Large-area pulsed laser deposition: Techniques and applications

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Pulsed laser deposition (PLD) has quickly emerged as a unique tool with which to grow high quality films of complex chemical compounds. It is estimated that at present the number of different materials which have been deposited by PLD now exceeds two hundred. Scientists have used this process primarily as a laboratory tool to deposit films of various compounds that are typically difficult to synthesize by other techniques, and then quickly evaluate the relevant material properties. Deposition techniques such as ion-beam, rf, or dc magnetron sputtering, electron-beam evaporation, molecular beam epitaxy, chemical vapor deposition, and metal organic chemical vapor deposition, have all achieved wide-spread acceptance as processes with which to grow various types of electronic and optical films. In order for PLD to emerge as a real production process, it must be demonstrated that PLD is capable of depositing material over useful substrate sizes with acceptable uniformity. PLD must also compete with more established growth techniques or provide film properties which are otherwise unobtainable with these alternatives. Furthermore, as with any commercial process, PLD will become viable only if it is cost effective. Due to its unique capabilities, it is expected that PLD will emerge as a production tool for several applications in the near future. However, such applications are expected to be relatively low volume with high value added to the end product by the laser-deposited film.

I. INTRODUCTION

This article will review three large-area techniques which have been used to scale the pulsed laser deposition (PLD) process to substrates ranging in size from 50 mm (2 in.) to 150 mm (6 in.) in diameter. Several issues related to scaling up PLD will be examined and some of the significant differences between these large-area approaches will be discussed. Furthermore, a few applications of PLD will be presented which point out some of the more salient features of this unique thin-film growth process.

II. LARGE-AREA PLD APPROACHES

It is well known that the ablation plume produced during laser deposition has a highly forward scattered and nonsymmetric flux distribution.[1,2] This fact initially led to a widespread belief that PLD could not be scaled up for any useful applications. However, over the last several years three large-area PLD approaches have been developed to deposit films over substrates at least 100 mm in diameter. These include two techniques utilizing laser beams which are focused (or imaged) down onto small-diameter rotating targets along a fixed optical path, as shown in Figs. 1(a) and 1(b). These approaches are referred to as ''offset'' (OS) PLD, and ''rotational/translational'' (R/T) PLD, respectively. In OS PLD the target is positioned such that the center of the ablation plume (which nominally leaves normal to the target surface), impinges near the outer edge of the rotating substrate. OS PLD was first described as an approach with which to deposit films over several small substrates simultaneously.[3] This technique easily accommodates substrate heating, and has been further used to grow high quality films of YBCO *in situ* on 50-mm-diam $LaAlO_3$ substrates.[4] Furthermore, the OS approach has been scaled up to deposit $Bi_4Ti_3O_{12}$ films over 100-mm-diam Si substrates for RAM applications.[5]

In the second static beam approach, R/T PLD, the substrate is both rotated and translated in a linear fashion as shown in Fig. 1(b). A computer controls both substrate motions such that uniform film properties are obtained. Due to the excursions required to coat large or multiple substrates, this approach does not readily lend itself to substrate heating. However, R/T PLD has been used for several years to deposit the thallium based high-temperature superconducting (HTS) precursor onto three 50-mm-diam $LaAlO_3$ substrates simultaneously, with excellent uniformity of all relevant film properties. This latter work represents the first real small-scale production system for the PLD process.[6,7] More recently, this approach has been used to deposit YBCO films onto 75-mm-diam $LaAlO_3$ substrates.[8]

An alternative to these two static-beam approaches uses computer-controlled laser-beam rastering over a large-diameter rotating target[9–12] as shown in Fig. 2. In this case a target at least one-half the diameter of the substrate is used, and its center is offset from the rotation axis of the substrate as shown. This arrangement allows large substrates to be coated with relatively small targets. Alternatively, a large target whose rotation axis is aligned with the substrate can be used. A focused laser beam is rastered over the target using a mirror held in a programmable kinematic mount. Thus, the laser beam can be programmed to dwell longer at target locations where the center of the ablation plume impinges on the outer edge of the rotating substrate. Laser beam rastering is the oldest of the large-area PLD approaches,[10] accommodates substrate heating,[11] and has been used recently to deposit films on 125-mm-diam substrates.[9] As will be discussed, this process has now been further scaled up to grow highly uniform oxide films on substrates that are 150 mm (6 in.) in diameter.

