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Pulsed laser deposition of crystalline garnet waveguides at a growth rate of 20 μm per hour

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A B S T R A C T

We report pulsed laser deposition of high-quality crystalline yttrium aluminium oxide and yttrium gallium oxide with a growth rate approaching 20 μm per hour by using an excimer laser operating at a repetition rate of 100 Hz. This result demonstrates the capability of PLD at 100 Hz for upscaling deposition speeds to a rate that is industrially relevant. In addition, we show that use of this high repetition rate can cause additional heating of the substrate, which in turn affects the film composition. This effect is used as an additional control parameter on the composition, and thus refractive index, of the grown material.

1. Introduction

Pulsed laser deposition (PLD) is a technique that uses laser pulses to ablate a target to form a plume of material which is then transferred to a substrate positioned nearby. PLD offers a variety of control parameters that can affect the thickness, quality and composition of the deposited material. As an example, the target composition, laser fluence and substrate temperature have been shown to affect the phase, stoichiometry and degree of crystallinity in a grown film. PLD has been used to deposit and grow a variety of materials, such as metal oxides [1], ferromagnetic materials [2], glasses [3] and, as discussed further in this work, crystalline optical materials [4,5].

Crystalline optical materials grown via PLD, have been shown to have very low propagation losses down to values of ~0.1 dB/cm [6], though one limitation on growing optical films with desired properties is the ability to obtain the correct stoichiometry. However, as demonstrated previously, by pre-compensating a target to counteract specific elemental losses during transfer or evaporation during growth on the substrate, stoichiometric deposition can be achieved [7].

Optical films made of garnet materials such as yttrium aluminium oxide ($Y_3Al_5O_{12}$/YAG) are useful as solid-state laser hosts owing to their properties of high thermal conductivity and being optically isotropic [8]. In particular, erbium-doped yttrium gallium oxide ($Y_3Ga_5O_{12}$/YGG) is of interest owing to its spectroscopic properties, which are desirable in applications for both CO_2 and methane gas detection in LIDAR (LIght Detection And Ranging) systems [9–11]. One hurdle for PLD being used as an industrial and commercially viable process for films of thickness > 10 μm, is the rate of deposition. In this work, we present the hetero-epitaxial growth of single-crystal garnets on ⟨100⟩-oriented YAG substrates via pulsed laser deposition using laser repetition rates from 20 Hz up to 100 Hz.

In this paper, we show that high-quality single-crystal garnets can be grown by PLD at a deposition rate of 20 μm per hour, and that, the increase in number of pulses when operating at 100 Hz contributes to heating of the substrate, which in turn affects the composition of the material in the grown film.

2. Experimental setup

Using a PLD setup discussed in previous work [12], laser pulses of ~20 ns duration from a Coherent COMPexPro 110 excimer, operating at 20–100 Hz, were focused into a stainless-steel vacuum chamber, which was back filled with oxygen, onto a target material at a pulse energy of 100 mJ. The focused laser pulses resulted in a fluence of 1.1 J cm^{-2} on the target, which was rotated by an offset cam using a DC motor, in order to increase utilization of the surface area. The targets were 5-mm thick, 50-mm diameter, hot-pressed ceramic discs consisting of YAG, Er(1%):YGG or YGG. We also added an additional 2.5% Al_2O_3 to the YAG target, 9.4% Ga_2O_3 to the Er(1%):YGG target, and 12.7% Ga_2O_3 to the YGG targets, in order to compensate for loss of Al and Ga respectively that routinely occurs during deposition.

The ablation plume produced was incident on surface-polished ⟨100⟩-orientated YAG substrates, of dimensions 10 mm × 10 mm × 1 mm, placed 6 cm away from the target. To enable crystalline growth of the

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deposited material, the substrates were heated using the output from a CO_2 laser (10.6 μm wavelength (IR), with a maximum output of 38 W), which was incident on the rear surface of the substrate [13]. The CO_2 laser beam had a 'top-hat' spatial intensity profile, produced using a ZnSe tetraprism. This method has been routinely used to minimize both undesirable heat sinking and the total power required to heat the substrates, thereby avoiding undesirable vacuum chamber heating [14]. The substrates used here were heated by a CO_2 laser power of 17.5 W, corresponding to a substrate temperature of ~600 °C, before deposition. All the garnet films, grown under the same conditions, though at different repetition rates of 20 Hz and 100 Hz for YAG and Er(1%):YGG, and 20, 40, 60, 80 and 100 Hz for YGG, were compared. The number of pulses for each deposition was kept constant at 36000, which corresponded to ~2 μm-thick films, sufficient for reliable X-ray diffraction (XRD) analysis, which was carried out using a Rigaku Smartlab X-ray diffractometer and Bruker D2 Phaser.

