Influence of Substrate Heat Treatment to the Structural Properties of Nd:YAG Thin Films Produced by Femtosecond Pulsed Laser Deposition

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The crystallinity, surface morphology and topography of the femtosecond pulsed laser deposited Nd:YAG film under in-situ and post deposition heat treatment were examined. Heat treatment improved the crystallinity of the film with the increase in reflecting planes shown in X-ray diffraction data. Scanning electron micrograph of the heat treated film surface indicates a small degree of melting. Surface topography of the as-deposited Nd:YAG film under AFM shows size variation to within few tens of nanometers indicating the generation of nanoparticles. Temperature dependence of the cross section area, height and surface roughness of the film was determined and explained by the volume free energy of the film. Our results demonstrate the novel use of femtosecond laser to ablate and deposit laser crystal as well as heat treatment to engineer the structural properties of the film.


Keywords: Laser methods; Growth; Surface structure, morphology, roughness, and topography

I. INTRODUCTION

Lasers with pulse duration in the nanosecond domain such as excimer and Nd:YAG lasers have been used in pulsed laser deposition (PLD) as an excitation source [1, 2]. Thin films of different materials such as superconductor [3, 4] and semiconductor [5] as well as devices [6, 7] have been fabricated by this technique. Recently, there is growing interest in the use of ultrafast femtosecond (fs) pulsed lasers for ablation and deposition [2, 8]. The fs laser pulse duration is much less than the electron-lattice interaction time as well as the electron heat conduction time [9, 10]. Heat generated in the process does not have enough time to propagate in the target due to its small thermal diffusion length [1, 9]. The thermal damage to the target is minimized, reducing the number of particulates deposited on the substrate [1, 9, 11] and improving the quality of the film.

One novel application of PLD is fabrication of thin film laser crystal, in which good optical, mechanical and thermal properties are needed. One of the most popular is the neodymium doped yttrium aluminium garnet (Nd:YAG) due to its spectral and lasing characteristics [12]. Solid state laser crystals are usually produced in bulk form. Nd:YAG or other crystals made into thin film can take advantage of the photon confinement resulting from its reduced dimension. Confinement occurs by the repeated internal reflection due to differences in the refractive index of the crystal with respect to its surrounding. It can result in a more efficient and lower onset of laser oscillation [13, 14]. Other innovative optoelectronic applications of thin film laser crystals include waveguide lasers [13–15] and microchip laser [16, 17].

Physical vapor deposition technique such as PLD have inherent limitations restricting the properties of the film produced [18]. As a result, heat treatment is usually employed to improve the quality of the deposited film [4, 19]. In general, there are 2 types of heat treatment. In-situ heat treatment refers to application of heat during deposition. In post-deposition heat treatment, heat is applied to the substrate and film after the deposition process. Thin films prepared by nanosecond pulsed laser with heat treatment must consider the large sized particulates produced [20, 21]. On the other hand, fs pulsed laser deposition results in a fewer number of particulates [1, 9, 11], making heat treatment investigations more controllable.

In this work, we investigate the characteristics and properties of Nd:YAG thin film on Si substrate fabricated by PLD using a fs Ti:Sapphire laser as the excitation source. The films are characterized and studied using X-ray diffraction analysis, atomic force microscopy and scanning electron microscopy. The effects of post deposition and in-situ substrate heat treatment on crystallinity, surface morphology and surface topography of the deposited Nd:YAG thin film are also examined.

II. EXPERIMENTAL

A 500 mW mode-locked fs Ti:Sapphire laser (Spectra Physics Tsunami) operating at 808 nm, ≈100 fs pulse duration and 80 MHz repetition rate ablates the Nd:YAG target and form the deposition flux. The target exhibits high absorption at this wavelength [12]. A rotating mount is provided for the target inside the vacuum chamber. The laser beam was focused on the target using a lens. The pressure inside the vacuum chamber was brought down using a combination of rotary pump and turbo-molecular pump. The thin film was deposited on silicon (110) and (111) substrates in a wide range of deposi-
tion period (2 hrs-6 hrs), deposition pressure (10^{-2} mbar-10^{-6} mbar) and target-to-substrate distance (2 cm-5 cm) with no background gas. In-situ heat treatment as well as post deposition heat treatment were implemented through radiative heating using a 1 kW halogen lamp located at the back of the substrate. A thermocouple sensor near the substrate monitors the temperature. The silicon substrates were processed using RCA cleaning method. The schematic diagram of the experimental set-up is shown in Fig. 1.

Surface morphology, crystallinity and surface topography of the deposited films were investigated using scanning electron microscope (Philips XL30), X-ray diffractometer (Bede D3 XRD) and atomic force microscope (NTMDT Solver Pro).

III. RESULTS AND DISCUSSION

Figure 2 shows the XRD spectrum of the (a) Nd:YAG target, (b) as-deposited Nd:YAG film, and (c) in-situ heat treated Nd:YAG film. The wavelength used in the X-ray diffractometer is 1.5406 Å with scan step size is 0.050°. The as-deposited films were grown at 3 cm target-to-substrate distance and 2 hrs deposition period while the in-situ heat treated films were deposited for 3 hrs at 2 cm target-to-substrate distance. Peaks corresponding to the (321) and (611) planes indicate that films have preferred crystallographic orientation on the Si substrate. Improvement in crystallinity of the in-situ heat treated films deposited at substrate temperature of $T_{sub} = 400\,^\circ C$ for 3 hrs (Fig. 2(c)) were observed with the appearance of the (422) reflecting plane. The number of reflecting planes increased with the improved mobility of the deposited particles to orient themselves. The additional energy from the heat treatment allowed atoms to adjust and relax in their corresponding lattice sites.

