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LETTERS

Pristine surface growth features on 100 Ma garnet phenocrysts: Interference imaging results

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ABSTRACT

Microstructures of crystal surfaces can reveal growth mechanisms. However, original surfaces of phenocrysts are rare because they are commonly covered with glass or rock matrix. In an unusual occurrence, microscopic growth features are clearly visible on original surfaces of 100 Ma melanite garnet phenocrysts of the Crowsnest Formation, Alberta, Canada. Such details as the steps of growth pyramids are clearly resolved at the 100 nm scale (vertically) in reflected- light Nomarski differential interference contrast (NDIC). However, because of the small step size (<0.5 micrometers), these features are not resolved in transverse section using a transmitted-light polarizing microscope. In transmitted-light microscopy, growth features of a thin-section appear indistinguishable from oscillatory zoning. These results suggest that, in general, oscillatory zoning may be surface growth forms seen in transverse section. Growth of the Crowsnest phenocrysts was both lateral and intermittent along the crystal face rather than continuously outward from the face, an observation of considerable significance to the theoretical modeling of natural crystal growth. This empirical study should alert other workers to the possibility of studying crystal growth using natural magmatic crystals. In particular, this unique Crowsnest material would repay study by diverse techniques.

INTRODUCTION

The samples in this work were collected some years ago by the author, in connection with a study of the Crowsnest Volcanics, Alberta, Canada, that are found about 50 miles north of Montana (Pearce 1967, 1970). The samples come from an outcrop on Alberta Highway no. 3, just west of Coleman, Alberta, Canada (Latitude 49° 30', Longitude 114° 30') and, regrettably, highway "improvement" has removed the part of the outcrop containing the garnets.

Andradite garnet, $Ca_3Fe_2(SiO_4)_3$ is cubic, yellow (becoming black with increasing Ti substitution, (variety melanite), and quite hard (7 on the Mohs scale). It has no cleavage and favors dodecahedral {110} and trapezohedral {211} crystal forms. The garnets in this study [see Pearce (1967) and Dingwell and Bearly (1985)] are rhombohedral dodecahedrons up to 5 mm across usually with 12 faces of the same size, although twinning is not uncommon, (Fig. 1). The crystal faces have an astonishing optical quality considering their great age (Lower/Upper Cretaceous boundary, 100 Ma) and geological complexity (burial metamorphism followed by uplift, erosion, and weathering). The samples come from a trachytic tuff bed in the lower part of the Formation. Sanidine, garnet, cognate rock fragments, and aegirine-augite crystals in a green matrix (probably devitrified glass) make up the important tuff bed. Considering the mineral composition and the explosive character of the volcanics (Pearce 1970, 1993), the liquid from which the garnet crystals grew was trachytic and contained volatiles (probably H_2O and CO_2) at a temperature of up to 1000 °C, but the pressure could not be well constrained. The liquid likely had a mantle-derived component, as implied by the garnet's low initial Sr-isotope ratio of 0.7038 (Pearce 1993), thus the pressure during the earliest growth of the garnet may have been high (approaching that of the mantle). The chemical composition of the magma was similar to Crowsnest trachytes (SiO_2 = 54.8, TiO_2 = 0.35, Al_2O_3 = 20.2, Fe_2O_3 = 3.00, FeO = 1.21, MnO = 0.165, MgO = 0.22, CaO = 3.58, Na_2O = 3.44, K_2O = 8.50, SrO = 0.10, BaO = 0.08, all in wt%).

It appears that the significant hardness and chemically inert character of the garnet allowed it to be released from its enclosing tuffaceous matrix during weathering with little damage to the original surface. As a result, the present surfaces of many samples contain textbook-like examples of growth forms, some predicted from theory (Burton and Cabrera 1949, Frank 1949, Burton et al. 1951) and known in the field of synthetic crystal growth (e.g., Verma 1953; Tiller 1991b), as well as from the interferograms of Tolansky (1960, 1968).

