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Angular relationships between host and exsolution lamellae and the use of the Mohr circle

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Abstract

The Mohr circle for strain provides a rapid, precise, and calculation-free method of predicting angles of exsolution of clinopyroxene or clinoamphibole lamellae from a knowledge of their lattice parameters and those of the host. Agreement between predicted and observed angles indicates that host and lamellae have maintained 'coherency' during cooling; disagreement, on the contrary, indicates loss of 'coherency.' As suggested by Dr. P. Robinson and others, this information may become very useful in unravelling the cooling history of these minerals.

Introduction

Robinson *et al.* (1971), Jaffe *et al.* (1972), and Ross *et al.* (1972) report clinoamphibole and clinopyroxene "001" and "100" exsolution lamellae which are in fact not oriented parallel to (100) and (001) as often assumed (*e.g.* Deer *et al.*, 1963). Robinson *et al.* (1971) show that observed orientations are satisfactorily explained in many cases by the requirement that lattices of coexisting phases be 'matched' along the planes of contact. In all cases, the planes of the lamellae contain [010]; only their angles with the *a* and *c* axes vary among occurrences. Robinson and his co-workers predict this exsolution angle from measured values of lattice parameters for host and lamellae; generally, their predictions agree well with observations. For finding the "plane of dimensional best fit" Robinson *et al.* first used large-scale graphic representations of the two lattices in (010) (their Fig. 4); they later used a computerized numerical method based on that representation. The purpose of this note is to show that the *Mohr circle for strain* (*e.g.* Nye, 1957) provides the same result without any calculation. In addition, the Mohr circle gives immediately the angular deviation of any lattice direction, [*u*0*w*], between the two lattices.

Coherency

Boundaries along which two phases have maintained full continuity of their lattices are called *coherent boundaries* (*e.g.* Kelly and Nicholson, 1963). By extension, lamellae whose boundaries are coherent are called *coherent lamellae* (Robin, 1974a). The clinopyroxene (or clinoamphibole) lamellae considered here would be *truly coherent* if the repeat distances in the two lattices, that of the host and that of the lamellae, were identical not only along the direction of the trace of the lamella on (010), but also along [010]. The results of Robinson *et al.* imply that the first condition is often satisfied; as to the second, they only state (p. 926) that "generally the *b* dimensions of host and lamellae are nearly identical," without indicating to what extent the matter was investigated. If *b* dimensions of host and lamellae were *not* equal, the lamellar boundaries would only be semi-coherent.

Much of the development presented here is applicable whether *b* dimensions of host and lamellae are equal (true coherency) or not (semi-coherency). However some statements are valid only if the boundaries are truly coherent. For simplicity of writing we shall distinguish between *strictly coherent* boundaries, for which true coherency is assumed, and, simply, *'coherent'* boundaries, where only the equality of repeat distances along the trace of the lamella on (010) is assumed.

The Mohr circle for strain

Lattice strain between host and lamella

Differences in lattice dimensions (say, between an augite and a pigeonite) define a *strain*, formally similar to a thermal expansion or an elastic deformation. Both augite and pigeonite are monoclinic, *b* being the diad axis in both; for symmetry reasons *b* is also a

