

July 15, 1993

## Epitaxial yttrium-iron-garnet films grown by pulsed-laser deposition

P. C. Dorsey

S. E. Bushnell

R. G. Seed

C. Vittoria

*Northeastern University*

---

### Recommended Citation

Dorsey, P. C.; Bushnell, S. E.; Seed, R. G.; and Vittoria, C., "Epitaxial yttrium-iron-garnet films grown by pulsed-laser deposition" (1993). *Electrical and Computer Engineering Faculty Publications*. Paper 44. <http://hdl.handle.net/2047/d20002215>



## Epitaxial yttrium iron garnet films grown by pulsed laser deposition

P. C. Dorsey, S. E. Bushnell, R. G. Seed, and C. Vittoria

Citation: *J. Appl. Phys.* **74**, 1242 (1993); doi: 10.1063/1.354927

View online: <http://dx.doi.org/10.1063/1.354927>

View Table of Contents: <http://jap.aip.org/resource/1/JAPIAU/v74/i2>

Published by the [American Institute of Physics](#).

---

### Related Articles

Optically driven method for magnetically levitating diamagnetic material using photothermal effect  
*J. Appl. Phys.* **111**, 023909 (2012)

Simultaneous observation of magnetostatic backward volume waves and surface waves in single crystal barium ferrite platelets with in-plane easy axis  
*J. Appl. Phys.* **111**, 023901 (2012)

The effect of Fe<sup>2+</sup> ions on dielectric and magnetic properties of Yb<sub>3</sub>Fe<sub>5</sub>O<sub>12</sub> ceramics  
*J. Appl. Phys.* **111**, 014112 (2012)

Direct observation of magnetic phase coexistence and magnetization reversal in a Gd<sub>0.67</sub>Ca<sub>0.33</sub>MnO<sub>3</sub> thin film  
*Appl. Phys. Lett.* **100**, 022407 (2012)

Epitaxial thin films of p-type spinel ferrite grown by pulsed laser deposition  
*Appl. Phys. Lett.* **99**, 242504 (2011)

---

### Additional information on *J. Appl. Phys.*

Journal Homepage: <http://jap.aip.org/>

Journal Information: [http://jap.aip.org/about/about\\_the\\_journal](http://jap.aip.org/about/about_the_journal)

Top downloads: [http://jap.aip.org/features/most\\_downloaded](http://jap.aip.org/features/most_downloaded)

Information for Authors: <http://jap.aip.org/authors>

## ADVERTISEMENT

**LakeShore Model 8404** developed with  
**TOYO Corporation**  
**NEW AC/DC Hall Effect System** Measure mobilities down to 0.001 cm<sup>2</sup>/V s

# Epitaxial yttrium iron garnet films grown by pulsed laser deposition

P. C. Dorsey, S. E. Bushnell, R. G. Seed, and C. Vittoria

*Department of Electrical and Computer Engineering, Northeastern University, Boston, Massachusetts 02115*

(Received 28 October 1992; accepted for publication 17 March 1993)

Epitaxial  $\text{Y}_3\text{Fe}_5\text{O}_{12}$  (YIG) films have been grown by the pulsed laser deposition (PLD) technique on (111) gadolinium gallium garnet substrates. The effect of substrate temperature and oxygen partial pressure on the structure, composition, and magnetic properties of the films was investigated and compared to liquid phase epitaxy YIG films. The results demonstrated that epitaxial YIG films could be prepared under a wide range of deposition conditions, but narrow linewidth ( $\Delta H \approx 1$  Oe) films were producible only at low oxygen partial pressures ( $\text{O}_2 < 250$  mTorr) and relatively high substrate temperatures ( $T_s > 800^\circ\text{C}$ ). Since the linewidth of single-crystal YIG is dominated by surface and volume defects and/or impurities, the narrow linewidth indicated that PLD is a viable technique for producing high-quality ferrite films for microwave device applications. In addition, under all deposition conditions (50–1000 mTorr and  $700\text{--}850^\circ\text{C}$ ) there is a uniaxial axis perpendicular to the film plane. However, at low oxygen pressure the uniaxial anisotropy energy constant  $K_u$  is negative while at high oxygen pressure  $K_u$  is positive.