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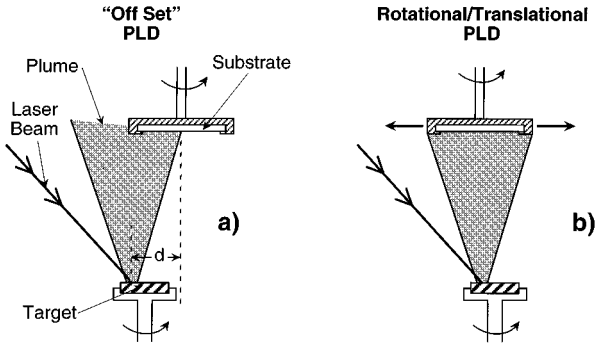

FIG. 1. (a) Schematic diagram of the "offset" large-area PLD approach. (b) Schematic diagram of the "rotation/translational" large-area PLD approach.

III. LARGE-AREA PLD RESULTS

Most applications of the PLD process will require that the films be deposited with a relatively high degree of thickness and compositional uniformity over a given substrate size. Figures 3(a), 3(b), and 3(c) display the normalized thickness profiles which have been obtained using one of the three PLD techniques described above. First, in Fig. 3(a), $Bi_4Ti_3O_{12}$ was deposited onto heated 100-mm-diam (100) Si substrates using the OS technique with a 7 cm target–substrate distance and a 3.2 cm offset distance d [defined in Fig. 1(a)]. Using a fluence of 2 J/cm^2 (248 nm, 25 ns pulse length) with a pulse repetition rate of 10 Hz and a background O_2 pressure of 200 mTorr, an average growth rate of 0.5 μm/h over the central 20 cm^2 portion of the substrate was obtained. The *maximum* variation in the film thickness (defined as the difference between the maximum and minimum values measured across the substrate divided by the average thickness) was found to be just 3% ($\pm$1.5%) over the central $\sim$50 mm portion of the substrate. At larger diameters the thickness starts to drop sharply, and dips below 50% at the substrates outer edge. It has been demonstrated that inserting a metal mask in front of the central region of the substrate during OS deposition can improve the thickness uniformity of the laser-deposited film.[4] However, the insertion of such a mask will clearly reduce the deposition rate as well as significantly alter the temperature of heated substrates.

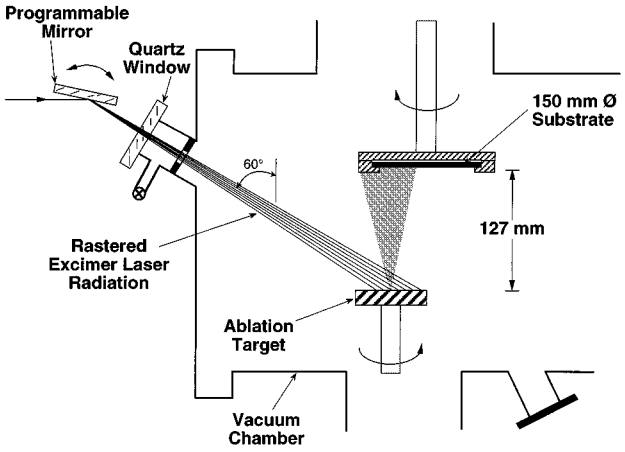

FIG. 2. Schematic diagram of large-area PLD utilizing laser beam rastering with target one-half the diameter of the substrate.

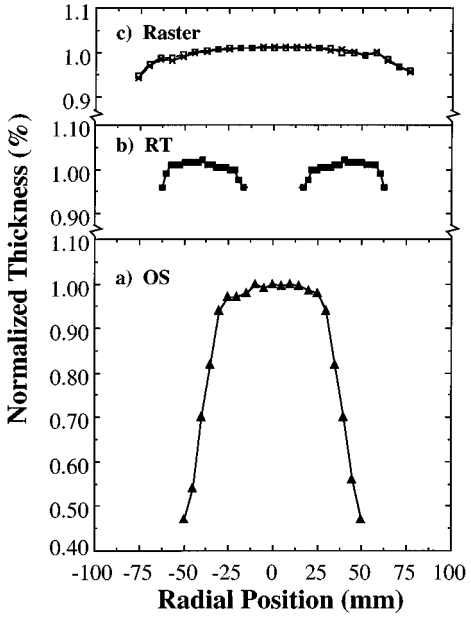

FIG. 3. (a) Thickness profile obtained over 100-mm-diam substrates using OS PLD. (b) The thickness profile obtained over a 50-mm-diam substrate held in a 125-mm-diam substrate holder using R/T PLD. (c) The thickness profile obtained in two mutually perpendicular directions over a 150-mm-diam substrate using laser beam rastering with a 75 mm target diameter.

