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Macroscopic strain in facetted regions of garnet crystals

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Morphological studies of Czochralski-grown gadolinium gallium garnet single crystals have revealed facetting characteristics identical to those observed in yttrium aluminium garnet. Accurate lattice parameter measurements made at various points within single crystals of these materials have been used to determine the strains associated with such facets. A possible explanation for the origin of the strain is proposed and the effect of facet imperfections upon the use of garnets in optical and magnetic thin film devices is discussed.

1. Introduction

Facets are an important defect in Czochralski-grown garnet single crystals intended for use as either substrates for magnetic garnet films (e.g. gadolinium gallium garnet – $Gd_3Ga_5O_{12}$) or as solid state lasers (e.g. yttrium aluminium garnet – $Y_3Al_5O_{12}$) since they correspond to a region of optical inhomogeneity [1-3]. For convenience, these materials are generally referred to as GGG and YAG respectively.

The purpose of the present paper is to report upon measurements of the strain induced by facet formation and upon similarities in facet morphology between GGG and YAG. The relevance of facet induced strain to the applications of garnet crystals is also discussed.

2. Experimental details

The results reported here were obtained using Czochralski-grown crystals of GGG and YAG prepared on a $\langle 111 \rangle$ axis from melts of stoichiometric composition [3, 4].

Macroscopic strain effects were observed through specimens of a standard 1 cm length, with ends polished flat and parallel, using both crossed polar and interferometric techniques. Accurate lattice spacings were measured on and off facetted regions by the Bond technique [5] using an (888) reflection and Cu radiation ($\lambda_{K\alpha_1} = 1.540562$Å). The advantage of this technique is that a relative accuracy of 1 part in 10^6 can be obtained between adjacent areas of crystal. The technique, as used in these experiments, yields values of the $d_{(111)}$ spacing in the crystal lattice which can be used to derive the unit cell parameter, a, the difference in unit cell dimension between facet and matrix, Δa, and e, the lattice strain defined as $\Delta a/a$.

3. Results

3.1. Facet morphology

Earlier work [3, 6] has shown that both {211} and {110} facets can form on the solid/liquid interface of Czochralski-grown YAG crystals. This work shows that the facet orientations in GGG crystals are identical. The facet configuration in $\langle 111 \rangle$ axis crystals is shown in Fig. 1. The three facets at A are of the type {211} and those

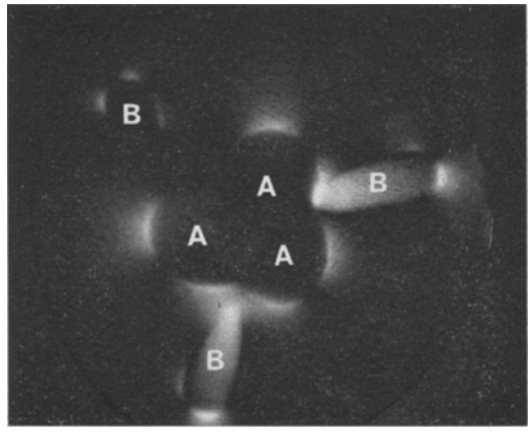

Figure 1 A transverse section of a $\langle 111 \rangle$ axis GGG crystal, viewed between crossed polaroids, showing {211} facets (A) and {110} facets (B). (Crystal diameter = 1.5 cm.)

at B are {110}. The facet orientation was determined by using growth striations to delinate the shape of the solid/liquid interface in (110) crystal sections and measuring the angle at which the facetted regions intersected the growth axis.

Other similarities with YAG are also apparent. For instance, the external morphology of ⟨111⟩ axis crystals is governed by the development of six {211} facets which form parallel to the growth axis and confer a hexagonal shape on the crystal. However, the shape is less recognizable in GGG crystals due to surface decompositional effects associated with the presence of a volatile component, Ga_2O_3. In common with YAG, interface facets can be removed by increasing the crystal rotation rate and thereby flattening the crystal interface so that no part is tangential to a possible facetting plane. A significant difference here is that under identical growth conditions, a planar interface is produced in GGG crystals at approximately half the crystal rotation rate required for YAG (80 rev/min for GGG: 150 rev/min for YAG). This is consistent with the relative interface shapes, YAG being the more convex with respect to the melt.

TABLE I (GGG)

	{110} facets	{211} facets	Matrix	Dislocated region
Lattice parameter (Å)	12.3838_8	12.3841_4	12.3824_0	12.3841_0
Δa	0.0014	0.0017	—	—

3.2. Facet strain

Lattice parameters have been measured on {211} and {110} facets and compared with unfacetted regions (matrix) in a number of crystals. The values for each facet type in a typical GGG crystal are given in Table I; these are mean values, with an error range of $\pm$ 0.000 09Å, derived from several measurements. The results indicate that the lattice parameter of both facet types exceeds that of the matrix but that a small difference exists between the two types of facet. Measurements on several crystals with matrix values within the range 12.3818 to 12.3826Å have shown that Δa always lies within the range 0.0011 to 0.0015Å; the corresponding lattice strain, e, is 0.000 09 to 0.000 12. The matrix values all lie within those reported for the homogeneity range of GGG [7].