## I. INTRODUCTION

The pulsed laser deposition (PLD) technique is emerging as a viable technique for preparing high-quality films of ferrite materials. Growth facilities are being set up to prepare ferrites of various compositions and structures. Although this technique appears to be promising based upon our earlier results,<sup>1</sup> we believe that there are still obstacles to overcome before this technique is worthy of further development. As in any new development, calibration and standardization runs are necessary in order to properly evaluate the technique itself. As such, we believe that the preparation of yttrium iron garnet (YIG) films on substrates of gadolinium gallium garnet (GGG) presents an ideal system to prepare for the purposes of calibration since there is an abundant amount of information in the literature about this system. For example, it is well known that high-quality films of YIG on GGG have been prepared by the liquid phase epitaxy (LPE) technique in the seventies.<sup>2</sup> At the very least we would like to compare our films prepared by PLD with the films prepared by the LPE technique.

In addition, although structural and morphological studies can determine the quality of magnetic thin films, these methods do not really supply a practical indication of the quality of the films in terms of suitability for microwave device applications because the magnetic properties (e.g., magnetic resonance linewidth) are really the important figures of merit. Single-crystal YIG thin films, however, present a unique opportunity to evaluate film quality. Since the intrinsic linewidth of YIG is very narrow (0.25–0.5 Oe), any broadening of the linewidth would be in large part due to surface or volume defects and/or impurities.<sup>3</sup> Thus, linewidth measurements can be used to determine the potential of the PLD technique for fabricating high-quality ferrite thin films in general.

In this paper, we report on the magnetic and structural characteristics of epitaxial YIG films prepared by PLD on

(111) GGG substrates at various ambient oxygen pressures and substrate temperatures  $T_s$ . These characteristics were then compared to those measured for LPE prepared YIG films. We find that for some special conditions (300–600 mTorr with corresponding substrate temperatures of  $700\text{--}850^\circ\text{C}$ ) the effective magnetization was measured to be  $\sim 1750$  G, which is the accepted value of saturation magnetization for pure YIG.<sup>4</sup> The FMR linewidth measured for these deposition conditions ranged from about 3 to 90 Oe, which is considerably larger than the  $\sim 0.5$  Oe value for pure YIG.<sup>5</sup> However, we measured narrow FMR linewidths on the order of 1 Oe or less for deposition conditions with oxygen partial pressure less than 250 mTorr and substrate temperature greater than  $800^\circ\text{C}$ . For all growth conditions a uniaxial magnetic anisotropy is induced.

This paper is organized such that in Sec. II the PLD technique as applied to YIG is described along with the experimental techniques used. The structure, composition, and magnetic properties of the PLD YIG films are then discussed in Sec. III. Finally, we arrive at conclusions from the experimental results in Sec. IV.

## II. FILM PREPARATION

YIG ( $\text{Y}_3\text{Fe}_5\text{O}_{12}$ ) thin films were prepared by the pulsed laser deposition technique using a KrF excimer laser ( $\lambda = 248$  nm) with a 20 ns pulse width in a vacuum chamber with a base pressure of  $3 \times 10^{-6}$  Torr. The laser was focused to an energy density of  $\sim 3$  J/cm<sup>2</sup> onto a standard polycrystalline YIG target which was rotated (6 rpm) during the deposition process. The material vaporized from the target was deposited onto 1 cm  $\times$  1 cm (111) single-crystal gadolinium gallium garnet (GGG) substrates bonded to a resistive heater positioned 4 cm away from the target. Films were grown at various substrate temperatures and oxygen partial pressures ranging from 700 to  $850^\circ\text{C}$  and 50 to 1000 mTorr, respectively. After

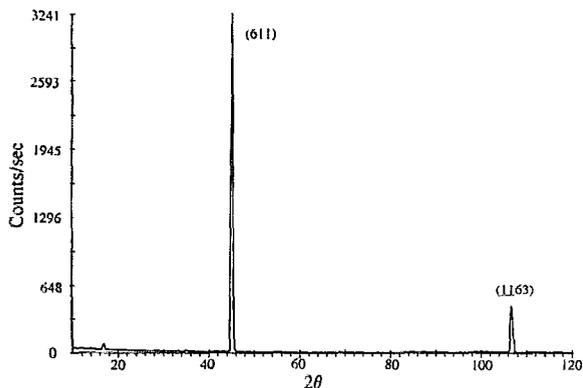


FIG. 1. Glancing incidence angle x-ray diffraction spectrum for an LPE YIG film on (111) GGG substrate.

