

# Structural analysis of twins in feldspars. I. Carlsbad twinning

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**Abstract:** In this article, we analyse the common substructure of the Carlsbad twin in terms of the (pseudo)-eigensymmetry of the crystallographic orbits. In the case of the monoclinic polymorphs, all the orbits in the highest subgroup compatible with the twin lattice possess a (pseudo)-eigensymmetry which contains a restoration operation corresponding to the twin operation, which implies a high degree of structural restoration. This structural restoration is realised for twin elements located at distance one half the length of the unit cell, which explains the high occurrence frequency of this twin and its occurrence as both contact and penetration twin, and as primary or synneusis twin. In the case of triclinic polymorphs, the common substructure is much less satisfactory and the Carlsbad twin is probably inherited from the monoclinic phases, accompanied by transformation twins, or by late-stage synneusis, where mechanical adherence on prominent faces plays a more fundamental role.

**Key-words:** albite; Carlsbad; hemitropy; orthoclase; synneusis; twinning; feldspar.

## 1. Introduction

Feldspars are tectosilicates with general formula  $M^{p+}Al_pSi_{4-p}O_8$ , where  $M = K^+$  (orthoclase),  $Na^+$  (albite),  $Ca^{2+}$  (anorthite),  $Ba^{2+}$  (celsian),  $NH_4^+$  (buddingtonite). They frequently occur as twins, either as reflection twins – the most common laws being Albite (0 1 0), Manebach (0 0 1) and Baveno (0 2 1) – or as rotation twins – mainly Ala [1 0 0], Pericline [0 1 0] and Carlsbad [0 0 1]; several other twin laws have been reported but are rarer or doubtful (Smith, 1974). From the petrological viewpoint, more fundamental is the composition plane, *i.e.* the plane representing the physical interface between the twinned individuals. Albite and Carlsbad have (0 1 0) as composition plane, Manebach and Ala (0 0 1), Pericline the rhombic section and Baveno (0 2 1). Twins with composition plane (0 1 0) are most typically represented in magmatic rocks, whereas in metamorphic rocks the predominance depends on the facies: twins with composition plane (0 0 1) or the rhombic section dominate at higher pressure, whereas those with composition plane (0 1 0) are more frequent at lower pressure, *i.e.* approaching magmatic conditions (Tobi, 1987).

Twins may occur following a phase transition (transformation twins), as the result of an external force (mechanical twins) or during crystal growth (growth twins) (Buerger, 1945). Albite and Pericline laws are possible only in triclinic feldspars, because in monoclinic polymorphs the (0 1 0) plane and the [0 1 0] axis are symmetry elements. When they occur as transformation

twins from monoclinic sanidine/orthoclase to triclinic microcline, they coexist and produce the well-known cross-hatched pattern observed under the polarising microscope, unless recrystallisation takes place (Akizuki, 1972). Mechanical twins occur as lamellar twinning in plagioclase in the amphibolite facies, with preference for composition plane (0 0 1) or the rhombic section, but are rare under low-grade metamorphism (Tobi, 1987). Growth twins occur as the result of a perturbation (impurities, dislocations or other defects) during the crystal growth, leading to so-called primary twins attributed to nucleation errors or to nucleation on the surface of a growing crystal (Kumar *et al.*, 2014), or following the oriented attachment of crystals with a precise mutual orientation (Nespolo, 2015); the oriented attachment, also known as synneusis (Vogt, 1921), may occur at any stage, including the post-growth (Nespolo & Ferraris, 2004). Synneusis is realised when nearby crystals drift together, become attached and adhere on their broader crystal faces. The phenomenon can involve: (1) crystals of the same phase; (2) crystals with the same composition but different structure; or (3) crystals with different composition. According to Vance (1969), in the first case there is normally a strong tendency to join either in parallel or twin orientation, whereas random attachment is rare. The reason is that a random attachment normally corresponds to an unstable atomic configuration at the interface which tends to evolve towards either a parallel growth or a twin. If both configurations are far from the relative orientation occurred at the moment of the attachment, an intermediate

orientation, corresponding to a less unstable interface, can sometimes be reached: the result is known as a plesiotwin (Nespolo *et al.*, 1999a). The second case involves oriented attachment of different polymorphs, which is a less probable event because they normally have different cell parameters; in case of polytypes, however, synneusis gives rise to allotwins (Nespolo *et al.*, 1999b), as in the example of ferriphlogopite from the Ruiz Peak (Nespolo *et al.*, 2000). The third case produces epitaxies. Synneusis is considered a common event in feldspars (Stull, 1979), but opinions diverge on whether it occurs early in crystallisation (Ross, 1957; Vance, 1969) or rather by post-growth attachment (Swanson, 1967). The latter phenomenon should be guided by the morphology of the mineral, rather than by crystallographic orientation, because late-stage oriented attachment has higher probability on large faces (Dowty, 1980a).

As pointed out by Tobi (1987), the genetic considerations are not sufficient to explain the difference in the characteristic patterns of twinning found in natural rocks: the underlying crystallographic causes must be understood. However, until recently a satisfactory *structural* investigation of twinned crystals was out of reach and only a *reticular* interpretation was possible (see Section 3 for details). The recent theoretical and computational advances have allowed to overcome that limitation and a general structural approach is now available, which exploits the analysis of the eigensymmetry of the crystallographic orbits building the structure of the mineral (Marzouki *et al.*, 2014a). We present in this article the results of that approach to the study of Carlsbad twins. In the next articles of the series, we will discuss the other established twins in feldspars.