METHODS

Nomarski Differential Interference Contrast (NIDC) is an interference technique and, as such, it is not diffraction-limited (i.e., wavelength) except in plan view. An NDIC "photograph" may have a resolution of 0.5 micrometers in plan (x-y) view (as in a normal photomicrograph). However, in the z-direction (height), the resolution is much higher, being only limited by the method of resolving the phase difference of the interference fringes. In the present case, in which we are recording an image of topography, we may have a photograph with micrometer resolution horizontally, which has nanometer resolution vertically. Thus, if we consider a step pyramid, we can measure it's height in micrometers to ± about 10% using the micrometer scale of the research microscope. We can also

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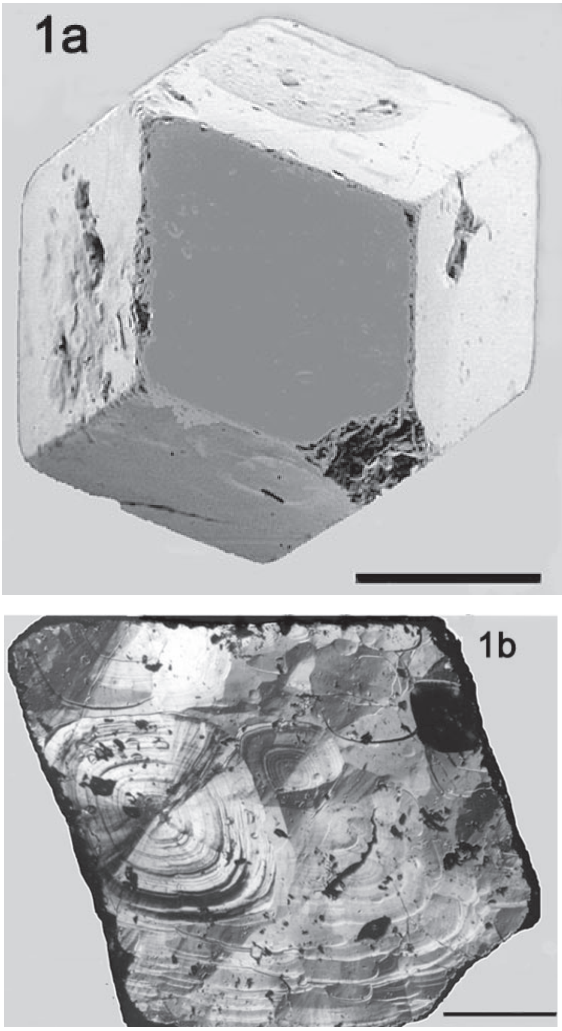

FIGURE 1. (a) SEM image of a 4 mm diameter Crowsnest garnet phenocryst. The crystal is euhedral and shows only {110} dodecahedral forms. Note the absence of detail in the SEM image owing to the extremely low relief of the growth features. Sample (G-20) is gold-plated. The scale bar is 1 mm. **(b)** Reflected light NDIC image of the central face of the garnet in Figure 1a (G-20, prior to gold-plating). The Nomarski technique exaggerates the apparent vertical relief by about 10 times. The surface is actually quite flat as it appears in Figure 1a. Two features are prominent in this view: stepped pyramids that, on close inspection, are invariably spiral in nature, and terraces with plate-like tops (not obviously spiral) and with steeper sides that are usually decorated with conical groups of striations (see Fig. 4). Scale bar is 500 m.

count the number of steps providing they are wider then the diffraction-limited resolution of the microscope (nominally 1 micrometer). For example, a pyramid 20 micrometers high, with 100 steps, has steps that average 200 ± 20 nm in height. This step size is smaller than optical microscopists normally can see and, in fact, the steps cannot be seen in transmitted light.

