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ELECTRON MICROSCOPY OF YTTRIUM ALUMINIUM GARNET (YAG)

K. H. G. Ashbee* and G. Thomas

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January 1968

ABSTRACT

A transmission electron microscopy study has been made of defects in yttrium aluminium garnet (YAG). Three kinds of planar defect were observed: antiphase boundaries generated by slip on {110} planes, antiphase domain boundaries separating regions of crystal which appear to have slightly different lattice parameter, and orientation twins.

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I. INTRODUCTION

Although the optical properties of yttrium aluminium garnet (YAG) have been extensively investigated, very little is known about its microstructure on an electron microscope scale. For example, it is not known whether dislocations in YAG are hollow, extended, or unextended. The purpose of the present work is to investigate this and other less obvious questions relating to the crystal perfection of YAG at high resolution.

The general chemical formula for garnet is $R'_3R''_2R'''_3O_{12}$. According to Menzer,[1] the crystal structure is cubic with space group Ia3d and the unit cell contains 8 molecular formula units, i.e. 160 ions. The ionic positions are:

R'	in 16(a)	0	0	0
R''	in 24(c)	1/4	1/8	0
R'''	in 24(d)	1/4	3/8	0
O^{2-}	in 96(h)	x	y	z

Each of the three R ion positions is surrounded by a different coordination polyhedron of oxygen ions. The polyhedron is an octahedron for R', a distorted cube for R'', and a tetrahedron for R'''. Although the oxygen ions in each case are equidistant or nearly equidistant from the central R ion, the edge lengths of any polyhedron are not equal.

In yttrium aluminium garnet (YAG), R' and R''' are both Al^{3+}, and R'' is Y^{3+}. Yoder and Keith[2] report that the lattice parameter of YAG is 12.01 ± 0.02Å, and that the parameters x, y, z are 0.04, 0.055 and 0.64 respectively.

II. EXPERIMENTAL

A transparent single crystal of yttrium aluminium garnet (YAG) was grown from 1/4" diameter sintered rod at the Materials Research Corporation, Orangeburg, New York, using the plasma float-zone technique developed by Class, Nesor and Murray.[3] This crystal was found to cleave fairly easily to give foils sufficiently thin for transmission electron microscopy.

The foils were examined in a Siemens electron microscope using projector pole piece two and an accelerating voltage of 100 kV. Occasionally, a foil would spontaneously recrystallize during irradiation by the electron beam. This produced large, thin areas containing several types of defects, not normally observed in as-received crystals.

III. ELECTRON DIFFRACTION

With the incident beam directed along a symmetry axis, a regular cross-grid of diffraction spots is obtained (Fig. 1). More generally, however, the electron diffraction pattern consists of several curved zones of spots (Fig. 2a, b). Such a pattern is a consequence of the large lattice parameter of YAG (and is formed as shown in Figure 3). The plane of Figure 3 contains the incident electron beam, the normal to the reciprocal lattice layers whose intersections with the Ewald sphere give rise to the zones of spots, and the principal axis common to all the zones of spots. The reciprocal lattice layers may be indexed by analyzing the spot pattern in the zero order zone and by making a scale drawing of Figure 3, the foil orientation can be determined very quickly. The non-equal spacings of the zones of spots are measured along the common axis (see Fig. 2a), transferred to

the scale drawing and projected back onto the Ewald sphere to give the points a, b, c, etc., in Figure 3. The set of equally spaced, parallel lines which pass through these points represent the reciprocal lattice planes producing the zones of spots. Their spacing may be used to check the indices assigned using the zero order spot pattern. The angle between the incident beam and the normal to the indexed reciprocal lattice planes is $\alpha = \tan^{-1} \frac{r}{R}$, where r is the radius of curvature (measured along the common axis) of the zero order zone of spots, and R is the radius of the Ewald sphere.

It is evident from Figure 3 that the curvature of the zones defines the sense of tilt, i.e. once the zero order spot pattern has been indexed, there is no ambiguity in deciding the sense of tilt.

IV. ELECTRON MICROSCOPY

Due to the very small grain size and the large number of reflections that are always excited (Figs. 1, 2), it has not yet been possible to study the observed defects in detail utilizing diffraction contrast. However, it is possible to draw some tentative conclusions as illustrated in the following.

