consideration may be described by a PO or by combinations of POs.

A PO is fully characterized by

(a) the transformation of space, and

(b) the layer which is to be transformed.

The transformation need not bring any other layer into coincidence with any part of the sturcture." In addition, their foot-note described that "the term 'equivalent' stands for 'congruent or enantimorphous' ".

The most important property of POs deducible from the above descriptions is that POs must be rigid motions, because POs transform layers into equivalent (=congruent or enantimorphous) layers. The rigid motion compatible with the vicinity condition (β) has the rotation part transforming invariantly the common two dimensional lattice between the equivalent layers. If the proposition that POs are rigid motions transforming invariantly the common two-dimensional lattice between equivalent layers is approved, the layer A con nat be brought by any POs to the layer B, since the rotation bringing the small squares of the layers A to the small squares of the layer B can not bring the lattice (the large square) to that of the layer B. The comment of the referee shown in Fig. 1 is denial to the proposition.

When we examine POs in the comment, we find out that the rotation part of a PO is effective in a certain domain (unit cell or structure unit). Hence, the rotation part of a PO consists of infinite number of rotations. Space-groupoid theorists do not consider the relation between the rotations, although the rotations should satisfy the two-dimensional translation symmetry. That the rotations satisfy the two-dimensional translation symmetry means that result by a "rotation of a layers around an axis parallel to $\boldsymbol{c}$ and passing its lattice point" is the same as that by the rotation around an axis parallel to $\boldsymbol{c}$ and passing another lattice point. When the results of such rotations are all the same as the result by the rotation around the axis

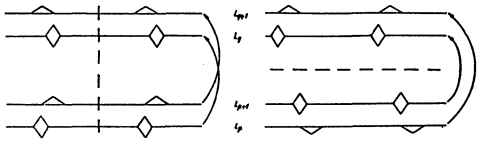
parallel $\boldsymbol{c}$ and passing the lattice origin, we say that the rotation transform invariantly the lattice. The rotations of POs described by the referee do not transform even the domain into the equivalent domains, since the rotations described by the referee tilt the large square in Fig. B. The only objects of the rotations are the points at apexes of the one of the small squares. In other words, space of space-groupoid theory consists of a definite number of points.

The terms in space-groupoid theory are quite different from those of space group theory. For example, "the same transformation" appeared in the following description of Dornberger-Schiff & Grell-Niemann (1961), "If two pairs of adjacent layers, e.g. L_p, L_{p+1} and L_q, L_{q+1} are equivalent then there exists

either a PO ${}_{p,q}a$ characterized by the same transformation as a PO ${}_{p+1,q+q+1}a$

or a PO ${}_{p+1,q}b$ characterized by the same transformation as a PO ${}_{p,q+1}b$.",

is quite different from ordinary meanings. Two symmetry operations having the same rotations and the same translations are said to be the same in space group theory. The examples of the same transformation of Dorberger-Schiff & Grell-Niemann (1961) are illustrated in Fig. 2. Two curves with arrows in Fig. 2 (a) or (b) indicates the same transformation. The two translation parts in Fig. 2 (b) are quite different. Judging from Fig. 2(b), two transformations are said to be the same if they have the same invariant position in OD structure theory.



Die Aequivalcnz crgibt sich
(a) aus den PDen ${}_{p,q}\tau$ ${}_{p+1,q+1}\tau$ (b) aus den PDen ${}_{p+1,q}\rho_{p,q+1}\rho$
Fig. 2, Fig. 4 of Dornberger-Schiff (1964)

Symmetry operations are made to be belonged to symmetry elements in OD structure or space-groupoid theory. Then a symmetry operation (a rigid motion of whole crystal structure) becomes an infinite set of symmetry operations belonging to own symmetry elements, since symmetry elements are invariant positions under a symmetry operation, and there exist an infinite number of symmetry elements in a crystal space for a symmetry operation. The atoms in OD structures are also made to be belonged to the symmetry elements. Then, the author's criticism that the rotation makes tilt the large square (lattice) becomes not effective, since the rotation makes tilt only a small square belonging to the symmtry element by the frame work of thinking of space-groupoid theorists.