In order to make observations of the growth forms in reflected light it was sufficient to clean the specimen (ultrasonic cleaning bath with alcohol) then select those samples with the reflective optical quality surfaces (as many as half the crystals). As garnet is cubic, opposing rhombohedral faces are subparallel (not quite parallel, owing to vicinal planes). Thus the crystals naturally sit on a microscope slide with the upper face “parallel” to the microscope stage; further mounting is not necessary. Reflected and transmitted light observations were made using a Nikon Optiphot–Pol with NDIC reflected light attachment, the same objectives being used for transmitted and reflected light. Photographs were made using a Nikon HFX-II and 35 mm Kodak T400CN black and white film.

OBSERVATIONS AND INTERPRETATIONS

A typical screw dislocation growth form (Figs. 1b, 2a, 2b, 3a, 3b) exhibits a stepped pyramidal shape with the steps 0.2 micrometers high (>150 unit cells) and 4 micrometers wide (>3000 unit cells). Typical dimensions of a pyramid are a height of several to 50 micrometers and a width at the base of several

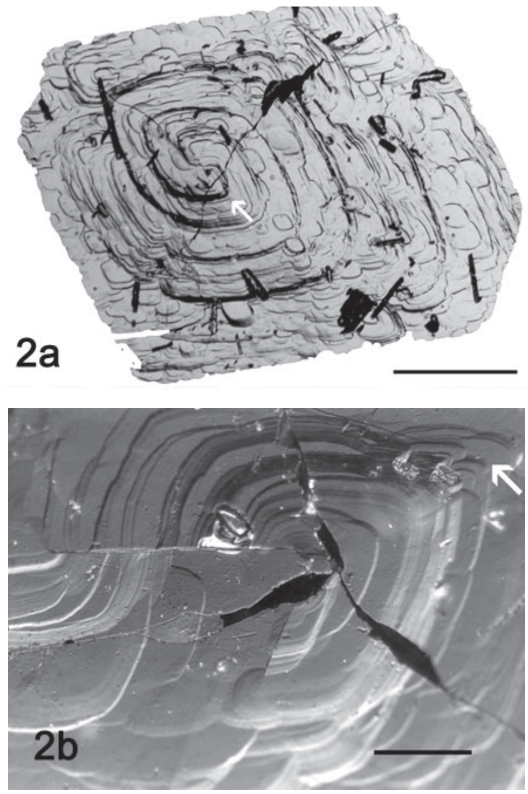

FIGURE 2. (a) Reflected light NDIC image of a {110} face of garnet G-11; overall width, 2200 m with a relief of 10 m. The “Greek key” appearance is due to the intergrowth of two spiral structures of the same sign of rotation. Note the absence of a prominent furrow in contrast of Figure 3a. The spiral surface is apparently “overgrown” with flat-topped, rounded, rhombohedral plates; close inspection shows that the plates are an integral part of the spiral edifice and may be due to local spiral centers. Scale bar is 500 m. **(b)** NDIC image of the top of the pyramid in Figure 2a (G-11). The pyramid terminates in two straight lines, which are parallel to the {110} forms of the rhombohedral face. Note the apparently different handedness of the steps about the horizontal straight line (showing screw symmetry as in Fig. 2a). These euhedral forms are very common on the larger pyramids. Scale bar is 50 m.