The dominant defects observed in the recrystallized grains are planar, as evidenced by the fringe patterns. Many of these defects are undoubtedly translation twins, i.e. surface defects terminated by partial dislocations. Several translation twins can be seen in Figure 4. Three different fault planes are present in some grains, in which case their orientations can be deduced from their projected widths and the angles between their intersections with the foil surface. Thus, in Figure 5, the translation twins

labelled A, B, and C are found to lie on {110} planes. No orientations other than {110} could be unequivocably assigned to any of the present surface defects.

To see if there are any obvious displacement vectors for translation twins on {110} planes, consider the garnet crystal structure. Since YAG is bcc, only one quarter of the unit cell needs to be considered and, since the oxygen ions are the largest (the ionic radius of O^{2-} is 1.40Å; cf. Y^{3+}, 0.93Å and Al^{3+}, 0.50Å) and the most numerous, it is instructive to focus attention on these. The oxygen ions are conveniently visualized in groups of six arranged as octahedra centered on the aluminium ions in 16(a); see Figure 6. Although the total Burgers vector is probably 1/2<111>, it is evident from Figure 6 that a very small rotation of an oxygen octahedron would allow it to settle at 1/4<111>. If the atomic displacements which constitute this small rotation can occur, a surface fault with low specific energy might be envisaged, since only yttrium and some aluminium (in 16(a)) ions would be misplaced. Such a defect might properly be called an antiphase boundary.

Electron diffraction suggests that some of the "fringe" boundaries may separate regions which differ by more than a rigid body translation. Examination of Figure 2, for example, reveals that some Kikuchi lines are split into doublets or triplets. This splitting is so small that it is usually masked when the Kikuchi line coincides with a diffraction spot, cf. lines K in Figure 2a, b. The magnitude of the splitting represents a lattice spacing difference of ~1% and suggests the presence of domains which are not exactly cubic.[4] Such a domain structure could be envisaged as a consequence of ordering of oxygen ions in such a way that the structure

becomes very slightly tetragonal. One domain could then differ from the next by choosing a different cube axis for the tetragonal distortion.

In postulating the presence of a domain structure, it should be noted that, since none of the ions in YAG carries a magnetic moment, the domains cannot be attributed to ordering of magnetic spins. Any ordering must be purely crystallographic in nature.

In addition to antiphase boundaries generated by slip, and antiphase domain boundaries separating regions of differently ordered crystal, planar defects were observed which exhibit a reversal of contrast in dark field. These are no doubt orientation twins, and examples can be seen in the grain labelled A in Figure 7.

ACKNOWLEDGMENTS

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APPENDIX

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A 1% difference in lattice parameter between adjacent antiphase boundaries is indicated by the splitting of Kikuchi lines, and may be detectable by x-ray diffraction. Indeed, supplementary spots were observed on single crystal x-ray oscillation patterns but not on rotation, precession or Weissenburg patterns. Each supplementary spot on the oscillation patterns can, however, be interpreted as a short section of continuum that lies either between a yttrium absorption edge limiting the spot inwards and an oscillation range cut-off which limits the spot outwards, or between a bromine absorption edge which limits the spot outwards and an oscillation range cut-off which limits the spot inwards. Consequently, it is concluded that, if yttrium aluminium garnet does consist of domains which are not quite cubic, then the distortion from cubic symmetry is too small to be detected by x-ray diffraction. A similar result applies to ordering in the Ta-C system.[4]

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3. W. Class, H. R. Nesor and G. T. Murray, "Crystal Growth", Proceedings of International Conference, Boston, 1966 (Pergamon 1967), p. 75.