## 2. Carlsbad twinning: an example of parallel hemitropy

The remarkable and systematic size difference between twinned and untwinned grains observed by Ingerson (1952) led Vance (1969) to conclude that contact and penetration Carlsbad twins originate as primary and synneusis twins, respectively. However, the size difference has also been explained by the effect of twinning on growth kinetics. The increase in growth rate was attributed to the so-called reentrant angle effect (Carstens 1968), but Kitamura *et al.* (1979) have demonstrated that this effect can only work under low supersaturation and only for perfect crystals containing no screw dislocations, whereas for real crystals, flattened or elongated morphology of twinned crystals is due to screw dislocation bundles concentrated in the composition plane.

In a twinned crystal there is a change of orientation from one individual (or domain) to the next one, but this change of orientation does not occur at random. First, the operation mapping the two individuals is a crystallographic operation in direct space (“Mallard’s law”: Friedel, 1926) called the *twin operation*; secondly, a significant part of the structure of one individual continues, more or

less unperturbed, across the interface – called the *composition surface* or, in case of a planar interface, *composition plane* – and builds up a common substructure. The latter constitutes the structural rationale for the occurrence of the twin (Marzouki *et al.*, 2014a).

Before the systematic categorisation of twinning in terms of the reticular theory (Friedel, 1926), twins were tentatively explained as arising from a two-fold rotation or *hemitropy* (from the Greek ημυ “half” and τροπία “to turn”) about a direction either perpendicular to the composition plane or about a direction contained in this plane (Haüy, 1801; see also Bravais, 1851). Accordingly, twins were classified as due to *normal hemitropy* or *parallel hemitropy*, with a few cases left out and assigned to a *complex hemitropy* possibly resulting from the action of both normal and parallel hemitropy. This old categorisation has been criticised by Friedel (1926) for at least two reasons:

- rotation twins in which the twin operation is a rotation of order higher than two have been discovered; although rarer, these twins cannot be explained in the framework of the hemitropy theory;
- reflection twins are not equivalent to rotation twins, even from a purely reticular viewpoint, unless the twin plane has a lattice direction exactly perpendicular to it, which in general is not the case; these twins again cannot be explained in the framework of the hemitropy theory.

Despite its lack of generality, the distinction between normal hemitropy and parallel hemitropy, which in the petrological literature are sometimes called “normal law” and “parallel law” (Takahashi, 2002), does make sense when dealing with rotation twins, because the relative orientation of the composition plane with respect to the twin axis implies a difference in the interpretation of the (pseudo)-eigensymmetry of the crystallographic orbits.

In case of normal hemitropy, as for the twins of staurolite studied by Marzouki *et al.* (2014b), the composition plane is perpendicular to the twin axis; its orientation is therefore determined by the twin element. In case of parallel hemitropy, instead, the orientation of the composition plane is not fixed a priori. In fact, infinitely many planes have the twin axis in common, and the orientation must be obtained by a morphological study of the twin. Carlsbad twins fall precisely in the category of parallel hemitropy, with twin axis  $[001]$ . They occur both as contact and as penetration twins; for the former, the composition plane is  $(010)$  (Wooster, 1981). Other possible composition planes are  $(110)$ ,  $(1\bar{1}0)$  (which belong to the same crystal form in monoclinic polymorphs), and  $(100)$ , but are very rare and the latter cannot occur as a result of synneusis, due to the fact that  $\{100\}$  is not a prominent form in any feldspar (Dowty, 1980a). The rare examples of the occurrence of Carlsbad twins with  $(100)$  composition plane (sometimes called “Carlsbad B”) show irregular composition surface with inclusions of other minerals and have been interpreted as a result of growth factors rather than structural continuity, because of the severe disruption or distortion at the twin boundary

(Dowty, 1980b). Even in these exceptional samples, (0 1 0) is most likely the original composition plane, as defined by Schaaque (1938a and b) and later refined by Curien and Kern (1957). Our analysis below is therefore applied to the Carlsbad twin based on (0 1 0) as twin plane.

### 3. Reticular vs. structural theory of crystal twinning

With the possible exception of late-stage synneusis, where the mechanical adherence on prominent faces may become a predominant factor, the occurrence frequency of a twin should be more or less directly related to the extent of a common substructure continuing across the composition surface. Until recently, investigators relied on the reticular (from the French *réseau*, “lattice”) theory of twinning, which determines the *sublattice* (as opposed to *substructure*) common to the individuals. The fraction of lattice nodes common to the individuals is taken as measure of the lattice *restoration*, expressed as the twin index, which is the reciprocal of that fraction; departure from exact overlap is measured by the obliquity or misfit (Friedel, 1926; Nespolo & Ferraris, 2007). Because the lattice expresses the periodicity of the crystal structure, a significant lattice restoration is a necessary condition for the formation of a twin; however, because a large, potentially infinite, number of crystal structures may have the same lattice type, the predictive power of the lattice restoration is limited; in particular, it cannot explain why twins with the same degree of lattice restoration have different occurrence frequency. For that purpose, we need the degree of *structural* restoration, *i.e.* we need to know the substructure that crosses more or less unperturbed the composition surface.