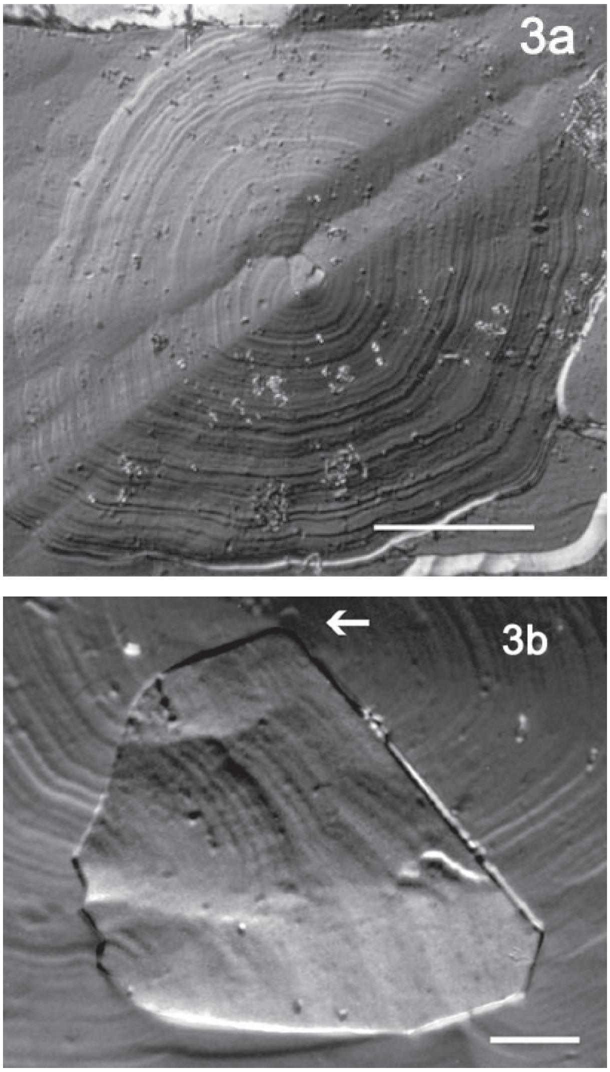

FIGURE 3. (a) Reflected light NDIC image of a 200 mm diameter complex, finely stepped spiral pyramid in sample G-6. The prominent furrow is thought to be the locus of intersection of two growth fronts. The centers of curvature (and growth) of the system (apparently 12 m apart) are sufficiently far apart for the left and right growth fronts to meet at an angle, which is replicated by successive growth steps to the top of the pyramid. The sinuous form of the furrow is thought to be due to irregular and unequal growth rates of the "left" and "right" crystallization fronts giving a zigzag interleaving along the base of the furrow. The smaller furrows, which end abruptly on the surface, are thought to be due to "captured" or "dominated" screw-related growth centers. The overlapping (interleaving) of the steps from each domain is similar to interlaced spirals depicted by Verma (1953) on a surface of SiC. Scale bar is 50 m. **(b)** NDIC image showing detail at the top of a growth pyramid in sample G-12. The arrow points to a 1.7 mm circular feature, which is the geometric center of the spiral, i.e., the point of emergence of the major screw axis. The large partly faceted plate, which touches the edge of the circular feature, shows {110} edges, and possibly one {100} edge just above the scale bar. Where the steps impact the plate on the left, note the change in the direction of curvature of the steps (180 rotational symmetry), consistent with a screw axis. Scale bar is 10 m.

hundred micrometers to over one mm, some pyramids being visible to the naked eye. The overall shape of the pyramid is flat conical (with an apex angle of about 174) and asymmetrical if near an edge. One or more obvious valleys typically occur on the pyramid slope (Figs. 1b, 3a). These valleys are thought to indicate the locus of the point of intersection of two growth fronts, possibly of different handedness and/or thickness (Tiller 1991b). The layers due to the different growth fronts do not meet along a simple curve but rather interleave (Fig. 3a), implying a variable and alternating growth rate. Nominal slopes of 3 (range <1 to 5) are common. In plan view, the form is rounded rhombohedral shape, as in Figures 1b, 2a, 2b, and 3a. Locally, under high magnification, the steps are seen to have an irregular scalloped shape (0.5 to 1 micrometer across), with the scallops convex away from the dislocation. I interpret this last feature to be a result of self-organization of the crystallization front as it sweeps across the crystal face. Irregularities in the crystallization front may be self-perpetuating (at least locally) as they affect the precipitation of the overlying layer, in this case, serving as a pre-existing template. Where closely stacked vertically, the irregularities yield striations as in Figure 4. The striations appear to form cone shapes with the apex toward the summit of the pyramid. The repetition involved in this phenomenon recalls oscillatory zoning, which has been found to have a non-linear dynamical (or chaotic) character in natural crystals (Higman and Pearce 1993; Holten et al. 1997).

