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Groupoid description of modular structures

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Modular structures are crystal structures built by subperiodic (zero-, mono- or diperiodic) substructures, called modules. The whole set of partial operations relating substructures in a modular structure build up a groupoid; modular structures composed of identical substructures are described by connected groupoids, or groupoids in the sense of Brandt. A general approach is presented to describe modular structures by Brandt's groupoids and how to obtain the corresponding space groups, in which only the partial operations that have an extension to the whole crystal space appear.

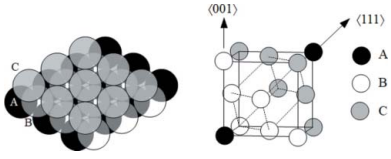
1. Introduction

The symmetry of a crystal structure is described by its space group, *i.e.* a group of isometries φ (distance-preserving transformations) in the three-dimensional Euclidean space E^3. The group formed by all the isometries of E^3 is known as the Euclidean group $\mathcal{E}(3)$; every space group is a subgroup of $\mathcal{E}(3)$. If we consider the subset of isometries that preserve the origin, we obtain the orthogonal group $\mathcal{O}(3)$, which is a subgroup of $\mathcal{E}(3)$; every point group is a subgroup of $\mathcal{O}(3)$. An isometry in E^3 is represented by a matrix–column pair $(\boldsymbol{W}, \boldsymbol{w})$, where, with respect to a suitable coordinate system, the matrix $\boldsymbol{W}$ represents the linear part of the operation (rotation, reflection, rotoinversion) and the vector $\boldsymbol{w}$ represents the translation part of the operation. This matrix–column pair can be synthetically represented by the Seitz notation $\{\boldsymbol{R}|\boldsymbol{v}\}$, where $\boldsymbol{R}$ is the Hermann–Mauguin symbol of the linear part and $\boldsymbol{v}$ is the translation part (Glazer *et al.*, 2014).

Space groups do not describe, in general, all symmetry properties of each and every possible periodic structure; additional isometries can often be found which express some features of the structure that cannot be found in the space-group description. This calls for a generalization of the concept of symmetry group. The possible generalizations of the symmetry group of a crystal pattern[1] can be classified as follows:

(i) Higher-dimensional groups (superspace groups): used to describe the symmetry of incommensurately modulated structures, composite crystals and quasicrystals, which appear aperiodic in E^3, yet produce a diffraction pattern with sharp Bragg peaks in the corresponding reciprocal space. Interpreting the crystal pattern in E^3 as a section of a periodic structure in a higher-dimensional Euclidean space (E^n, $n > 3$) explains the sharp Bragg peaks.

(ii) Non-Euclidean groups. Some aperiodic structures can be described as periodic without the need for a higher-



[1] We remind the reader that a crystal pattern is the generalization of a crystal structure which abstracts from the atomic nature.

dimensional space, but by changing the geometry of the space (Kocian, 2013). This is the case for incommensurately modulated structures and composite crystals, which bear a correspondence with periodic structures, to which they are reduced when bringing to rational values the amplitude of the modulation wave (modulated structures) or the ratio of the cell parameters of the structures building the composite crystals. It is not possible to adopt this approach to quasicrystals, because they are intrinsically aperiodic; for them a description in terms of a modulation of a basic periodic structure or a composition of two or more substructures is either inappropriate or impossible (Lifshitz, 2003).

(iii) Chromatic groups. Atoms of the same chemical species and occupying corresponding positions in different unit cells may need to be differentiated on the basis of a physical state (*e.g.* their spin). To express this state, a colour is associated to the atom (Litvin, 2013). Similarly, the operation mapping the orientation of domain states in a twin is also described as a chromatic operation, relating domain states bearing a different 'colour', each colour corresponding to an orientation in space (Nespolo, 2004, 2019).

(iv) Restriction of the domain of action of the isometries, which leads to algebraic structures more general than a group.

Generalizations (i) and (iii) are well known and have been collectively called 'metacrystallographic groups' (Opechowski, 1986). Generalization (ii) has received increased interest in recent years (see also, *e.g.*, Pedersen & Hyde, 2017) but the results are still in the embryonic state with respect to potential applications. We are particularly interested in (iv), which has been extensively studied in the past but has remained somewhat esoteric among crystallographers.

By removing the requirement that the isometries φ act everywhere in the crystal space, we obtain an algebraic structure more general than a group. In a space group, all the isometries are global (or total) operations, which means that when they are applied to any building block of the crystal structure (atoms, molecules, polyhedra, synthons, layers *etc.*) one obtains an identical building block somewhere else in the crystal structure. This condition is actually too restrictive for a complete description of a crystal structure; in general, there exist isometries that do not act on the whole crystal structure

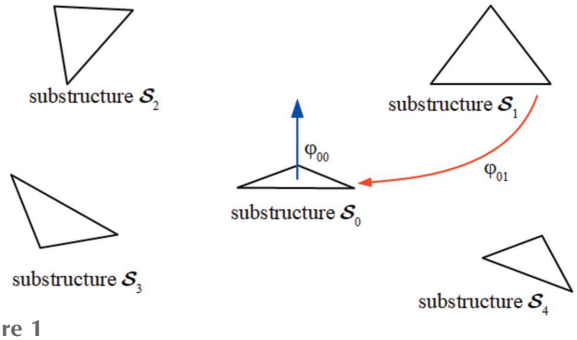


Figure 1
Model of a unit cell composed of five identical substructures S_0 to S_4 with a different configuration (orientation and position in space). φ_{00} is an operation mapping S_0 onto itself (symmetry operation of S_0: a local operation); φ_{01} is an operation mapping S_1 onto S_0 (a partial operation) (modified after Sadanaga *et al.*, 1980).

but only on part of it. These isometries relate subsets of chemically equivalent building blocks. In macromolecular crystallography, they are often, although incorrectly, called 'non-crystallographic symmetry operations'; in general they are perfectly crystallographic (obeying the restrictions on the order of the operation imposed by the existence of a Bravais lattice), but their domain of action is restricted to some but not all of the building blocks (Nespolo *et al.*, 2008). These isometries are called *partial* operations and their composition forms an algebraic structure known as a *groupoid* (Higgins, 1971; Brown, 1987). When the building blocks are of the same type, so that partial operations can relate any pair of substructures, then the structure is described by a connected groupoid, originally introduced by Brandt (1927). The application of space groupoids to the study of layer structures has been the subject of intensive investigation by the so-called OD ('order–disorder') school, from the late 1950s to the early 1980s (see, *e.g.*, Dornberger-Schiff & Grell-Niemann, 1961). A more general approach was earlier introduced in Japan, but, with only one exception, was left incomplete; and, with only one exception (Kato, 1999), is today almost completely forgotten [see a recent review by Umayahara & Nespolo (2020)]. The current article presents the application of groupoids to the description and analysis of modular structures in general, not limited to layers, and the guidelines to obtain the subset of global operations that build up the space group of the structure. A brief glossary of the symbols used in the next sections is given in Appendix *A*. In particular, small Greek letters like φ and ϕ represent isometries; capital Greek letters such as Φ and Γ are used for sets of isometries which do not form a group; a set of isometries which forms a group is indicated by a capital Latin script letter: $\mathcal{N}, \mathcal{G}, \mathcal{C}$.

2. Partial operations in crystallography

The concept of a partial operation in a crystal pattern can be easily visualized in E^3 with the help of Fig. 1, representing the unit cell of a crystal pattern composed of identical modules that, following the previous literature [see a review by Umayahara & Nespolo (2020)], will also be called *substructures*. For the sake of simplicity, the substructures in this figure are finite (in the specific case, they are drawn as isosceles triangles), but this restriction will be removed later. Triangles with different orientations occur in the unit cell (where orientations inclined with respect to the plane of the figure result in deformed triangles). Each substructure is labelled as S_p, with p indexing the substructures in a unit cell; here $p = 0$, $n - 1$ ($n = 5$ in the figure). The content of the unit cell of S can thus be written as the (set-theoretical) union of substructures:

$$S = \cup_p S_p.$$

A mapping from S_p to S_q will be written as φ_{qp}: $S_p \nrightarrow S_q$, where we use $\nrightarrow$ as the symbol for a partial function, we call S_p the *source* and S_q the *target* of φ_{qp} and we have $\varphi_{qp}S_p = S_q$ (note the order of the indices p and q: because the mappings are applied from the left, we put the index of the source right and the index of the target left). We call φ_{qp} a *partial operation*

because it acts on the substructure S_p, but not on any other substructure S_r with $r \neq p$. The whole set of partial operations $S_p \not\to S_q$ is indicated by Φ_{qp}. A partial operation φ_{qp} is the *restriction* of an isometry $\{\boldsymbol{R}|\boldsymbol{v}\}$ of E^3 to the space occupied by S_p. One can also define the reverse, an *extension* $\overline{\varphi}^*_{qp}$ of the partial operation φ_{qp} to the whole three-dimensional space, by simply 'forgetting' about the fact that φ_{qp} is restricted to the substructure S_p. The star in $\overline{\varphi}^*_{qp}$ indicates that the isometry is in general not valid for the whole structure.