Dowty (1980b) and Wooster (1981) have provided a qualitative estimation of the structural restoration in twinned feldspars but for a convincing explanation we need a quantitative evaluation of the degree of such restoration. This can be obtained on the basis of the (pseudo)-eigensymmetry of the crystallographic orbits building the crystal structure of the individual. A crystallographic orbit is the set of atoms which are equivalent to one atom in the asymmetric unit under the operations of the space group of the structure. Let  $\mathcal{E}(O_i)$  – often abbreviated as  $\mathcal{E}_i$  – be the eigensymmetry of the *i*th orbit  $O_i$ , *i.e.* the group of all motions mapping the orbit  $O_i$  to itself. The crystal structure  $S$  is the set-theoretical union of the orbits:  $S = \cup_i O_i$ , whereas the space group  $\mathcal{G}(S)$  of the structure  $S$  is the intersection of the eigensymmetries of all crystallographic orbits:  $\mathcal{G}(S) = \cap_i \mathcal{E}(O_i)$ . The eigensymmetry  $\mathcal{E}_i$  of an orbit  $O_i$  is a supergroup of the space group  $\mathcal{G}(S)$  of the structure; when it is a proper supergroup,  $\mathcal{E}_i$  may contain operations whose linear part (the rotation, reflection or rotoinversion component) is the same as the twin operation. When that is the case, the orbit  $O_i$  crosses the composition surface of the twin unperturbed (or *almost* unperturbed if  $\mathcal{E}_i$  is allowed to contain *pseudo*-symmetries of  $O_i$ ) and contributes to the common substructure defined

above. The operations of the eigensymmetry group  $\mathcal{E}_i$  are space-group operations, which contain, besides the linear component, a translational component (possibly zero); these structural counterparts of the twin operation are called the *restoration operations*. Concretely, if we take the structure of one individual and apply to it a restoration operation, we get the same structure in a different, non-equivalent orientation. When we overlap these two structures we can recognise three types of situations:

1. some atoms of one individual overlap, at least approximately, atoms of the other individual that play the same structural role (*e.g.* cations centring coordination polyhedra of the same type in the two individuals);
2. some atoms of one individual may overlap, at least approximately, atoms of the other individual that do not play the same structural role (*e.g.* cations centring different types of polyhedra, or cations and anions);
3. some atoms of one individual do not overlap any atom of the other individual.

Atoms in situation 1 correspond to crystallographic orbits whose eigensymmetry contains a restoration operation and build up the common substructure of the twin. If this common substructure represents a significant portion of the whole structure, it justifies the occurrence of the twin.

The analysis of the (pseudo)-eigensymmetry of crystallographic orbits for the study of twins has been applied with success to several well-known twins in minerals, such as melilite (Marzouki *et al.*, 2014a), staurolite (Marzouki *et al.*, 2014b), aragonite (Marzouki *et al.*, 2015), marcasite (Nespolo & Souvignier, 2015a), cassiterite and rutile (Nespolo & Souvignier, 2015b) and forsterite (Azevedo & Nespolo, 2017). These are all reflection twins, with the exception of the two twins of staurolite, which are rotation twins by normal hemitropy. For reflection twins, the composition plane is normally, but not always, parallel to the twin plane. In normal hemitropy, its orientation is known *a priori* but its position is not fixed by symmetry: the composition plane can be shifted along the twin axis without changing the restoration accuracy. In parallel hemitropy, the opposite is true: the orientation of the composition plane is not known *a priori*, but its position is determined by symmetry arguments; the eigensymmetry of the orbits building the common substructure contains a restoration operation and its geometric element (*i.e.* the rotation or screw axis) should lie in the composition plane.

In this article we present the eigensymmetry analysis of the Carlsbad twin as the first case study of parallel hemitropy. To provide a quantitative evaluation of the degree of structural restoration across the composition plane we need to use the experimental structural data of feldspars. We have chosen the orthoclase structure reported at 293 K by Angel *et al.* (2013) and the monoclinic albite structure reported at 980 °C by Winter *et al.* (1979). The cell parameters and the fractional atomic coordinates depend on the chemical composition, in particular the substitution in the alkali site, and the external

conditions (temperature, pressure), but these differences do not affect the conclusions, they only modify slightly the numerical results presented below.

#### 4. Eigensymmetry of the crystallographic orbits of orthoclase and monoclinic albite and its application to the Carlsbad twin

The twin operation in Carlsbad twins is a two-fold rotation about the  $[001]$  direction; accordingly, the restoration operation which can lead to a common substructure in this twin is a two-fold rotation or screw-rotation about the  $c$  axis of the mineral. Figure 1 shows a polyhedral view of the structure of orthoclase (the structure of monoclinic albite would look exactly the same, given the minimal differences), which is built by a three-dimensional framework of eight-member rings of tetrahedra with a topology described by the code UUUUDDDD and UDDUDUUD (Smith & Rinaldi, 1962) where the M cations find place. The structure is monoclinic holohedral, with space group of type  $C2/m$ ; there is no lattice plane exactly perpendicular to the  $[001]$  direction so that twinning is by reticular pseudo-merohedry. The lattice plane close to perpendicularity, which defines a small supercell on which the twin lattice is built, is  $(10\bar{2})$ : the twin index is 2 and the obliquity depends on the cell parameters. For the structures reported by Angel *et al.* (2013) and by Winter *et al.* (1979), which we use in the following, the cell parameters are  $a = 8.575 \text{ \AA}$ ,  $b = 12.984 \text{ \AA}$ ,  $c = 7.2008 \text{ \AA}$ ,  $\beta = 116.028^\circ$  and  $a = 8.274 \text{ \AA}$ ,  $b = 12.991 \text{ \AA}$ ,  $c = 7.144 \text{ \AA}$ ,  $\beta = 116.13^\circ$  respectively; the obliquity is  $1.21^\circ$  and  $0.56^\circ$ , respectively (calculations performed with the software *geminography*: Nespolo & Ferraris, 2006).