Do these growth features represent the usual growth of garnets in the Crowsnest magma or are they the result of unusual growth conditions prior to eruption of the magma during ascent or perhaps due to increased cooling rate in a sub-volcanic environment? While it is not possible to completely rule out the latter case, careful examination of thin sections (Fig. 5), shows remarkable triangular features, as in Figure 6. These tri-

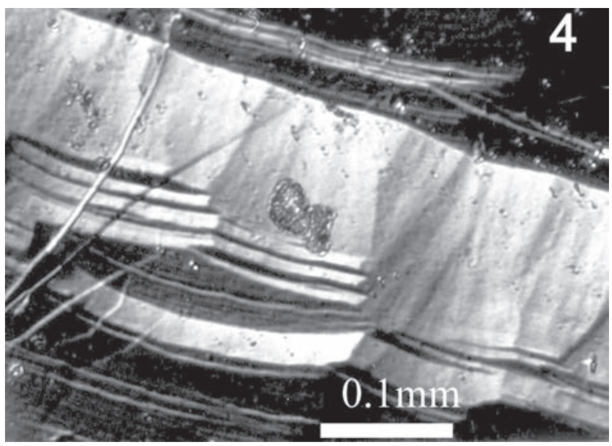

Figure. 4 This NDIC image shows the striations on the sides of plate-like growth forms of sample G-12 (Fig. 3b). The "steep" slopes are vicinal planes. These features are down slope from the screw center. Note the triangular (cone-shaped) appearance of aggregations of striations. The fact that they terminate upward in the structure suggests they are growth features using underlying layers as a template, thus becoming smaller as growth proceeds. The striations have the same magnitude of spacing (about 1 m) as undulations on the outermost (and lowermost) growth fronts seen under high magnification (600×).

angular microstructures are the correct shape and size to be consistent with sections cut through growth pyramids. Many of these triangles appear asymmetrical and flat-topped rather than strictly pyramidal. This shape could be due to the apparent dip problem owing to the arbitrary angle of the section but, in addition, not all the pyramids are symmetrical, particularly when they are near a [110] edge. These surface growth features appear to make up entire zones of the oscillatory-zoned crystals. The zones are about 5 times thicker than individual pyramids.

DISCUSSION

Given that screw layers are theoretically one unit thick, the large size of the growth steps (ca., 150 unit cells) seems unusual (but not unknown; see Verma 1953; Tiller 1991a). The presence of plates of rounded rhombohedral shape making up part of the growth pyramids seems notable; it is not clear if they are screw-related features. These plates appear closely intergrown with the spiral features but also resemble “pill-box” and “pancake” features depicted by Tiller (1991a) and Lasaga (1998), respectively.

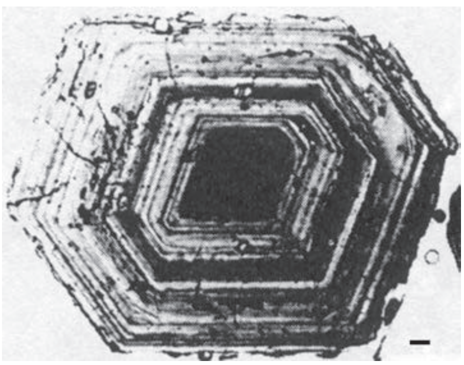

FIGURE 5. Transmitted, plane-light photomicrograph of a 30 m thick section of a garnet phenocryst. The overall composition is andradite (melanite). The oscillatory zoning is mainly due to variations in Fe and Ti. Growth features are not readily seen in transmitted light because of their low topological relief (1:100). Scale bar is 100 micrometers.