Representation of POs by the form of hull and kernel described by Sadanaga & Ohsumi (1979) and discussed by the author (1983) is given by the following form,

$$h_p{}^{-1}=\delta(-p)\ (\boldsymbol{E}, \boldsymbol{p})\ (\boldsymbol{B}_p, \boldsymbol{b}_p) \tag{1}$$

and

$$h_p = (\boldsymbol{B}_p^{-1}, -\boldsymbol{B}_p\boldsymbol{b}_p)\,(\boldsymbol{E}, -\boldsymbol{p})\,\delta(p) \tag{2}$$

where

$$\boldsymbol{p}=p\boldsymbol{c} \quad \text{and} \quad \delta(p)\delta(-q)=1 \quad \text{for} \quad p=q, \qquad \delta(p)\delta(-q)=0 \quad \text{for} \quad p\neq q.$$

The number of the elements of the hull of an space-groupoid becomes infinite. Since we can not obtain h_p from X-ray crystallographical method, we can not determine the hull. Hence, we can conclude that space-groupoid theory is not suitable for interpretation of X-ray differaction phenomena by OD structures.

3. Discussion

Poper (1959) required a condition for an empirical scientific system: *it must be possible for an empirical scientific system to be refuted by experience.* When this condition is applied to space-groupoid theory, we must be able to calculate X-ray diffraction intensity from a space-groupoid and the coordinates of symmetrically independent atoms in a layer for an OD structure.

Since space-gropoid structures of Sadanaga & Ohsumi (1979) are more typical than OD structures, let us examine correspondence between diffracted wave amplitude and POs of them by means of elementary X-ray crystallography. When an atom of ith kind in a unit cell is at (x, y, z), there are another atom of the same kind at $(x+n_1, y+n_2, z+n_3)$, where n_1, n_2 and n_3 are integers. The contribution of the atom to $\boldsymbol{h}$th diffracted wave is

$$f_i\exp[2\pi i\{h(x+n_1)+k(y+n_2)+l(x+n_3)\}]$$

and the total contribution of the atoms translatively equivalent to the ith atom in the structure becomes

$$A_i = F_i \cdot F_{lat}$$

where

$$F_i = f_i\exp\{2\pi i(hx+ky+lz)\}$$

and

$$F_{lat} = \sum_{n_1}\sum_{n_2}\sum_{n_3} \exp\{2\pi i(hn_1+kn_2+ln_3)\}.$$

The total contribution of the atoms in the structure becomes

$$F_{total} = \sum_{n_1} A_i$$

$$= (\sum_i F_i)\cdot F_{lat}$$

$$= F\cdot F_{lat},$$

and F is called the structure factor. The reason of treating only structure factors for ordinary structures is that F_{lat} is invariant by space group symmetry operations, in other words, lattices are invariant by space group operations. This is the most important basic law of X-ray crystallography. If we wish to extend the operations to non-space-group operations, we must examine the lattice invariance by the operations at first. In space-groupoid structures, there are some atoms of the same kind, which are equivalent to each other with respect to space-groupoid theoretically but not equivalent to each other with respect to space group symmetry. If the atomic position of one of such atoms is $(x+n_1, y+n_2, z+n_3)$, there is a PO transforming the position to $(x'+n_1, y+n_2, z'+n_3)$. In this case, eventhough (x, y, z) is transformed to (x', y', z'), (n_1, n_2, n_3) is required to be invariant. If this requirement is satisfied, F_{lat} becomes invariant by the transformation. However this requirement is nonsense, since (x, y, z) and (n_1, n_2, n_3) should be transformed by the same form. OD structure theorists say that POs are transformation of space, but their space has singular points which are not moved by the transformation. In conclusion, vicinity condition can not be compatible with POs described by space-groupoid theorists.

It is necessary to introduce statistics into intensity equation for calculation of the intensity of X-ray diffraction intensity of so-called OD structures. Space-groupoid theory may be useful if statistical intensity equation can be derived from the theory. But, OD structure theorists depend on statistical intensity equation which can not be derived from space-groupoids for the calculation. In addition, space-groupoid theory can not give any reasonable interpretation for the existence of many polytypes of close-packed structures. Sequential occurrence of stacking vectors should be considered connectedly with POs. Dornberger-Schiff (1964) discussed "Anahl Z der mit NB verträglichen Lage einer Schicht L_{p+1} bei vorgegebener Lage der Nachbarschicht L_p". The author does not believe that the derivation of the number Z is reasonable, because "Fortsetzung" is meaningless in the author's representation. We can easily see that the number Z is insufficient to define a polytype of close-packed structures.

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