Figure 3(b) shows the normalized thickness profile obtained from films of the (2122) TlCaBaCuO HTS precursor deposited by the R/T PLD technique onto 50-mm-diam (100) $LaAlO_3$ substrates.[6,7] This film was deposited using a fluence of 4 J/cm^2 (248 nm), with a 50 Hz pulse repetition rate, a background O_2 pressure of 20 mTorr, and a target–substrate distance of 5 cm, yielding an average growth rate of 1.8 μm/h over an area of 127 cm^2. In this case, the thickness profile was measured across one of three 50-mm-diam substrates which were all held in a 125-mm-diam fixture during deposition. The thickness profile obtained from this substrate was measured in the radial direction of the substrate holder. The *maximum* variation in film thickness across the 50-mm-diam substrate was found to be 6% ($\pm$3%). Assuming the deposited film thickness is symmetric about the center of the substrate holder, these data were reflected about the holders center in order to depict a thickness profile which should be obtainable from a single 125-mm-diam substrate.

Figure 3(c) displays the normalized film thickness obtained from a Y_2O_3 film deposited onto a 150 mm (6 in.) diameter Si substrate using laser beam rastering over a 75-mm-diam target with a 12.7 cm throw. The target was offset 45 mm from the rotation axis of the substrate, as shown in Fig. 2. The film was deposited using a fluence of 3.5 J/cm^2 (248 nm and 18 ns pulse length) in an Ar background gas pressure of 7 mTorr with a 100 Hz repetition rate yielding an average growth rate of 0.4 μm/h. The final film thickness, 0.75 μm, was measured with an ellipsometer at 50 locations, 25 each in two mutually perpendicular directions and the normalized film thickness profile is shown in Fig. 3(c). The *maximum* variation over the 150-mm-diam substrate was

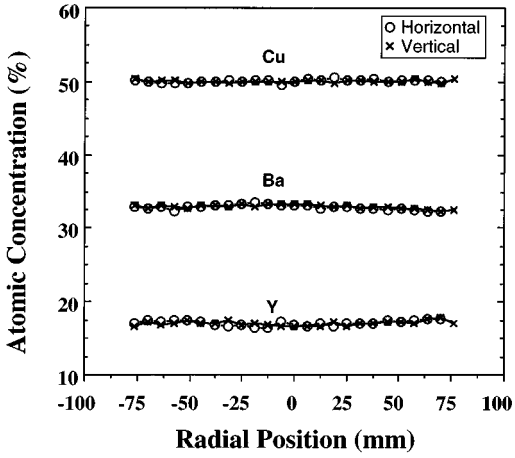

FIG. 4. The composition profile of a YBCO film grown by PLD on a 150-mm-diam Si substrate as measured by EDXA. The data were measured in two mutually perpendicular directions of the substrate as shown.

found to be just 7.1%. However, neglecting the four end points, each located at a radial location of 74.5 mm (taken 0.5 mm from the substrate edge), the *maximum* variation in film thickness was found to be only 4.6%, or $\pm 2.3\%$.

While the thickness uniformity is a relevant issue, film composition may in fact be more important for most PLD applications. Figure 4 shows the composition profile of a 2-μm-thick YBCO film laser deposited onto a 150-mm-diam Si substrate using beam rastering over a 90-mm-diam stoichiometric target. YBCO was chosen as it provided three elements whose composition could be easily mapped using energy dispersive x-ray analysis (EDXA). This film was deposited onto an unheated substrate using a fluence of 3 J/cm^2 (248 nm) at 100 Hz in an O_2 pressure of 2 mTorr. Using a 12.7 cm throw distance the average growth rate over a 182 cm^2 area with these conditions was 1 μm/h. EDXA was used to map the film composition at 50 different locations (every 6.35 mm) across the substrate surface, 25 points in each of two mutually perpendicular directions. The nominal film composition was defined as $Y_1Ba_2Cu_3O_7$ at the substrate's center for the EDXA calculations. The *maximum* variation in film composition, defined simply as the difference between the maximum and minimum atomic concentration values obtained from the EDXA data, for the Y, Ba, and Cu cations were 1.4, 1.2, and 0.8 at. % (± 0.7, ± 0.6, and ± 0.4 at. %), respectively, across the 150-mm-diam substrate. In comparison, the standard errors for the EDXA data were found to be ± 1.48, ± 0.17, and ± 0.36 at. % for the Y, Ba, and Cu species, respectively. Matching the levels of uniformity demonstrated by PLD with laser beam rastering for both thickness and composition for a complex chemical compound such as YBCO would be difficult for most of the alternative PVD techniques mentioned previously. The R/T PLD approach has also demonstrated excellent compositional uniformity after a high-temperature thallination step. Using EDXA to map the composition of a film deposited onto a 50-mm-diam substrate held in a 125-mm-diam holder, maximum variations between 1.0 and 2.8 at. % for the Tl, Ba, Ca, and Cu elements were obtained.

