

quenching in an inert atmosphere [13]. With poor thermal contact a high-conductivity substrate is wasted.

### Acknowledgements

The author is grateful to Dr J. A. Leake for useful discussions, to Professor R. W. K. Honeycombe for the provision of laboratory facilities and to the Science Research Council for financial support in the form a research studentship.

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Received 12 February  
and accepted 22 March 1974

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### LPE growth of garnets from the barium oxide-boric oxide flux

Thin films of magnetic garnet are important for both magneto-optic and magnetic "bubble" domain applications. The films are commonly grown from solution in a  $\text{PbO-B}_2\text{O}_3$  flux onto non-magnetic garnet substrates by the liquid phase epitaxy (LPE) dipping method, as reported by Levinstein *et al.* [1]. In order to provide the required properties, bubble domain garnet materials are necessarily multicomponent, e.g.  $\text{Eu}_{0.6}\text{Y}_{2.4}\text{Fe}_{3.9}\text{Ga}_{1.1}\text{O}_{12}$  [2].

However,  $\text{PbO-B}_2\text{O}_3$  fluxes have a number of undesirable features; they are toxic, corrosive and slightly volatile at the growth temperature. To obtain garnet as the primary phase, the  $(\text{Fe} + \text{Ga})$ :rare earth ratio must be greater than 12:1, which rapidly increases for subsequent growth runs from the same melt and also lowers the garnet solubility by the "common-ion effect". Lead is incorporated into the garnet lattice on the rare earth (dodecahedral) sites, particularly at lower growth temperatures. The incorporation of lead is detrimental to both the optical and magnetic properties of the garnet and because of its large ionic radius, affects the epilayer/substrate lattice mismatch. Furthermore the ratio of the Ga:Fe segregation coefficient

is approximately 2.0 and varies with both temperature [1] and growth rate [3].

Linares [4] has grown yttrium iron garnet (YIG) from the  $\text{BaO-B}_2\text{O}_3$  flux and has shown that it is congruently saturating (i.e. garnet can be crystallized when the Fe:Y ratio is the stoichiometric 5:3) when the molar ratios of  $\text{BaO:B}_2\text{O}_3$  are between 67:33 and 58:42. Recently Hiskes and Burmeister [5] have demonstrated the use of this flux for garnet LPE. The advantages of  $\text{BaO-B}_2\text{O}_3$  fluxes are that they are non-volatile, less corrosive and have both a higher garnet solubility and temperature coefficient of solubility. Divalent barium has a large ionic radius (1.35 Å) and is, therefore, unlikely to be accommodated in the garnet lattice.

According to the phase diagram of Levin and McMurdie [6], the melting points of  $\text{BaO-B}_2\text{O}_3$  fluxes vary almost linearly between 950°C for the 58:42 composition to 1100°C for the 67:33 composition. Compositions with lower  $\text{B}_2\text{O}_3$  contents have even higher melting points. Low growth temperatures are desirable since the epilayers produced have a higher growth-induced magnetic anisotropy, which is essential for the formation of stable bubble domains, especially as the trend towards smaller bubbles continues.

The melt composition used is given in Table I.

TABLE I

Constituent	Weight (g)
Eu <sub>2</sub> O <sub>3</sub>	8.90
Y <sub>2</sub> O <sub>3</sub>	26.86
Fe <sub>2</sub> O <sub>3</sub>	28.06
Ga <sub>2</sub> O <sub>3</sub>	10.45
BaO	149.50
B <sub>2</sub> O <sub>3</sub>	34.20

BaO:B<sub>2</sub>O<sub>3</sub> are in a molar ratio of 60:40. The garnet liquidus temperature is 1048°C and increases by 6.5°C for each gram of garnet added. Since BaO is extremely hygroscopic and cannot be easily dehydrated, Analar BaCO<sub>3</sub> was used as the starting material. B<sub>2</sub>O<sub>3</sub> was BDH optran grade and the other oxides were Johnson-Matthey 3N or better.

Using this melt, epitaxial layers were grown onto (111) oriented slices of gadolinium gallium garnet. The vertical dipping method was used with substrates rotating in a horizontal plane. For undercoolings of 20°C, highly reproducible growth was achieved. Growth rates were about 10% lower than those reported for the PbO—B<sub>2</sub>O<sub>3</sub> flux [7] for comparable undercoolings and substrate rotation rates. Magnetisation measurements showed the ratio of the Ga:Fe segregation coefficients to be close to unity and hardly affected by changing the growth rate, in accordance with the interpretation based on the Burton, Prim and Slichter model [8].

The barium content in the epilayer was below the level of detection of electron microprobe analysis (0.2% by weight), except at pitted regions in the surface (Fig. 1) where up to 1% was detected. This analysis was supplemented by X-ray fluorescence spectroscopy which indicated the average barium content through the whole of the epilayer was 0.02% by weight.

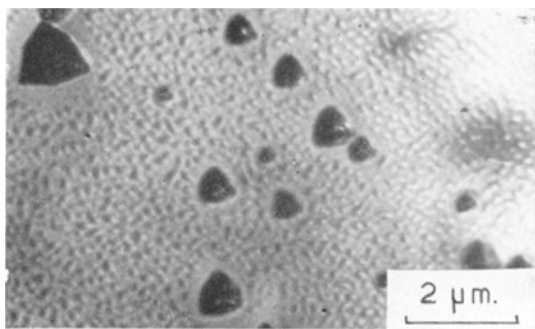


Figure 1 Surface defects caused by adhesion of melt droplets.