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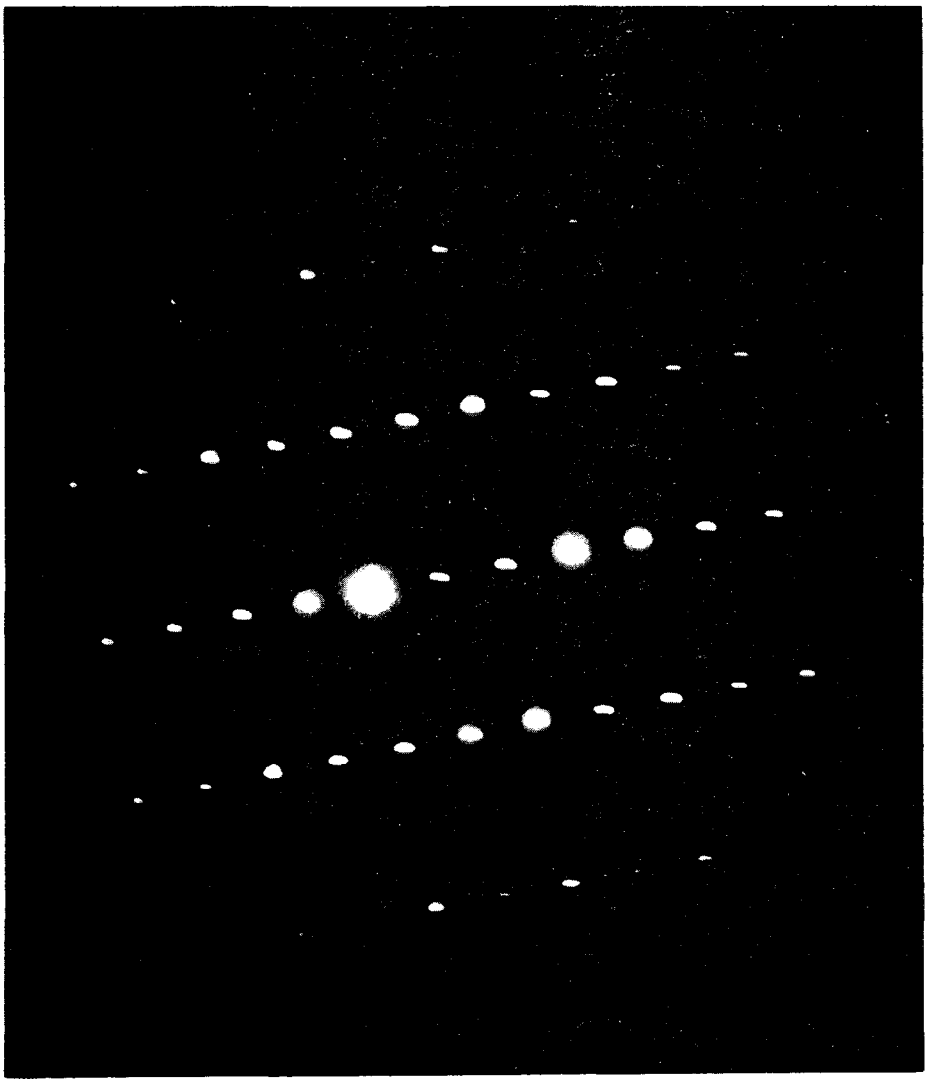

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Fig. 1 Symmetrical electron diffraction pattern.