Not only is the uniformity of film composition over the substrate surface a concern when using any growth technique, but the reproducibility of film composition for successive depositions needs to be addressed as well. Furthermore, certain applications of PLD will require relatively thick films, and the question of composition gradients in the growth direction become an issue. One such application which will require thick films is microwave circulators based on ferrite materials such as $Y_3Fe_5O_{12}$ (YIG). Thick YIG films were deposited over 3-cm-diam polycrystalline YIG substrates using a target–substrate distance of only 4 cm. The laser beam was rastered over the outer radii of a 50-mm-diam target located under the substrate. Average deposition rates of 10 μm/h were obtained using a fluence of 4 J/cm^2 (248 nm) with a 100 Hz repetition rate in an O_2 pressure of 2 mTorr. No attempt was made to heat the substrate during deposition, but it is estimated that the substrate temperature rose well over 100 °C due to the high rate of ferrite film condensation. Figure 5 shows a scanning electron microscope (SEM) micrograph obtained from the cross section of an as-deposited thick YIG film grown on a polycrystalline YIG substrate. The vertical white bar in the figure represents a length of 106 μm, and the interface between the fully dense film and substrate is clearly visible. EDXA was used to measure the cross-section's composition first at four locations within the bulk YIG substrate, and then at eleven points within the deposited film each point taken at 10 μm intervals (Fig. 6). A laser-deposited YIG film whose composition was determined by Ruthford backscattering spectroscope (RBS) served as a standard for the EDXA measurements. Variations in composition of the YIG substrate were found to be larger than that obtained within the laser-deposited YIG film, which had *maximum* variations of 2.3 and 5.0 at. % for the Y and Fe, respectively. The magnitude of these fluctuations are likely caused by sample charging and statistical variations in the x-ray signal for oxygen, both of which will affect the

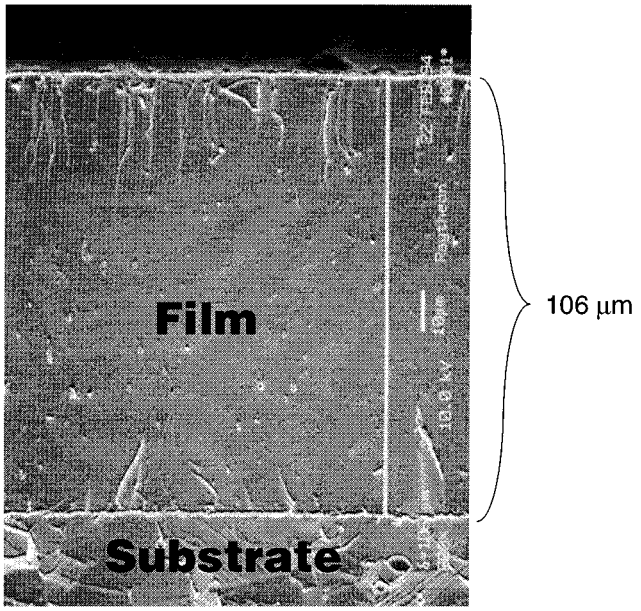

FIG. 5. Scanning electron micrograph of the cross section of a 106-μm-thick YIG film which was grown by PLD on a polycrystalline YIG substrate.

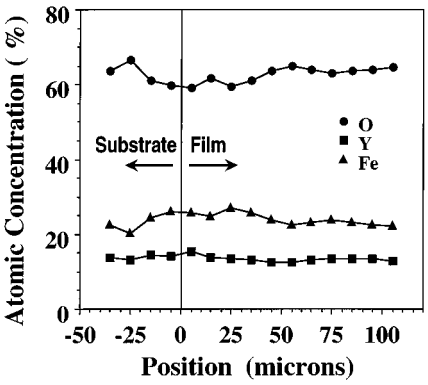

FIG. 6. The composition of the cross section of the YIG substrate and film shown in Fig. 5 as measured by EDXA.

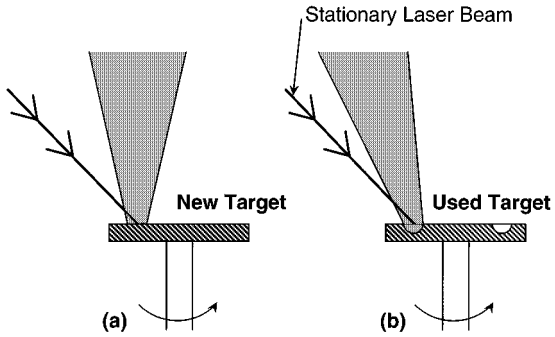

FIG. 7. (a) The ablation plume as shown when a laser beam impinges onto a virgin target surface. (b) The ablation plume as shown when a static position laser beam impinges on the trench which has been etched into the target surface by this type of laser deposition process.

calculated composition. However, there appear to be no concentration gradients for either Y or Fe in the direction of film growth. This fact has important implications for commercial applications of *thin* films (~ 1 μm thick) since it illustrates that the film composition remains constant even when large amounts of target material have been removed. Thus, using laser beam rastering with the PLD process should produce films with the same composition from run to run. It should be mentioned that the substrate shown in Fig. 5 spontaneously cracked a few minutes after removal from the vacuum chamber, even though the deposition parameters (pressure and fluence) were chosen to yield a relatively low film stress (-150 MPa$<\sigma$).

