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Orientation of phase and domain boundaries in crystalline solids: discussion

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Abstract

In reply to a paper by Fleet (1982) on the calculation of the orientations of phase and domain boundaries, the different models which have been developed for these calculations are compared. In spite of different theoretical approaches, it is shown that all models imply the calculation of very similar quantities. The use of these models for the determination of feldspar intergrowth orientations is evaluated. As the elastic anisotropy of feldspars is low, it can be shown that the results of the calculations using the different models are very similar; they are even identical for plagioclase exsolution. The predicted orientations of exsolution lamellae in alkali-feldspars should not be very different using the various models and they should approach the observed values if physically appropriate data are used. Detailed comments on diagonally associated cryptoperthites are presented, with reference to phase coherency and morphological evolution observed by transmission electron microscopy.

Formation of interface boundaries during exsolution processes

During exsolution from homogeneous crystals, precipitates are formed which generally have specific lattice and interface orientations with regard to the host crystal lattice. The driving force for exsolution is the reduction in free energy due to chemical separation, and this is opposed by positive free energy terms due to the creation of interfaces between the two compositional domains. It is generally admitted that the preferred interface orientation corresponds to a minimum in the latter free energy terms, which involve a surface term proportional to the interface area and a strain term proportional to the volume of strained lattice. However, no one is at present able to calculate the total interface free energy with precision. The relative values of the surface and strain terms will depend on the lattice coherency of the precipitate/matrix interface. For coherent precipitates the strain energy is dominant, whereas for incoherent precipitates the surface energy is dominant.[1] Below a certain thickness, the ratio volume/interface surface is small and, consequently, coherent precipitates are energetically more favorable. During coarsening, coherent precipitates may lose coherency.

In the case of similar parallel lattices, several models have been proposed which *rank* the energy of interface orientations, but do not attempt to calculate the total interfacial energy. All models calculate the strain or misfit between planes of different orientations in the two lattices, but they differ in the way that they treat elastic stiffness. In a first group of models (Bollmann and Nissen, 1968; Bonnet and Durand, 1972; Robinson et al., 1971, 1977; Fleet, 1982) the only input data are the lattice parameters of the two phases. In a second group of models the elastic energy necessary to maintain coherency between the two lattices is minimized; the input data are the lattice parameters as well as the elastic stiffness coefficients (Cahn, 1968; Willaime and Brown, 1974).

In a recent paper on the calculation of the orientations of phase and domain boundaries in crystalline solids, Fleet (1982) presented a three-dimensional lattice misfit model and compared it to previous models of Bollmann and Nissen (1968) and Willaime and Brown (1974), as applied specifically to the calculation of interface orientations in feldspar intergrowths. Fleet contested the validity of the model and some of the conclusions in the paper by

[1] In very special cases, coherency can be obtained without any strain in the crystal, i.e., by a small rotation of the lattices; then both the surface energy term and the strain energy term are small.

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Willaime and Brown, to which we here reply, and we also correct several errors in Fleet's paper as applied to feldspars.

Comparison of the models—strain between the two lattices

The different models so far proposed use different theoretical approaches, but the calculated interface orientations may be compared easily. For each model, it is postulated that the two crystals have a boundary which is an equivalent (*hkl*) plane in the two lattices. In low symmetry crystals, it is rarely possible to find such a plane in which structurally equivalent vectors have the same length in the two lattices. This implies that a strain will be necessary to obtain exact coincidence on this plane between the two crystals.

Choosing orthogonal axes in the boundary plane, this strain may be expressed by a two-dimensional strain tensor whose components ϵ_{ij} can be calculated from the strain free unit-cell parameters of the two crystals. This strain tensor is indeed calculated in Fleet's model (F), in the Willaime-Brown model (WB), as well as in the Bollmann-Nissen model (BN). The models differ in the way their authors use the components of this tensor to determine the optimal boundary.