A special case of partial operation is a mapping φ_{pp} from S_p to itself, which will be called a *local operation*. Local operations stand out because they form groups: the groups $\mathcal{N}_p$ of the symmetry operations of the substructures S_p. For $p \neq q$ the groups $\mathcal{N}_p$ and $\mathcal{N}_q$ are disjoint ($\mathcal{N}_p \cap \mathcal{N}_q = \emptyset$, where $\emptyset$ is the empty set) because the partial operations act on different substructures. $\overline{\mathcal{N}}_p$ denotes the group of extensions of the elements in $\mathcal{N}_p$. All $\overline{\mathcal{N}}_p$ are subgroups of $\mathcal{E}(3)$ and share at least one common element, the identity 1. If two different substructures S_p and S_q ($p \neq q$) are isometric,[2] $\overline{\mathcal{N}}_p$ and $\overline{\mathcal{N}}_q$ are conjugate subgroups of $\mathcal{E}(3)$ (and therefore of the same affine type[3]), since for an isometry φ_{qp} mapping S_p to S_q one has $\overline{\mathcal{N}}_q = \overline{\varphi}^*_{qp}\overline{\mathcal{N}}_p\overline{\varphi}^{*-1}_{qp}$. However, $\overline{\mathcal{N}}_p$ and $\overline{\mathcal{N}}_q$ are in general different groups, since the extensions of symmetry operations of S_p do not necessarily map S_q on itself (and vice versa) and therefore the geometric elements of the symmetry operations of S_p and S_q are in general differently oriented and/or located in space.[4]

The composition of two partial operations φ_{qp}: $S_p \not\to S_q$ and φ_{rq}: $S_q \not\to S_r$ is again a partial operation $\varphi_{rq}\varphi_{qp} \in \Phi_{rp}$, having source S_p and target S_r (note how, by our choice of the order of the indices, the compatibility of the two partial operations corresponds to the inner index being the same). In particular, if $p = r$, the composition $\varphi_{pq}\varphi_{qp}$ is a local operation contained in $\mathcal{N}_p$.

As already remarked, a partial operation φ_{qp} acts only on S_p; this also applies when $p = q$, *i.e.* for local operations and, although counterintuitive, even to the identity operations (note the plural): each S_p has its own local identity operation 1_{pp}, which is the restriction of the global identity operation $\{\boldsymbol{I}|\boldsymbol{o}\}$ to S_p.

The inverse operation φ^{-1}_{qp} of a partial operation φ_{qp} relates S_q to S_p, since $\varphi_{qp}S_p = S_q$ implies $\varphi^{-1}_{qp}S_q = \varphi^{-1}_{qp}\,\varphi_{pq}S_p = S_p$; hence we have $\varphi^{-1}_{qp} \in \Phi_{pq}$.

In general, applying the extension $\overline{\varphi}^*_{qp}$ of a partial or local operation φ_{qp} to a substructure S_r with $r \neq p$ will result in a fictitious substructure S_t^* that is not part of S and may either overlap the area occupied by some substructure S_s or fall into a void between the substructures. In the most extreme case, only the substructure S_p from which $\overline{\varphi}^*_{qp}$ is induced is mapped to a substructure (namely S_q) in S. In the opposite extreme, it may happen that the application of $\overline{\varphi}^*_{qp}$ to each substructure S_r gives a substructure in S: for each r one has $\overline{\varphi}^*_{qp}\,S_r = S_s$ for some s. Besides the identities 1_{pp}, which are all extended to the identity operation 1 of the structure, an instructive example is the case where the substructures S_p are layers in the same orientation: translation symmetry of one layer is actually a translation symmetry for each of the layers. In such a case, $\overline{\varphi}^*_{qp}$ is a *global* (or *total* in the literature on order–disorder structures, OD for short) symmetry operation for the whole modular structure S and this is indicated by omitting the star, *i.e.* denoting the extension by $\overline{\varphi}_{qp}$. The global operations $\overline{\varphi}_{qp}$ form the ordinary space group of the modular structure S.

The model in Fig. 1 represents the content of a unit cell, in which a finite number of identical finite substructures occur in different positions and orientations. Let us now act on it with a set of translations. Two possible scenarios arise depending on whether or not a set of linearly independent translations that are global operations can be found for each dimension of the space:

(i) In the negative case, the system is not periodic in at least one of the dimensions of the space; it may correspond to a disordered stacking of modules, like in the so-called 'disordered polytypes' (Guinier *et al.*, 1984), or to an amorphous solid; the whole set of partial (including local) operations can also characterize a 'snapshot' of a liquid or of a gas: the additional variable *time*, whose effect can be seen as modifying the configuration of the substructures, is frozen at the moment the set of partial operations is specified.

(ii) In the positive case, the system is periodic along all the dimensions of the space. Because these translations are global operations, their composition with further operations φ will result in partial (when φ is partial) or global operations (when φ is global).

In the following we restrict our attention to discretely periodic structures, built of a single kind of isometric substructure. The number of substructures is therefore countably infinite and can be indexed by the integers. Moreover, if the substructures are not finite but infinite in one (rods or chains) or two directions (sheets or layers), they cannot be *contained* in a finite unit cell. One can nevertheless *identify* a finite number of substructures that intersect a unit cell. The structure S is periodic in E^3 and the substructures S_p are subperiodic in this space, namely nonperiodic (finite), monoperiodic or diperiodic. The directions of missing periodicities are three, two or one, respectively, and it is along these directions that the substructures are stacked.

The example shown in Fig. 1 is clearly too simple to describe all possible crystal patterns composed of substructures. Two generalizations have to be considered:

(i) Patterns built by substructures that are themselves composed of substructures, *i.e.* $S_p = \cup_q S_{pq}$, where S_{pq} is the qth sub-substructure building the pth substructure; in this case,

[2] Two (sub)structures are called *isometric* if they can be brought into perfect coincidence by an isometry (*i.e.* a mapping preserving distances and angles). It is somewhat unfortunate that the same term *isometric* is often used as a synonym of 'cubic'.

[3] We remind the reader that two groups are of the same crystallographic type if they are conjugate by an isometry of the first kind (handedness-preserving) and of the same affine type if they are conjugate by any isometry (possibly of the second kind, *i.e.* handedness-reversing). The difference between crystallographic and affine types shows up only in the case of chiral groups: the two groups forming an enantiomorphic pair are conjugate only by an isometry of the second kind.

[4] The geometric element of a symmetry operation is the point (for inversions), line (for rotations and screw rotations) or plane (for reflections and glide reflections) about which the operation is performed. For rotoinversions it is the corresponding rotation axis together with the inversion centre on this line.

there will be: (*a*) local operations mapping S_p onto S_p and S_{pq} onto S_{pq} (*i.e.* fixing the sub-substructures within S_p); (*b*) partial operations mapping S_p to S_p, but S_{pq} to S_{pr} (*i.e.* permuting the sub-substructures within S_p); (*c*) double partial operations mapping S_{pq} to S_{rs} (*i.e.* moving the sub-substructures of S_p to a different substructure S_r).

(ii) Patterns built by more than one kind of substructure; the symmetry group of a substructure is only conjugate to the symmetry group of another substructure if it is of the same kind; furthermore, there are only partial operations between substructures of the same kind, so that the crystal pattern can be seen as the intergrowth of disjoint modules of different kinds. This case has to be treated in terms of disconnected groupoids according to Ehresmann (Taylor, 1977), which are more general than Brandt groupoids (the latter are the connected components of the former: for a more formal definition, see Appendix *B*). Applications to allotwins and incomplete (allo)twins have been presented by Nespolo (2019).

An example of point (i) above can be found in the modular structure of sepiolite (Nespolo *et al.*, 2018) built by stacking modules eight-rod thick, where each module can be further subdivided into three modules two-rod thick with additional atoms at the boundary. The two modular interpretations correspond to a different fineness in the analysis of a crystal structure.

3. The algebraic structure of a groupoid of partial operations

We assume that all substructures constituting the modular structure are isometric. This means that the partial operations are transitive on the substructures S_p, *i.e.* that for any pair p, q there exists at least one partial operation φ_{qp} mapping S_p to S_q. In this case, any operation mapping S_p to S_q can be decomposed into a two-step operation, first mapping S_p to S_r and then S_r to S_q:

$$\varphi_{qp} = \varphi_{qr}\varphi_{rp} = \varphi_{rq}^{-1}\varphi_{rp}. \quad (1)$$

The whole set of local operations Φ_{pp} for each substructure S_p forms the symmetry group $\mathcal{N}_p$ of the substructure. If φ_{qp} is any isometry mapping S_p to S_q, then $\mathcal{N}_q = \varphi_{qp}\,\mathcal{N}_p\varphi_{qp}^{-1}$, *i.e.* $\mathcal{N}_p$ and $\mathcal{N}_q$ are conjugate under φ_{qp}. As a consequence, the Hermann–Mauguin symbols (expressed in a standard setting) of the two groups are either the same or belong to an enantiomorphic pair.

The whole set of partial operations Φ_{qp} mapping S_p to some different substructure S_q does not form a group, since it is not closed under composition of operations. Let us now focus our attention on a specific substructure that will be used as reference and indicate it as S_0. We can collect all the operations mapping *any* substructure to S_0, including S_0 itself:

$$\cup_p \Phi_{0p} = M_0. \quad (2)$$

The set M_0 contains the group $\mathcal{N}_0$ of local operations of S_0 and we denote the set $\cup_{p\neq 0}\Phi_{0p}$ of partial operations mapping any S_p different from S_0 onto S_0 by H_0, so that $M_0 = \mathcal{N}_0 \cup H_0$. The sets $\mathcal{N}_0$, M_0 and H_0 have received different names by different authors; here we follow Dixon (1963) in calling $\mathcal{N}_0$ the *nucleus* and M_0 the *mixed group* [after the German term *Mischgruppe* (Loewy, 1927)]; H_0 is called the *shell* [for a brief discussion about the history and choice of these names see Nespolo (2019)]. The nucleus is a finite group if the substructure is nonperiodic, or a subperiodic group (in particular, rod or layer group) if the substructure possesses a periodicity in a subspace of the crystal pattern space. Despite being called a mixed group, M_0 is *not* a group: only its nucleus is. M_0 can nevertheless be decomposed into cosets with respect to the nucleus $\mathcal{N}_0$, fully analogous to a group being decomposed into cosets with respect to a subgroup. For that, fix for every substructure S_p a partial operation $\phi_{0p} \in \Phi_{0p}$ mapping S_p to S_0. Then, for an arbitrary partial operation φ_{0p} also mapping S_p to S_0 one has $\varphi_{0p}\phi_{0p}^{-1} \in \mathcal{N}_0$ and thus $\varphi_{0p} = \varphi_{00}\phi_{0p}$ for some $\varphi_{00} \in \mathcal{N}_0$. In short, one has $\Phi_{0p} = \mathcal{N}_0\phi_{0p}$ and we can decompose M_0 into right cosets with respect to N_0:

