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3.3. Twinning of crystals

By Th. Hahn and H. Klapper

In this chapter, the basic concepts and definitions of twinning, as well as the morphological, genetic and lattice classifications of twins, are presented. Furthermore, twin boundaries are discussed extensively. The effect of twinning in reciprocal space, *i.e.* on diffraction and crystal-structure determinations, is treated in Section 3.3.11. In the literature, the concept of twinning is very often used in a non-precise or ambiguous way. In order to clarify the terminology, this chapter begins with a section on the various kinds of crystal aggregates and intergrowths; in this context twinning appears as a special intergrowth of two or more crystals with well defined crystallographic orientation relations.

3.3.1. Crystal aggregates and intergrowths

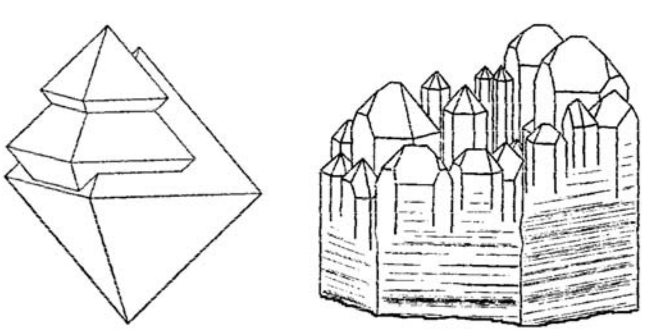
Minerals in nature and synthetic solid materials display different kinds of *aggregations*, in mineralogy often called *intergrowths*. In this chapter, we consider only aggregates of crystal grains of the same species, *i.e.* of the same (or nearly the same) chemical composition and crystal structure (*homophase* aggregates). Intergrowths of grains of different species (*heterophase* aggregates), *e.g.* heterophase bicrystals, epitaxy (two-dimensional oriented intergrowth on a surface), topotaxy (three-dimensional oriented precipitation or exsolution) or the paragenesis of different minerals in a rock or in a technical product are not treated in this chapter.

(i) *Arbitrary intergrowth*: Aggregation of two or more crystal grains with arbitrary orientation, *i.e.* without any systematic regularity. Examples are irregular aggregates of quartz crystals (*Bergkristall*) in a geode and intergrown single crystals precipitated from a solution. To this category also belong untextured polycrystalline materials and ceramics, as well as sandstone and quartzite.

(ii) *Parallel intergrowth*: Combination of two or more crystals with parallel (or nearly parallel) orientation of all edges and faces. Examples are dendritic intergrowths as well as parallel intergrowths of spinel octahedra (Fig. 3.3.1.1*a*) and of quartz prisms (Fig. 3.3.1.1*b*). Parallel intergrowths frequently exhibit re-entrant angles and are, therefore, easily misinterpreted as twins.

Two possible reasons for the formation of parallel intergrowths are mentioned:

(*a*) A smaller crystal has set down in parallel orientation on a growth face of an already existing crystal of the same species and has further grown together with its host. Fig. 3.3.1.1(*a*) suggests such a mechanism.



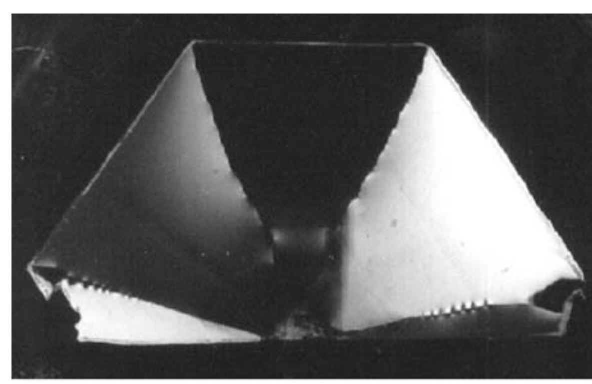
(*a*) (*b*)

Fig. 3.3.1.1. Parallel intergrowth (*a*) of spinel octahedra and (*b*) of hexagonal quartz prisms. Part (*a*) after Phillips (1971, p. 172), part (*b*) after Tschermak & Becke (1915, p. 94).

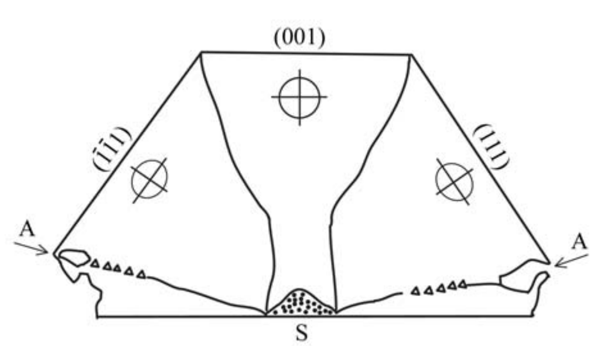
(*b*) The growth of one or several faces of a crystal is inhibited by a layer of impurities or by foreign particles. By a local 'breaking down' of these obstacles, several parallel individual crystals may appear and grow together during further growth. This mechanism is suggested for Fig. 3.3.1.1(*b*).