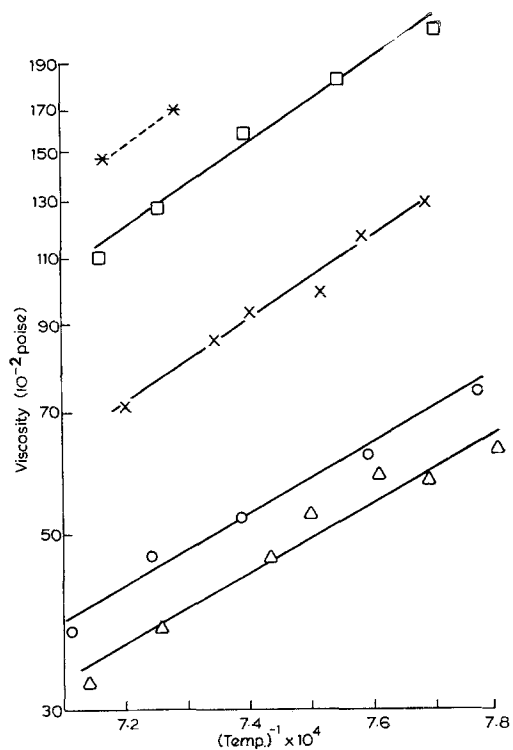


Figure 2 Viscosity of various composition versus the reciprocal of the absolute temperature.

- BaO (60), B<sub>2</sub>O<sub>3</sub> (40);
- BaO (60), B<sub>2</sub>O<sub>3</sub> (40) + 15 wt % YIG;
- \* BaO (60), B<sub>2</sub>O<sub>3</sub> (40) + 25 wt % YIG;
- × BaO (51), B<sub>2</sub>O<sub>3</sub> (34), BaF<sub>2</sub> (15) + 15 wt % YIG;
- △ BaO (30), B<sub>2</sub>O<sub>3</sub> (20), BaF<sub>2</sub> (50) + 15 wt % YIG.

However, the barium-based melt was found to have one serious disadvantage. Because of its high viscosity, droplets adhered to the epilayer surface on withdrawal from the melt. Even substrate rotation rates of 2000 rpm immediately after dipping failed to completely remove these. On cooling, rapid growth continued under the residual melt producing surface defects (Fig. 1). These defects were detrimental to both bubble mobility and stability.

It has been reported by Lawrence *et al.* [9] that small additions of BaF<sub>2</sub> lower the viscosity of the BaO—B<sub>2</sub>O<sub>3</sub> flux. In order to determine the optimum addition of BaF<sub>2</sub>, a viscosity study was undertaken. Essentially the method used was that of Lillie [10] and Margules [11], in which a crucible containing the melt was rotated at a constant rate and the viscous torque imparted on a solid platinum cylinder suspended in the melt measured by an optical lever. A rotation rate of 20 rpm was used and the apparatus calibrated

with water/glycerol mixtures of known viscosity.

The results obtained are shown in Fig. 2, with viscosity plotted on a logarithmic scale against the reciprocal of absolute temperature. The absolute accuracy is  $\pm 10\%$ . The BaO:B<sub>2</sub>O<sub>3</sub> flux (60:40) has a viscosity of 0.6 P at 1050°C. The addition of 15% by weight of YIG dramatically increases this to 1.9 P. Although BaF<sub>2</sub> addition decreases the viscosity, the effect is not substantial. The results should be compared with those for a typical growth solution using the PbO—B<sub>2</sub>O<sub>3</sub> flux, which has a viscosity of  $7 \times 10^{-2}$  P at 1050°C [12]. Since the flux consisting of BaO:BaF<sub>2</sub>:B<sub>2</sub>O<sub>3</sub> in a molar ratio of 30:50:20 had the lowest viscosity, this composition was used for epitaxial growth but even so the adhesion of melt droplets to the epilayer surface was still apparent.

Although BaO—B<sub>2</sub>O<sub>3</sub> fluxes have a number of desirable features for the epitaxial growth of garnet, their high viscosity makes the removal of residual melt from the epilayer surface extremely difficult. Unless more effective ways of removing this excess melt can be devised, the use of the BaO—B<sub>2</sub>O<sub>3</sub> flux for garnet epitaxy is severely limited.

### Acknowledgements

The authors would like to thank Mr J. D. C. Wood for useful discussions, and the Science Research Council for financial support.

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Received 22 February  
and accepted 4 March 1974

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### Fibre diameter measurement by laser diffraction

One problem involved in studying the tensile properties of fine fibres is determining the effective cross-section. For smooth, parallel-sided fibres of circular section the problem reduces to one of diameter determination; the diameters involved for carbon fibres, for example, are of the order 7 to 10  $\mu\text{m}$ . Two methods have been employed to date: firstly, the use of a pre-calibrated image shearing eye-piece on an optical microscope for direct measurement [1, 2], and secondly, the interpretation of the diffraction pattern from an incident laser beam [3, 4] — this latter being illustrated in Fig. 1. Both methods allow a rapid determination of the required diameter.

A comparison between diameters as deter-

mined both by laser diffraction and on an optical projection microscope showed [4] the existence of a difference of the order of  $\frac{1}{3} \mu\text{m}$  with the laser value being the smaller. Whilst the difference was not significant in that work [4], this article reports a preliminary study to understand the origin of the difference.

The simplest interpretation of the laser diffraction pattern of a fibre can be understood as follows. The pattern created by an infinitely thin lath of width  $d_s$  is the same as that of a slit of the same width. It is a series of maxima and minima on either side of a central maximum as indicated in Fig. 1. The separation  $\delta_s$  between the first corresponding pair of minima is given in the Fraunhofer approximation by the relation

$$\delta_s = \frac{2\lambda_s}{d_s} \quad (1)$$

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