The surface topography of the as-deposited film fabricated at 3 hrs deposition time, $\approx 10^{-5}$ mbar deposition pressure and 5 cm target-to-substrate distance are shown in Fig. 4. The image was taken using tapping mode AFM with Si tip and corrected by a linear fit. A line scan (Fig. 4-bottom) taken along the horizontal white line in the image, reveals that the variation in the height of deposited particles in the observation area to be within few tens of nm, indicating production of nanoparticles. The short pulse duration of the femtosecond laser causes the ablation area to experience high strain rate with minimal heat transfer to the neighboring lattice site [1, 9]. Superheated layer is not fully formed, reducing the micron-sized
particulates from the ejected liquid droplets [9]. However, the material can be ablated due to the fragmentation as a
effect of isochoric heating decomposition in the high strain
ablation site [9, 22] resulting to ejection of the nanoparticles
that has been observed in the AFM image. This
production of the nanoparticles can be utilized to investig-
ate in detail the influence of the substrate heating effect
in the deposited film.

Nd:YAG thin films were prepared with
in-situ
and post
deposition substrate heat treatment at 3 hrs deposition
time, \(10^{-4}\) mbar deposition pressure and 2 cm target-to-
substrate distance. The temperature was increased from
room temperature to the desired substrate temperature
and brought back to room temperature after 3 hrs of heat
treatment.

Figures 5(a), 5(b) and 5(c) shows the atomic force micro-
graph of the films deposited with in-situ substrate heat
treatment of temperatures 300°C, 450°C and 600°C, re-
spectively. The surface topography of the films subjected
to post deposition substrate heat treatment of 400°C, 500°C and 600°C are shown in Figs. 5(d), 5(e) and 5(f).

The scan area of all the images are 10 \(\mu\)m \(\times\) 10 \(\mu\)m. The images are corrected by a linear fit. Both in-situ and post
deposition heat treatment of films results to decrease in
the size of the deposited particles with temperature in-
crease compared to films without heat treatment. More
islands are also found in the post deposition heat treated
films compared to the in-situ heat treated films. This
behaviour is traced to the volume free energy per unit
volume of the clustered particles given by the equation

\[
\Delta G_V = -\frac{kT}{\Omega} \ln \left( \frac{P}{P_e} \right),
\]

where \(k\) is the Boltzmann’s constant, \(T\) the absolute tem-
perature, \(\Omega\) the atomic volume of the film atoms, \(P\)
the pressure of the incoming ablated atoms in the surface
of the substrate and \(P_e\) the vapour equilibrium pressure
[23, 24]. Decreasing \(\Delta G_V\) would result to a decrease in
the total free energy barrier for cluster formation that can
induce nucleation and island growth [23, 24]. Equation (1) dictates that
volume free energy of the as-deposited film would depend
solely on the deposition pressure. However, the temper-
ature term becomes more significant if substrate heating
effects are considered.

To compare the two substrate heating mechanism, we
introduce \(P = xP_0\), in which \(P_0\) the inherent pressure in
the film and substrate when there is no deposition and \(x\)
is a positive valued coefficient (\(x \geq 1\)). The repetition
rate of the femtosecond laser is high (\(=80\) MHz) so the
pressure \(P\) can be assumed to be quasi-continuous. We
also introduce \(P_e = \beta P_1\), in which \(P_1\) is the vapour pres-
FIG. 5. Surface topography of films subjected to in-situ heat treatment with temperatures of (a) 300°C, (b) 450°C and (c) 600°C as well as films subjected to post deposition heat treatment with temperatures of (d) 400°C, (e) 500°C and (f) 600°C. The dimension of each image is 10 μm × 10 μm. The scale bar at the bottom of (c) indicates the height of films subjected to in-situ heat treatment and the scale bar at the bottom of (f) indicates the height of the films subjected to post deposition treatment.
heat treatment. The results are shown in Fig. 6(b), measured as the root-mean-square of the height of the deposited film for both heat treatments. Different substrate heat treatment has varied effects on the properties of the film. It is therefore an effective engineering method to improve the quality of the film.

IV. CONCLUSION

We demonstrated the pulsed laser deposition of Nd:YAG laser crystal on Si substrate using femtosecond Ti:Sapphire laser. Nanoparticles are observed in the surface of the thin film. The crystallinity of the deposited Nd:YAG target can be improved by heat application, as manifested by the increased number of reflecting planes. Clustering and coalescence of the heat treated deposited particles are observed in the scanning electron micrograph, since the adatoms receive additional energy from the heater. The effects of the in-situ and post deposition substrate heat treatment in the deposited film have also been investigated. The results showed that flattening of the thin film is more apparent at post deposition heat treatment than in-situ substrate heat treatment due to the volume free energy of the film. Both heat treatments are found to be effective measures on engineering the surface topography and roughness of the deposited film in the microscopic level.

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