In this context the term *mosaic crystal* must be mentioned. It was introduced in the early years of X-ray diffraction in order to characterize the perfection of a crystal. A mosaic crystal consists of small blocks (size typically in the micron range) with orientations deviating only slightly from the average orientation of the crystal; the term 'lineage structure' is also used for very small scale parallel intergrowths (Buerger, 1934, 1960*a*, pp. 69–73).

(iii) *Bicrystals*: This term is mainly used in metallurgy. It refers to the (usually synthetic) intergrowth of two single crystals with a well defined orientation relation. A bicrystal contains a *grain boundary*, which in general is also well defined. Usually, homophase bicrystals are synthesized in order to study the structure and properties of grain boundaries. An important tool for the theoretical treatment of bicrystals and their interfaces is the *coincidence-site lattice* (CSL). A brief survey of bicrystals is given in Section 3.2.2; a comparison with twins and domain structures is provided by Hahn *et al.* (1999).



(*a*)



(*b*)

Fig. 3.3.1.2. (*a*) Optical anomaly of a cubic mixed (K,NH_4)-alum crystal grown from aqueous solution, as revealed by polarized light between crossed polarizers: (110) plate, 1 mm thick, horizontal dimension about 4 cm. (*b*) Sketch of growth sectors and their boundaries of the crystal plate shown in (*a*). The {111} growth sectors are optically negative and approximately uniaxial with their optical axes parallel to their growth directions ⟨111⟩ [birefringence Δn up to 5×10^{-5}; Shtukenberg *et al.* (2001)]. The (001) growth sector is nearly isotropic ($\Delta n < 10^{-6}$). Along the boundaries A between {111} sectors a few small {110} growth sectors (resulting from small {110} facets) have formed during growth. S: seed crystal.

(iv) *Growth sectors and optical anomaly*: Crystals grown with planar faces (habit faces), *e.g.* from vapour, supercooled melt or solution, consist of regions crystallized on different growth faces (Fig. 3.3.1.2). These growth sectors usually have the shapes of pyramids with their apices pointing toward the nucleus or the seed crystal. They are separated by *growth-sector boundaries*, which represent inner surfaces swept by the crystal edges during growth. In many cases, these boundaries are imperfections of the crystal.

Frequently, the various growth sectors of one crystal exhibit slightly different chemical and physical properties. Of particular interest is a different optical birefringence in different growth sectors (optical anomaly) because this may simulate twinning. A typical example of this optical anomaly is shown in Fig. 3.3.1.2. Since this phenomenon has sometimes been misinterpreted as twinning, it is treated in detail in the *Extended note* below.

(v) *Translation domains*: Translation domains are homogeneous crystal regions that exhibit exact parallel orientations, but are displaced with respect to each other by a vector (frequently called a *fault vector*), which is a fraction of a lattice translation vector. The interface between adjoining translation domains is called the 'translation boundary'. Often the terms *antiphase domains* and *antiphase boundaries* are used. Special cases of translation boundaries are stacking faults. Translation domains are defined on an atomic scale, whereas the term parallel intergrowth [see item (ii) above] refers to macroscopic (morphological) phenomena; *cf.* Note (7) in Section 3.3.2.4.

(vi) *Twins*: A frequently occurring intergrowth of two or more crystals of the same species with well defined *crystallographic* orientation relations is called a *twin* (German: *Zwilling*; French: *macle*). Twins form the subject of the present chapter. The closely related topic of *Domain structures* is treated in Chapter 3.4.

In 1975, J. D. H. Donnay and H. Takeda even proposed a new name for the 'science of twinning': *geminography* [as reported by Nespolo & Ferraris (2003) and by Grimmer & Nespolo (2006)]. A complete review of the history and the various theories of twinning, together with an extensive list of references, is provided in a recent monograph (in French) by Boulliard (2010). In addition, it contains an extraordinarily large set of beautiful colour photographs of many natural twins.