$$M_0 = \cup_p \mathcal{N}_0\phi_{0p} \quad (3)$$

where ϕ_{0p} are the chosen coset representatives. Given any partial operation φ_{0p} belonging to the shell H_0, one can always define the inverse operation φ_{0p}^{-1}; however, this operation does not belong to the shell, since its target is S_p and not S_0. The inverse operation φ_{0p}^{-1} nevertheless plays an important role in the analysis of the complete algebraic structure of the crystal pattern S, because it transports back the geometric properties of S_0 to S_p. Since we have already chosen for each source S_p an operation ϕ_{0p} that maps S_p to the target S_0, the obvious choice for a mapping in the opposite direction is the inverse operation ϕ_{0p}^{-1}. Note that the chosen operations ϕ_{0p} also give a canonical choice for an operation ϕ_{qp} mapping S_p to S_q, namely the composite operation $\phi_{qp} = \phi_{0q}^{-1}\phi_{0p}$ which maps S_p via S_0 to S_q. As already remarked, any φ_{qp} mapping S_p to S_q conjugates $\mathcal{N}_p$ to $\mathcal{N}_q$ by $\mathcal{N}_q = \varphi_{qp}\,\mathcal{N}_p\varphi_{qp}^{-1}$ and in the opposite direction by $\mathcal{N}_p = \varphi_{qp}^{-1}\mathcal{N}_q\varphi_{qp}$. In particular, we have $\mathcal{N}_q = \phi_{0q}^{-1}\mathcal{N}_0\phi_{0q}$. The reference nucleus $\mathcal{N}_0$ therefore determines the type and Hermann–Mauguin symbol of all the nuclei.

Recall that $\Phi_{0p} = \mathcal{N}_0\phi_{0p}$, that is, every $\varphi_{0p} \in \Phi_{0p}$ can be written as $\varphi_{0p} = \varphi_{00}\phi_{0p}$ for some $\varphi_{00} \in \mathcal{N}_0$. Analogously (taking inverses), every $\varphi_{q0} \in \Phi_{q0}$ can be written as $\varphi_{q0} = (\varphi'_{00}\phi_{0q})^{-1} = \phi_{0q}^{-1}\varphi_{00}'^{-1}$ for some $\varphi'_{00} \in \mathcal{N}_0$. Since $\varphi_{00}'^{-1}\varphi'_{00} \in \mathcal{N}_0$, an arbitrary operation $\varphi_{qp} \in \Phi_{qp}$ that maps the substructure S_p to S_q can be written as $\varphi_{qp} = (\phi_{0q}^{-1}\varphi_{00}'^{-1})(\varphi_{00}\phi_{0p}) = \phi_{0q}^{-1}(\varphi_{00}'^{-1}\varphi_{00})\phi_{0p} \in \phi_{0q}^{-1}\mathcal{N}_0\phi_{0p}$. Thus, the union $\Gamma = \cup_{p,q}\Phi_{qp}$ over all partial operations of S, which is called the *space groupoid* of S (Sadanaga, 1978), can be decomposed with respect to the nucleus $\mathcal{N}_0$ as:

$$\Gamma = \cup_{p,q}\phi_{0q}^{-1}\mathcal{N}_0\phi_{0p}. \quad (4)$$

Equation (4) represents the decomposition of Γ in terms of $\mathcal{N}_0$. It can be made explicit in a tabular form, which is itself infinite. However, if we limit p and q to the substructures included in a full period, *i.e.* identified in a single unit cell, equation (4) can be written modulo the lattice translations so that the groupoid can be expressed in a finite tabular form (Table 1). If n is the number of substructures S_p within a

Table 1
Tabular expression of the groupoid in equation (4).

$M_0 =$	$\mathcal{N}_0$	$\cup$	$\mathcal{N}_0\phi_{01}$	$\cup$	$\mathcal{N}_0\phi_{02}$	$\cup$	$\ldots$	$\cup$	$\mathcal{N}_0\phi_{0p}$	$\cup$	$\ldots$	$\cup$	$\mathcal{N}_0\phi_{0n}$
$\phi_{01}^{-1}M_0 =$	$\phi_{01}^{-1}\mathcal{N}_0$	$\cup$	$\phi_{01}^{-1}\mathcal{N}_0\phi_{01}$	$\cup$	$\phi_{01}^{-1}\mathcal{N}_0\phi_{02}$	$\cup$	$\ldots$	$\cup$	$\phi_{01}^{-1}\mathcal{N}_0\phi_{0p}$	$\cup$	$\ldots$	$\cup$	$\phi_{01}^{-1}\mathcal{N}_0\phi_{0n}$
$\phi_{02}^{-1}M_0 =$	$\phi_{02}^{-1}\mathcal{N}_0$	$\cup$	$\phi_{02}^{-1}\mathcal{N}_0\phi_{01}$	$\cup$	$\phi_{02}^{-1}\mathcal{N}_0\phi_{02}$	$\cup$	$\ldots$	$\cup$	$\phi_{02}^{-1}\mathcal{N}_0\phi_{0p}$	$\cup$	$\ldots$	$\cup$	$\phi_{02}^{-1}\mathcal{N}_0\phi_{0n}$
$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$
$\phi_{0n}^{-1}M_0 =$	$\phi_{0n}^{-1}\mathcal{N}_0$	$\cup$	$\phi_{0n}^{-1}\mathcal{N}_0\phi_{01}$	$\cup$	$\phi_{0n}^{-1}\mathcal{N}_0\phi_{02}$	$\cup$	$\ldots$	$\cup$	$\phi_{0n}^{-1}\mathcal{N}_0\phi_{0p}$	$\cup$	$\ldots$	$\cup$	$\phi_{0n}^{-1}\mathcal{N}_0\phi_{0n}$

Table 2
Tabular expression of the groupoid in equation (4) where the mixed groups and nuclei are explicitly pointed out.

$M_0 =$	$\mathcal{N}_0$	$\cup$	$\mathcal{N}_0\phi_{01}$	$\cup$	$\mathcal{N}_0\phi_{02}$	$\cup$	$\ldots$	$\cup$	$\mathcal{N}_0\phi_{0p}$	$\cup$	$\ldots$	$\cup$	$\mathcal{N}_0\phi_{0n}$
$M_1 =$	$\mathcal{N}_1\phi_{10}$		$\mathcal{N}_1$	$\cup$	$\mathcal{N}_1\phi_{12}$	$\cup$	$\ldots$	$\cup$	$\mathcal{N}_1\phi_{1p}$	$\cup$	$\ldots$	$\cup$	$\mathcal{N}_1\phi_{1n}$
$M_2 =$	$\mathcal{N}_2\phi_{20}$	$\cup$	$\mathcal{N}_2\phi_{21}$		$\mathcal{N}_2$	$\cup$	$\ldots$	$\cup$	$\mathcal{N}_2\phi_{2p}$	$\cup$	$\ldots$	$\cup$	$\mathcal{N}_2\phi_{2n}$
$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$	$\ldots$
$M_n =$	$\mathcal{N}_n\phi_{n0}$	$\cup$	$\mathcal{N}_n\phi_{n1}$	$\cup$	$\mathcal{N}_n\phi_{n2}$	$\cup$	$\ldots$	$\cup$	$\mathcal{N}_n\phi_{np}$	$\cup$	$\ldots$	$\cup$	$\mathcal{N}_n$

translation period of S along the direction(s) of missing periodicity of the substructure, the indices p and q run over the interval $[0, n-1]$. It can easily be seen that the rth line of Table 1 represents the mixed group M_r of the rth substructure, *i.e.* those partial operations that map any of the substructures to the rth substructure S_r. Clearly, collecting together the partial operations of Γ according to their target substructure S_r, one has

$$\Gamma = \cup_r M_r. \tag{5}$$

In fact, remembering that ϕ_{0r}^{-1} maps S_0 to S_r, we immediately obtain

$$M_r = \cup_p \Phi_{rp} = \cup_p \phi_{0r}^{-1}\mathcal{N}_0\phi_{0p} = \phi_{0r}^{-1}M_0.$$

Moreover, using the fact that $\phi_{0r}^{-1}\mathcal{N}_0\phi_{0r} = \mathcal{N}_r$, one sees that $\phi_{0r}^{-1}\mathcal{N}_0\phi_{0p} = \phi_{0r}^{-1}\mathcal{N}_0\phi_{0r}\phi_{0r}^{-1}\phi_{0p} = \mathcal{N}_r\phi_{0r}^{-1}\ \phi_{0p}$ and hence

$$M_r = \cup_p \mathcal{N}_r\phi_{0r}^{-1}\phi_{0p} = \cup_p \mathcal{N}_r\phi_{rp} \tag{6}$$

which is precisely the same as the rth line in the above decomposition, but expressed with respect to the nucleus $\mathcal{N}_r$ contained in M_r instead of the reference nucleus $\mathcal{N}_0$.

The diagonal terms in Table 1 are the nuclei of the substructures. The off-diagonal terms give the partial operations relating any pair of different substructures. The decomposition in (4) can thus be rewritten as in Table 2. This representation of the groupoid Γ has been called *OD matrix* by Grell (1998), with specific reference to the symmetry description of OD structures built by one kind of layer. Because Γ is obtained from M_0 by multiplying (from the left) with the elements ϕ_{0r}^{-1}, M_0 is called the *generating mixed group of the groupoid*.