Each of the large-area PLD approaches described above have been used to grow films of HTS materials over large areas.[4,6–11] The electrical properties of these films such as T_c, J_c, normal state dc resistance, and microwave surface resistance have found to be quite uniform for all three of the large-area PLD techniques. It should be stressed that the properties of HTS thin films depends critically on a wide variety of deposition parameters. The demonstrated ability to grow uniform films of high quality HTS materials over large substrates is relevant since it indicates that obtaining uniform film properties of other potentially less complex materials should be a relatively easy task using the PLD process.

While both static beam and rastering approaches have demonstrated the ability to deposit uniform films over large substrate sizes, the basic difference between these approaches has some important implications for the condition of the ablation target surface topography, which in turn can effect deposited film properties. The static large-area PLD approaches typically utilize 25-mm-diam rotating ablation targets with laser beam focused down to a small spot ($\sim 1\times 3$ mm) close to the target's outer edge. When the deposition process starts with a new target the plume is ejected normal to the target surface, as shown in Fig. 7(a). However, as the deposition proceeds, the laser beam etches a trench in the target surface with each laser pulse. The formation of this trench can have several deleterious effects on the film growth process. First, as the trench forms, the irradiated surface area increases, and thus the on-target fluence is reduced which can have significant effects on the characteristics of the deposition process as well as film properties. Furthermore, since the ablation plume leaves the target in a direction normal to the *local* surface, the plume starts to tilt back toward the incident laser beam, as shown in Fig. 7(b). The plume angle changes continually throughout the film growth process, and will continually alter the deposition rate as well as other film properties. This effect also makes it difficult to predict the final film thickness and thickness uniformity from one deposition to the next. As an example, a YBCO film was deposited onto a 75-mm-diam Si substrate using a static laser beam focused 9 mm from the center of a virgin 25-mm-diam YBCO target using 90 K laser pulses. The centers of rotation of both the substrate and target were aligned. After deposition, the substrate was removed from the chamber and the film thickness profile was measured and is displayed as in Fig. 8, curve (a). This ablation target was then irradiated for another 180 K laser pulses using the same fluence and pressure, simulating two more ''identical'' depositions. Finally, another film was deposited onto a second 75-mm-diam substrate using a final 90 K laser pulses with the same parameters. The thickness profile of this second film is shown in Fig. 8, curve (b). Clearly evident is the fact that the thickness profile changed significantly and the deposition rate dropped to 32% of its initial value. While the thickness uniformity appears to have improved, the bulk of the film is now comprised of mostly off-axis (nonstoichiometric) mate-

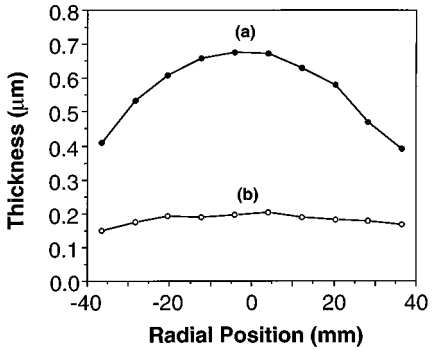

FIG. 8. Curve (a): the thickness profile, shown as (●), obtained from a rotating 75-mm-diam Si substrate using 90 K laser pulses with a static laser beam and a new 25-mm-diam YBCO target. Curve (b) the thickness profile, shown as (○), obtained across a rotating 75-mm-diam substrate deposited using a final 90 K laser pulses with a static laser beam after this target had been previously etched by 270 K pulses.

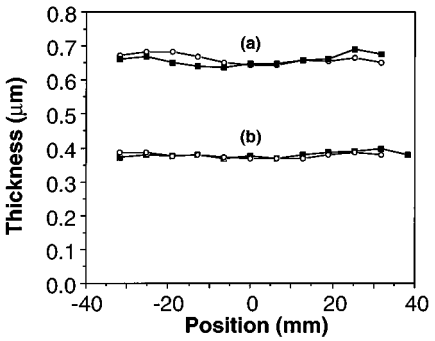

FIG. 9. (a) The thickness profile obtained across a rotating 75-mm-diam substrate using 240 K laser pulses with laser-beam rastering and a new 50-mm-diam target. (b) The thickness profile obtained across a rotating 75-mm-diam substrate using 240 K pulses with laser beam rastering after this target was previously etched by nearly 10^6 laser pulses. (○): Horizontal direction; (■): vertical direction.

rial. The change in deposition rate and film uniformity are primarily due to dynamic tilting of the ablation plume caused by the macroscopic topographical changes in the target surface. In order to minimize these effects researchers typically remove the ablation target after each run and polish the surface with emery paper. This process wastes significant amounts of target material, can possibly contaminate the target, and requires a loadlock if an ultrahigh vacuum (UHV) type PLD systems is used. Local rastering of the laser beam over small diameter targets can help, but it will not eliminate the need for frequent target resurfacing. As applications demand larger substrate sizes and/or thicker films target trenching will become even more of a problem for both of the static beam PLD approaches.