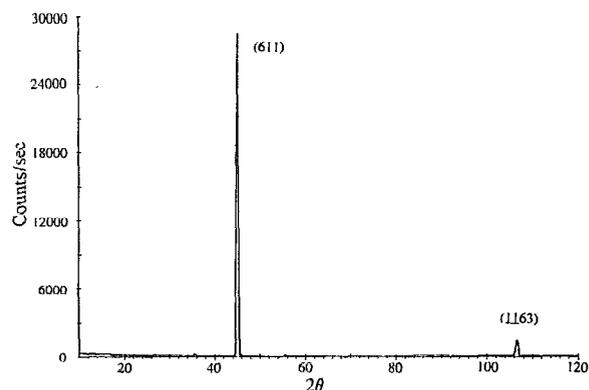


FIG. 2. Glancing incidence angle x-ray diffraction spectrum for a PLD YIG film on (111) GGG substrate prepared at 850 °C and 250 mTorr of oxygen.

deposition the oxygen partial pressure in the vacuum chamber was raised to near atmosphere and the films were then cooled to room temperature. The thickness of the films was measured by both profilometer and scanning electron microscopy techniques to be  $\sim 1 \mu\text{m}$  with a deposition rate of 5.5 Å/s.

Structural characterization of the films was performed using glancing incidence angle x-ray diffraction (GIA XRD) from a  $\text{Cu } K_{\alpha}$  source at an incidence angle of  $2^{\circ}$  operating at 50 kV and 150 mA. Scanning electron microscopy (SEM) was used to determine the surface morphology of the films for comparison with magnetic measurements. Energy dispersive spectroscopy (EDS) provided composition information on the films with respect to the target. Ferrimagnetic resonance (FMR) was performed using a conventional Varian *E*-line spectrometer operating at 9.53 GHz with a cavity resonating in the  $\text{TE}_{102}$  mode in addition to dc magnetic measurements using a vibrating sample magnetometer (VSM).

### III. RESULTS AND DISCUSSION

The GIA XRD experiments were used to investigate the phase and epitaxy of the PLD YIG films. By matching the GIA XRD spectrums of the PLD YIG films with the standard YIG powder diffraction file, it was determined that at all deposition conditions the films were single phase  $\text{Y}_3\text{Fe}_5\text{O}_{12}$ . Epitaxy of the films was qualitatively determined by comparing the GIA XRD spectrum of the PLD YIG films against the GIA XRD spectrum of a high-quality single-crystal LPE YIG film on a (111) GGG substrate, shown in Fig. 1. The LPE YIG film exhibits two prominent diffraction peaks corresponding to the (611) and (1163) planes of YIG. Typically, these peaks were also the major diffraction peaks present in the GIA XRD spectrums of the PLD YIG films, as shown in Fig. 2 for a PLD YIG film deposited at 850 °C and 250 mTorr.

There were two cases where the GIA XRD spectrums of the LPE YIG film and PLD YIG films were markedly different. In the first case, two films deposited at 800 °C and at oxygen partial pressures of 100 and 250 mTorr, respectively, had GIA XRD spectrums which contained almost

all of the diffraction lines in the standard YIG powder diffraction file, although the relative intensities were different, thus indicating that these two films were both textured polycrystalline. In the second case, films deposited at high oxygen partial pressures ( $> 250$  mTorr) and low substrate temperatures ( $< 800$  °C) exhibited additional strong YIG diffraction peaks as shown in Fig. 3, which indicates the presence of YIG crystallites with other orientations. It was subsequently found that films deposited at a high oxygen partial pressure and low substrate temperature had large micron-sized droplets on the surface of the films.