The global operations obtained as an extension of a local operation φ_{pp} in each nucleus build the *core* of the groupoid:

$$\mathcal{C} = \cap_p \overline{\mathcal{N}}_p \tag{7}$$

Thus, only those operations of each nucleus that have an extension to all other nuclei end up in the core $\mathcal{C}$. These operations leave invariant all the substructures S_p simultaneously so that $\mathcal{C}$ is a normal subgroup of the space group $\mathcal{G}$. The global operations in $\mathcal{C}$ do not exchange any substructure (since they leave all of them fixed); therefore $\mathcal{C}$ is a subperiodic group, like the nuclei themselves. Therefore, in analogy with the construction of a factor group from a group and one of its normal subgroups (see Appendix *C*), we can construct the factor groupoid $\Gamma/\mathcal{C}$, whose elements are the right cosets $\mathcal{C}\varphi_{qp}$. In general, the product $\mathcal{C}\varphi_{qp}\mathcal{C}\varphi_{sr}$ is not defined because $\mathcal{C}\varphi_{sr}$ maps S_r to S_s, but φ_{qp} needs not be defined on S_s. In order to enforce that all the products $\varphi_{qp}\varphi_{sr}$ for all p, q, r, s exist, one is required to construct the extension $\overline{\varphi}_{qp}^{*}$ for each partial or local operation φ_{qp}. In Section 6 we present the analysis of a concrete case of this.

4. From groupoid to space group

The union of the mixed groups is the groupoid Γ of the structure [equation (6)]. We denote by $\overline{M}_p$ the set of extensions of the partial operations in the mixed group M_p. Let us now consider those extensions $\overline{\varphi}_{pq}$ of partial operations that are extensions obtained from each of the mixed groups, *i.e.* if $\overline{\varphi}_{pq}$ has representation $(\boldsymbol{W}, \boldsymbol{w})$ as a matrix–column pair, then each $\overline{M}_p$ also contains the operation $(\boldsymbol{W}, \boldsymbol{w})$. The set of operations with this property is just the intersection of all $\overline{M}_p$ and forms the space group $\mathcal{G}$ of S or, as we are going to see, the subgroup of it imposed by the arrangement of the substructures:

$$\mathcal{G} = \cap_p \overline{M}_p. \tag{8}$$

In fact, taking the intersection of the $\overline{M}_p$ means identifying those partial operations φ_{qp} that are restrictions of a global operation [represented by the matrix–column pair $(\boldsymbol{W}, \boldsymbol{w})$] to the different modules S_p. These operations may be classified into three types:

(i) Local operations φ_{pp} that can be extended to each $\overline{\mathcal{N}}_q$. These are isometries that are symmetry operations for each of the substructures S_p and therefore also leave the whole structure S invariant.

(ii) Local operations φ_{pp} whose extension occurs in one or more $\overline{\mathcal{N}}_q$ and which is the extension of an operation in the shell of each of the other mixed groups. These isometries leave invariant the structure S as well as (at least) one substructure

S_p, while mapping the other substructures S_q to different substructures S_r. The geometric element of such an operation lies within the subspace occupied by the substructures fixed by the operation.

(iii) Partial operations φ_{qp} whose extension is the extension of an operation in each shell but not of an operation in any nucleus. These are isometries that leave invariant the structure S but none of the substructures; they instead map every substructure S_p onto a different substructure S_q. The geometric element of such an operation does not fully lie within the subspace occupied by any of the substructures.

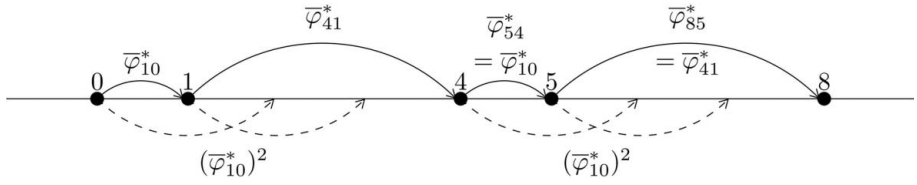


Figure 2
Example of a superposition structure, obtained from a one-dimensional structure with atoms at $x = 0$, $x = 1$ periodically repeated with cell parameter $a = 4$ Å. The translations of lengths $0 + 4n$, $1 + 4n$ and $3 + 4n$ for an integer n are partial operations, but the composition of two translations is not induced by any partial operation.

The partial operations simply determine the way in which the substructures are arranged in the structure, but do not impose additional restrictions on their relative positions, when these are not fully established by the partial operations, as illustrated by the example in Section 6. When some degree of freedom is left in the relative positions of the substructures, special cases may occur that increase the symmetry of the structure to a supergroup $\mathcal{G}_S$ of $\mathcal{G}$: this is not an inherent feature of the topology itself but an additional constraint which it may or may not fulfil. When it is fulfilled, additional operations Φ_a may result from this constraint which extend $\mathcal{G}$:

$$\mathcal{G}_S = \langle \mathcal{G}, \Phi_a \rangle \tag{9}$$

where the angle brackets denote the group of products of elements from $\mathcal{G}$ and Φ_a. In the important case of the close-packing of spheres, discussed in detail in Section 6, the additional constraint that the spheres of neighbouring layers are in contact increases the symmetry from rhombohedral to cubic.

5. Superposition structures

The structural model of a modular structure is easily obtained in a 'bottom-up' procedure from the knowledge of the structure of the modules and of the partial operations acting on them; the real structure may then differ from the structural model obtained in this way by some degree of deformation. In fact, the logical process of building a structural model makes use of ideal, undistorted modules; these, in the real structure, may actually undergo some degree of deformation ('desymmetrization': Ďurovič, 1979). A 'top-down' approach can however also be adopted, where the structural model can be obtained from a common prototype.

As we have seen, partial operations φ_{qp} whose extensions $\overline{\varphi}_{qp}$ are valid for the whole crystal space define the space group of the structure S. We have also seen that we can artificially extend *all* the partial operations to global operations $\overline{\varphi}^*_{qp}$, producing additional substructures S^*_t, which do not make part of the structure S.

The whole set $\{\overline{\varphi}^*_{qp}\}$ of extended operations does not necessarily form a group, since the composition $(\overline{\varphi}^*_{qp})(\overline{\varphi}^*_{sr})$ might not be induced by any partial operation. As an abstract example, take a subperiodic structure along a single line with atoms at $x = 0$, $x = 1$ and then periodically repeated with cell parameter $a = 4$ Å (Fig. 2). There are partial translations of lengths $0 + 4n$, $1 + 4n$ and $3 + 4n$ for an integer n, but the composition of two translations by 1, *i.e.* a translation by 2, is not induced by any partial operation. The composition of extensions can be repeated until the group $\mathcal{G}^*$ consisting of all products of the extensions $\overline{\varphi}^*_{qp}$ for all p and q is obtained. The group $\mathcal{G}^*$ is not necessarily a space group, notably if the translational parts of the partial operations are not commensurate with the lattice of the structure or non-crystallographic rotations are generated. When the group $\mathcal{G}^*$ is applied to all substructures, a *superposition structure*

$$\mathbb{S} = (\cup_p S_p) \cup (\cup_t S^*_t) \tag{10}$$

consisting of real and fictitious substructures is obtained. This superposition structure is in general fictitious because it is atomically overpopulated and contains atomic distances and angles that are physically and chemically unrealistic. However, it is the prototype of all possible structures S built from the substructures S_p by the partial operations φ_{qp}.

6. Example: cubic close-packing of spheres

Structures based on the close-packing of spheres are obtained by stacking a diperiodic layer according to the well known A–B–C mode; the coordination polyhedra generated by the stacking can host various cations and the structure can be described by an alphanumerical symbol that indicates fractional occupancy of each polyhedron [for a recent review, see Umayahara & Nespolo (2018)]. The substructures are diperiodic layers of spheres, whose symmetry is expressed by the diperiodic group $P(6/m)mm$ [notation after Dornberger-Schiff (1959)] or $p6/mmm$ [notation after Kopský & Litvin (2010)]: this is the nucleus of the mixed group. The partial operations are stacking operations mapping one layer onto the next one and can be chosen to be translations. Because two successive layers cannot be stacked directly one on top of the other if the result has to be a compact structure, the translational components of the partial operations in the plane of the layer are $\frac{1}{3}$ and $\frac{2}{3}$ along a and b, whereas the component in the direction perpendicular to the layer is $1/n$, where n is the number of layers in the period of the polytype. In other words, the partial operations are of type 0, 0, m/n (for $m > 1$), $\frac{2}{3}$, $\frac{1}{3}$, m/n and $\frac{1}{3}$, $\frac{2}{3}$, m/n, where n is the number of layers in the period of the polytype and m ($1 \leq m \leq n$) is the sequential number for each layer.

Table 3
Construction of the space groupoid of the ABC close-packing of identical spheres.

$P(6/m)mm$	$P(6/m)mm\ t(\frac{2}{3}\frac{1}{3}\frac{1}{3})$	$P(6/m)mm\ t(\frac{1}{3}\frac{2}{3}\frac{2}{3})$
$t(\frac{2}{3}\frac{1}{3}\frac{1}{3})^{-1}P(6/m)mm$	$t(\frac{2}{3}\frac{1}{3}\frac{1}{3})^{-1}P(6/m)mm\ t(\frac{2}{3}\frac{1}{3}\frac{1}{3})$	$t(\frac{2}{3}\frac{1}{3}\frac{1}{3})^{-1}P(6/m)mm\ t(\frac{1}{3}\frac{2}{3}\frac{2}{3})$
$t(\frac{1}{3}\frac{2}{3}\frac{2}{3})^{-1}P(6/m)mm$	$t(\frac{1}{3}\frac{2}{3}\frac{2}{3})^{-1}P(6/m)mm\ t(\frac{2}{3}\frac{1}{3}\frac{1}{3})$	$t(\frac{1}{3}\frac{2}{3}\frac{2}{3})^{-1}P(6/m)mm\ t(\frac{1}{3}\frac{2}{3}\frac{2}{3})$

The stacking operations simply determine the topology of the layer sequence but do not restrict the vertical separation between diperiodic layers of spheres. In other words, the close-packing is not an inherent feature of the stacking itself but an additional constraint which may or may not be present. When it is present, then additional operations φ_a ('a' for 'additional') may result from this constraint which extend the group of operations obtained from the stacking itself.

