

A lamellar texture with chemical contrast in grandite garnet from Nevada

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Abstract

Detailed investigations of the peculiar lamellar texture in an iridescent garnet from Adelaide, Nevada, described previously, were made by SEM, EPMA, AEM and X-ray diffraction methods. Observations by SEM showed that the lamellar texture was due to compositional variation. Quantitative analyses by EPMA and AEM and X-ray diffraction experiments revealed that the lamellar texture was composed of alternate Fe-rich and Fe-poor lamellae with small differences in chemical composition and cell parameter. The most probable interpretation of the lamellae is an exsolution texture, although the configuration of the lamellae is somewhat different from exsolution textures previously described.

Introduction

Iridescent garnet from the Adelaide mining district, Nevada, U.S.A., has been described by Ingerson and Barksdale (1943). They found two types of lamellar textures in the iridescent garnet under a polarizing microscope, designated “octahedral lamellae” and dodecahedral lamellae, because these lamellae were parallel to “{111}” and {110}, respectively, in their observation. They concluded on optical observations that iridescence of the garnet was caused by the “octahedral lamellae” and that the “octahedral lamellae” were polysynthetic twins.

The origin of iridescence has, however, been ascribed recently to submicroscopic twinning parallel to crystal surfaces with a periodicity of about 1000 Å (Hirai and Nakazawa, 1982). The “octahedral lamellae” show rather strange shapes and such a lamellar texture has not been found in any other grandite garnet. The subject of this paper is to clarify the nature of the “octahedral lamellae”. The dodecahedral lamellae of Ingerson and Barksdale are merely oscillatory compositional zoning common in grandite garnet (Lessing and Standish, 1973;

Murad, 1976) and are not dealt with at length here.

Experimental

Observation by polarizing microscope

The samples used in this paper were provided by J. D. Barksdale through F. P. Okamura. Detailed description of the garnet has been made by Ingerson and Barksdale (1943).

Thin sections parallel to a crystal surface of {110} and normal to [001] were prepared from a dodecahedral crystal. Miller indices used in this paper are referred to pseudo-cubic symmetry for convenience. In the thin section normal to the [001] well defined zones parallel to crystal surface were observed under crossed nicols (Fig. 1a). The zonal texture corresponds to the dodecahedral lamellae of Ingerson and Barksdale's observations, and is oscillatory compositional zoning due to the compositional changes during crystal growth. The compositional variation was confirmed by electron probe microanalysis (EPMA).

The “octahedral lamellae” can be seen in several compositional zones (Fig. 1). The elongated directions of the lamellae are rather variable, often wavy