The presence of droplets as a result of using the PLD technique has been reported on extensively in the literature and is usually attributed to either the segregation of a second phase in the film or to laser target interaction parameters such as laser energy density, the optical absorption coefficient or target morphology.<sup>7-9</sup> A second phase in the PLD YIG films can be ruled out, though, since GIA XRD results showed the films to be single phase. In addition, the composition of these droplets was the same as the surrounding film area as determined using EDS; however, it is likely that the additional YIG diffraction peaks at high oxygen partial pressure were due to the presence of these droplets. The effect of the oxygen partial pressure on the

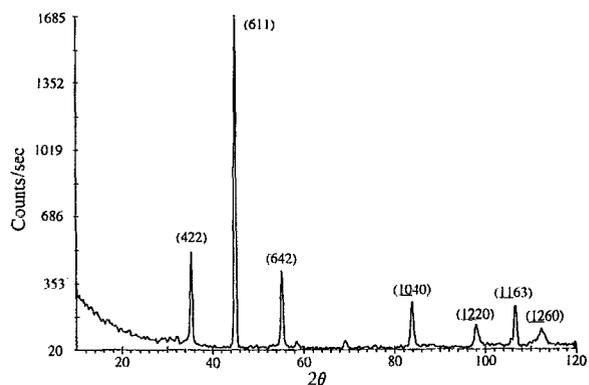


FIG. 3. Glancing incidence angle x-ray diffraction spectrum for a PLD YIG film on (111) GGG deposited at 750 °C and 1000 mTorr.

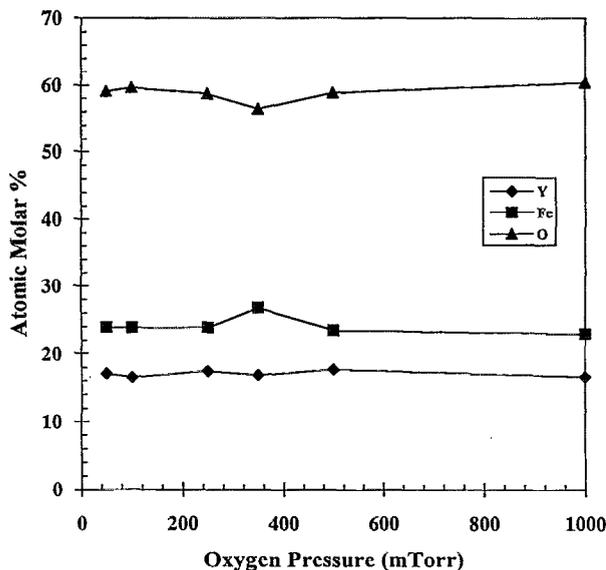


FIG. 4. Composition of PLD YIG films prepared at 750 °C as a function of oxygen partial pressure.

number and size of the droplets, which is much more pronounced than the effect due to substrate temperature, may be due to the expansion of oxygen gas within the porous target during the laser heating process. The result would be to eject particulates or splash material from the target and onto the film.<sup>10</sup> Although we were not able to confirm the mechanism for droplet formation at high oxygen partial pressures, the reduction of particulates with a reduction in oxygen partial pressure has been previously reported for YBCO superconducting films.<sup>11</sup>

The composition, measured using EDS, as a function of oxygen pressure for films prepared at 750 °C is shown in Fig. 4. Only one substrate temperature is shown for clarity since the behavior of the composition at other substrate temperatures was similar. The composition was calculated by comparing the EDS spectrums of the films to an EDS spectrum of the polycrystalline  $Y_3Fe_5O_{12}$  target. First, the target EDS spectrum was normalized to the film spectrum; both spectra were then background filtered and peak intensity ratios were taken. Finally, elemental concentrations were calculated from the peak ratios and the known composition of the target. An average  $X^2$  value of 0.69 between the target and film spectra indicated very good correlation between the target composition and the composition of the films at all deposition conditions. The composition as a function of oxygen pressure is within a few atomic molar percent of stoichiometric with respect to the target and is not dependent on the oxygen partial pressure or the substrate temperature. This is consistent with the x-ray diffraction data, which showed no other phases present in the films.