For easy comparison between these three models, it is convenient to use the strain tensor in its diagonal form. This is obtained through a suitable choice of reference axes in the boundary plane, i.e. parallel to the principal strains.

The strain tensor

$$\epsilon_{ij} = \begin{bmatrix} \epsilon_{11} & \epsilon_{12} \\ \epsilon_{12} & \epsilon_{22} \end{bmatrix}$$

is transformed into

$$\epsilon = \begin{bmatrix} \epsilon_1 & 0 \\ 0 & \epsilon_2 \end{bmatrix} \text{ with } \epsilon_{11} + \epsilon_{22} = \epsilon_1 + \epsilon_2$$

As demonstrated by Willaime and Brown (1974, p. 32; see also Willaime, 1981) either ϵ_1 and ϵ_2 in the plane of minimum strain have the same sign (and there is one plane orientation of minimum strain), or one of them is zero (and there are two different plane orientations with the same minimum strain).

Bollmann and Nissen (1968: optimal phase boundary model derived from the 0-lattice theory) minimized a parameter, P, which varies in a monotonic fashion with the surface energy:

$$P = \frac{b_1}{d_1} + \frac{b_2}{d_2} \qquad (1)$$

where b_1 and b_2 are the Burgers vectors of dislocations of spacing d_1 and d_2. These arrays of misfit dislocations do not necessarily exist in the boundary plane, but, if they did exist, they would compensate for the differences in parameters between the two lattices. Using the strain tensor notation

$$P\,(BN) = \epsilon_1^2 + \epsilon_2^2 \qquad (2)$$

Fleet (1982: lattice misfit model) generalized to three dimensions the previous two-dimensional model of Robinson et al. (1971, 1977). In this model, the optimal interface orientation is obtained by minimizing a strain, the "area misfit" P (F), between equivalent planes in the two lattices:

$$P\,(F) = |\epsilon_{11}| + |\epsilon_{22}| \qquad (3)$$

In Fleet's (1982) paper, no indication is given about the orientation of the reference axes; we assume that the axes are parallel to the principal two-dimensional strains in the boundary plane. Then

$$P\,(F) = |\epsilon_1| + |\epsilon_2| \qquad (4)$$

(If the choice of the axes is different, this relation is still valid when ϵ_{11} and ϵ_{22} are both positive or negative).

For comparison with the results obtained with his own model, Fleet (1982) calculated a "coherent elastic strain energy" by setting elastic strain components equal to the corresponding principal two-dimensional strains. This procedure does not measure the elastic energy necessary for maintaining coherency between the lattices, because it does not take into account the necessary deformation of the lattices perpendicular to the boundary plane. The value calculated by Fleet is

$$W\,(F) \propto 1/2\,[c_{11}\,\epsilon_1^2 + c_{22}\,\epsilon_2^2 + 2\,c_{12}\,\epsilon_1\,\epsilon_2] \qquad (5)$$

c_{ij} being the elastic stiffness coefficients of the material expressed in the chosen reference axes.

Cahn (1968) and Willaime and Brown (1974, coherent elastic model) determined the optimal boundary plane by minimizing the elastic strain energy necessary to maintain coherency between the two crystals.

In this procedure, the three dimensional strain resulting from a two-dimensional coherency is calculated, taking into account the fact that stress components outside of the plane of the interface are zero. Therefore the elastic strain energy is (Willaime and Brown, 1974, p. 321)

$$W\,(WB) = 1/2 \sum_{ij} c_{ij}\,\epsilon_i\,\epsilon_j, \text{ with } i = 1, 2 \text{ and } j = 1,2,3,4,5 \qquad (6)$$

If the variation of the c_{ij} with the boundary orientation is much smaller than the variation of the ϵ_i, the elastic anisotropy of the medium is negligible and the elastic energy can be calculated as if the medium were isotropic, i.e., $c_{11} = c_{22}$, $c_{12} = c_{13} = c_{21} = c_{23}$ and $c_{14} = c_{15} = c_{24} = c_{25} = 0$.