Table 1 gives the fractional atomic coordinates of orthoclase and monoclinic albite in the asymmetric unit of  $C2/m$ , as well as the (pseudo)-eigensymmetry of the crystallographic orbits defined by these atoms, computed with the routine PSEUDO (Capillas *et al.*, 2011) at the Bilbao Crystallographic Server (Aroyo *et al.*, 2006). By  $d_{\min}$  we denote the minimal distance between the position to which a chosen atom in a crystallographic orbit  $O_i$  is mapped under the restoration operation and the nearest atom in  $O_i$ . When  $d_{\min} = 0$  for all atoms in  $O_i$ ,  $\mathcal{E}(O_i)$  is rigorously realised. If the eigensymmetry is only approximate, then  $d_{\min} > 0$  and its value is a measure for the quality of the quasi-restoration. Using different data, either from isostructural compounds or from varying experimental conditions (temperature, pressure) results in small differences in the values of  $d_{\min}$  due to the slight differences in the cell parameters and fractional coordinates, as can be seen by comparing the results for orthoclase and monoclinic albite in Table 1. The conclusions and interpretation with respect to the formation of the twins are, however, unaffected.

All the crystallographic orbits composing the structure of orthoclase and monoclinic albite possess a (pseudo)-eigensymmetry which is a proper supergroup of the space group of the minerals; however, the additional symmetry

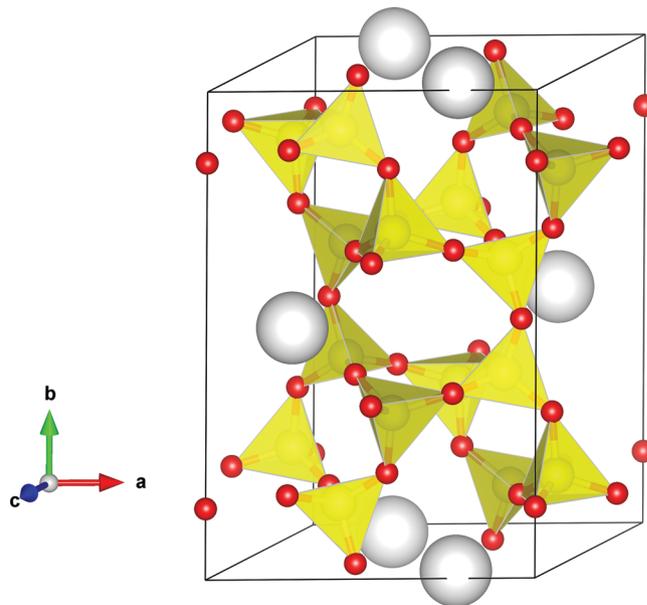


Fig. 1. The structure of orthoclase. Tetrahedra are centred on Si/Al and K occupies cavities defined by eight tetrahedra. This and the following figures are drawn with VESTA (Momma and Izumi, 2011).

with respect to  $C2/m$  contains the expected restoration operation only for two of the eight orbits (the T1- and the O4-orbit), which are also the only two orbits for which the deviation from the metric symmetry is within acceptable limits (last column of Table 1). For these two orbits we thus find the restoration operation required to explain the formation of the twin; they account for 31% of the structure (16 atoms out of 52, considering the multiplicities of the Wyckoff positions: see Table 1). This is already a significant contribution when considering the whole unit cell, but of particular interest is the region close to the composition plane, as we are going to discuss in detail. The other six orbits, instead, do not possess a restoration operation in their (pseudo)-eigensymmetry; in fact, under the basis transformation necessary to realise the pseudo-symmetry, the  $[001]$  direction of orthoclase/monoclinic albite is transformed to a direction which is not a symmetry direction of the corresponding lattice. For these other orbits we therefore need to analyse the (pseudo)-eigensymmetry of *suborbits* (*i.e.* orbits under the action of a subgroup), according to the following procedure, already explained in detail in our previous reports:

- identify the unit cell of the twin lattice: this is built on the twin axis and the lattice plane (quasi)-perpendicular to it;
- identify the maximal subgroup  $\mathcal{H}$  of the space group of the individual  $\mathcal{G}$  compatible with the twin lattice: this is obtained by taking the intersection of  $\mathcal{G}$  and its conjugate obtained through the twin operation, *i.e.*  $\mathcal{H} = \mathcal{G} \cap t\mathcal{G}t^{-1}$ ;
- split the crystallographic orbits of  $\mathcal{G}$  into orbits under the action of  $\mathcal{H}$ : going from a group to a subgroup, a crystallographic orbit in general splits into two or more orbits (Wondratschek, 1993);

Table 1. Fractional coordinates of orthoclase  $\text{KAlSi}_3\text{O}_8$  (Angel *et al.*, 2013) and monoclinic albite  $\text{NaAlSi}_3\text{O}_8$  (Winter *et al.*, 1979) and pseudo-eigensymmetry of the corresponding crystallographic orbits. Basis transformation and origin shift are expressed from  $C2/m$  to the (pseudo)-eigensymmetry group (group-to-supergroup relation).  $d_{\min}$  is the minimal distance between atoms related by the operations in the pseudo-eigensymmetry: when  $d_{\min} = 0$  the higher eigensymmetry is exact. The angular deviation from the eigensymmetry lattice (last column) is not considered in the calculation of  $d_{\min}$ , which is therefore approximated. The orientation of the twin axis corresponds to the  $[001]$  direction in  $C2/m$ , to  $[1\bar{1}0]$  in  $Fmmm$  and to  $[11\bar{1}]$  in  $I4/mmm$ , neither of which is a symmetry direction for the lattice corresponding to the orbit eigensymmetry. For the T1 and O4 orbits, the twin axis corresponds to the  $[010]$  direction, which is a symmetry direction of the orthorhombic lattice. These are also the only two orbits for which the deviation from the metric orthorhombic symmetry is within acceptable limits.