Ferrimagnetic resonance measurements were performed on the films in order to determine the effective magnetization,  $4\pi M_{\text{eff}}$ , the effective Landé  $g$  factor,  $g_{\text{eff}}$ , and the FMR linewidth,  $\Delta H$ , for the YIG films. The mea-

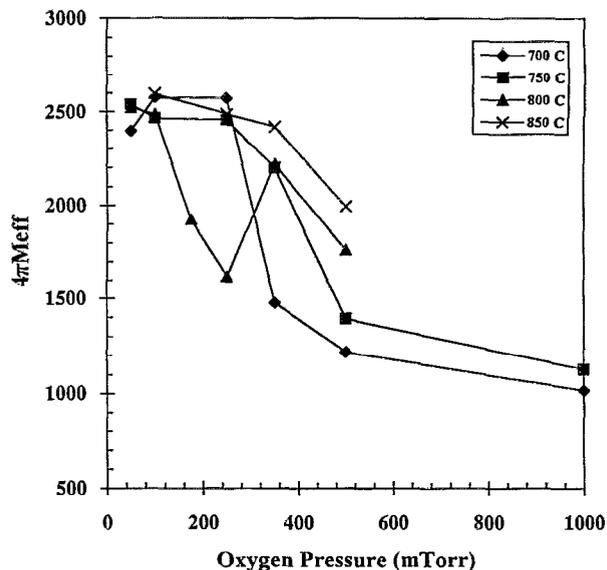


FIG. 5.  $4\pi M_{\text{eff}}$  as a function of oxygen partial pressure and substrate temperature calculated from the in-plane and perpendicular resonant field values measured using standard FMR cavity technique.

surements were performed with the dc magnetic field applied perpendicular ( $H_{\perp}$ ) and parallel ( $H_{\parallel}$ ) to the plane of the film. The resonant equations for such a geometry can be written as

$$(\omega/\gamma) = H_{\perp} - 4\pi M_{\text{eff}},$$

$$(\omega/\gamma)^2 = H_{\parallel} (H_{\parallel} + 4\pi M_{\text{eff}}),$$

where  $4\pi M_{\text{eff}} = 4\pi M_s - H_A$  and  $H_A$  is the uniaxial anisotropy field,  $2K_u/M$ .  $4\pi M_{\text{eff}}$  and  $g_{\text{eff}}$  were calculated by substituting the measured resonant field values for the perpendicular ( $H_{\perp}$ ) and parallel ( $H_{\parallel}$ ) experimental conditions into the above equations. The  $4\pi M_s$  and  $g_{\text{eff}}$  values for YIG in the literature are 1750 G and 2.0, respectively.<sup>4</sup> The calculated values for  $4\pi M_{\text{eff}}$  are shown in Fig. 5 with  $g_{\text{eff}} = 2.0 \pm 0.04$  over the range of deposition conditions. The results indicate a strong dependence on oxygen partial pressure and a lesser dependence on substrate temperature. Since YIG has relatively small cubic anisotropy terms,  $4\pi M_{\text{eff}}$  should approximately equal  $4\pi M_s$ . The variations of  $4\pi M_{\text{eff}}$  from 1750 G were assumed to be due to  $H_A$  and not  $4\pi M_s$ , since the x-ray diffraction data showed the films to be single-phase YIG. The curve at 800 °C deviates from the overall trend at 100 and 250 mTorr such that  $4\pi M_{\text{eff}}$  is much closer to the expected value of  $4\pi M_s$ . It was discussed previously that the films grown under these deposition conditions were polycrystalline with some degree of orientation. This indicates that the changes in  $4\pi M_{\text{eff}}$  are in large part due to a magnetocrystalline anisotropy field, which agrees with our earlier assumptions. Therefore, at low oxygen partial pressures  $H_A$  was found to be negative. This implies that the easy axis of magnetization is in the film plane. However, at high oxygen partial pressures  $H_A$  was positive, implying an easy axis