Consequently:

The "isotropic elastic strain energy" calculated by Willaime and Brown (6) becomes

$$W\,(WB) \propto \tfrac{1}{2}\,c_{11}\,(\epsilon_1^2 + \epsilon_2^2) + \tfrac{1}{2}\,c_{12}\,(2\,\epsilon_1\,\epsilon_2 + \epsilon_1\,\epsilon_3 + \epsilon_2\,\epsilon_3),$$

and assuming a constant-volume deformation, $\epsilon_1 + \epsilon_2 + \epsilon_3 = 0$, then:

$$W\,(WB) \propto \tfrac{1}{2}\,(c_{11} - c_{12})\,(\epsilon_1^2 + \epsilon_2^2) \text{ and we will note:}$$
$$P\,(WB) = \epsilon_1^2 + \epsilon_2^2 \text{ in the following.} \quad (7)$$

The "isotropic elastic strain energy" calculated by Fleet (5) becomes

$$W\,(F) \propto \tfrac{1}{2}\,c_{11}\,(\epsilon_1^2 + \epsilon_2^2) + c_{12}\,\epsilon_1\,\epsilon_2.$$

In a feldspar glass (Willaime and Brown, 1974, Table 2), $c_{11} \sim 4\,c_{12}$, therefore

$$W\,(F) \propto \tfrac{1}{2}\,c_{11}\,(\epsilon_1^2 + \epsilon_2^2 + 1/2\,\epsilon_1\,\epsilon_2) \text{ and we will note}$$
$$P'\,(F) = \epsilon_1^2 + \epsilon_2^2 + 1/2\,\epsilon_1\,\epsilon_2, \text{ in the following.} \quad (8)$$

From the above, it can be seen that all the models determine the optimal boundary by minimizing very comparable quantities (for the special case of an elastically isotropic medium).

Bollmann and Nissen minimized

$$P\,(BN) = \epsilon_1^2 + \epsilon_2^2. \quad (2)$$

Fleet minimized $P\,(F) = |\epsilon_1| + |\epsilon_2|$; it follows that he also minimized

$$P^2\,(F) = \epsilon_1^2 + \epsilon_2^2 + 2\,|\epsilon_1\,\epsilon_2| = \epsilon_1^2 + \epsilon_2^2 + 2\,\epsilon_1\,\epsilon_2 \quad (9)$$

(because, as seen above ϵ_1 and ϵ_2 never have opposite signs).

In his calculation of elastic energy, Fleet minimized

$$P'\,(F) = \epsilon_1^2 + \epsilon_2^2 + 1/2\,\epsilon_1\,\epsilon_2. \quad (8)$$

Willaime and Brown minimized

$$P\,(WB) = \epsilon_1^2 + \epsilon_2^2. \quad (7)$$

Comparison of the models. Effect of elastic anisotropy

The lattice misfit model of Fleet (1982) and the models of Bollmann and Nissen (1968), Bonnet and Durand (1972) and Robinson et al. (1971, 1977) do not work where the misfit is isotropic, i.e., where the strain ϵ_{ij} does not vary with the interface orientation. They cannot be applied to exsolution in cubic crystals and they also fail to distinguish between *h* and *k* indices in hexagonal, tetragonal and trigonal crystals.

The coherent elastic energy model works for all crystals, including cubic ones. It is particularly powerful where the misfit anisotropy is small and the medium is elastically highly anisotropic; results obtained using the coherent elastic model should be correct, whereas those calculated using the other models should be much less precise. On the other hand, in a medium with low elastic anisotropy, there would be no special advantage in using the coherent elastic energy model, but it would still give suitable results.

When several models are available, a determination of the best one depends on the criteria used. The best one may be the easiest to use or it may be the one that is the nearest to physical reality; it may be the one for which the calculated values are closest to the observed values. In light of the above, the paper by Fleet (1982) will now be discussed with special reference to the validity of the comparison he made between two of the previous models for the case of feldspar intergrowths.

