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tion was enhanced in that region by the temperature rise due to radiation absorption.

The intense light at the focal point in all of the photographs should not be thought of as occurring at the time the schlieren photograph was taken. This light lasts only a short time after the laser pulse, but since the Kerr-cell shutter was on the flash-lamp side of the test cell (Fig. 6) any light in the test area would be recorded on the time-exposed film regardless of when it happened.

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[5]These fluids have typical extinction coefficients of about 0.25 cm^{-1} at 1.06 μ and are therefore considered to be weakly absorbing.
[6]The laser beam direction is from left to right in all of the photographs.

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Growth of High-Quality Garnet Thin Films from Supercooled Melts

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A new dipping technique is described for producing uniform-thickness low-defect-density magnetic garnet films suitable for bubble-domain devices. The technique is based on the observation that melts having the appropriate flux and garnet composition can be supercooled over a large temperature range and will remain supercooled, permitting the growth at constant temperature of epitaxial magnetic garnet films.

Since the first publication of Bobeck *et al.*[1] on the use of low-moment rare-earth garnets for bubble-domain-device applications, a considerable effort has been directed toward the growth of epitaxial films of these materials. Liquid-phase epitaxial growth (LPE) of YIG had been reported previously by Linares.[2] Shick *et al.*[3] used an LPE technique to produce garnet films capable of in excess of 100-step operation as a shift register. In this letter we report on a new technique for liquid-phase epitaxial growth of garnet films. The technique is based on the observation that melts having the appropriate composition of flux and garnet can be supercooled over a large temperature range and will remain in the supercooled state permitting the growth at constant temperature of single-crystal epitaxial magnetic garnet films. The isothermal dipping process is simple and rapid, and has produced films having defect densities of less than 10 defect/cm^2 with a uniformity of thickness of $\pm 2.5\%$ over an area of 0.6 cm^2.

The apparatus employed for the growth of the epitaxial garnet films is shown in Fig. 1. It consists of a platinum-wound resistance furnace supported by a platform which can be raised or lowered. A platinum crucible containing a mixture of the appropriate oxides of the flux and garnet is positioned by a pedestal in the constant-temperature zone of the furnace. A Pt—Pt 10% Rh thermocouple is immersed in the bath to monitor the bath temperature. A control couple also Pt—Pt 10% Rh is positioned as shown in Fig. 1. The substrate, an approximately 20-mil-thick slab of the appropriate nonmagnetic garnet, is held so that it will enter the bath in the vertical position. The substrate is attached to a ceramic rod which is held in a fixed position above the furnace. With the substrate in place the furnace is raised to position the substrate above the melt. This position is maintained for approximately 5 min in order for the sample temperature to approximate the temperature of the bath. The substrate is then inserted into the bath by raising the furnace the required distance. The growth time required to obtain a film of the thickness desired for a particular bubble application depends upon the degree of supercooling employed, and the temperature at which the particular degree of supercooling has been achieved. Using the melt composition shown in Table I a 10-min growth time at 945 °C (~30° supercooling) produces a 4-μm-thick film. At the completion of the growth cycle the sample is slowly withdrawn from the bath. The flux solution does not wet the substrate during the withdrawal and no flux remains on the sample.

In order to establish the appropriate range of growth temperature for a new garnet composition the following procedure is employed. Approximately 9% by weight of the appropriate garnet oxides are dissolved in a flux having a 50/1 weight ratio of PbO to B_2O_3. The solution is heated to about 1100 °C and held for several hours in order to equilibrate the melt. The temperature of the melt is lowered to approximately