An interesting result is obtained from the comparison of lattice parameter data for GGG with that of YAG (Table II). This shows that Δa values are approximately the same in both materials.

TABLE II (YAG)

	{211} facets	Matrix
Lattice parameter (Å)	12.0099_0	12.0085_0
Δa	0.0014	—

Facet-free crystals frequently contain regions of high dislocation density [4]. The macroscopic strain field associated with such a region is clearly defined in the optical micrograph of Fig. 2. Table I gives values of lattice parameter computed from measurements made within this region and on the unstrained matrix. These values indicate a positive lattice dilation of about 0.0015Å around the heavily dislocated region.

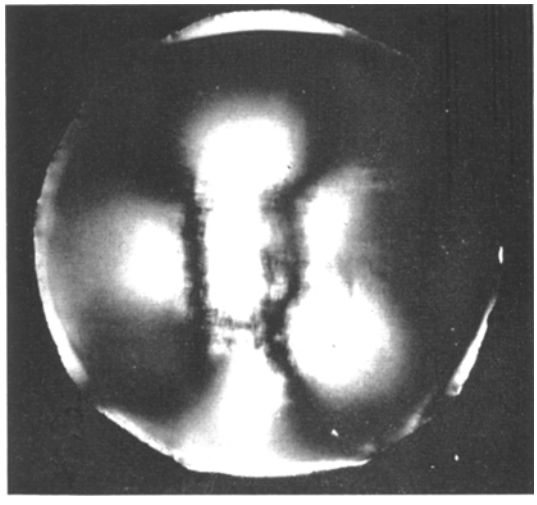

Figure 2 A transverse section of a facet-free ⟨111⟩ axis GGG crystal, viewed between crossed polaroids, showing the strain associated with a high dislocation density in the crystal core. (Crystal diameter = 1.5 cm.)

4. Discussion

The lattice parameter changes ($\sim$ 0.001Å) are significantly lower than the changes (0.005 to 0.012Å) deliberately induced between a GGG substrate and its magnetic layer in order to produce the anisotropy necessary for supporting bubble domains. Thus the anisotropy change due to facet strain will be small and difficult to distinguish from that due to other variables, e.g. layer thickness. In consequence facet strain is unlikely to significantly affect the character-

istics of magnetic bubble domains at low frequencies. At high frequencies, however, the operating tolerances are much reduced and anisotropy changes of this order may then prove significant.

The macroscopic strain associated with heavily dislocated regions is apparently also a small effect. However, such regions are composed of individual dislocations and arrays which have associated strain fields comparable in size to the bubble domains (5 μm diameter). Such regions are therefore far more likely than facets to impair the mobility of bubble domains.

The facets are less significant in substrates than in optical use where they produce changes in refractive index of between 1 part in 10^4 and 1 part in 10^5. In a YAG/Nd^{3+} laser rod (typically 5 cm long), changes of this magnitude produce optical path differences which are comparable to the operating wavelength (1.06 μm).

Several mechanisms can be postulated to explain facet strain. The principal mechanisms are (i) impurity segregation, (ii) changes in Ga/Gd or Y/Al ratio, (iii) oxygen segregation, and (iv) dislocations, and all are capable of producing lattice parameter changes of the observed magnitude. Impurities are an unlikely cause as facets have been observed in YAG crystals where no impurity element is present in concentrations > 1 ppm [8]. Dislocations must also be excluded as they do not form in facetted crystals [4]. Hence (ii) and (iii) are the most likely possibilities. The similarities in the strain level between facet and matrix in both YAG and GGG suggest that the same mechanism operates in both materials, in which case, the common element in the two materials is oxygen and mechanism (iii) is the most likely one. This would also explain the insensitivity of Δa to the bulk lattice spacing in GGG which is determined by differences in the ratio Ga_2O_3:Gd_2O_3 in the charge material. Changes in oxygen concentration within an oxide are almost impossible to detect directly. The present work provides indirect evidence that oxygen concentration differences can account for facet strains in garnet single crystals. Oxygen segregation would explain the presence of facet strain in very pure undoped crystals.

5. Conclusions

It has been shown that facet morphology in GGG and YAG is identical. This is consistent with the isostructural characteristics of the two compounds, both having cubic symmetry. The observation that the facet strain is the same in both materials is in keeping with the suggestion that such strain is caused by an oxygen segregation effect. The magnitude of the strain has less significance for applications of garnet crystals as substrates than as optical components.

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