Application to feldspar intergrowths

Fleet (1982) applied his model to feldspar intergrowths, which are of great complexity as three exsolution regions occur in plagioclase and one in the alkali feldspars, the latter being complicated, however, by possible symmetry changes in both exsolved phases.

Effect of elastic anisotropy

The feldspars are of low symmetry and rather strong anisotropy of compositional expansion and small elastic anisotropy. They are thus not a good test for distinguishing between the two groups of models, and the calculated orientations are very similar in most cases. This was clearly pointed out by us (Willaime and Brown, 1974, p. 236), as the elastic anisotropy plays only a *secondary* role (see also Fleet, 1981, p. 66). This, however, in no way allows one to suggest that elastic anisotropy plays *no role at all* in the calculation of the orientations of coherent lamellae, as maintained by Fleet.

Physical significance of the models

The question is whether there are theoretical reasons for asserting that one model more closely approaches physical reality than the others. As pointed out by Fleet (1981, p. 67, ". . . there are conceptual problems in equating coherent elastic strain energy directly with interface energy," but certainly neither Cahn (1968) nor ourselves (1974) considered that the two could be equated. Nevertheless, if the exsolution is coherent, which is common at least in the *early* stages when the lamellar orientation is determined, then a coherent model will be physically more realistic (Tullis, 1975; Robin, 1974). If, on the other hand, the lamellae are incoherent from the *start* of the exsolution, the optimal boundary model of Bollmann and Nissen (1968), which minimized the energy of a dislocation array, seems more appropriate (Nevertheless, it must be noted that, from the results of the last section, these two models are strictly equivalent when the elastic anisotropy is negligible.)

Before using a model for calculating the orientation of interfaces in anisotropic materials, it may be of interest to examine the degree of coherency of the interfaces at the beginning of and during exsolution. If one restricts discussion to fine-scale intergrowths (i.e. to the early stages of exsolution), such as plagioclase/plagioclase intergrowths and cryptoperthites, the evidence available suggests (almost) complete coherency. All diagonally associ-

ated cryptoperthites examined by Brown et al. (1983) are fully coherent, as shown by high-resolution microscopy (Brown and Parsons, 1984; Brown, 1983). The common occurrence of strained cryptoperthites also speaks in favor of coherency. No report of dislocations along plagioclase/plagioclase interfaces has been published. Only two reports of dislocations, one periodic, are known in cryptoperthites, both An-rich, the high An-content blocking diffusion and interface rotation at low temperature (Brown and Willaime, 1974; Brown and Parsons, unpublished data). Coarser intergrowths, such as microperthites and macroperthites, have rarely been studied by TEM and little is known about their degree of coherency except for two microperthites with periodic dislocations (Gandais et al., 1974; Aberdam, 1965). The presence of periodic dislocations shows that such interfaces are semicoherent (Brown and Parsons, 1984), and for such cases, the coherent elastic model is still appropriate as well as the Bollmann-Nissen model.

Comparison between observed and calculated orientations

Fleet claimed that his model gave calculated orientations closer to the observed ones than the coherent elastic model. It must be pointed out that his calculation procedure for elastic strain energy (5) differs from that defined in the coherent elastic model (6), which could invalidate his conclusions. Furthermore, a table comparing sets of data calculated with both models to *observed* orientations is essential to justify the better precision of one or the other, preferably with *angular positions* rather than indices which are hard to compare. Fleet did not give such a table, so that the reader is unable to judge for himself.

Fleet (1982, p. 932) is correct when he supposes some imprecision in measurements made from TEM observations, but he should cite the authors he suspects to have rounded the indices of the observed orientations in the diagonal association to make them consistent with the calculated boundaries of Willaime and Brown (1974). The diagonal association was observed by Brown et al. (1972) and the orientations easily measured (on (001) cleavages and not (010) as reported by Fleet); they published their results before extending Cahn's model to crystals of low symmetry.