Starting from an A layer, the next layer can be B or C and from then on each layer can be either A, B or C. The superposition structure has A, B and C at each occupied height k/n (k and n integers) so that the period of the superposition structure along the stacking direction is one layer. The unit cell of the superposition structure is easily obtained by promoting the partial operations to global operations, and is one-layer thick. By extending the layer group $P(6/m)mm$ by the operations above, one gets the group $\mathcal{G}^* = H6/mmm$;[5] the basis vectors with respect to those of the n-layer polytype are **a**,**b**,**c**/n. Because hH is a non-conventional unit cell, $\mathcal{G}$ can also be described as $P6/mmm$ whose basis vectors, with respect to those of the n-layer polytype, are however (2**a** + **b**)/3, (−**a** + **b**)/3, **c**/n. The use of the non-conventional unit cell hH simplifies the comparison of polytypes.

One of the most common polytypes is 3C, which corresponds to a face-centred cubic structure and is obtained by a monotonous ABC stacking of layers. The A $\not\to$ B generating partial operation is the translation $t(\frac{2}{3}\frac{1}{3}\frac{1}{3})$, which has an extension in B $\not\to$ C as well as in C $\not\to$ A. The A $\not\to$ C generating partial operation is $t(\frac{1}{3}\frac{2}{3}\frac{2}{3})$, which has an extension in B $\not\to$ A as well as in C $\not\to$ B. The structure of the groupoid is therefore straightforward to obtain (Table 3).

$P(6/m)mm$ is the nucleus of layer A with origin at 000. From this, one obtains the nuclei of layers B and C by conjugating it with the stacking operations (which have simple inverses): $t(\overline{\frac{2}{3}\frac{1}{3}\frac{1}{3}})\ P(6/m)mm\ t(\frac{2}{3}\frac{1}{3}\frac{1}{3})$ and $t(\overline{\frac{1}{3}\frac{2}{3}\frac{2}{3}})\ P(6/m)mm\ t(\frac{1}{3}\frac{2}{3}\frac{2}{3})$. The nucleus of the B layer is $P(6/m)mm$ with an origin shift of $\frac{2}{3}\frac{1}{3}\frac{1}{3}$, *i.e.* on the B sphere, and the nucleus of the C layer is $P(6/m)mm$ with an origin shift of $\frac{1}{3}\frac{2}{3}\frac{2}{3}$, *i.e.* on the C sphere.

The operations shown in bold in Table 4 are operations that have an extension in common with the three nuclei. For example, $\{3^+_{001}|000\} = \varphi_{2,AA}$, $\{3^+_{001}|\bar{1}00\} = \varphi_{2,BB}$, $\{3^+_{001}|\bar{1}\bar{1}0\} = \varphi_{2,CC}$, so that $\overline{\varphi}_{2,AA} = \overline{\varphi}_{2,BB} = \overline{\varphi}_{2,CC} = \{3^+_{001}|rs0\}$, where r, s are any integer values. These operations lie in the core of the groupoid, see equation (7), which in this case is the layer group $\mathcal{C} = P(3)m1$. The other operations in the same lines of Table 4 also have an extension to the whole structure S, as can be easily seen from the fact that they differ by a lattice translation: they are therefore also extended to global operations. They occur in the shell of each mixed group and are therefore operations mapping physically distinct layers. Finally, operations in lines Nos. 7, 8, 9, 13, 14 and 15 again can be extended to global operations but occur in the nucleus of one mixed group and in the shell of the two other mixed groups: they fix one layer and interchange the other two layers. The space-group type generated by adding all these operations to the core $P(3)m1$ is $R\bar{3}m$, with the origin on the A sphere, a result that could be expected from the fact that the ABC stacking of spheres is topologically isomorphic to a rhombohedral lattice, whose symmetry is precisely $R\bar{3}m$. However, in a rhombohedral lattice there is a degree of freedom: the c/a ratio between the cell parameters of the conventional hexagonal cell, which translates into the unrestricted α angle of the rhombohedral primitive cell. When building a compact stacking of spheres with the rhombohedral topology such a degree of freedom does not exist, because the spheres are in contact. The height of a tetrahedron whose edge is a_H is $(\frac{2}{3})^{1/2}a_H$ and the period in an ABC stacking is three layers so that the c parameter of the conventional cell is $6^{1/2}a_H$. The axial transformation from the hexagonal cell with edges a_H and $c_H = 6^{1/2}a_H$ gives a rhombohedral cell with cell parameters $a_R = a_H$ and $\alpha = 60°$, which is simply the primitive cell of a lattice whose conventional cell is cF with cell parameter $a_F = 2^{1/2}a_R = 2^{1/2}a_H$ and where $a_R = a_H$ is the diameter of the sphere. As a consequence, the $R\bar{3}m$ symmetry obtained from the groupoid analysis and characteristic of an unrestricted packing of diperiodic layers of spheres is promoted to $Fm\bar{3}m$ when the layers are brought into contact. This additional symmetry does not come from the ABC topology: the operations in the coset decomposition of $Fm\bar{3}m$ with respect to $R\bar{3}m$ are therefore considered additional symmetry operations. The coordinate transformation from $R\bar{3}m$ in hexagonal axes to $Fm\bar{3}m$ via the intermediate rhombohedral axes $(\frac{1}{2}01/\frac{11}{22}1/0\frac{1}{2}1)$, transforming 000, $\frac{2}{3}\frac{1}{3}\frac{1}{3}$ and $\frac{1}{3}\frac{1}{3}\frac{2}{3}$ to 000, $0\frac{11}{22}$ and $\frac{11}{22}1$, respectively, correspond to position 4a in $Fm\bar{3}m$ and to the three spheres black (A), white (B) and grey (C) in Fig. 3. The change in the multiplicity of the Wyckoff position comes from the change in the multiplicity of the unit cell: the transformation matrix has determinant $\frac{3}{4}$, the inverse matrix – which transforms the axes – has determinant 4/3 and thus the cubic unit cell has a volume 4/3 times bigger than the hR unit cell in hexagonal axes, which corresponds to

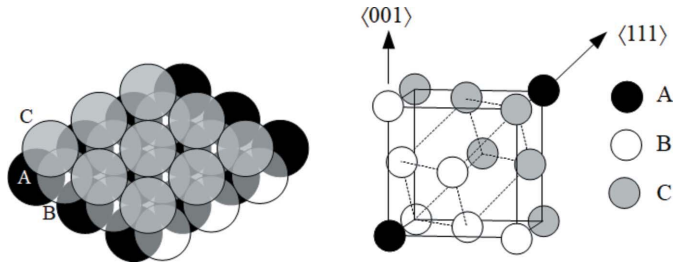


Figure 3
Cubic close-packing of spheres (ABC) seen along the direction of the stacking (left) and in perspective (right) where the cubic unit cell is drawn.

[5] We remind the reader that the unit cell of type H is a non-conventional unit cell with centring vectors $t(\frac{2}{3}\frac{1}{3}0)$ and $t(\frac{1}{3}\frac{2}{3}0)$.

Table 4
Construction of the space groupoid of the ABC close-packing of identical spheres.

Operations are expressed by their Seitz symbol. M_0, M_1 and M_2 are the three mixed groups. The operations in bold are local operations that have extensions to the whole structure S: they define the core $\mathcal{C}$ of the groupoid.