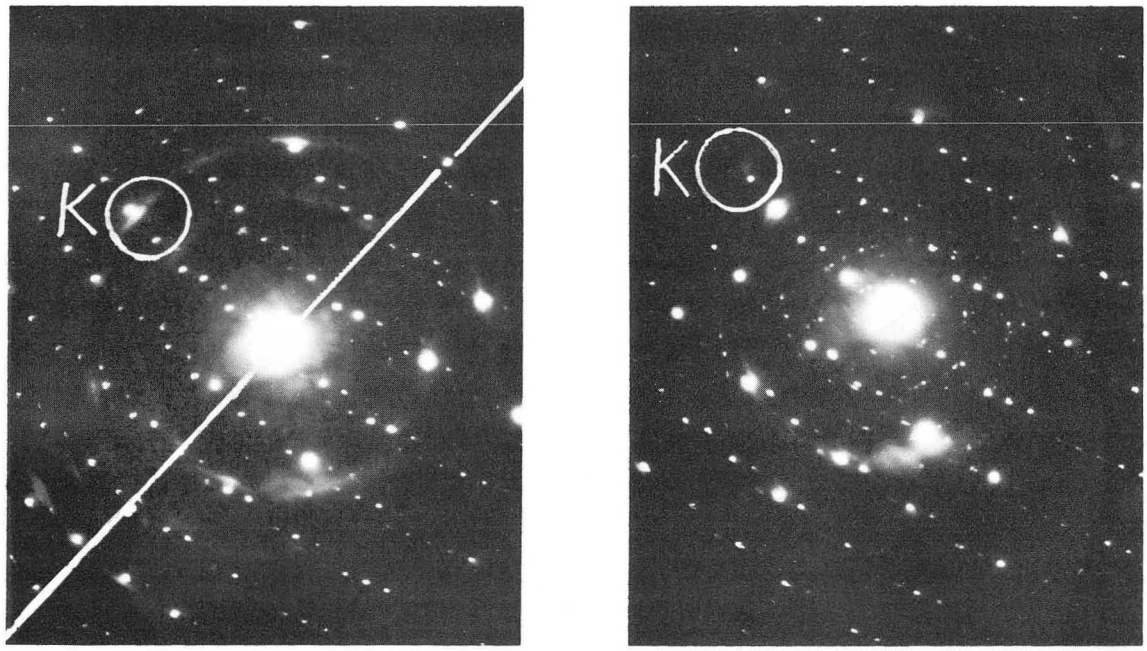

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Fig. 2 Unsymmetrical electron diffraction patterns.