On the other hand, rastering the laser beam over a large-diameter target eliminates the macroscopic trenching and leads to much more reproducible deposition rates and thickness profiles. Figure 9 shows the thickness profile as measured in two mutually perpendicular directions of a phosphor film deposited onto a rotating 75-mm-diam Si substrate using a virgin 50-mm-diam Tb:YAGG target. This film was deposited with 240 K laser pulses using a fluence of 1 J/cm^2 (248 nm) in a background O_2 pressure of 2 mTorr. The substrate was then removed from the chamber, and the target was irradiated again for another 720 K pulses (representing three more runs) using the same deposition parameters. A second substrate was placed in the chamber and a film was again deposited with 240 K pulses using the same deposition conditions. The thickness profile obtained in two mutually perpendicular directions for this latter substrate is also depicted in Fig. 9 as curve (b). In this case the thickness profile has remained virtually unchanged. However, the film thickness and thus, the deposition rate has dropped to about 57% of its initial value. This drop in deposition rate is not due to gross changes in the plume angle, but is caused by much smaller changes in the target topography, i.e., the microscopic formation of cone like structures on the target's surface.[2] The formation of these cones reduces the average laser fluence, but once they have formed over the entire target surface, the fluence reaches a steady state value and the deposition rates remain constant. Subsequent depositions with such targets yield both reproducible deposition rates and film thickness profiles. Thus, when using laser beam rastering target polishing or loadlocks are not required, and target utilization and film purity are greatly enhanced. It should be pointed out that the cones are formed on the target surface with both static and rastered laser beams, and have been associated with the particulates found in laser-deposited films.[2] The formation of the cones will be much quicker with small diameter targets (with or without rastering) due to the fact that there is a smaller available area to irradiate. However, the normalized particle densities obtained from films of the same compounds deposited with either static or rastered laser beams have been found to be similar in magnitude.

IV. LARGE-AREA APPLICATIONS

One interesting application of the PLD process is the growth of yttria (Y_2O_3) coatings on CdTe or CdZnTe substrates.[9] With the proper thickness, these films act as antireflective coatings and enhance the sensitivity of backside illuminated HgCdTe infrared (IR) diode arrays. While there are several ways with which to deposit yttria films such as rf magnetron or ion-beam sputtering, as well as electron beam evaporation, PLD can offer several advantages over these techniques. The advantage for this particular application is the fact that PLD can be a *low-temperature* process. Low-temperature processing is critical to the fabrication of the HgCdTe diode arrays as process temperatures much above 75 °C causes Hg diffusion which in turn produces vacancies in the HgCdTe lattice, altering device performance. Large-area PLD based on laser beam rastering has been used to deposit Y_2O_3 films over several rectangular (20 by 30 mm) (111)*b* CdTe and CdZnTe substrates simultaneously. The yttria films were deposited with 248 nm radiation using a fluence of 3.5 J/cm^2, a repetition rate of 100 Hz, in a pressure of 7 mTorr of Ar with a 12.7 cm throw, yielding a growth rate of 1 μm/h. Special temperature monitors attached to the substrates indicated that the substrate did not rise above 72 °C during deposition. On the other hand, monitors placed on similar substrates indicated that the substrate temperature exceeded 120 °C for Y_2O_3 deposition with both rf magnetron sputtering (5 cm throw distance and 150 W of rf power) and electron-beam evaporation (91 cm throw and 3 Å/s deposition rate). Both CdTe and CdZnTe substrates are extremely fragile, and the Y_2O_3 films are deposited onto the backside of the substrates after the formation of the HgCdTe diode arrays. Thus, the substrates cannot be clamped or thermally greased down to water-cooled holders to reduce substrate temperature when using any deposition technique. X-ray powder diffraction scans of these low temperature laser-deposited Y_2O_3 films indicates that they display bcc polycrystalline texture. In contrast, films deposited with the other PVD techniques were amorphous. The adherence of the laser-deposited films were also superior, they resisted chemical attack much better than the alternative PVD films in subsequent diode array processing, and they displayed excellent lift-off properties. Furthermore, the adherence of the yttria films to the CeTe and CdZnTe substrates depended strongly