Orbit	Wyckoff position	Fractional coordinates		(Pseudo)-eigensymmetry	Basis transformation	Origin shift	$d_{\min}$ (Å)	$\alpha/\beta/\gamma$ (°)	
		Orthoclase	Monoclinic albite					Orth	Mon
M	4i	0.28476,	0.27890,	<i>Fmmm</i> (69)	$\mathbf{a}+2\mathbf{c},\mathbf{a},\mathbf{b}$	0,0,½	0.5894	90/	90/
		0,	0,				0.6619	90/	90/
T1	8j	0.13825	0.13780	<i>Immm</i> (71)	$-\mathbf{a}-\frac{1}{2}\mathbf{c},\frac{1}{2}\mathbf{c},\mathbf{b}$	0,0,0	0.4662	80.11	81.22
		0.00932,	0.00810,				0.4567	90/	90/
		0.18481,	0.17840,					90/	90/
T2	8j	0.22407	0.22330	<i>Fmmm</i> (69)	$\mathbf{a}+2\mathbf{c},\mathbf{a},\mathbf{b}$	0,0,0	0.6324	88.79	89.45
		0.70912,	0.69590,				0.4120	90/	90/
		0.11770,	0.11630,					90/	90/
O1	4g	0.34449	0.34200	<i>Fmmm</i> (69)	$\mathbf{a}+2\mathbf{c},\mathbf{a},\mathbf{b}$	0,0,0	0	80.11	81.22
		0,	0,				0	90/	90/
		0.14607,	0.13930,					90/	90/
O2	4i	0	0.00000	<i>Fmmm</i> (69)	$\mathbf{a}+2\mathbf{c},\mathbf{a},\mathbf{b}$	0,0,0	0.0815	80.11	81.22
		0.6379,	0.60630,				0.5866	90/	90/
		0,	0,					90/	90/
O3	8j	0.2853	0.28350	<i>I4/mmm</i> (139)	$\frac{1}{2}\mathbf{a}+\frac{1}{2}\mathbf{b}+\mathbf{c},$ $\frac{1}{2}\mathbf{a}-\frac{1}{2}\mathbf{b}+\mathbf{c},\mathbf{a}$	0,0,½	0.6433	80.11	81.22
		0.82665,	0.82330,				0.6566	82.99/	83.81/
		0.14651,	0.13500,					82.99/	83.81/
O4	8j	0.22770	0.22520	<i>Immm</i> (71)	$-\mathbf{a}-\frac{1}{2}\mathbf{c},\frac{1}{2}\mathbf{c},\mathbf{b}$	0,0,0	0.5564	89.33	90.05
		0.03510,	0.02570,				0.4058	90/	90/
		0.31147,	0.30440,					90/	90/
O5	8j	0.25930	0.25350	<i>Fmmm</i> (69)	$\mathbf{a}+2\mathbf{c},\mathbf{a},\mathbf{b}$	0,0,0	0.5564	88.79	89.45
		0.18081,	0.18910,				0.2209	90/	90/
		0.12583,	0.12290,					90/	90/
		0.40697	0.40490					80.11	81.22

– search for the crystallographic orbits  $O_i$  under  $\mathcal{H}$  for which the (pseudo)-eigensymmetry  $\mathcal{E}(O_i)$  contains a symmetry operation whose linear part coincides with the twin operation and thus identifies the restoration operation.

The unit cell of the Carlsbad twin lattice is defined by the  $[001]$  direction and the  $(10\bar{2})$  plane quasi-perpendicular to it. The two shortest directions in this plane are  $[201]$  and  $[010]$ , which, together with the  $[001]$  twin axis, constitute the columns of the transformation matrix from the basis vectors  $(\mathbf{abc})_I$  of the untwinned orthoclase/monoclinic albite crystal to those of the Carlsbad twin  $(\mathbf{abc})_T$  (I=individual, T=twin):

$$(\mathbf{a} \ \mathbf{b} \ \mathbf{c})_I = \begin{pmatrix} 2 & 0 & 0 \\ 0 & 1 & 0 \\ 1 & 0 & 1 \end{pmatrix} = (\mathbf{a} \ \mathbf{b} \ \mathbf{c})_T,$$

*i.e.*  $\mathbf{a}_T\mathbf{b}_T\mathbf{c}_T = 2\mathbf{a}_I+\mathbf{c}_I,\mathbf{b}_I,\mathbf{c}_I$ . The  $[201]$  direction of orthoclase/monoclinic albite is transformed to the  $[100]$  direction of the twin lattice; the two other basis vectors

are in common. The cell parameters of the twin lattice are  $a_T = 15.414 \text{ \AA}$ ,  $b_T = 12.984 \text{ \AA}$ ,  $c_T = 7.2008 \text{ \AA}$ ,  $\beta_T = 91.21^\circ$  for orthoclase and  $a_T = 14.857 \text{ \AA}$ ,  $b_T = 12.991 \text{ \AA}$ ,  $c_T = 7.144 \text{ \AA}$ ,  $\beta_T = 90.56^\circ$  for monoclinic albite; the unit cell of the twin lattice is *B*-centred, monoclinic but pseudo-orthorhombic with a deviation of  $\beta$  which corresponds precisely to the obliquity. The advantage of taking an unconventional *mB* unit cell lies in the fact that it clearly shows the orthorhombic pseudo-symmetry of the lattice, which would be somewhat camouflaged by the standard  $P2/m$  setting. The unit cell of the twin lattice is twice larger than that of the individual and contains therefore  $52 \times 2 = 104$  atoms. The highest space-group type compatible with this lattice is  $\mathcal{H} = \mathcal{G}_1 \cap \mathcal{G}_2$  where  $\mathcal{G}_1$  and  $\mathcal{G}_2$  are the space groups of the two individuals in their respective orientations.  $\mathcal{G}_2$  is of the same type as  $\mathcal{G}_1$  and differs from it in the orientation of the basis vectors: it is the conjugate of  $\mathcal{G}_1$  by the twin operation. In our case,  $\mathcal{H}$  is of type  $B2/m$ , which is an unconventional setting of  $P2/m$  (Nespolo & Aroyo, 2016). The International Tables for Crystallography (2016) do not list this setting, so we list in Table 2 explicitly the positions obtained by adding the