As previously noted, the elastic anisotropy of feldspars is low and the calculations of coherent elastic energy would not be very different using elastic stiffness coefficients of a feldspar crystal or using those of an isotropic feldspar glass. Therefore the differences in the calculated optimal boundaries for the different models should be mainly related to the difference in the parameters P (BN), P^2 (F), P′ (F) and P (WB) defined in a previous section.

With certain sets of lattice parameters two plane orientations have the same minimum strain; as mentioned previously, this implies either ϵ_1 or ϵ_2 equal to zero. In this case, the parameters P (BN), P^2 (F), P′ (F) and P (WB) are strictly equal ($= \epsilon_1^2$ or $= \epsilon_2^2$) and the calculation by the three methods should give the same results. This situation occurs for the calculation of exsolution lamellar orientation in *plagioclases*. In fact there is very good agreement between observed orientations and those calculated using the different models (Nissen, pers. comm., 1972; Willaime and Brown, 1974; Fleet, 1982).

For alkali feldspar exsolution, there is only one lamellar orientation in most cases. The parameters P^2 (F) and P′ (F) differ from P (BN) and P (WB), and therefore they may give different calculated orientations that are of interest to compare with the observed ones.

Whichever model is used, the choice of input data is important and errors in the lattice parameters (or in the elastic stiffness coefficients) will affect the calculated orientations. There are indeed some problems in the choice of input data for perthite intergrowths, except for those in which both phases are monoclinic. All TEM studies on cryptoperthites (except rare monoclinic/monoclinic ones) have shown that either the Ab-rich phase alone or the Or-rich phase as well are triclinic. In such cases the triclinic phases are twinned in such a way as to show that both phases were *monoclinic* at the beginning of exsolution—the overall diffraction symmetry is monoclinic. We are thus not free to use any combination of input data, but must take into account the morphological development of the intergrowth. This explains the choice of input data of Bollmann and Nissen (1968) and of Willaime and Brown (1974). Fleet (1982), however, took no account of such constraints and attempted to use his calculations to disprove deductions made by us from *TEM observations*. We will illustrate this by a discussion of only the diagonal association, as the diagonal association develops from the normal perthite texture, and braid perthite develops from the diagonal association (Ramberg, 1972; Lorimer and Champness, 1973; Willaime et al. 1976; Brown et al., 1983; Brown and Parsons, 1984). All stages in this sequence are given by Brown and Parsons (1984).

In normal cryptoperthites (moonstones) lamellae of periodically albite or pericline twinned albite occur with lamellae of monoclinic Or-rich feldspar. It was clearly shown by Bollmann and Nissen (1968) that agreement between calculated and observed orientations was good only when they took account of the periodic twins. This was confirmed by our calculations (Willaime and Brown, 1974). The overall monoclinic symmetry shows that twinning occurred *after* exsolution, as does the relationship between twin width and lamellar thickness (Willaime and Gandais, 1972; McLaren, 1974; Willaime et al., 1976). Calculations by Fleet (1982, Table 1, normal perthite, numbers 2, 2A, 3 and 3A) using cell parameters of sanidine and untwinned anorthoclase or high albite have no applicability to perthites and it is *singularly inappropriate* to have compared the misfit model and coherent elastic model using only such physically unrealistic input data (Fleet, 1982, calculation 2, Table 1, and Fig. 5).

The situation is, however, worse for the diagonal

association. This texture consists of periodically albite twinned low albite lamellae which may pinch and swell and parallel-sided zig-zag lamellae of maximum microcline. The interface between the two is oblique to b^*, each zig (and each zag) corresponding to a single microcline orientation and to an average monoclinic albite twinned albite. The indices of the interface cannot be chosen freely—($\bar{6}\bar{6}1$) and ($\bar{6}61$) are equivalent in the overall monoclinic symmetry but *not* for the triclinic microcline. The indices of the interface must be given relative to the triclinic microcline lattice. From dark-field and high resolution studies (Brown, unpublished data) the indices correspond to ($\bar{6}\bar{6}1$), the same result obtained, but not clearly stated, by Brown et al. (1972), and definitely not to ($\bar{6}61$).