Extended note: Optical anomaly, Curie's principle and dissymmetry

The phenomenon of *optical anomaly* [*cf.* Section 3.3.1(iv) and Fig. 3.3.1.2] can be explained as follows: as a rule, impurities (and dopants) present in the solution are incorporated into the crystal during growth. Usually, the impurity concentrations differ in symmetry-*non-equivalent* growth sectors (which belong to different crystal forms), leading to slightly changed lattice parameters and physical properties of these sectors. In mixed crystals these changes often arise from a partial ordering of the mixing components parallel to the growth face [example: (K,NH_4)-alum, *cf.* Fig. 3.3.1.2; Shtukenberg *et al.*, 2001]. Optical anomalies may occur also in symmetry-*equivalent* growth sectors (which belong to the same crystal form) owing to their different growth directions: as a consequence of growth fluctuations, layers of varying impurity content are formed parallel to the growth face of the sector ('growth striations'). This causes a slight change of the interplanar spacing normal to the growth face. For example, a cubic NaCl crystal grown on the {100} cube faces from an aqueous solution containing Mn ions consists of three pairs of (opposite) growth sectors exhibiting a slight tetragonal distortion with tetragonality 10^{-5} along their $\langle 100\rangle$ growth directions and hence is optically uniaxial (Ikeno *et al.*, 1968)[1].

An analogous effect of optical anomaly may be observed in crystals grown from the melt on rounded interfaces with planar facets of prominent habit faces (*e.g.* of melt-grown synthetic garnets). Owing to different growth mechanisms on round and facet interfaces (rough growth and growth by supercooling, respectively), the incorporation of impurities or dopants is usually different on the two types of interfaces (*e.g.* Hurle & Rudolph, 2004). The regions crystallized on the rounded faces and on the different facets correspond to different growth sectors and may exhibit optical anomalies.

Although the phenomenon of *optical anomaly* closely resembles all features of twinning, it does not belong to the category 'twinning' because it is not an intrinsic property of the crystal species, but rather the result of different growth conditions (or growth mechanisms) on different faces of the same crystal ('growth anisotropy'). It is the consequence of the well known Curie principle[2] (Curie, 1894; Chalmers, 1970) which describes (as an 'effect') the reduction of the symmetry ('dissymmetry') of an object (crystal) under an external influence ('cause') which itself exhibits a symmetry. It says, in terms of group theory, that the point-group symmetry G_{CF} of the crystal under the external influence (field) F is the intersection of the symmetry G_C of the crystal without field and the symmetry G_F of the influence without crystal:

$$G_{CF} = G_C \cap G_F,$$

i.e. G_{CF} is a (proper or improper) subgroup of both groups G_C and G_F. In the example of the optical anomaly of the {111} growth sectors of (K,NH_4)-alum (Fig. 3.3.1.2) the crystal point group is $G_C = 2/m\bar{3}$ and the symmetry of the cause 'growth in direction [111]' is $G_F = \infty m$ (symmetry of a stationary cone, *cf.* ITA, Table 10.1.4.2). The intersection symmetry [*i.e.* the symmetry of the (111) growth sector] is $G_{CF} = 3m$ ('dissymmetry') with the threefold axis along the growth direction [111] of this sector. This leads to a reduction of the isotropic optical birefringence of the 'undisturbed' cubic alum crystal to an uniaxial birefringence of its {111} growth sectors.

A very early review of the optical anomaly of crystals with many examples was published in 1891 by von Brauns. An actual review, treating the 'historical' observations and various interpretations (starting with Brewster, 1818) as well as the modern aspects of optical anomalies, is presented by Kahr & McBride (1992). A similar, very comprehensive review is contained in the monograph of Shtukenberg *et al.* (2007).

3.3.2. Basic concepts and definitions of twinning

Because twinning is a rather complex and widespread phenomenon, several definitions have been presented in the literature. Two of them are quoted here because of the particular engagement of their authors in this topic.

George Friedel (1904; 1926, p. 421): *A twin is a complex crystalline edifice built up of two or more homogeneous portions of the same crystal species in contact (juxtaposition) and oriented with respect to each other according to well-defined laws.*

These laws, as formulated by Friedel, are specified in his book (Friedel, 1926). His 'lattice theory of twinning' is discussed in Sections 3.3.8 and 3.3.9 of the present chapter.

Paul Niggli (1919, 1920/1924/1941): *If several crystal individuals of the same species are intergrown in such a way that all analogous faces and edges are parallel, then one speaks of parallel intergrowth. If for two crystal individuals not all but only some of the (morphological) elements (edges or faces), at least two independent ones, are parallel or antiparallel, and if such an intergrowth due to its frequent occurrence is not 'accidental', then one speaks*

[1] The relative lattice-parameter changes associated with these phenomena are usually smaller than 10^{-4} and cannot be detected in ordinary X-ray diffraction experiments. They are, however, accessible by high-resolution X-ray diffraction (Bowen & Tanner, 1998; Lal, 1993; Robert *et al.*, 1981).

[2] Also called the Neumann–Curie principle.