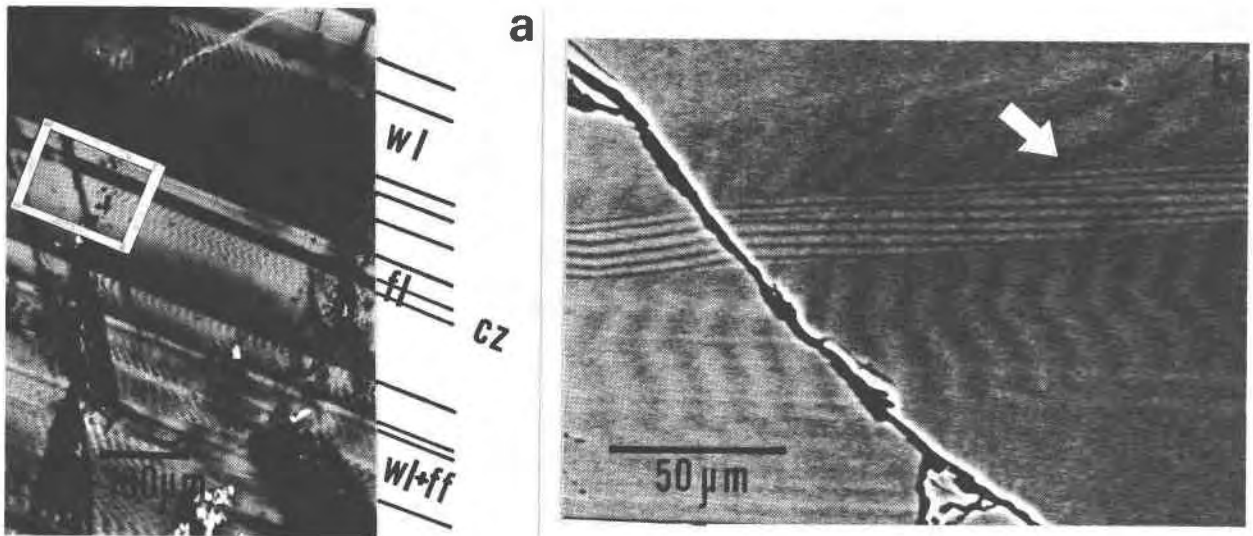


Fig. 1. (a) Optical micrograph of a thin section sliced normal to an axis of [001]. Well defined compositional zones run parallel to a crystal surface. "Octahedral lamellae" are distributed throughout several compositional zones: they are wavy or sigmoid (wl) in some zones and almost straight (fl) in other zones. A square shows the area where a back-scattered electron image (BEI) was taken.

In some wavy part of the "octahedral lamellae", finer texture-like fringes (ff) are also seen, cutting the wavy lamellae. These fringes drift and change their shape with tilting of the thin section. Because compositional difference was not detected in BEI for these fringes (Fig. 1b), they may not be intrinsic but an optical phenomenon probably caused by superposition of the wavy lamellae and the submicroscopic periodic twinning which is the source of the iridescence (Hirai and Nakazawa, 1982). (b) BEI of the area shown in Figure 1a. The contrast coincides completely with the lamellar texture observed by a polarizing microscope. Any contrast corresponding to the finer fringes (ff) was not detected.

or sigmoid, (wl, Fig. 1a) and sometimes straight (fl, Fig. 1a), and are not exactly parallel to {111} (Ingerson and Barksdale, 1943). Some preferential orientations are, however, observed to be parallel to symmetrically equivalent {110}. The preferential orientations can be seen in the thin section prepared parallel to a crystal surface of {110} (Fig. 2a).

The lamellar texture consists of alternation of anisotropic and almost isotropic lamellae. In rare cases, the isotropic lamellae show incomplete extinction under crossed nicols: faint brightness remains under rotation of the microscope stage. The incomplete extinction may be due to scattering of light at the boundaries between the lamellae. Average thickness of the lamellae is about ten microns. The isotropic lamellae are larger than the anisotropic ones. The ratio of the thicknesses of two kinds of lamellae is constant within a compositional zone, but it varies among zones.

To confirm that the texture is produced by chemical differences, and not simply an optical effect, back-scattered electron images (BEI) by SEM in the two-detector mode should be informative. If the lamellae have any chemical differences, this should appear as an atomic number contrast on BEI (Rob-

inson and Nickel, 1979; Hall and Lloyd, 1981). The results will be described in the next section.

SEM, EPMA and analytical electron microscope (AEM) analyses

BEIs of the areas shown in Figure 1a and 2a were obtained by SEM (JOL-50A) and were reproduced in Figure 1b and 2b. Fluorescent X-ray images for Al show that the dark area of BEIs have relatively high concentration of Al (Fig. 1c). For another specimen, element concentration maps for Fe and Al were drawn by EPMA (JXA-50A). These maps indicated clearly an inverse relationship between concentration of Fe and Al across the lamellae, which implies substitution of trivalent iron.

Quantitative analyses by EPMA indicated that the compositions are An_{98} and An_{90} for Fe-rich and Fe-poor lamellae, respectively. The thicknesses of the Fe-rich and the Fe-poor lamellae analyzed by EPMA are 8–10 and 2–4 microns, respectively. These thicknesses are near the limit of the spatial resolution of EPMA. The analytical electron microscope (AEM), which has much better spatial resolution (*e.g.*, Lorimer and Cliff, 1976), was, therefore, required for more accurate measurements.