	M_0			M_1			M_2		
No.	$P(6/m)mm$	$P(6/m)mm$ $t(\frac{2}{3}\frac{1}{3}\frac{1}{3})$	$P(6/m)mm$ $t(\frac{1}{3}\frac{2}{3}\frac{2}{3})$	$t(\frac{2}{3}\frac{1}{3}\frac{1}{3})^{-1}$ $P(6/m)mm$	$t(\frac{2}{3}\frac{1}{3}\frac{1}{3})^{-1}P(6/m)mm$ $t(\frac{2}{3}\frac{1}{3}\frac{1}{3})$	$t(\frac{2}{3}\frac{1}{3}\frac{1}{3})^{-1}P(6/m)mm$ $t(\frac{1}{3}\frac{2}{3}\frac{2}{3})$	$t(\frac{1}{3}\frac{2}{3}\frac{2}{3})^{-1}P(6/m)mm$	$t(\frac{1}{3}\frac{2}{3}\frac{2}{3})^{-1}P(6/m)mm$ $t(\frac{2}{3}\frac{1}{3}\frac{1}{3})$	$t(\frac{1}{3}\frac{2}{3}\frac{2}{3})^{-1}P(6/m)mm$ $t(\frac{1}{3}\frac{2}{3}\frac{2}{3})$
1	**{1|000}**	$\{1\|\frac{1}{3}(211)\}$	$\{1\|\frac{1}{3}(122)\}$	$\{1\|\frac{1}{3}(\overline{2}\overline{1}1)\}$	**{1|000}**	$\{1\|\frac{1}{3}(\overline{1}11)\}$	$\{1\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{1\|\frac{1}{3}(1\overline{1}\overline{1})\}$	**{1|000}**
2	**$\{3^+_{001}\|000\}$**	$\{3^+_{001}\|\frac{1}{3}(\overline{1}11)\}$	$\{3^+_{001}\|\frac{1}{3}(\overline{2}12)\}$	$\{3^+_{001}\|\frac{1}{3}(\overline{2}11)\}$	**$\{3^+_{001}\|\overline{1}\overline{1}00\}$**	$\{3^+_{001}\|\frac{1}{3}(\overline{4}21)\}$	$\{3^+_{001}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{3^+_{001}\|\frac{1}{3}(\overline{2}11)\}$	**$\{3^+_{001}\|\overline{1}\overline{1}0\}$**
3	**$\{3^-_{001}\|000\}$**	$\{3^-_{001}\|\frac{1}{3}(\overline{1}21)\}$	$\{3^-_{001}\|\frac{1}{3}(1\overline{1}2)\}$	$\{3^-_{001}\|\frac{1}{3}(\overline{2}11)\}$	**$\{3^-_{001}\|\overline{1}\overline{1}0\}$**	$\{3^-_{001}\|\frac{1}{3}(\overline{1}21)\}$	$\{3^-_{001}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{3^-_{001}\|\frac{1}{3}(\overline{2}41)\}$	**$\{3^-_{001}\|0\overline{1}0\}$**
4	$\{2_{001}\|000\}$	$\{2_{001}\|\frac{1}{3}(\overline{2}11)\}$	$\{2_{001}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{2_{001}\|\frac{1}{3}(\overline{2}11)\}$	$\{2_{001}\|\frac{1}{3}(\overline{4}20)\}$	$\{2_{001}\|\frac{1}{3}(\overline{3}31)\}$	$\{2_{001}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{2_{001}\|\frac{1}{3}(\overline{3}31)\}$	$\{2_{001}\|\frac{1}{3}(\overline{2}40)\}$
5	$\{6^-_{001}\|000\}$	$\{6^-_{001}\|\frac{1}{3}(1\overline{1}1)\}$	$\{6^-_{001}\|\frac{1}{3}(212)\}$	$\{6^-_{001}\|\frac{1}{3}(\overline{2}11)\}$	$\{6^-_{001}\|\frac{1}{3}(\overline{1}20)\}$	$\{6^-_{001}\|\frac{1}{3}(001)\}$	$\{6^-_{001}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{6^-_{001}\|\frac{1}{3}(0\overline{3}1)\}$	$\{6^-_{001}\|\frac{1}{3}(1\overline{1}0)\}$
6	$\{6^+_{001}\|000\}$	$\{6^+_{001}\|\frac{1}{3}(121)\}$	$\{6^+_{001}\|\frac{1}{3}(\overline{1}12)\}$	$\{6^+_{001}\|\frac{1}{3}(\overline{2}11)\}$	$\{6^+_{001}\|\frac{1}{3}(\overline{1}10)\}$	$\{6^+_{001}\|\frac{1}{3}(\overline{3}01)\}$	$\{6^+_{001}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{6^+_{001}\|\frac{1}{3}(00\overline{1})\}$	$\{6^+_{001}\|\frac{1}{3}(\overline{2}10)\}$
7	$\{2_{110}\|000\}$	$\{2_{110}\|\frac{1}{3}(12\overline{1})\}$	$\{2_{110}\|\frac{1}{3}(21\overline{2})\}$	$\{2_{110}\|\frac{1}{3}(\overline{2}11)\}$	$\{2_{110}\|\frac{1}{3}(\overline{1}\overline{1}2)\}$	$\{2_{110}\|00\overline{1}\}$	$\{2_{110}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{2_{110}\|00\overline{1}\}$	$\{2_{110}\|\frac{1}{3}(1\overline{1}4)\}$
8	$\{2_{100}\|000\}$	$\{2_{100}\|\frac{1}{3}(1\overline{1}1)\}$	$\{2_{100}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{2_{100}\|\frac{1}{3}(\overline{2}11)\}$	$\{2_{100}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{2_{100}\|\overline{1}11\}$	$\{2_{100}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{2_{100}\|0\overline{1}1\}$	$\{2_{100}\|\frac{1}{3}(\overline{2}44)\}$
9	$\{2_{010}\|000\}$	$\{2_{010}\|\frac{1}{3}(\overline{2}11)\}$	$\{2_{010}\|\frac{1}{3}(\overline{1}\overline{1}2)\}$	$\{2_{010}\|\frac{1}{3}(\overline{2}11)\}$	$\{2_{010}\|\frac{1}{3}(\overline{4}22)\}$	$\{2_{010}\|\overline{1}0\overline{1}\}$	$\{2_{010}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{2_{010}\|\overline{1}11\}$	$\{2_{010}\|\frac{1}{3}(\overline{2}14)\}$
10	$\{2_{1\bar{1}0}\|000\}$	$\{2_{1\bar{1}0}\|\frac{1}{3}(\overline{1}21)\}$	$\{2_{1\bar{1}0}\|\frac{1}{3}(\overline{2}12)\}$	$\{2_{1\bar{1}0}\|\frac{1}{3}(\overline{2}11)\}$	$\{2_{1\bar{1}0}\|\frac{1}{3}(\overline{3}32)\}$	$\{2_{1\bar{1}0}\|\frac{1}{3}(\overline{4}23)\}$	$\{2_{1\bar{1}0}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{2_{1\bar{1}0}\|\frac{1}{3}(\overline{2}43)\}$	$\{2_{1\bar{1}0}\|\frac{1}{3}(\overline{3}34)\}$
11	$\{2_{120}\|000\}$	$\{2_{120}\|\frac{1}{3}(\overline{1}1\overline{1})\}$	$\{2_{120}\|\frac{1}{3}(12\overline{2})\}$	$\{2_{120}\|\frac{1}{3}(\overline{2}11)\}$	$\{2_{120}\|\frac{1}{3}(\overline{3}0\overline{2})\}$	$\{2_{120}\|\frac{1}{3}(\overline{1}\overline{1}3)\}$	$\{2_{120}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{2_{120}\|\frac{1}{3}(\overline{2}13)\}$	$\{2_{120}\|\frac{1}{3}(00\overline{4})\}$
12	$\{2_{210}\|000\}$	$\{2_{210}\|\frac{1}{3}(21\overline{1})\}$	$\{2_{210}\|\frac{1}{3}(1\overline{1}2)\}$	$\{2_{210}\|\frac{1}{3}(\overline{2}11)\}$	$\{2_{210}\|\frac{1}{3}(00\overline{2})\}$	$\{2_{210}\|\frac{1}{3}(\overline{1}23)\}$	$\{2_{210}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{2_{210}\|\frac{1}{3}(\overline{1}13)\}$	$\{2_{210}\|\frac{1}{3}(0\overline{3}4)\}$
13	$\{\bar{1}\ \|000\}$	$\{\bar{1}\ \|\frac{1}{3}(\overline{2}11)\}$	$\{\bar{1}\ \|\frac{1}{3}(\overline{1}22)\}$	$\{\bar{1}\ \|\frac{1}{3}(\overline{2}11)\}$	$\{\bar{1}\ \|\frac{1}{3}(\overline{4}22)\}$	$\{\bar{1}\ \|\overline{1}11\}$	$\{\bar{1}\ \|\frac{1}{3}(\overline{1}22)\}$	$\{\bar{1}\ \|\overline{1}11\}$	$\{\bar{1}\ \|\frac{1}{3}(\overline{2}44)\}$
14	$\{\bar{3}^+_{001}\|000\}$	$\{\bar{3}^+_{001}\|\frac{1}{3}(1\overline{1}\overline{1})\}$	$\{\bar{3}^+_{001}\|\frac{1}{3}(21\overline{2})\}$	$\{\bar{3}^+_{001}\|\frac{1}{3}(\overline{2}11)\}$	$\{\bar{3}^+_{001}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{\bar{3}^+_{001}\|00\overline{1}\}$	$\{\bar{3}^+_{001}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{\bar{3}^+_{001}\|0\overline{1}1\}$	$\{\bar{3}^+_{001}\|\frac{1}{3}(\overline{1}\overline{1}4)\}$
15	$\{\bar{3}^-_{001}\|000\}$	$\{\bar{3}^-_{001}\|\frac{1}{3}(12\overline{1})\}$	$\{\bar{3}^-_{001}\|\frac{1}{3}(1\overline{1}\overline{2})\}$	$\{\bar{3}^-_{001}\|\frac{1}{3}(\overline{2}11)\}$	$\{\bar{3}^-_{001}\|\frac{1}{3}(\overline{1}\overline{1}2)\}$	$\{\bar{3}^-_{001}\|\overline{1}0\overline{1}\}$	$\{\bar{3}^-_{001}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{\bar{3}^-_{001}\|00\overline{1}\}$	$\{\bar{3}^-_{001}\|\frac{1}{3}(\overline{2}14)\}$
16	$\{m_{001}\|000\}$	$\{m_{001}\|\frac{1}{3}(21\overline{1})\}$	$\{m_{001}\|\frac{1}{3}(12\overline{2})\}$	$\{m_{001}\|\frac{1}{3}(\overline{2}11)\}$	$\{m_{001}\|\frac{1}{3}(00\overline{2})\}$	$\{m_{001}\|\frac{1}{3}(\overline{1}\overline{1}3)\}$	$\{m_{001}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{m_{001}\|\frac{1}{3}(1\overline{1}3)\}$	$\{m_{001}\|\frac{1}{3}(00\overline{4})\}$
17	$\{\bar{6}^-_{001}\|000\}$	$\{\bar{6}^-_{001}\|\frac{1}{3}(\overline{1}1\overline{1})\}$	$\{\bar{6}^-_{001}\|\frac{1}{3}(\overline{2}12)\}$	$\{\bar{6}^-_{001}\|\frac{1}{3}(\overline{2}11)\}$	$\{\bar{6}^-_{001}\|\frac{1}{3}(\overline{3}0\overline{2})\}$	$\{\bar{6}^-_{001}\|\frac{1}{3}(\overline{4}23)\}$	$\{\bar{6}^-_{001}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{\bar{6}^-_{001}\|\frac{1}{3}(\overline{2}13)\}$	$\{\bar{6}^-_{001}\|\frac{1}{3}(\overline{3}34)\}$
18	$\{\bar{6}^+_{001}\|000\}$	$\{\bar{6}^+_{001}\|\frac{1}{3}(\overline{1}21)\}$	$\{\bar{6}^+_{001}\|\frac{1}{3}(1\overline{1}2)\}$	$\{\bar{6}^+_{001}\|\frac{1}{3}(\overline{2}11)\}$	$\{\bar{6}^+_{001}\|\frac{1}{3}(\overline{3}32)\}$	$\{\bar{6}^+_{001}\|\frac{1}{3}(\overline{1}23)\}$	$\{\bar{6}^+_{001}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{\bar{6}^+_{001}\|\frac{1}{3}(\overline{2}43)\}$	$\{\bar{6}^+_{001}\|\frac{1}{3}(\overline{0}34)\}$
19	**$\{m_{110}\|000\}$**	$\{m_{110}\|\frac{1}{3}(\overline{1}21)\}$	$\{m_{110}\|\frac{1}{3}(\overline{2}12)\}$	$\{m_{110}\|\frac{1}{3}(\overline{2}11)\}$	**$\{m_{110}\|\overline{1}\overline{1}0\}$**	$\{m_{110}\|\frac{1}{3}(\overline{4}21)\}$	$\{m_{110}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{m_{110}\|\frac{1}{3}(\overline{2}41)\}$	**$\{m_{110}\|\overline{1}\overline{1}0\}$**
20	**$\{m_{100}\|000\}$**	$\{m_{100}\|\frac{1}{3}(\overline{1}11)\}$	$\{m_{100}\|\frac{1}{3}(122)\}$	$\{m_{100}\|\frac{1}{3}(\overline{2}11)\}$	**$\{m_{100}\|\overline{1}00\}$**	$\{m_{100}\|\frac{1}{3}(\overline{1}11)\}$	$\{m_{100}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{m_{100}\|\frac{1}{3}(\overline{2}11)\}$	**$\{m_{100}\|000\}$**
21	**$\{m_{010}\|000\}$**	$\{m_{010}\|\frac{1}{3}(211)\}$	$\{m_{010}\|\frac{1}{3}(1\overline{1}2)\}$	$\{m_{010}\|\frac{1}{3}(\overline{2}11)\}$	**$\{m_{010}\|000\}$**	$\{m_{010}\|\frac{1}{3}(\overline{1}21)\}$	$\{m_{010}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{m_{010}\|\frac{1}{3}(\overline{1}11)\}$	**$\{m_{010}\|0\overline{1}0\}$**
22	$\{m_{1\bar{1}0}\|000\}$	$\{m_{1\bar{1}0}\|\frac{1}{3}(121)\}$	$\{m_{1\bar{1}0}\|\frac{1}{3}(212)\}$	$\{m_{1\bar{1}0}\|\frac{1}{3}(\overline{2}11)\}$	$\{m_{1\bar{1}0}\|\frac{1}{3}(\overline{1}10)\}$	$\{m_{1\bar{1}0}\|\frac{1}{3}(001)\}$	$\{m_{1\bar{1}0}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{m_{1\bar{1}0}\|\frac{1}{3}(00\overline{1})\}$	$\{m_{1\bar{1}0}\|\frac{1}{3}(1\overline{1}0)\}$
23	$\{m_{120}\|000\}$	$\{m_{120}\|\frac{1}{3}(1\overline{1}1)\}$	$\{m_{120}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{m_{120}\|\frac{1}{3}(\overline{2}11)\}$	$\{m_{120}\|\frac{1}{3}(\overline{1}20)\}$	$\{m_{120}\|\frac{1}{3}(\overline{3}31)\}$	$\{m_{120}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{m_{120}\|\frac{1}{3}(0\overline{3}1)\}$	$\{m_{120}\|\frac{1}{3}(\overline{2}40)\}$
24	$\{m_{210}\|000\}$	$\{m_{210}\|\frac{1}{3}(\overline{2}11)\}$	$\{m_{210}\|\frac{1}{3}(\overline{1}12)\}$	$\{m_{210}\|\frac{1}{3}(\overline{2}11)\}$	$\{m_{210}\|\frac{1}{3}(\overline{4}20)\}$	$\{m_{210}\|\frac{1}{3}(\overline{3}01)\}$	$\{m_{210}\|\frac{1}{3}(\overline{1}\overline{2}2)\}$	$\{m_{210}\|\frac{1}{3}(\overline{3}31)\}$	$\{m_{210}\|\frac{1}{3}(\overline{2}10)\}$