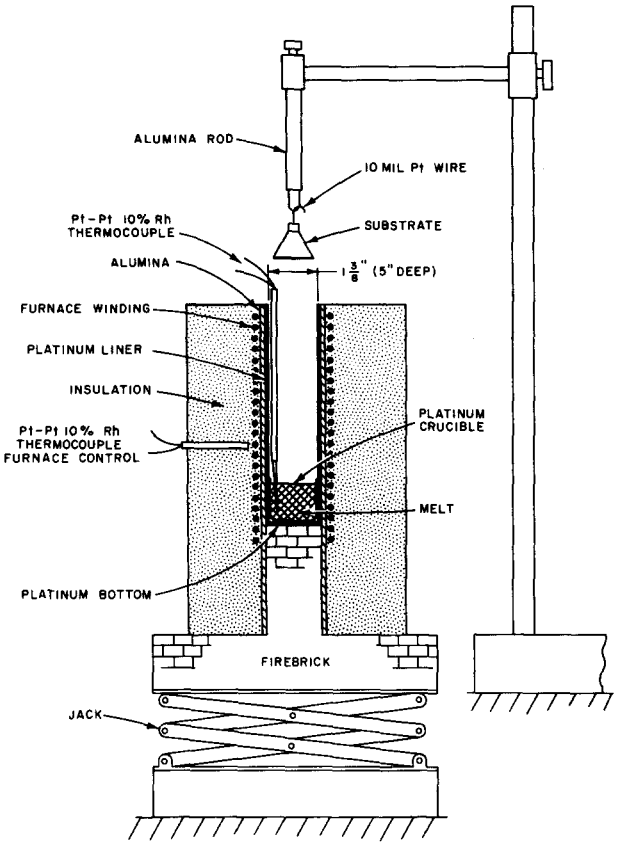

FIG. 1. Apparatus for epitaxial film growth.

970 °C. A substrate is then inserted and the bath temtemperature is lowered to 890 °C over a period of about 10 min. If growth occurs the sample is then examined by optical microscopy to determined if etching has occurred at the interface between the film and the substrate. If surface irregularities such as etch pits are present, the insertion temperature was above the saturation temperature of the magnetic garnet. On subsequent runs the insertion temperature is lowered while the withdrawal temperature of 890 °C is held constant. The maximum insertion temperature at which no apparent substrate attack occurs represents the upper temperature limit for growth for the particular bath composition employed. To further establish the upper temperature limit for growth, the furnace is equilibrated at a temperature 5° above the temperature at which no apparent attack occurred and a sample is inserted for 5 min at this temperature. If no growth occurs, the furnace temperature is lowered several degrees and the run is repeated with the furnace temperature held constant. This procedure is repeated until a temperature is obtained at which growth of a film occurs on the substrate. The highest temperature at which growth of a film is observed to occur is assumed to be the saturation temperature for the melt composition employed.

To establish the useful lower limit of growth temperature, the bath is equilibrated at successively lower temperatures and the surface of the melt is examined for particle nucleation. The lowest temperature which can be achieved before self-nucleation of the garnet becomes apparent defines the lower-growth-temperature limit. The presence of precipitates on the melt surface results in improper drainage of the flux from the sample on withdrawal from the melt. If flux remains on the sample after removal from the bath, irregular growth occurs during the cooling and a nonflat surface results.

We have found that at low concentrations of garnet dissolved in the flux (less than 9% by weight of the rare-earth oxides and iron oxide) the saturation temperature is below 970 °C. We have also found that when the saturation temperature is low and the ratio of rare-earth oxide to iron oxide is such that only garnet can form from solution, then homogeneous nucleation of the garnet phase is difficult and as a result the melts will supercool over an extended temperature range. In the absence of a disturbing influence such as the introduction of a substrate, supercooling of 120 °C has been achieved. For less extreme degrees of supercooling, 10 to 40 °C, the melts will remain supercooled for periods as long as 48 h. Substrates can be inserted into the melts in this supercooled condition without causing homogeneous nucleation in the melt. Garnet nucleates and grows on the substrate but nowhere else in the melt. This permits growth under isothermal conditions within the bath.

The film thickness as a function of time for different degrees of supercooling for $Eu_1Er_2Fe_{4.3}Ga_{0.7}O_{12}$ on $Gd_3Ga_5O_{12}$ substrates when the bath composition shown in Table I is employed is given in Table II. The saturation temperature was found to be 970 ±5 °C for this composition. The growth rate approximately doubles with each 10° increase in supercooling up to 30° of supercooling.