centring vector  $(\frac{1}{2}, 0, \frac{1}{2})$  that are relevant to the Carlsbad twin. The splitting scheme of the occupied Wyckoff positions in  $C2/m$  to those in  $B2/m$  under the transformation  $2\mathbf{a} + \mathbf{c}, \mathbf{b}, \mathbf{c}$  is  $4g \rightarrow 4i + 4j$ ;  $4i \rightarrow 4m + 4n$ ;  $8j \rightarrow 2 \times 8o$ , obtained through the WYCKSPLIT routine at the Bilbao Crystallographic Server (Kroumova *et al.*, 1998). The fractional coordinates of orthoclase and monoclinic albite in the asymmetric unit of  $B2/m$  are given in Table 3, where the orbits are numbered sequentially, keeping a record of the original labelling in  $C2/m$  ( $M \rightarrow M1$  and  $M2$ ;  $T1 \rightarrow T11$  and  $T12$ ; *etc.*). All the crystallographic orbits in  $B2/m$  have  $Bmmm$  (pseudo)-eigensymmetry,<sup>1</sup> with varying deviations from perfect symmetry, in some cases the origin is in common, whereas for some orbits an origin shift of  $\frac{1}{4}, 0, \frac{1}{4}$  from  $B2/m$  to  $Bmmm$  is necessary (Table 4). When no shift of the origin is required, the restoration operations are  $2\ 0, 0, z$  and, combining with the  $B$ -centring vector,  $2_1\ \frac{1}{4}, 0, z$ ; when instead an origin shift is required, the restoration operations are  $2_1\ 0, 0, z$  and  $2_1\ \frac{1}{4}, 0, z$ . The deviations  $d_{\min}$ , defined as in Table 1, lie between 0 (O11 and O12), meaning that the orbits have exact  $Bmmm$  eigensymmetry, and  $\sim 0.5$  Å (O21 and O22), which is still less than half the ionic radius (both Pauling and Shannon radii).

It turns out that half of the 16 independent crystallographic orbits in  $B2/m$  are quasi-restored by  $2\ 0, 0, z$  as restoration operation and the other half by  $2_1\ 0, 0, z$  (the four orbits T11, T12, O41 and O42 are even quasi-restored by both, but we stick to the restoration operation with the lower value of  $d_{\min}$ ). A closer analysis shows that for each of the 8 orbits under  $C2/m$ , one of the (sub)orbits under the action of  $B2/m$  is quasi-restored by  $2\ 0, 0, z$  and the other by  $2_1\ 0, 0, z$ . Strikingly, each of the orbits quasi-restored by  $2\ 0, 0, z$  is fully contained in the region with  $y$  between  $-0.25$  and  $0.25$ , which we will call region I, and all orbits quasi-restored by  $2_1\ 0, 0, z$  are contained in the region with  $y$  between  $0.25$  and  $0.75$ , which we will call region II. This means that the part of the structure contained in region I is fully restored by the restoration operation  $2\ 0, 0, z$ , with a maximal deviation of  $0.4846$  Å (orthoclase) and  $0.2808$  Å (monoclinic albite) (see Table 4). The geometric element of this restoration operation is located in the centre of region I. Similarly, the part of the structure contained in region II is fully restored by the restoration operation  $2_1\ 0, 0, z$ , again with maximal deviation  $0.4846$  Å (orthoclase) and  $0.2808$  Å (monoclinic albite). Note that the geometric element of  $2_1\ 0, 0, z$  is not contained in region II, consequently it maps region II to the region with  $y$  between  $-0.75$  and  $-0.25$ . However, composing  $2_1\ 0, 0, z$  with the translation by  $0, 1, 0$  results in the restoration operation  $2_1\ 0, \frac{1}{2}, z$  which has its geometric element in the centre of region II and maps this region to itself.

Figures 2 and 3 show the overlap of the structures of two individuals (atoms in white and black, respectively) for the regions I and II obtained by the restoration operations  $2\ 0, 0, z$ , and  $2_1\ 0, \frac{1}{2}, z$ , respectively. The parts of the structure shown in the figures are from  $y = -0.25$  to  $y = 0.25$  for

Table 2. Wyckoff positions in the  $P2/m$  and in the  $B2/m$  settings of the space group relevant to Carlsbad twin.

$P2/m$		$B2/m: (\frac{1}{2}, 0, \frac{1}{2})+$	
Wyckoff position	Coordinates	Wyckoff position	Coordinates
4o	$x, y, z$	8o	$x, y, z$
2n	$x, \frac{1}{2}, z$	4n	$x, \frac{1}{2}, z$
2m	$x, 0, z$	4m	$x, 0, z$
2j	$\frac{1}{2}, y, 0$	4j	$\frac{1}{4}, y, \frac{3}{4}$
2i	$0, y, 0$	4i	$0, y, 0$

Table 3. Orthoclase and monoclinic albite fractional coordinates in the  $B2/m$  setting of the twin lattice. First line: orthoclase; second line: monoclinic albite.