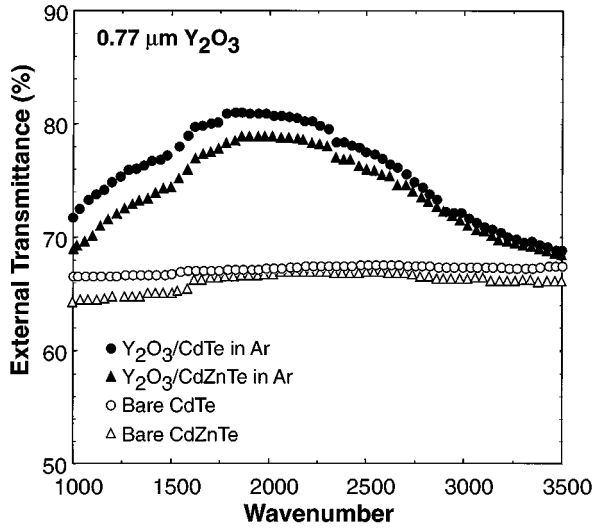

FIG. 10. The transmission characteristics of (111)*b* CdTe and CdZnTe substrates with and without 0.77-mm-thick yttria films deposited by PLD.

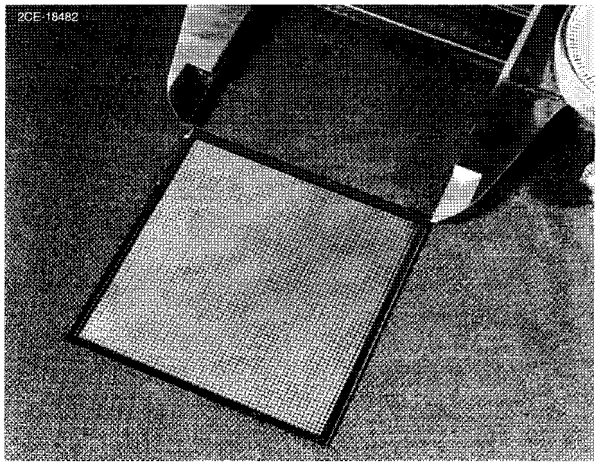

FIG. 11. A photograph of a 50-mm-sq RGB thin-film phosphor screen deposited by PLD and a lift-off process.

on the deposited film stress. However, by varying both the background gas pressure and on-target laser fluence the Y_2O_3 film stress could be tailored to yield high quality, well adhered films with excellent anti-reflective (AR) properties as well as low normalized particulate densities[2] (65 particles/cm^2/Å). All the particulates found with the SEM were below 1 μm in diameter. Figure 10 shows the IR transmission characteristics of (111)*b* oriented CdTe and CdZnTe substrates with and without a 0.77 μm Y_2O_3 laser-deposited AR coating. As noted, the coatings increased the IR transmission for these substrates from 65% to 80% (the amount predicted theoretically for this material). The adherence and IR transmission characteristics of these laser deposited Y_2O_3 films were found to be as good or better than those produced by electron-beam evaporation, rf, and dc magnetron sputtering. Furthermore, the PLD process provided higher deposition rates with significantly lower substrate temperatures making it much more attractive for this application. Finally, the IR diode arrays being processed are expensive, and the value of product which can go through the chamber in one Y_2O_3 deposition can exceed $250K, indicating a high level of confidence in this large-area PLD process.

Another interesting application for the PLD process is the growth of thin-film phosphor materials for use in flat panel displays (FPDs) based on field emission array (FEA) technology.[13,14] Thin-film phosphors will offer several advantages over conventional powder phosphor screens. Again, due to the ability to grow films at low temperatures, PLD can be used with photo resist and a standard lift-off process, and full color red-green-blue (RGB) thin-film screens can be fabricated as shown in Fig. 11. To form this screen, PLD with laser beam rastering was employed using 50-mm-diam targets of $Y_{2.82}Eu_{0.18}Ga_2Al_3O_{12}$ (Eu:YAGG) for red, $Y_{2.82}Tb_{0.18}Ga_2Al_3O_{12}$ for green, and $Y_{2.80}Tm_{0.20}Ga_2Al_3O_{12}$ for blue, respectively. These YAGG films were deposited to a thickness of 2 μm one at a time onto a photolithographically patterned 75-mm-diam (1102) sapphire substrate. The undesired material was removed using liftoff after each deposition, and the process was repeated for the next color. After all three materials were deposited the substrate was annealed in oxygen at 1400 °C for 3 h. An Al film was then deposited over the phosphor for charge dissipation, the screen was cut to size, and then placed inside a CRT test apparatus to measure the efficiency. A luminous efficiency of 1.2 lm/W was obtained with a screen brightness of over 1000 cd/m^2 using a screen potential of 20 kV, as shown in Fig. 12. For comparison, modern TV screens using powder phosphors provide about 550 cd/m^2 with total RGB efficiency of about 20 lm/W at 27 kV. Monochrome screens up to 8 μm in thickness have also been fabricated and tested, indicating that in order to obtain high brightness and improved efficiency thicker YAGG films are necessary.