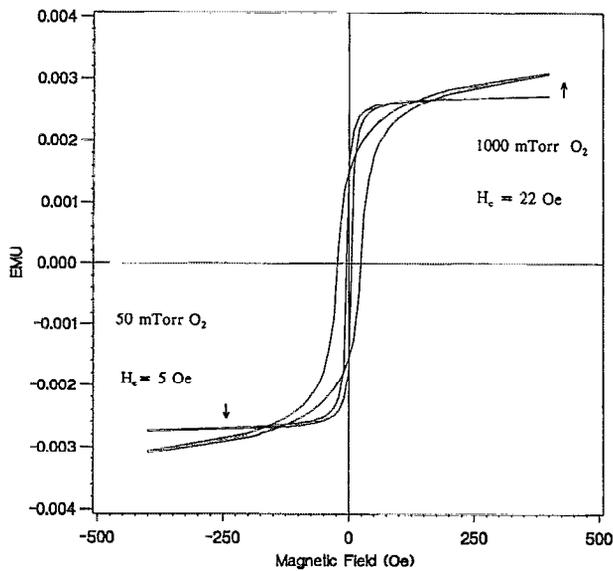


FIG. 6. VSM hysteresis curves for two films prepared at different oxygen partial pressures with the dc magnetic field applied in the film plane. Both films were prepared at 750 °C.

of magnetization perpendicular to the film plane. In either case, the symmetry axis is perpendicular to the film plane.

In order to verify the existence of an induced anisotropy field at low and high oxygen partial pressures, respectively, hysteresis curves were measured for the films using a VSM with the dc magnetic field applied parallel and perpendicular to the film plane. In the perpendicular case, the “knee” of the hysteresis curve is approximately equal to  $4\pi M_{\text{eff}}$  and exhibited the same dependence on oxygen partial pressure as discussed previously for the FMR results. The coercive field  $H_c$  for this case was about 15 Oe for all the films. If the previous assumption about the uniaxial axis is correct, then there should also be a change in the magnetic field required to saturate samples prepared at different oxygen pressures with the dc magnetic field applied in the plane of the film. When the easy axis of magnetization is in the film plane, there are no demagnetizing fields or anisotropy fields to overcome in order to saturate the film; therefore, the hysteresis curve should be relatively square. On the other hand, if the easy axis of magnetization is perpendicular to the film plane, then the magnetic field required to saturate the sample should increase while the squareness decreases. This is consistent with the hysteresis measurements which showed an increase in the magnetic field required for saturation with an increase in oxygen partial pressure. The two hysteresis curves shown in Fig. 6 for films prepared at 50 and 1000 mTorr are illustrative of the effect.

The FMR linewidth  $\Delta H$  of the films measured in the experiments was found to vary with oxygen partial pressure and substrate temperature. In general, at high oxygen pressures and low substrate temperatures the linewidth was largest ( $\cong 90$  Oe), while at low pressures and high substrate temperatures the linewidth was significantly reduced to  $\sim 1$  Oe (see Fig. 7); thus indicating an improve-

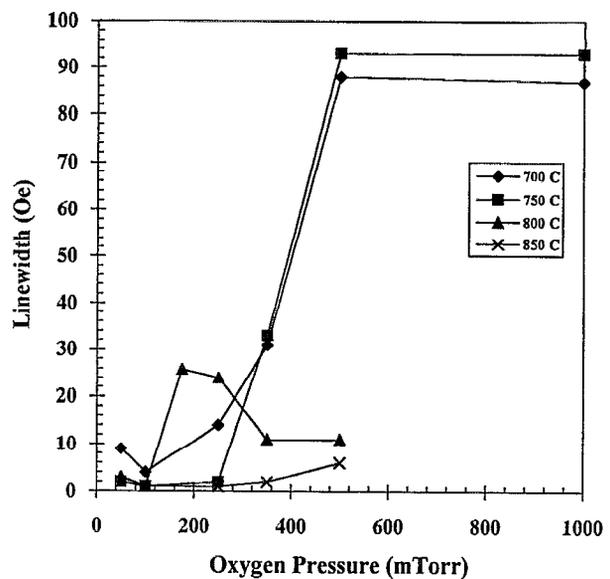
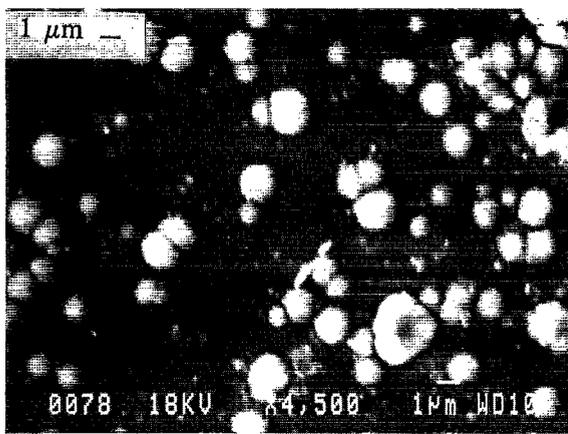