Atom	Wyckoff position	x	y	z
M1	4m	0.14238	0	0.99587
		0.13945	0	0.99835
M2	4n	0.39238	$\frac{1}{2}$	0.74587
		0.38945	$\frac{1}{2}$	0.74835
T11	8o	0.00466	0.18481	0.21941
T12	8o	0.00405	0.17840	0.21925
		0.25466	0.68481	0.96941
T21	8o	0.25405	0.67840	0.96925
		0.35456	0.11770	0.98993
T22	8o	0.34795	0.11630	0.99405
		0.60456	0.61770	0.73993
O11	4i	0.59795	0.61630	0.74405
		0	0.14607	0
O12	4j	0	0.13930	0
		$\frac{1}{4}$	0.64607	$\frac{3}{4}$
O21	4m	$\frac{1}{4}$	0.63930	$\frac{3}{4}$
		0.31895	0	0.96635
O22	4n	0.30315	0	0.98035
		0.56895	$\frac{1}{2}$	0.71635
O31	8o	0.55315	$\frac{1}{2}$	0.73035
		0.41333	0.14651	0.81438
O32	8o	0.41165	0.13500	0.81355
		0.66333	0.64651	0.56438
O41	8o	0.66165	0.63500	0.56355
		0.01755	0.31147	0.24175
O42	8o	0.01285	0.30440	0.24065
		0.26755	0.81147	0.99175
O51	8o	0.26285	0.80440	0.99065
		0.09041	0.12583	0.31657
O52	8o	0.09455	0.12290	0.31035
		0.34041	0.62583	0.06657
		0.34455	0.62290	0.06035

Fig. 2 and from  $y = 0.25$  to  $y = 0.75$  for Fig. 3, *i.e.* half of a unit cell around the planes  $x, 0, z$  and  $x, \frac{1}{2}, z$  that contain the geometric elements of the restoration operations.

The same analysis (details omitted for brevity) on triclinic plagioclases shows a much lower degree of structural restoration, despite a good reticular restoration (twin index 2 and obliquity about  $1^\circ$ ), as shown in Section 4. In particular, the oxygen atoms are not restored by the Carlsbad twin operation. The occurrence of simple Carlsbad twins in triclinic feldspars is therefore an unlikely event from the

<sup>1</sup> $Bmmm$  is an unconventional setting of  $Cmmm$  adopted here to keep the same basis vectors for  $\mathcal{H}$  and  $\mathcal{E}(O_i)$ .

Table 4. Restoration operations for the crystallographic orbits in Table 3. In all cases the eigensymmetry is  $Bmmm$ , unconventional setting of No. 65, with basis vectors parallel to those of the twin lattice. In the case of the restoration operation  $2\ 0,0,z$ , the origin of  $Bmmm$  coincides with that of  $B2/m$ , in the case of the restoration operation  $2_1\ 0,0,z$  it is shifted by  $\sqrt{4},0,\sqrt{4}$ . The orbits are located in two distinct regions, region I with  $y$  between  $-0.25$  and  $0.25$  and region II with  $y$  between  $0.25$  and  $0.75$ . The deviation  $d_{\min}$  is defined as in Table 1, in all cases only the best restoration is given.

Orbit	Wyckoff position	Region	$d_{\min}$ (Å)		Restoration operation
			Orthoclase	Monoclinic albite	
M1	$4m$	I	0.0595	0.0236	$2\ 0,0,z/2_1\ \frac{1}{4},0,z$
M2	$4n$	II	0.0595	0.0236	$2_1\ 0,0,z/2\ \frac{1}{4},0,z$
T11	$8o$	I	0.1437	0.1203	$2\ 0,0,z/2_1\ \frac{1}{4},0,z$
T12	$8o$	II	0.1437	0.1203	$2_1\ 0,0,z/2\ \frac{1}{4},0,z$
T21	$8o$	I	0.1450	0.0850	$2\ 0,0,z/2_1\ \frac{1}{4},0,z$
T22	$8o$	II	0.1450	0.0850	$2_1\ 0,0,z/2\ \frac{1}{4},0,z$
O11	$4i$	I	0	0	$2\ 0,0,z/2_1\ \frac{1}{4},0,z$
O12	$4j$	II	0	0	$2_1\ 0,0,z/2\ \frac{1}{4},0,z$
O21	$4m$	I	0.4846	0.2808	$2\ 0,0,z/2_1\ \frac{1}{4},0,z$
O22	$4n$	II	0.4846	0.2808	$2_1\ 0,0,z/2\ \frac{1}{4},0,z$
O31UO51	$8o$	I	0.2751	0.1837	$2\ 0,0,z/2_1\ \frac{1}{4},0,z$
O32UO52	$8o$	II	0.2751	0.1837	$2_1\ 0,0,z/2\ \frac{1}{4},0,z$
O41	$8o$	II	0.1188	0.1336	$2_1\ 0,0,z/2\ \frac{1}{4},0,z$
O42	$8o$	I	0.1188	0.1336	$2\ 0,0,z/2_1\ \frac{1}{4},0,z$