the change in the multiplicity of the Wyckoff position from 3*a* in $R\bar{3}m$ to 4*a* in $Fm\bar{3}m$.

7. Discussion and relation to OD theory

The groupoid analysis of modular structures allows an in-depth analysis and understanding of the structural relations between the modules building the structure. It also provides a key for structure prediction and structure design from the knowledge of the structure of the modules and of the partial operations mapping different modules. The starting point is, however, the choice of the modules, which in general is not unique. As mentioned in Section 3, a substructure S_p can sometimes be further subdivided into sub-substructures S_{pq}. The opposite process is also possible: a set of modules can be considered as a single, larger module. Last but not least, a structure can sometimes be subdivided into different modules of the same size (Grell, 1984). There exists indeed, in general, a certain degree of ambiguity in the choice of the modules, which is, however, not arbitrary. The following natural guidelines apply:

(i) Chemically identified modules (*e.g.* modules with stronger intra-module than inter-module bondings) are usually the starting point in a modular analysis.

(ii) An alternative choice, which does not respect the chemical identity of the module, is sometimes useful to reduce the number of *types* of modules and simplify the groupoid description (see, *e.g.*, Durovic, 1974).

(iii) Splitting a module into small building blocks may be useful when these smaller blocks also occur in other structures, *e.g.* in a polysomatic series (Nespolo *et al.*, 2018).

(iv) Conversely, sets of modules can be regarded as a single module if this perspective brings some advantage, for example when different types of modules occurring regularly in the structure can be regarded as a single type of module.

A different approach to the groupoid analysis of modular structures was introduced by Dornberger-Schiff & Grell-Niemann (1961) under the name order–disorder (OD) theory, which is the theory of a subclass of modular structures built of diperiodic layers, though generalizations to rods and blocks have been proposed (Dornberger-Schiff, 1964; Belokoneva, 2005). OD structures fulfil the vicinity condition (VC), which

states that equivalent sides of equivalent layers contact in such a way that pairs of adjacent layers are geometrically equivalent. In OD structures, the stacking of layers can be periodic ('ordered') or nonperiodic ('disordered'), but the stacking mode is restricted to arrangements that respect the geometric equivalence. Periodic OD structures are polytypes, but the opposite is not necessarily true. All possible stacking arrangements of an OD structure are locally (at least up to pairs of layers) equivalent. They are therefore, if desymmetrization and interatomic interactions beyond one layer width are ignored, also energetically equivalent, which explains the high likelihood of OD structures featuring twinning or stacking disorder. The choice of OD layers is in general not unique and does not necessarily follow crystallo-chemical considerations (Grell, 1984).

In OD theory, all operations acting on layers are designated as partial operations (POs). The local symmetry operations mapping a layer onto itself are specified as λ-POs (where λ stands for layer), partial operations relating different layers as σ-POs. The λ-POs and σ-POs correspond to φ_{pp} and φ_{qp} in our approach, respectively, and define the groupoid of the OD structure, which is called an OD groupoid because it only allows those POs that preserve the geometric equivalence of layer pairs characterizing an OD structure. As before, if OD structures are built from different types of layers, the OD groupoid is disconnected and composed of one connected component per type of layer.

The group Γ^* generated by the extensions of all POs of a modular structure has been called superposition group in the OD literature (Fichtner, 1977*a*). The corresponding superposition structure $\mathbb{S}$ is generally known as the family structure, because all members of an OD family can be derived from it. Since this does not hold for non-OD polytype families, we do not use the term in this context. On the other hand, the term superposition structure is sometimes used in a more restricted sense in the OD literature, since it only designates superpositions of polytypes related by translation (Ďurovič, 1994). However, the family structure can not always be constructed by superposition of translated copies of a given polytype.

All possible stacking arrangements of the same layers which preserve the geometric equivalence of layer pairs, *i.e.* all possible OD structures built by the same type of layers, defines an OD family: this is a family of structures. All OD groupoids that describe stacking arrangements built according to the same symmetry principle (Fichtner, 1979), *i.e.* the same type of layers and the same type of orientation relationship between layers, belong to the same OD groupoid family. OD groupoid families abstract from particular stacking arrangements and metric parameters. They thus play the role of the 230 space-group types in classical crystallography and are represented by a symbol that is constructed on the same principles as the Hermann–Mauguin symbol for space groups and contains a number of parameters; these can be seen as a generalization of the Hermann–Mauguin symbols. In general, there is an infinity of OD groupoid families. For the special case of OD structures built of a single type of layers and all layers possessing the same two-dimensional translation lattice, 400 OD groupoid families have been identified (Fichtner, 1977*b*). The OD groupoids corresponding to a specific OD family can be obtained by assigning precise values to the parameters occurring in the symbol, the cell parameters of a layer and the layer widths (Fichtner, 1979).