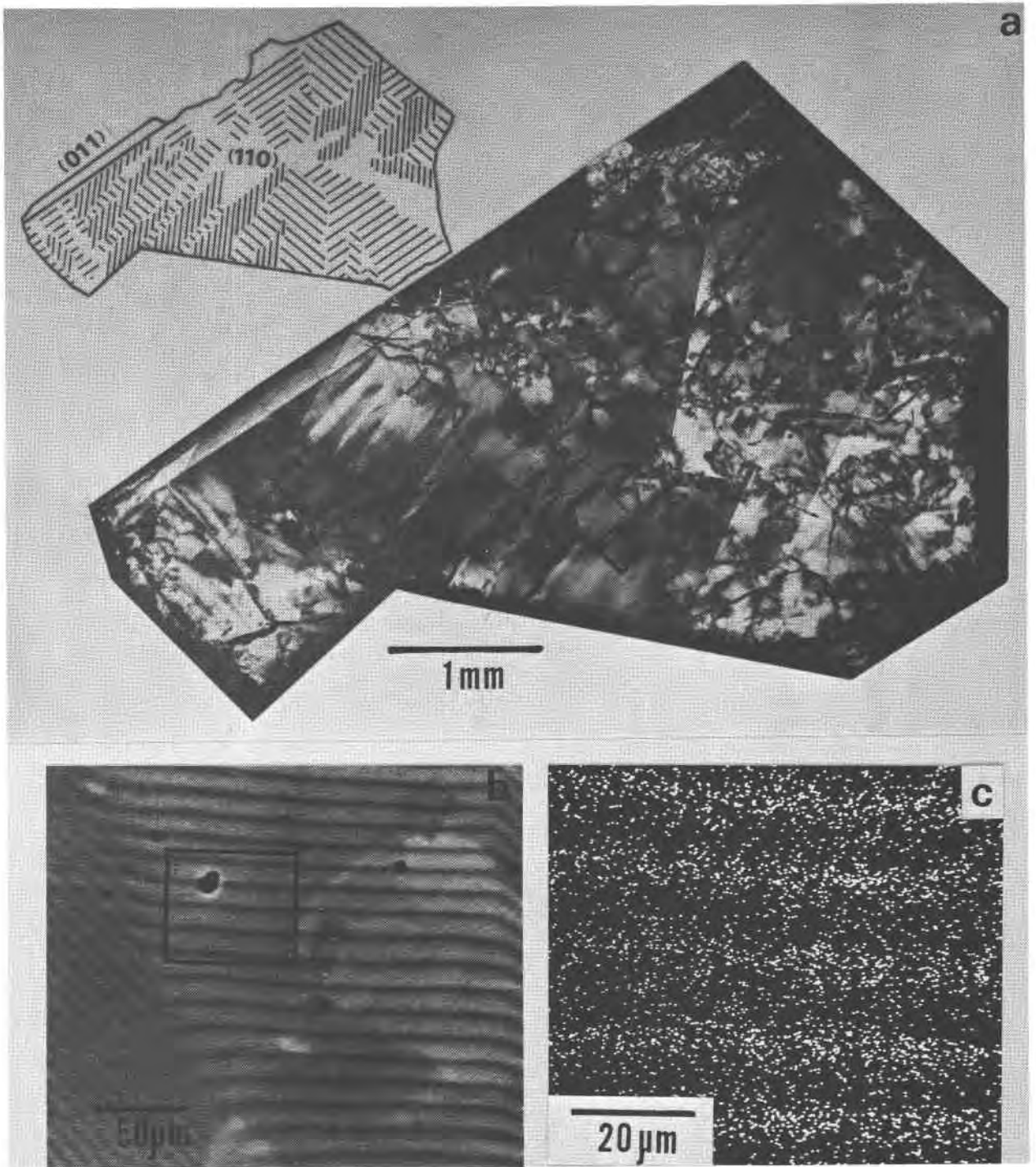


Fig. 2. (a) A polarizing micrograph of a thin section parallel to a crystal surface of $\{110\}$. The preferential orientation of the lamellae, parallel to $\{110\}$, may easily be recognized. A square shows the area where BEI was taken. (b) BEI of the area shown in Figure 2a. Fluorescent X-ray image was obtained from the area shown by a square. (c) Fluorescent X-ray image of Al. The dark areas in Figure 2b correspond to the parts of relatively higher concentration of Al.