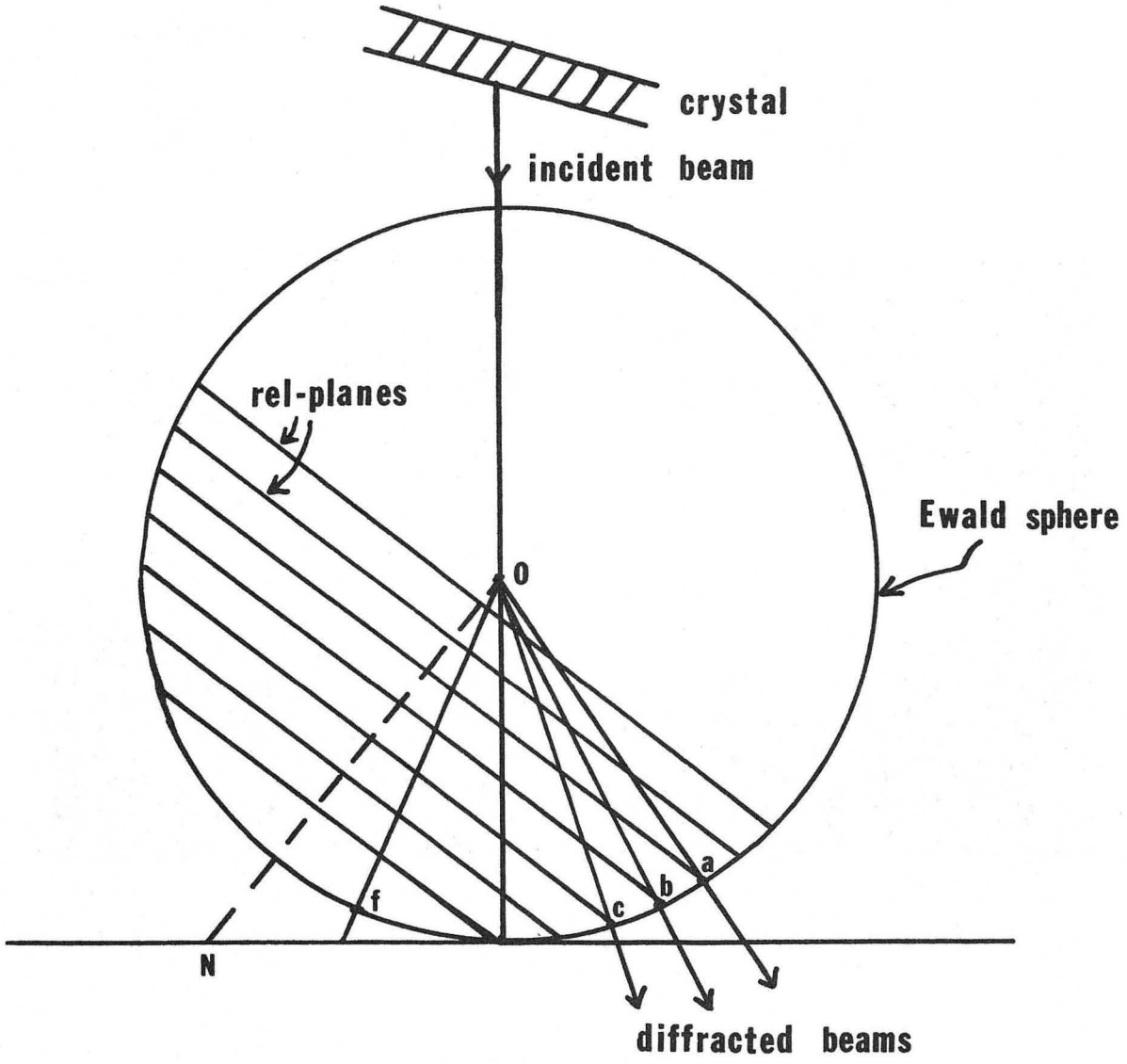

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Fig. 3 Reciprocal lattice construction used to index unsymmetrical electron diffraction patterns, using the Laue zone construction (exaggerated).