The OD groupoid family of the close-packing of spheres is described by the symbol:

$$\begin{Bmatrix} P & m & m & m & (6/m) & m & m & m \\ & & & & \begin{pmatrix} 2_2 \\ 3_3 \\ \overline{3} \\ 6_6 \\ \overline{6} \\ n_{r,s} \end{pmatrix} & & & \\ & 2_{r-\frac{s}{2}} & 2_{-\frac{r}{2}+s} & 2_{\frac{r}{2}+\frac{s}{2}} & & 2_{\frac{s}{2}} & 2_{\frac{r}{2}} & 2_{\frac{r}{2}-\frac{s}{2}} \\ & n_{\frac{s}{2},2} & n_{2,\frac{r}{2}} & n_{2,\frac{r}{2}-\frac{s}{2}} & & n_{2,r-\frac{s}{2}} & n_{2,-\frac{r}{2}+s} & n_{2,\frac{r}{2}+\frac{s}{2}} \end{Bmatrix}$$

The first line indicates the symmetry of the layers (λ-POs). The bottom indicates the operations relating two adjacent layers (σ-POs) in one possible stacking arrangement. A seven-placed symbol is used because the σ-POs are in general different in the six special directions of the hexagonal layer lattice. The symbol $n_{r,s}$ is used in the OD literature to indicate a glide reflection with non-standard translation component: the *International Tables for Crystallography* would rather use the symbol $g(2r,2s,0)$ (or the result of a permutation, depending on the orientation of the plane). Similarly, symbols like 2_p, 3_p *etc*. indicate a twofold, threefold *etc*. screw rotation with screw component $q/2$, $q/3$ *etc*. For the OD family of sphere packings, r and s adopt the values $(r, s) = (2/3, 4/3)$, so that the symbol simplifies to

$$\begin{Bmatrix} P & m & m & m & (6/m) & m & m & m \\ & & & & \begin{pmatrix} 2_2 \\ 3_3 \\ \overline{3} \\ 6_6 \\ \overline{6} \\ n_{\frac{2}{3},\frac{4}{3}} \end{pmatrix} & & & \\ & 2 & 2_1 & 2_1 & & 2_{\frac{2}{3}} & 2_{\frac{1}{3}} & 2_{-\frac{1}{3}} \\ & n_{\frac{2}{3},2} & n_{2,\frac{1}{3}} & n_{2,-\frac{1}{3}} & & c_2 & n_{2,1} & n_{2,1} \end{Bmatrix}$$

For this special subset of the OD groupoid family, given a layer, the adjacent layers can be placed in two ways, corresponding to the well known A–B–C stacking modes of the close sphere packings.

Whereas OD groupoid family symbols are often rather informative, in a case like the present one, the symbol which describes a groupoid family containing the groupoid in Table 4 is not particularly helpful.

APPENDIX *A*
Glossary

S: a modular crystal structure built by subperiodic (zero-, one- or two-periodic) substructures (modules) S_p, where p is an integer index enumerating the different modules in *S*.

φ_{qp}: a partial operation mapping the pth substructure (source) S_p onto the qth substructure (target) S_q; φ_{qp} acts only on S_p and produces only S_q; when $p = q$, the source and the target coincide and the partial operation φ_{pp} represents a symmetry operation of the pth substructure, that is called a local operation.

Φ_{qp}: the set $\{\varphi_{qp}\}$ of partial operations mapping S_p to S_q.

$\mathcal{N}_p$: the set Φ_{pp} considered as the group of local operations that form the symmetry group of S_p; $\mathcal{N}_p$ is called a nucleus.

ϕ_{0p}: one of the operations in Φ_{0p} that has been selected and used as coset representative in the decomposition of the mixed group M_0 with respect to the nucleus $\mathcal{N}_0$.

$\overline{\varphi}^*_{qp}$: an operation obtained when the partial operation φ_{qp} is extended to act on the whole crystal space; it does not necessarily correspond to a global symmetry operation of the full modular structure S; if it does, this is denoted as $\overline{\varphi}_{qp}$ (omitting *).

S^*_t: a fictitious substructure obtained by applying the extension $\overline{\varphi}^*_{qp}$ of some partial operation φ_{qp} to some S_r (* being used to indicate that the substructure is fictitious).

$\mathbb{S}$: a superposition structure composed of all the real substructures S_p and fictitious substructures S^*_t obtained when the group generated by the extensions of all partial operations φ_{qp} of the modular structure is applied to all of its substructures.

APPENDIX *B*

The notion of groupoid used in this article corresponds to Brandt's definition (Brandt, 1927) which generalizes the notion of a group by replacing the binary group operation by a partial composition. According to this definition, a groupoid is a set Γ together with a composition * mapping a pair (α, β) in $\Gamma \times \Gamma$ to an element $\alpha^*\beta$ in Γ, but without the requirement that * is defined on all pairs in $\Gamma \times \Gamma$, such that the following axioms are fulfilled:

(i) *Associativity*: if $\alpha^*\beta$ is defined and $\beta^*\gamma$ is defined, then also $(\alpha^*\beta)^*\gamma$ and $\alpha^*(\beta^*\gamma)$ are defined and are equal.

(ii) *Unit elements*: for each α in Γ there exists a unique left unit element ϵ and a unique right unit element ϵ' such that $\epsilon^*\alpha$ and $\alpha^*\epsilon'$ are defined and $\epsilon^*\alpha = \alpha$ and $\alpha^*\epsilon' = \alpha$ holds.

(iii) *Inverse elements*: each α in Γ has an inverse element α^{-1} such that $\alpha^*\alpha^{-1}$ and $\alpha^{-1}*\alpha$ are defined and such that $\alpha^*\alpha^{-1} = \epsilon$ is the left unit of α and $\alpha^{-1}*\alpha = \epsilon'$ is the right unit of α.

As a consequence of theses axioms, two elements α and β can be composed if the right unit of α coincides with the left unit of β. In view of his applications, Brandt added the transitivity axiom given below which was later eliminated by C. Ehresmann in his more general approach to groupoids (see Ehresmann, 1957).

(iv) *Transitivity*: if ϵ and ϵ' are two unit elements in Γ, there exists an element α in Γ which has ϵ as its left and ϵ' as its right unit element.

In the modern perspective, groupoids are considered in terms of category theory. Without going into the formal details of categories, the basic notions reflect why groupoids are suitable for the description of modular structures. In this language, a groupoid consists of a set of objects and a set of morphisms between these objects. For a morphism φ mapping object p to object q, p is called the source and q the target of φ. Two morphisms φ and φ' can be composed if and only if the target of φ coincides with the source of φ'. In that case, the composition $\varphi'^*\varphi$ has as its source the source of φ and as its target the target of φ'. The axioms imposed on the groupoid are now as follows:

(i) The composition of morphisms is associative (if it is defined).

(ii) For every object p there exists an identity morphism i_p which has p as its source and target, such that $\mathrm{i}_p{}^*\varphi = \varphi$ for every morphism φ with target p and $\varphi'^*\mathrm{i}_p = \varphi'$ for every morphism φ' with source p.

(iii) For every morphism φ with source p and target q, there is an inverse morphism φ^{-1} with source q and target p and such that $\varphi^{-1}*\varphi = \mathrm{i}_p$ and $\varphi^*\varphi^{-1} = \mathrm{i}_q$.

The transitivity axiom of Brandt requires that for any pair (p, q) of objects there exists a morphism having p as its source and q as its target. Note that a group is the special case of a groupoid with a single object.

In the context of modular structures, the objects are identified with the substructures S_p and the morphisms are the partial operations. Local operations are morphisms having the same source and target.

APPENDIX *C*

A group $\mathcal{G}$ can be decomposed in terms of any of its subgroups $\mathcal{H}$ by forming left cosets $g\mathcal{H} = \{gh | h \in \mathcal{H}\}$ or right cosets $\mathcal{H}g' = \{hg' | h \in \mathcal{H}\}$:

$$\mathcal{G} = g_1\mathcal{H} \cup g_2\mathcal{H} \cup g_3\mathcal{H} \cup \ldots \cup g_n\mathcal{H} \cup \ldots \qquad (11)$$

$$\mathcal{G} = \mathcal{H}g'_1 \cup \mathcal{H}g'_2 \cup \mathcal{H}g'_3 \cup \ldots \cup \mathcal{H}g'_n \cup \ldots \qquad (12)$$

where g_i is not contained in $g_j\mathcal{H}$ for any $i \neq j$ and similarly g'_i is not contained in $\mathcal{H}g'_j$ for any $i \neq j$. The elements g_i are called the left coset representatives and correspondingly the g'_i are called the right coset representatives of $\mathcal{G}$ with respect to $\mathcal{H}$. If the number of cosets is finite, it is called the index of $\mathcal{H}$ in $\mathcal{G}$. This is in particular the case for a finite group $\mathcal{G}$ of order $|\mathcal{G}|$; in this case the index is just the ratio $|\mathcal{G}|/|\mathcal{H}|$ of the orders of $\mathcal{G}$ and $\mathcal{H}$. In general, the decompositions (11) and (12) are not identical; however, if $g\mathcal{H}g^{-1} = \mathcal{H}$, *i.e.* if $\mathcal{H}$ is a normal subgroup of $\mathcal{G}$, then each left coset $g_j\mathcal{H}$ is equal to one of the right cosets $\mathcal{H}g'_j$. If $\mathcal{H}$ is normal in $\mathcal{G}$, one can define a multiplication on the cosets by multiplying the respective coset representatives: $(g_i\mathcal{H})(g_j\mathcal{H}) = g_ig_j\mathcal{H}$. The fact that $\mathcal{H}$ is a normal subgroup makes sure that this product does not depend on the chosen coset representatives, and since $g_i^{-1}\mathcal{H}$ is the inverse element of $g_i\mathcal{H}$ with respect to this multiplication, the cosets form a group, called the factor group (or quotient group) $\mathcal{G}/\mathcal{H}$ of $\mathcal{G}$ by $\mathcal{H}$.

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