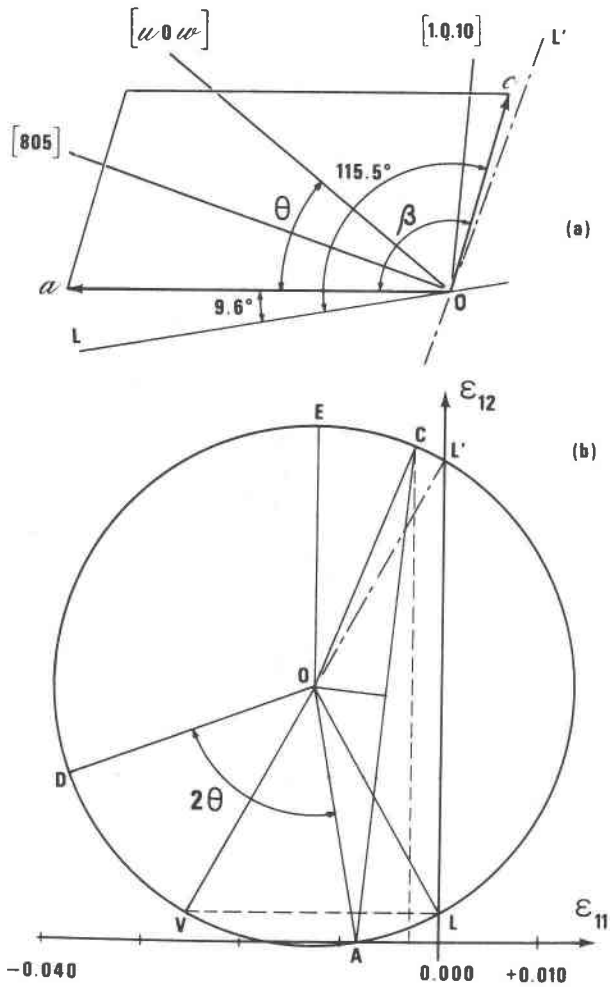

Fig. 1. (a) *a c* plane of clinopyroxene unit-cell. Parameters are those of an augite crystal described by Jaffe and Jaffe (1973) and cited by Robinson *et al.* (1971, Table 3, example 4 and Table 5). (b) Mohr circle (MC) for the lattice strain between the augite host and the pigeonite lamellae. Construction is explained in Appendix A. Points on the MC correspond to lattice directions in the crystal: A and C correspond to [100] and [001] respectively; E corresponds approximately to [1.0.10], which is thus the lattice direction that undergoes the largest angular deviation between the two lattices; [805] (V on the MC) on the contrary has the same direction in the two lattices.

diad axis for the strain tensor. In practice this means that the *b* axis is a principal direction of strain, the two other principal directions therefore lying in (010). Like stress, strain can be represented on a Mohr diagram, a particularly convenient representation when dealing with a plane which, like (010), contains two principal directions.

Mohr circle (MC)

Figure 1 shows the relationship between directions in the lattice, positions of representative points on the MC, and strains. The example used is that of a host augite and "001" pigeonite lamellae from the Hudson Highlands (Jaffe and Jaffe, 1973, cited by Robinson *et al.*, 1971, Tables 3 and 5). Lattice parameters for the augite are (Fig. 1a):

$$a_a = 9.77_6 \text{ Å}, \; c_a = 5.25_2 \text{ Å}, \; \beta_a = 105°55'; \quad (1a)$$

for the pigeonite:

$$a_p = 9.69_5 \text{ Å}, \; c_p = 5.23_6 \text{ Å}, \; \beta_p = 108°33'. \quad (1b)$$

The *b* dimensions are equal:

$$b_a = b_p = 8.89_0 \text{ Å}. \quad (1c)$$

Taking the host lattice as reference state, the lattice strain in (010) is completely defined by

$$\frac{\Delta a}{a} = \frac{a_p - s_a}{a_a} = \frac{-0.08_1}{9.77_6} = -0.008_3, \quad (2a)$$

$$\frac{\Delta c}{c} = \frac{0.01_6}{5.25_2} = -0.003_0, \quad (2b)$$

$$\Delta\beta = \beta_p - \beta_a = 2°38' = 0.050_0 \text{ rad}. \quad (2c)$$

The MC for strain (Fig. 1b) is easily constructed from the above data (Appendix A). The significance of the MC can be explained as follows. If any lattice direction, say [*u*0*w*], makes an angle θ with the *a* axis (Fig. 1a), point D which represents this direction on the MC (Fig. 1b) makes an angle of arc 2θ with point A, representing the *a* axis. In the Mohr coordinates the abscissa of D is equal to the longitudinal strain along [*u*0*w*], that is, the relative increase of the repeat distance along [*u*0*w*] in the lamella with respect to that in the host. The *ordinate difference* between any two points, say A and D, is equal to the *change* in angle (in radians) between the two corresponding directions, here [100] and [*u*0*w*], when comparing this angle in the lamella and in the host. The choice of an origin for the ordinates is somewhat arbitrary, and different origins can be thought of as being equivalent to different solid rotations of one lattice with respect to the other (Appendix B).