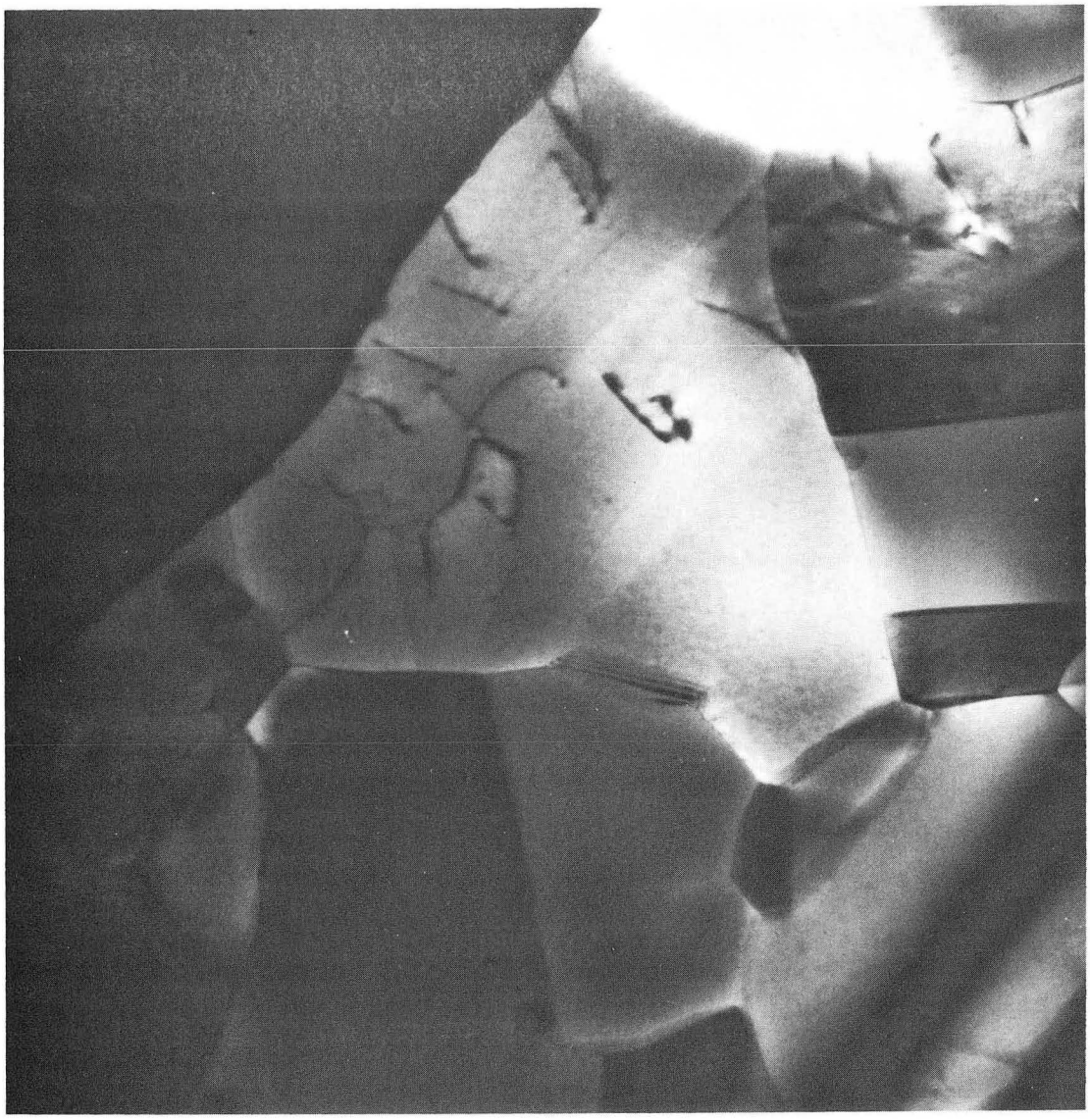

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Fig. 4 Fringe patterns due to antiphase boundaries created by gliding dislocations.