FIG. 7. Differential linewidth  $\Delta H$  as a function of oxygen partial pressure and substrate temperature measured using standard FMR cavity technique.

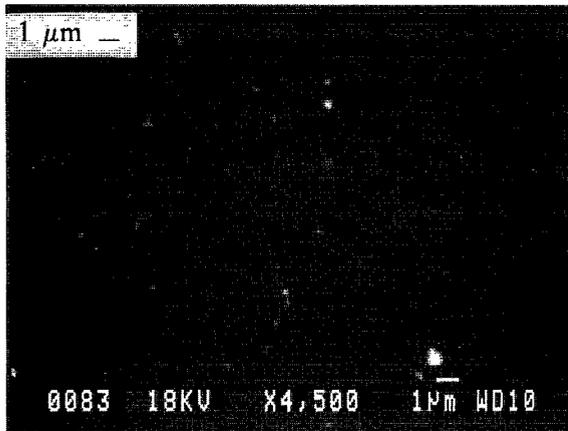
ment in crystal quality. The linewidth was possibly narrower, but due to poor dc magnetic field stability the ultimate linewidth measurable was limited by our FMR system. For example, the linewidth of the LPE YIG films was also found to be  $\sim 1$  Oe using the same experimental setup, which agrees well with linewidth values in the literature.<sup>4-6</sup> The narrowest linewidths were measured at oxygen partial pressures below about 250 mTorr. The dependence of the linewidth on pressure is consistent with the formation of the micron-sized droplets on the surfaces of the films at pressures above 250 mTorr, as evidence by the SEM micrograph in Fig. 8(a). At oxygen partial pressures below 250 mTorr, the number and size of the droplets on the film surfaces was significantly reduced, as seen in Fig. 8(b). The linewidth also in general decreases with an increase in substrate temperature which also exhibits a decrease in droplet size and density on the film surface. These droplets would presumably play the same role as pits in increasing the linewidth and in large part explain the change in linewidth with pressure and temperature. Similar to the graph of  $4\pi M_{\text{eff}}$  as a function of oxygen partial pressure, the linewidth curve for films prepared at 800 °C and at oxygen pressures of 100 and 250 mTorr also deviated from the overall trend. This is to be expected since polycrystalline materials have a larger linewidth than single crystals of the same material.

#### IV. CONCLUSION

Epitaxial YIG films can be grown under a wide range of conditions by PLD. Narrow linewidth films, however, which implies high-quality single crystals free of volume and surface defects require a low oxygen partial pressure and high substrate temperature during the deposition process. YIG films prepared under these deposition conditions



(a)



(b)

FIG. 8. (a) Surface SEM micrograph of PLD YIG film prepared at 750 °C and 500 mTorr O<sub>2</sub>. (b) Surface SEM micrograph of PLD YIG film prepared at 750 °C and 100 mTorr O<sub>2</sub>.

exhibited linewidths which compared well with LPE prepared YIG films. In both cases, the narrowest linewidth measured was  $\sim 1$  Oe; this is slightly larger than the narrowest value reported in the literature for pure single-crystal YIG.<sup>6</sup>

Since the composition data and x-ray diffraction data showed the films to be stoichiometric and single phase under all deposition conditions investigated, it was assumed that the saturation magnetization was in agreement with the bulk value of 1750 G and did not vary with deposition conditions. Variations in the resonant condition and fields necessary for saturation were found to be due to a growth-induced uniaxial anisotropy field which varied in direction and magnitude as a function of ambient oxygen pressure and substrate temperature. At low oxygen partial pressures the maximum  $H_A$  was measured to be  $-850$  Oe while at high oxygen pressures  $H_A = +650$  Oe.