The MC in Figure 1b intersects the ordinate axis in two points, L and L'; this means that there is no longitudinal strain along the corresponding lattice directions, or, in other words, that *repeat distances* along these directions are the same for both phases. L and L' are therefore the directions of "dimensional best fit" sought by Robinson *et al.* The angles between the L direction and the *a* and *c* axes are 9.6° and 115.5° respectively (half the angles which L makes with A and C respectively, on the MC). Op-

tical measurements of the angle between the actual "001" lamellae and the c axis give 116° ± 1° (Robinson *et al.*, 1971, Table 3). Like the numerical calculations by Robinson *et al.*, the Mohr circle construction thus demonstrates that the "001" pigeonite lamellae are 'coherent' within the augite host. Because $b_a = b_p$ (Equation 1c) the lamellae are in fact *strictly coherent.*

Angular deviation of lattice directions

Although L differs from $[30\bar{1}]$ by only 0.3°, (and from $[32.0.\overline{11}]$ by 0.003°!) it is not a rational lattice direction. By the very nature of coherency, the lattice direction L, and the lattice plane defined by L and [010], have the same orientation in the host and in the "001" lamellae. Such is not the case for other lattice directions $[u0w]$. The angular deviation of $[u0w]$ between the two lattices, which we can call $\delta[u0w]$, can be read directly on the MC: it is equal to the difference between the ordinate of D and that of L; more simply, it is equal to the ordinate of D if L is taken as origin of the ordinate axis (Appendix B). Thus the angular deviation of the a axis is:

$$\delta[100] = 0.0030 = 3.4';$$
$$\text{similarly } \delta[001] = 0.0470 = 2°\ 41.6'.$$

The lattice direction which undergoes the largest deviation is represented by point E on the MC, close to [1.0.10], with an angular deviation of 0.0490 = 2°48.5′. In contrast, the lattice direction represented by V, close to [805], undergoes no angular deviation,[1] as V has the same ordinate as L.

Discussion

Assumption of infinitesimal strain

Representation of strain by a MC implies that strain is infinitesimal. Errors introduced by this approximation lie, roughly, in equating cos 0.052 to 1, where 0.052 is the diameter of the MC. The error is therefore 0.125 percent. This is smaller than the uncertainty in the components of the strain, which is between 3 percent (Eq. 2b) and 0.5 percent (Eq. 2c). The error arising from the above uncertainty in the X-ray data can itself be estimated empirically by redrawing the MC for slightly different values of the data in Equations 2. For example, if 0.0083 (Eq. 2a) is replaced by 0.0081, the predicted angle of the lamellae with the c axis becomes 115.3° instead of 115.5°. Both predictions lie well within the limits of accuracy of optical measurements of exsolution angles (≃ 1°).

Applications

The Mohr-circle construction, like the numerical method of Robinson *et al.*, can serve to establish whether or not host and lamellae are 'coherent': they are 'coherent' if predicted and observed angles agree, they are not coherent if prediction and observation disagree. When exsolution lamellae between clinopyroxenes first form, it is energetically advantageous for them to maintain 'coherency.' The stress-free (single phase) lattice parameters of coexisting clinopyroxenes are such that there always are two directions along which 'coherency' can be maintained with no elastic distortion[2] (and consequently no elastic strain energy) and in this manner the high interfacial energy of incoherent boundaries can be saved. However, as Robinson *et al.* point out, changes in compositions of the coexisting phases (*solvus* effect) and differences in thermal strain (*thermal expansion* effect) during cooling change the angles of 'stress-free coherency.' The directions of existing lamellae are set and cannot be changed, and therefore, one of the following may happen:

1. *'Coherency' is not lost,* for example, as a result of rapid cooling. Lamellae may become elastically strained and acquire lattice parameters which are anomalous compared to those of homogeneous crystals of the same composition; if these anomalous parameters are used, the MC will still predict the observed exsolution angle. The lattice parameters of the two differently oriented sets of lamellae may be detectably different after cooling, because thermal contraction is not isotropic. Lattice parameters of the "100" pigeonite lamellae in the augite studied by Jaffe and Jaffe (1973) thus differ significantly from those of the "001" lamellae given in equation 1b. In such a case, the lattice parameters of the "001" la-

[1] Robin (1974 a,b) called such an undeviated lattice direction the *characteristic vector* of the coherent boundary. The longitudinal strain along this direction is equal to the abscissa of V on the MC, *i.e.* −0.0255. In the present case of strict coherency, this longitudinal strain is also equal to the volumetric strain between the two lattices.

[2] In case of strictly coherent clinopyroxenes, the maintenance of equal b parameters in the host and the lamellae does require some nonhydrostatic stresses, mostly in the lamellae, which therefore acquire some elastic strain energy. That strict coherency is nevertheless maintained in some cases means that loss of coherency is either thermodynamically more costly or is prohibited for kinetic reasons.

mellae can only serve to predict the exsolution angle for the "001" lamellae themselves.