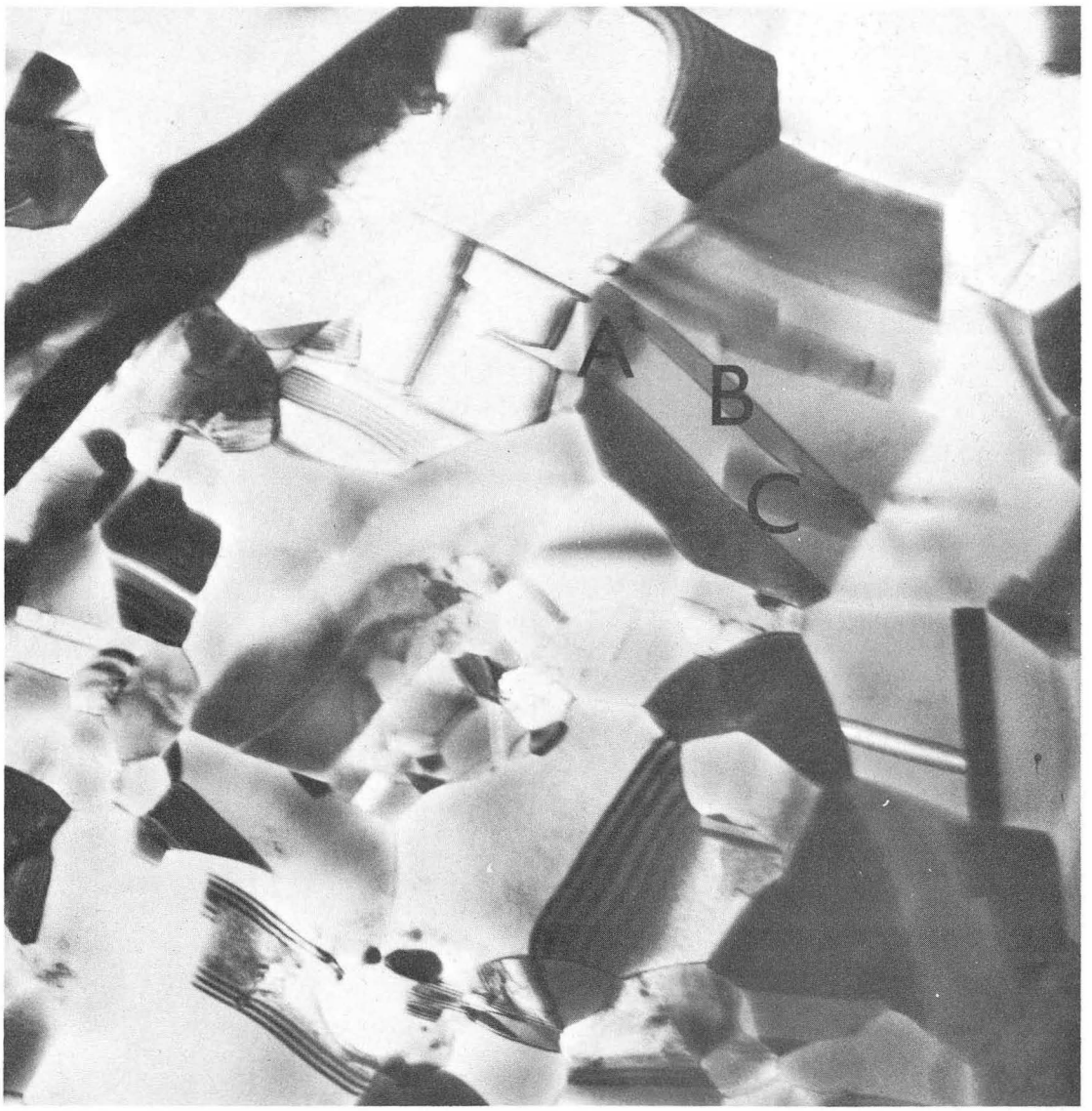

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Fig. 5 Antiphase boundaries on {110} planes.

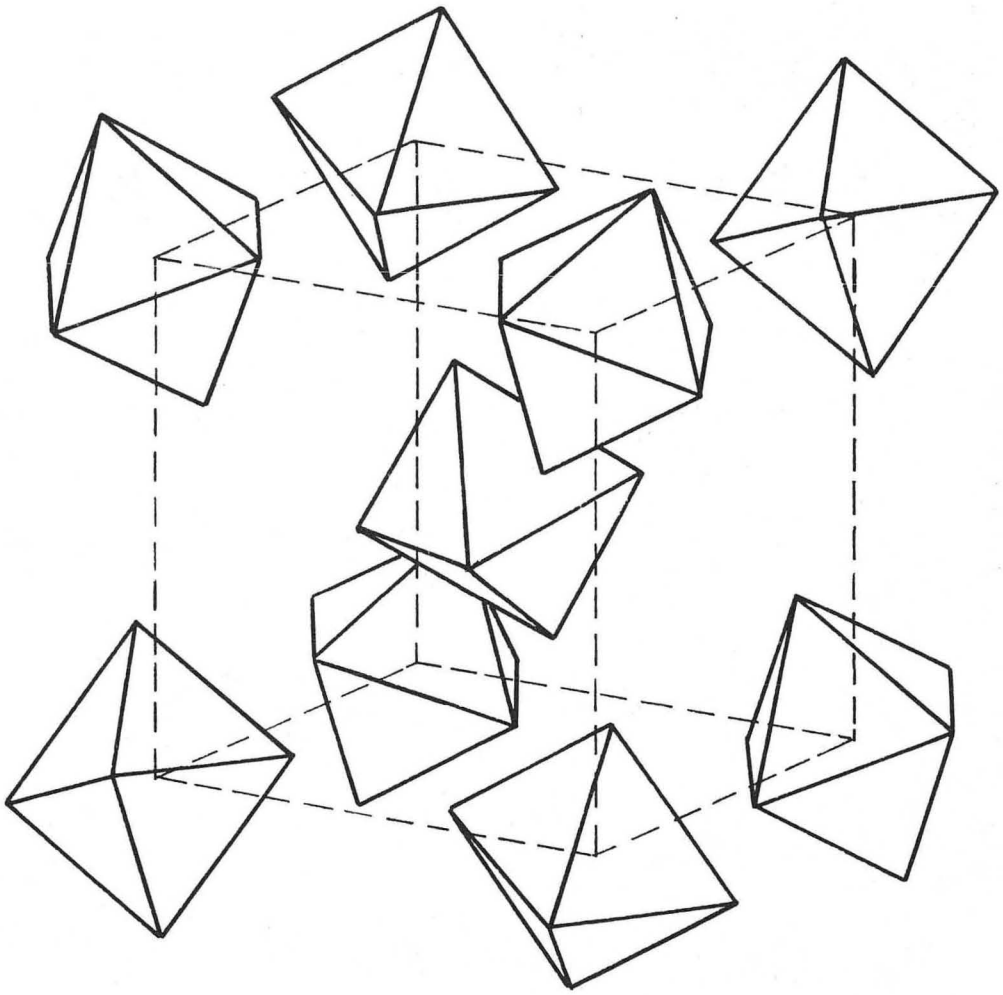

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Fig. 6 Oxygen octahedron model of the garnet structure (see text).