V. CONCLUSIONS

The PLD process is still being explored by research groups worldwide as a unique deposition tool which can deposit an array of complex materials for a variety of useful applications. As these processes are transitioned from the laboratory to the market place the techniques which can deposit large-area films will then be rigorously tested. Three

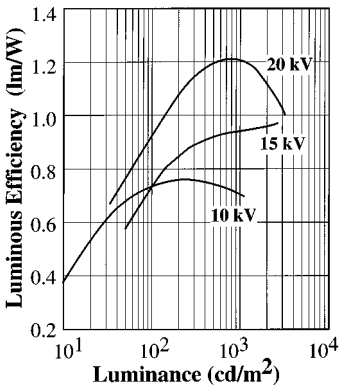

FIG. 12. The extrinsic luminous efficiency of a 2-μm-thick RGB thin-film phosphor screen deposited by PLD as a function of screen luminance (brightness) and anode potential.

approaches have been used to scale up the PLD process to substrates of at least 100 mm in diameter. While the rotational/translational approach provides excellent uniformity over large areas, the two static beam approaches will be hampered by the dynamic changes in target topography, and thus are not likely to become useful for commercial production systems. Laser beam rastering over large-diameter targets appears to offer the best approach for obtaining large-area films with predictable and reproducible growth rates and film qualities. It has also been shown that PLD is compatible with temperature sensitive materials and that, unlike sputtering, PLD can deposit patterned films using photoresist and lift-off processing. Furthermore, PLD has now demonstrated the capability to deposit highly uniform films over 150 mm (6 in.) diameter substrates. Variations over the useable area of 150-mm-diam substrates were found to be only $\pm 2.3\%$ and ± 0.5 at. % for thickness and composition, respectively. These latter data presently represent the state of the art for the laser-deposition process, and clearly indicates that PLD can be scaled to sizes compatible with mainstream semiconductor processing equipment. Finally, the limits of the PLD process have not yet been reached, and it is expected that similar uniformity over even larger substrate sizes will be demonstrated in the near future.

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[1]J. A. Greer and H. J. Van Hook, Mater. Res. Soc. Symp. Proc. **169**, 463 (1990).
[2]*Pulsed Laser Deposition of Thin Films*, edited by D. B. Chrisey and G. K. Hubler (Wiley, New York, 1994).
[3]K. B. Errington and N. J. Ianno, Mater. Res. Soc. Symp. Proc. **191**, 115 (1990).
[4]S. R. Foltyn, R. E. Muenchausen, R. C. Dye, X. D. Wu, L. Luo, D. W. Cooke, and R. C. Taber, Appl. Phys. Lett. **59**, 1374 (1991).
[5]H. Buhay, S. Sinharoy, M. H. Francombe, W. H. Kasner, J. Tavlacchio, B. K. Park, N. J. Doyle, D. R. Lampe, and M. Polinsky, Proc. Integrated Ferroelectrics **1**, 213 (1992).
[6]M. D. Robinson, M. M. Eddy, B. J. L. Nilsson, W. L. Olson, E. J. Smith, K. H. Young, G. V. Negrete, and R. B. Hammond, presented at The International Conference on Metallurgical Coatings and Thin Films, San Diego, 1991 (unpublished).
[7]E. J. Smith, M. T. Smith, M. M. Eddy, G. G. Firpo, B. J. L. Nilsson, B. F. Zuck, and D. D. Strother, presented at The Fall Meeting of the Materials Research Society, Boston, MA, 1992 (unpublished).
[8]M. Lorenz, H. Hochmuth, H. Borner, D. Natusch, and K. Kreher, Mater. Res. Soc. Symp. Proc. **341**, 189 (1994).
[9]J. A. Greer and M. D. Tabat, Mater. Res. Soc. Symp. Proc. **341**, 87 (1994).
[10]J. A. Greer, in *Superconductivity and Applications*, edited by H. S. Kwok, Y. Kao, and D. Shaw (Plenum, New York, 1989), pp. 117–126.
[11]J. A. Greer, J. Vac. Sci. Technol. A **10**, 1821 (1992).
[12]J. A. Greer, Proc. SPIE **1835**, 21 (1992).
[13]J. A. Greer, H. J. Van Hook, H. Q. Nguyen, M. D. Tabat, G. Gammie, and P. Koufopoulos, in SID'94 Digest, 827 (1994).
[14]J. A. Greer, H. J. Van Hook, M. D. Tabat, H. Q. Nguyen, G. Gammie, and P. F. Koufopoulos, Mater. Res. Soc. Symp. Proc. **345**, 281 (1994).