2. *'Coherency' is lost,* as perhaps during slow cooling. Stresses which otherwise would slowly increase as a result of the solvus and thermal expansion effects are thus relieved. Further growth of these lamellae would require a migration of now incoherent boundaries; this migration appears to be difficult, and another set of lamellae develops instead which has the new angle of 'stress-free coherency.' This second set may in turn lose 'coherency' during further cooling. Ross *et al.* (1972) report examples of such multiple exsolutions in which characteristically only the third (and last) set of lamellae has preserved coherency.

Our present knowledge of thermal expansion in the pyroxene quadrilateral and our understanding of the kinetics of 'coherency' loss are not sufficient to give more than qualitative information on relative cooling rates. As our knowledge improves, analyses such as the one presented here may tell us much about cooling histories of minerals and rocks.

Appendix A: construction of the Mohr circle

After the coordinate axes are drawn, with an appropriate scale, the procedure is as follows:
Point A (*a* axis) is positioned, with an arbitrary ordinate of zero[3]:

$$\begin{cases} x = -0.0083, \text{ (from Eq. 2a)} \\ y = 0. \end{cases}$$

Point C (*c* axis) is also marked, with

$$\begin{cases} x = -0.0030 \text{ (from Eq. 2b)}, \\ y = +0.050 \text{ (from Eq. 2c)}. \end{cases}$$

The MC must satisfy the following requirements:
(i) it must go through A and C;
(ii) its angle of arc AC (counted clockwise, as is $\beta = \angle aOc$ in Fig. 1a) must be equal to 2β given in Equation 1a.

The center of the MC is therefore at the intersection of the perpendicular bisector of AC and of a line through A making an angle

$$|\mathrm{CAO}| = \beta - 90° = 15° 55'.$$

The MC can thus be drawn, with a radius $|\overline{OA}| = |\overline{OC}|$. An ambiguity may arise in deciding whether C should be drawn above or below A. For an angle $\beta = \angle aOc$ counted clockwise (the present case), C is above A if $\Delta\beta$ (Eq. 2c) is positive, C is below A if $\Delta\beta$ is negative. This rule would be reversed if arc AC = 2β were counted counterclockwise on the MC.

[3] The significance of this choice of origin for the ordinate axis is discussed in Appendix B.

Appendix B: relationship between the choice of origin of ordinate in the Mohr coordinates and a rotational component of strain

Commonly, the origin of ordinates in the Mohr coordinates is chosen such that the center of the MC has an ordinate of zero. The ordinate of a point D, corresponding to a direction [*u*0*w*], is then equal to the change in angle between [*u*0*w*] and the principal directions of strain; it is in fact the tensor shear strain. Such an origin is convenient when we know, or are only interested in, the components of non-rotational, symmetric strain.

Other origins, however, are preferable in the present case. The X-ray data are reported in terms of β angle; therefore the strain of the lattice is known, in part, in terms of $\Delta\beta$ (Eq. 2c). If, as was done in Appendix A and Figure 1b, we choose point A [100] as having an ordinate of zero, then the ordinate of point D [*u*0*w*] on the MC is equal to the change in angle between [*u*0*w*] and [100]. Thus, in particular, the ordinate of C, corresponding to [001], must be equal to $\Delta\beta$; this fact was used in the construction given in Appendix A.

In effect, choosing the origin of ordinates at A is equivalent to Robinson *et al.*'s Figure 4, in which those authors have drawn the lattices of the host and of the lamellae with their *a* axes in coincidence. The angular deviation of a lattice direction [*u*0*w*] between the two lattices as drawn in that figure is equal to the ordinate of D, representing [*u*0*w*], on the MC.

In the crystals, the *a* axes of the host and of the lamellae do not coincide, rather the lattice direction L has the same orientation in the two coherent lattices (see main text). The ordinate difference between point D and point L on the MC thus gives the real angle between [*u*0*w*] in the lamellae and [*u*0*w*] in the host. In other words, if we now take L as the origin of ordinates, the ordinate of any point on the MC is equal to the actual angular deviation of the corresponding lattice direction between the host and the lamellae. This angular deviation is, of course, zero for L.

In effect, choosing this new origin for the ordinate axis is equivalent to the small rotation of the superimposed lattices made by Robinson *et al.* to reduce the misfit to zero. More generally any change in origin for the ordinates is equivalent to a solid body rotation about [010] of one lattice with respect to the

other. Such changes in origin do not affect the determination of L.

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