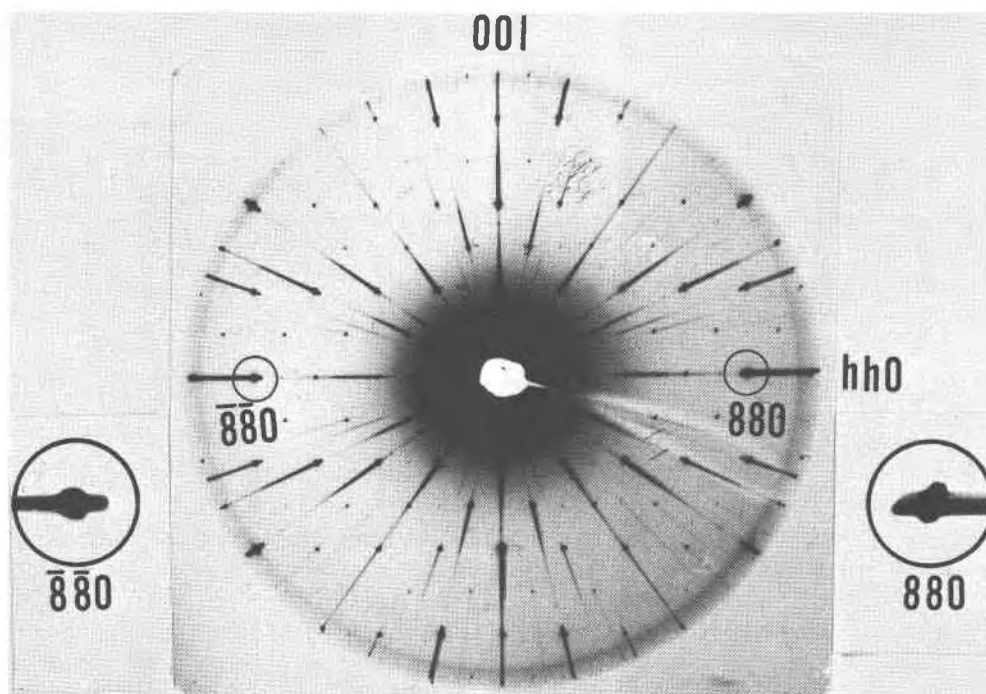


Fig. 3. X-ray precession photograph showing two reciprocal nets misoriented relative to each other. Because a reciprocal net of stronger spots is placed parallel to the film plane, the other weaker net is asymmetrical with respect to the center of the photograph. Insets are parts of enlarged precession photograph around a Friedel pair marked by circles, indicating that the weaker net is very slightly larger in reciprocal space.

Specimens for AEM were prepared by ion-thinning techniques from the thin sections prepared for the polarizing microscope. Analyses were carried out at accelerating voltage of 200 kV with beam diameter of about 200 Å (JEOL-200CX with KEVEX-7000). The average compositions measured were $\text{An}_{92}\text{Gr}_7\text{Sp}_1$ and $\text{An}_{88}\text{Gr}_{10}\text{Sp}_2$ for Fe-rich and Fe-poor lamellae, respectively. The small difference measured for these lamellae might be partially related to some technical reasons such as randomization or loss of elements by ion-spattering (Wehner and Anderson, 1970) or by converged electron beam for analysis (Cliff *et al.*, 1976), but because of the difference in observed cell parameters, described later, the difference in composition is probably real.