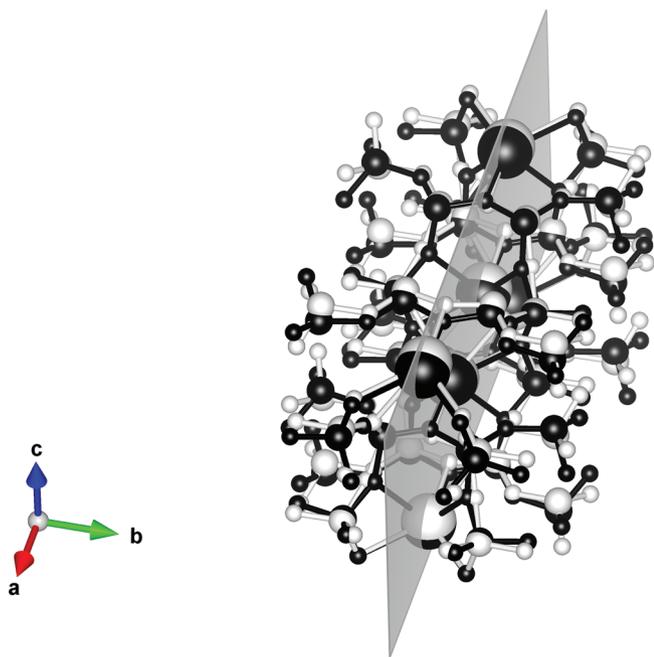


Fig. 2. The structure of the two individuals of orthoclase related by the  $2\ 0,0,z$  restoration operation. White and black atoms belong to different individuals. The structure spans half of a unit cell, from  $y = -0.25$  to  $y = 0.25$ . The grey plane is located at  $x,0,z$  and contains the geometric element of the restoration operation.

structural viewpoint and may actually be a witness of late-stage synneusis. Carlsbad twinning may also be inherited from the monoclinic polymorph, but in that case it should be accompanied by a transformation twinning occurring at the phase transition, unless of course recrystallisation took place. As a matter of fact, “complex laws”, in particular the Albite-Carlsbad twinning, are quite common in plagioclases (Gorai, 1957; Tribaudino *et al.*, 1995), an observation that supports our interpretation.

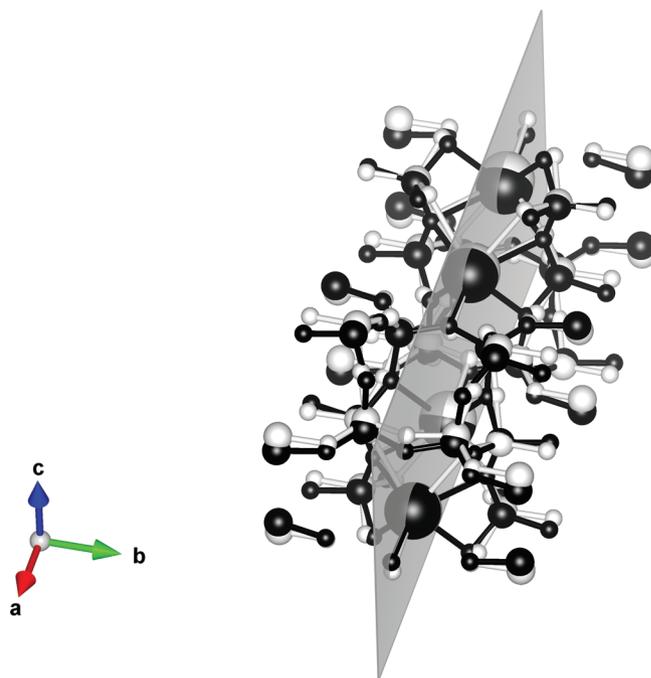


Fig. 3. The structure of the two individuals of orthoclase related by the  $2_1\ 0,\frac{1}{2},z$  restoration operation. Same conventions as in Fig. 2. The structure spans half of a unit cell, from  $y = 0.25$  to  $y = 0.75$ . The grey plane is located at  $x,\frac{1}{2},z$  and contains the geometric element of the restoration operation.

## 5. Conclusions and perspectives

The analysis of the (pseudo)-eigensymmetry of the crystallographic orbits of orthoclase and monoclinic albite explains the high occurrence frequency of the Carlsbad twin because of the existence of a large substructure common to the individuals in the respective orientations. The operation  $2\ 0,0,z$  quasi-restores all the atoms within the slab going from  $y = -1/4$  to  $y = 1/4$  containing at its

centre the geometric element of  $2\ 0,0,z$  within the composition plane  $x,0,z$ . The next slab of the same width ( $b_T/2$ ), located between  $y = 1/4$  and  $y = 3/4$  is not restored by  $2\ 0,0,z$ , but by the different restoration operation,  $2_1\ 0,1/2,z$ , which similarly has its geometric element contained in the composition plane  $x,1/2,z$  at the centre of the slab.

This means that every  $b_T/2$ , or  $\sim 6.5\ \text{\AA}$ , a change of orientation corresponding to the twin operation of the Carlsbad twin leads to an excellent structural match across the composition plane. From the genetic viewpoint, the opposite statement is more interesting: for positions of the composition plane at small distances, we can find an extensive common substructure, although the restoration operation having its geometric element in the composition plane may vary depending on the location of the composition plane. As a consequence, the change in orientation can occur repeatedly, leading to polysynthetic twinning, which was reported in TEM investigations of synthetic Sr-rich feldspars (Tribaudino *et al.*, 1995) and is ubiquitous in samples annealed directly from gel (Benna *et al.*, 1995). It may also occur at different positions in physically different sites of the growing crystal, resulting in a composition surface that is not necessarily planar: the result would be a penetration twin, rather than a contact twin; as a matter of fact, Carlsbad twins are known to occur more frequently as penetration twins than as contact twins. It would also explain the migration of twin boundaries as a local response allowing the individual experiencing the higher elastic strain energy to expand at the expense of the other individual, which may eventually lead to detwinning (DeVore, 1970).

In a forthcoming article we will apply the same analysis to other twin laws confirmed to occur in feldspars.

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