Calculation by Fleet (1982, Table 1, numbers 4, 5 and 5A) using *untwinned* high or intermediate albite have no applicability to the early stages of diagonally associated cryptoperthites as can be shown by TEM observations—no cryptoperthite has ever been found which has untwinned albite lamellae, either parallel or oblique to b^*, in any Or-rich matrix whatever. Furthermore, Fleet's calculations give two minima, but no sign of two orientations has ever been seen. Only one of the six calculated minima (number 5b) is even close to the observed orientation of ($\bar{6}\bar{6}1$). Fleet does not give the calculated minima for the phases actually found, namely low albite and maximum microcline. In addition, the poles to the faces ($66\bar{1}$), ($63\bar{1}$), ($\bar{6}61$) and ($\bar{6}31$) are wrongly plotted in his Figure 5, as are ($\bar{8}61$) and ($86\bar{1}$) in his Figure 6—clearly ($\bar{6}01$), ($\bar{6}31$), ($\bar{6}61$), and (010) lie in the [106] zone.

The coherent elastic model, on the other hand, gives only one calculated minimum which varies from near ($\bar{6}01$), to near ($\bar{7}\bar{2}1$), to near ($\bar{8}\bar{5}1$) as the Or-rich phase varies from orthoclase through intermediate microcline to maximum microcline, in very good agreement with observed orientations.

To sum up for the diagonal association, it is erroneous for Fleet (1982, p. 934) to conclude that his analysis ". . . . indicates quite positively that the fine albite twin lamellae in the Na-feldspar *post-date* intergrowth development" (Fleet's italics), as this is contradicted by his own calculations (five out of six of his minima are in the wrong place), by TEM observations (McLaren, 1974; Willaime et al., 1976; Brown and Parsons, 1984) and by the impossibility, following Fleet's chronology, to suggest any explanation for the periodicity of the twin-lamellae in the diagonal association configuration. This periodicity is easily explained if twinning occurs at the stage of a normal perthite configuration (($\bar{6}01$) lamella orientation): the twin period is related to the width of the Ab-rich lamella (Willaime and Gandais, 1972; McLaren, 1974). If the twinning occurs in an already existing Ab-rich domain of variable thickness, it will depend on this thickness; the greater the local thickness of the Ab-rich lamella, the greater will be the width of the twins (see, for example, McLaren, 1974, Fig. 22, which shows the variation of the twin width in a lens-shaped Ab-rich lamella in an Or-rich feldspar).

Conclusions

In conclusion, Fleet's paper brings nothing new to our understanding of feldspars, as by his own admission calculated orientations using either model are similar in most cases. In spite of this, Fleet comes to conclusions which "quite positively" contradict our own conclusions, not for plagioclase or normal moonstone but for more complex cryptoperthites. As shown above, these differences come not from the models but from the choice of input data which are inappropriate for diagonally associated cryptoperthites as shown by TEM observations.

Therefore, Fleet's calculations cannot be used for a rigorous comparison of the different models. The coherent elastic model (Cahn, Willaime-Brown) is physically the most realistic if coherency exists and is maintained between the lattices during exsolution, whereas the Bollmann-Nissen model is the best if the lamellae are incoherent from the start of exsolution. Fleet's model cannot be related to any physical mechanism, but seems useful as it is simple. Nonetheless, these three models give very similar results because, in spite of differences in the initial principles, they calculate very similar parameters. Future progress in our knowledge of exsolution in minerals will probably not depend on the choice of a model, but rather on the choice of suitable input data, and on high quality observations on geologically well chosen specimens.

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