We also observed that the saturation magnitization of the grown films is a function of the temperature of growth. The lower the temperature of growth the higher the $4\pi M$ observed. The difference in $4\pi M$ between the film grown at 955 °C and the one grown at 945 °C was 40 G. This increase in $4\pi M$ with decreasing growth temperature is probably caused by a temperature dependence of the Ga distribution coefficient. The lower the growth temperature the less Ga is incorporated into the grown film. It follows

TABLE I. Melt composition employed for the growth of $Eu_1Er_2Fe_{4.3}Ga_{0.7}O_{12}$.

Melt composition	Growth
Eu_2O_3	0.63
Er_2O_3	1.35
Fe_2O_3	10
Ga_2O_3	0.7
PbO	140.0
B_2O_3	2.8

TABLE II. Film thickness vs time and degree of supercooling for films grown from a melt of the composition of Table I.

Degree of supercooling (°C)	Growth time (min)	Film thickness (μ)
10	5	0.6
10	10	1.3
20	5	1.0
20	10	1.95
30	5	2.15
30	10	4.00
40	5	2.55
40	10	4.85

that in order to grow films of uniform thickness the substrate and bath must be uniform in temperature, and in order to achieve a constant value of moment the growth temperature must be constant. The ideal manner to achieve these requirements is by growth at constant temperature in a supercooled melt. The degree of thickness and moment uniformity which can be achieved is a function of how close the system can be maintained in a truly isothermal condition.

Employing the technique of growth at constant temperature in supercooled melts, high-quality films having the composition $Eu_1Er_2Fe_{4.3}Ga_{0.7}O_{12}$ have been grown. Films have been grown having zero defects in a 1-cm^2 area. The majority of defects present in the films can be identified with observable substrate defects, such as inclusions, or scratches introduced by improper substrate preparation.

Films have been grown which have variations in thickness of $\pm 2.5\%$ or less over a 0.6-cm^2 area. A detailed evaluation of the thickness uniformity, and associated uniformity of properties of films grown by this technique, are described by Hagedorn *et al.*[5] Employing films grown in this manner, Bobeck[6] has successfully operated a 10 000-bit shift register using a film 4.7 μ thick which supports 6-μ-diam bubbles.

This technique is not restricted to the growth of the garnet composition described in this letter. Using the PbO-B_2O_3 flux, 15 garnet compositions have been successfully grown from supercooled melts at constant temperature.[7] Growth by supercooling is not restricted to the PbO-B_2O_3 system. Garnet films having the composition $Eu_1Er_2Ga_{0.7}Fe_{4.3}O_{12}$ have been grown from supercooled melts in which the flux employed is Bi_2O_3 with 10% by weight V_2O_5 added.

We have described a new technique for growing uniform-thickness low-defect magnetic garnet films which are suitable for bubble-domain devices. This technique is based on the fact that melts having the appropriate flux and garnet composition can be supercooled over a large temperature range and will remain supercooled permitting the growth of epitaxial magnetic films at constant temperature. The technique has the virtue of being simple to employ, and requires on the order of only 10 to 20 min to produce high-quality epitaxial films. It is likely that epitaxial films of material other than garnet can be grown by this technique.

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Supercurrent Tunneling Junctions with Tellurium Barriers*

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Uniformly distributed supercurrents have been observed in Josephson-like junctions using tellurium barriers with thicknesses up to 800 Å. Dependences of the maximum zero-voltage current on temperature and thicknesses are different from those of the normal-metal and oxide-barrier junctions. Typical junction resistances are comparable with those of oxide-barrier junctions.

Effects similar to those predicted by Josephson[1] have been observed in sandwich structures of greatly differing characteristics. Anderson and Rowell[2] first observed Josephson's predicted tunneling of electron pairs through an oxide barrier. Such barriers are typically 20 Å thick. In the opposite extreme,