The exact origin of the uniaxial magnetic anisotropy observed in the PLD YIG films could not be determined

from the experimental results of this study. In general, garnet films can have a uniaxial magnetic anisotropy attributable to both a stress term and a growth induced term which are affected by such factors as lattice mismatch, doping concentration, and supercooling.<sup>12-15</sup> Typically the stress-induced term is less significant than the growth-induced term and indeed in the case of the PLD YIG films, annealings at 700, 800, and 900 °C for 2 h each produced no change in the magnetic properties of the films. This agrees with our earlier assumption based on the GIA XRD results that the changes in  $4\pi M_{\text{eff}}$  as a function of oxygen partial pressure were growth induced. However, in order to understand the role of oxygen partial pressure, which has the greatest effect on the magnitude of the uniaxial magnetic anisotropy field, more detailed microstructural studies of the PLD YIG films are required.

The results of this study demonstrate that the PLD technique can produce high-quality ferrite thin films. However, there are other crucial steps which need to be taken in order to achieve PLD ferrite films applicable for microwave devices, such as the potential for growing thicker films by PLD and the problem of producing films of a uniform thickness over large areas. If PLD is capable of overcoming these problems, then indeed PLD is an advantageous technique for preparing ferrite films.

#### ACKNOWLEDGMENTS

The authors would like to thank Dr. H. L. Glass for generously providing the (111) GGG substrates and R. Schmidt of M/A-COM, Inc. for providing the YIG targets used in the experiments. We would also like to thank the Office of Naval Research for supporting this work.

- <sup>1</sup> P. C. Dorsey, R. Seed, C. Vittoria, C. A. Carosella, D. B. Chrisey, P. Lubitz, and J. S. Horwitz, *IEEE Trans. Magn.* **MAG-28**, 3216 (1992).
- <sup>2</sup> R. C. Linares, R. B. McGraw, and J. B. Schroeder, *J. Appl. Phys.* **36**, 2884 (1965).
- <sup>3</sup> M. Sparks, *Ferromagnetic-Relaxation Theory* (McGraw-Hill, New York, 1964).
- <sup>4</sup> W. H. Von Aulock, *Handbook of Microwave Ferrite Materials* (Academic, London, 1965).
- <sup>5</sup> R. C. LeCraw, E. G. Spencer, and C. S. Porter, *Phys. Rev.* **110**, 1311 (1958).
- <sup>6</sup> C. Vittoria, P. Lubitz, P. Hansen, and W. Tolksdorf, *J. Appl. Phys.* **57**, 3699 (1985).
- <sup>7</sup> R. K. Singh, D. Bhattacharya, and J. Narayan, *Appl. Phys. Lett.* **61**, 483 (1992).
- <sup>8</sup> E. Fogarassy, C. Fuchs, A. Slaoui, and J. P. Stoquert, *Appl. Phys. Lett.* **57**, 664 (1990).
- <sup>9</sup> D. H. A. Blank, R. P. J. Ijsselsteijn, P. G. Out, H. J. H. Kuiper, J. Flokstra, and H. Rogolla, *Mater. Sci. Eng. B* **13**, 67 (1992).
- <sup>10</sup> J. T. Cheung and H. Sankur, *CRC Crit. Rev. Solid State Mater. Sci.* **15**, 63 (1988).
- <sup>11</sup> H.-U. Habermeier, *Mater. Sci. Eng. B* **13**, 1 (1992).
- <sup>12</sup> P. Hansen, K. Witter, and W. Tolksdorf, *J. Appl. Phys.* **55**, 1052 (1984).
- <sup>13</sup> P. Hansen, C.-P. Klages, and K. Witter, *J. Appl. Phys.* **60**, 721 (1986).
- <sup>14</sup> P. Hansen, K. Witter, and W. Tolksdorf, *Phys. Rev. B* **27**, 6608 (1983).
- <sup>15</sup> A. H. Boeck, E. G. Spencer, L. G. Van Uitert, S. C. Abrahams, R. L. Barns, W. H. Grodkiewicz, R. C. Sherwood, P. A. Schmidt, D. H. Smith, and E. M. Walters, *Appl. Phys. Lett.* **17**, 131 (1970).