The ubiquitous presence of furrows on spiral growth features is unusual and may indicate that many spiral growth centers can grow simultaneously in the same growth mound without annihilation. A similar conclusion was reached by Bauser (in Tiller 1991a) for a feature in artificially grown material. Successive growth layers appear to use pre-existing layers as a template, stopping growth just short of the lower, earlier, edge thus yielding a sloping vicinal plane (as in the analogous “cones” in Fig. 4). The common occurrence of straight [110] edges (forming rhombohedral angles) at the tops of pyramids is problematic (Fig. 3b), as is the observed ability of a spiral growth front to form a straight edge between two {110} faces.

Observed in transmitted-light microscopy, the growth features are an indistinguishable part of the oscillatory zoning; in fact, they appear to make up the zones. The unit of growth in these crystals is the screw-related surface growth layer, which is about 150 unit cells thick. Major oscillations in chemistry of the layers occur after >1000 layers have been deposited as pyramidal and other shapes. In particular, the observation, in thin section, that oscillatory zones are about five times thicker than nearby identifiable pyramids suggests that there are at least two processes at work during growth: one is surface and screw-dislocation related at a scale of ~150 unit cells; and the other is a dynamic process within the liquid producing longer range chemical oscillations in a slower time-frame.

Comparison of these garnet results with those of volcanic plagioclase from some Caribbean islands (Pearce and Kolisnik 1990; Pearce 1994) suggests that, in general, oscillatory zoning may be surface growth forms seen in transverse section. If so, growth of phenocrysts must be lateral and intermittent along the crystal face rather than continuously outward from the face, an observation of considerable significance to the theoretical modeling of natural crystal growth (Anderson 1983).

The observations in this work point out the possibility of studying crystal growth surfaces using natural magmatic crystals. Some of these observations would seem to require further work and, as the Crowsnest material appears unique, the writer is willing to share this material with other researchers, especially those with access to novel types of microscopy.

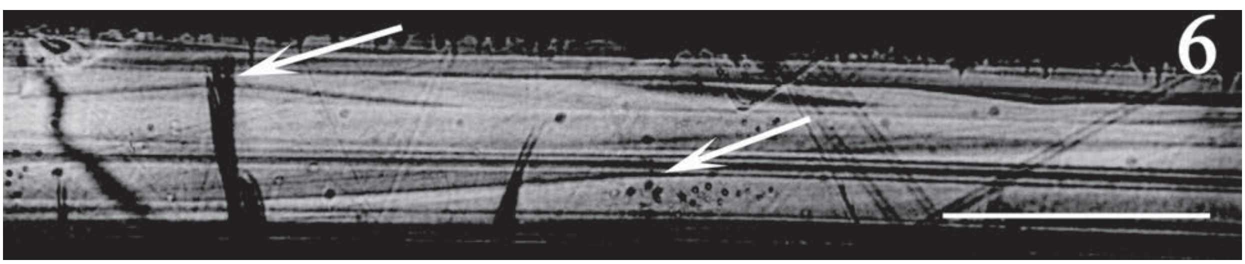

FIGURE 6. Digitally enhanced photomicrograph of a thin section of oscillatory zoned Crowsnest melanite garnet. The horizontal light-colored zone shows structures in cross section compatible with the shape of sections cut through growth pyramids of the same dimensions as those of the surface of the garnets. The arrows point to the tops of two pyramids. The shallow angles of the slopes are similar to those of the growth pyramids. If these features are sections of growth pyramids dating from earlier surfaces, then many of the earlier forms were asymmetrical with only one side and flat topped. These growth forms are difficult to recognize in thin section because of their low aspect ratio (1:100) and resulting shallow angles. This image has been digitally enhanced (contrast, brightness, and sharpness) but the overall geometrical shape is preserved. Scale bar is 100 micrometers.

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