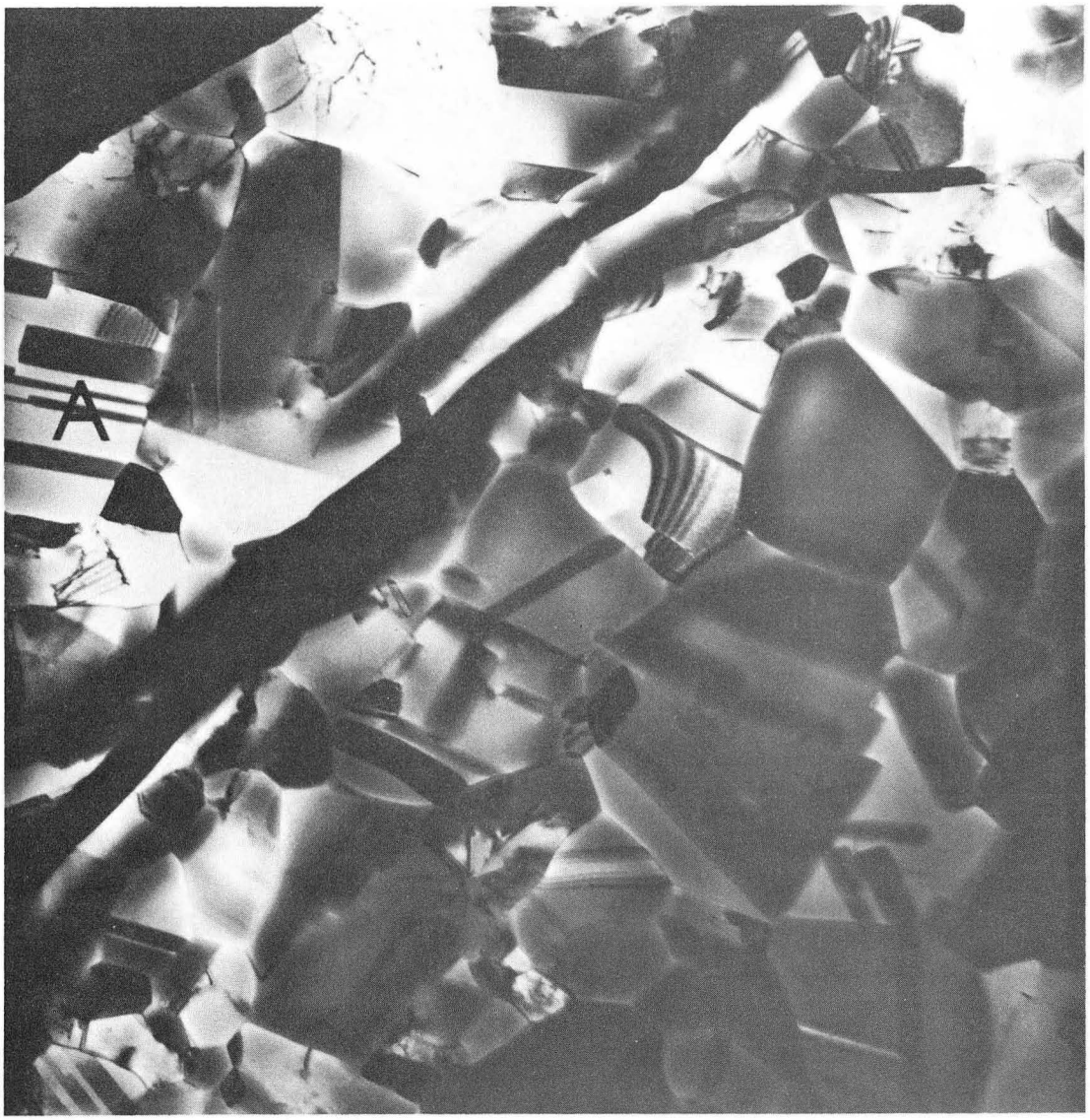

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Fig. 7 Orientation twins.

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