Preliminary heating experiments were carried out at 600°C for twenty-five days and at 1100°C for one day. Homogenization of composition in the lamellae texture could not be confirmed, but a decomposition product, hematite, was observed mainly along Fe-rich lamellae. This is consistent with the phase relations in grandite garnet at high temperature (Huckenholz *et al.*, 1974; Shoji, 1975).

X-ray diffraction study

All diffraction spots observed in precession photographs are split (Fig. 3). The main spots and the associated ones could be assigned to those diffracted from Fe-rich and Fe-poor lamellae, respectively, comparing the intensities of diffraction spots and the volumes of the lamellae. The shaded areas for $\mu = 25^\circ$ (Buerger, 1964) on the precession photograph are doubled, which indicates that two superposed reciprocal lattice planes are not in the same orientation. Since one reciprocal net with stronger spots was positioned parallel to the film plane, the other with weaker spots was consequently asymmetrical with respect to the center of the film. Taking misorientation into account, very slight difference in d -spacing can be seen from the relative positions of the two lattices (Fig. 3). The difference in unit cell parameter is estimated to be about 0.02 Å, assuming provisionally that the lattices are cubic. This corresponds to a compositional difference of about 10 mol%, consistent with that measured by EPMA.

Discussion

Nature of the lamellar texture

All results by SEM, EPMA, AEM and X-ray diffraction indicated consistently that the lamellar texture consists of alternative lamellae with small differences in chemical composition and cell parameter.

As seen in Figure 1a, the lamellae are often wavy or sigmoid (wl) and sometimes straight (fl). Some parts of the wavy lamellae are almost normal to the compositional zoning, and others are almost parallel to them. Still others are oblique or intermediate between these two orientations. Since the normal lamellae and the oblique lamellae are continuously distributed in orientation, these lamellae are considered to be formed by a single mechanism. The occurrence of some straight lamellae which are parallel to compositional zones seems to suggest that they are oscillatory zoning formed by layer growth. These straight lamellae (fl) are, however, regarded as an extreme type of wavy lamelle because some wavy lamellae gradually change their orientations and become nearly parallel to compositional zoning (Fig. 1b, shown as an arrow) and are continuous into straight lamellae. On the other hand, some of the wavy lamellae are almost normal to compositional zones, which seems to suggest that the lamellae were produced by cellular growth (*e.g.*, Tiller, 1966). The mechanism of cellular growth can, however, not explain the orientation of the oblique and parallel lamellae. Similarly, the mechanism of layer growth cannot explain the orientation of the oblique and normal lamellae. Another mechanism should, therefore, be considered to explain the whole lamellar texture.

The observation that some lamellae extend through several compositional zones may be an indication that the lamellae were formed not during but after the crystal growth. It might, therefore, be inferred that the lamellar texture are due to subsolidus decomposition, although grandite garnet has been stated to form complete solid solution (Winchell and Winchell, 1964; Deer *et al.*, 1965; Novak and Gibbs, 1971) and thermodynamic calculation by Ganguly (1976) gave negative free energy of mixing between andradite and grossular. A tweed-like texture was recently reported as a texture suggesting a subsolidus decomposition in some grandite garnets (Hirai and Nakazawa, 1982). The tweed-like texture is composed of lamellae and matrix with different compositions and cell parameters. The shape of the

present lamellar texture is different from that of the tweed-like texture, but the compositional difference and the width of lamellae are of the same order. The lamellar texture observed in the present iridescent garnet can thus be interpreted as another type of exsolution texture, of a somewhat unique configuration (see Fig. 1a). The possibility that the straight lamellae (fl) may only be oscillatory zoning cannot be entirely ruled out.

Including the present observation, the exsolution-like lamellae with compositional difference have rather commonly been found in grandite garnets from several localities (Hirai and Nakazawa, 1982; Wada *et al.*, 1978). A remaining problem in the final identification of grandite exsolution texture is that homogenization of composition by heating has not yet been achieved. Another is that an appropriate explanation of the small compositional difference in lamellae pairs has not been made. It is, however, certain that these textures have a compositional difference and that they were formed after crystal growth.

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