3. Results

3.1. Depositions at 20 Hz and 100 Hz

3.1.1. Crystallinity

X-ray diffraction spectra of the YAG and Er(1%):YGG films grown at 20 Hz and 100 Hz are shown in Fig. 1. The signal corresponding to the (400) peak from the YAG substrate at ~29.76°, allows us to correctly determine the location of the (400) YGG and (400) YAG 2θ peaks from the grown samples, by making sure the (400) YAG 2θ peak from the substrate lines up to its database value of 29.76° [15]. Since the erbium dopant concentration is only 1% of the yttrium atoms in Er(1%):YGG, we use the (400) YGG peak as a point of reference for simplicity. The FWHM values of the peaks are similar when grown at 20 Hz compared with 100 Hz for both YAG (0.080° and 0.065°, respectively) and Er(1%):YGG (0.036° and 0.031°, respectively) films. For both materials, the 100-Hz XRD peak has a smaller 2θ value than the 20-Hz peak. This is attributed to the reduced Al and Ga content of the respective films, as already noted in [16], which occurs as a result of evaporation from the hot substrate. However, we do not think that the trend observed here for a reduced FWHM value for films grown at 100 Hz is systematic, and at this stage do not have any reason to suspect improved crystal quality for such a higher repetition rate growth.

3.1.2. Surface analysis

Measurements using a Zescope optical profiler to determine the number of particles of height > 20 nm over a 100-μm × 100-μm area were taken along with roughness measurements to analyze surface quality, while the surface profiler enables the thickness of the materials to be determined. As an example of the surface morphology measurements taken of the films, Fig. 2 displays a 1-dimensional slice of the 2-dimensional optical profile data taken of the surface of YAG films grown at 20 Hz and 100 Hz, showing the greater surface roughness of the film grown at 20 Hz compared with 100 Hz. Table 1 displays a comparison of the particulate count, roughness and thickness of YAG and Er(1%):YGG films grown at 20 Hz and 100 Hz. As seen from the table, for both targets, the number of particulates is fewer and the roughness is lower for films grown at 100 Hz, compared with the films grown at 20 Hz. The thickness of the samples is ~2 μm for all samples, indicating that (for the same total number of pulses) thickness is repetition rate independent. It can also be seen that growth rates of ~20 μm per hour are achievable at 100 Hz.

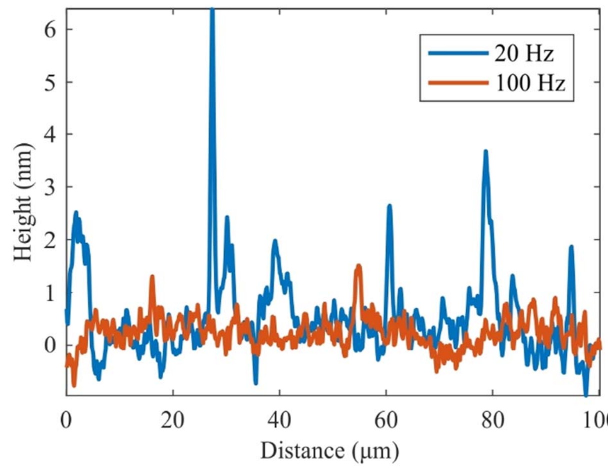

Fig. 2. A 1-dimensional slice of the 2-dimensional optical profile data of the surface of YAG films grown at 20 Hz (blue solid-line) and 100 Hz (orange solid-line). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3.2. Repetition rate analysis

To confirm the trend in higher deposition rate leading to a systematic decrease in 2θ, depositions were carried out at 20, 40, 60, 80 and 100 Hz. In this instance, we used an undoped YGG target, but with

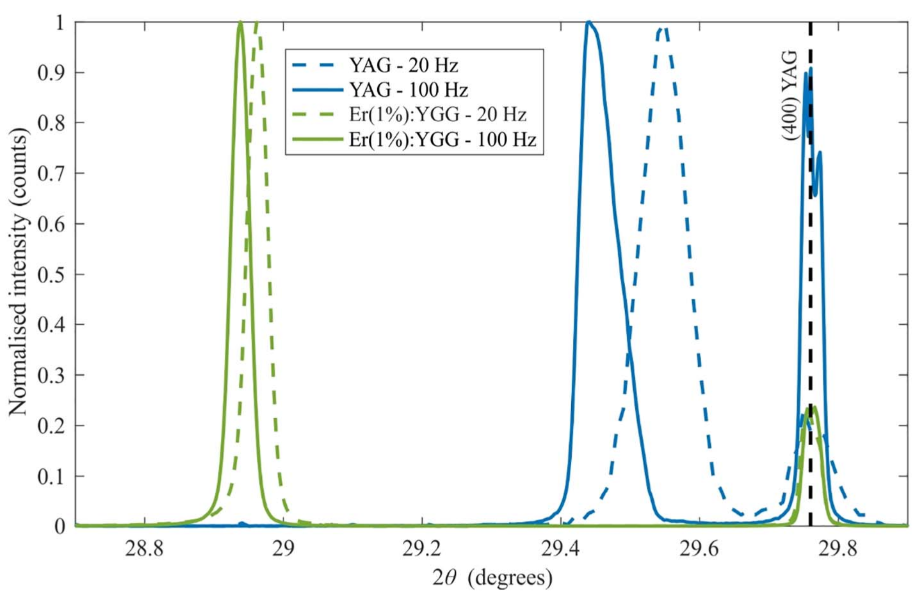

Fig. 1. XRD spectra of YAG film grown at 20 Hz (blue dashed-line) and 100 Hz (blue solid-line), and Er(1%):YGG film grown at 20 Hz (green dashed-line) and 100 Hz (green solid-line). The black dashed-line indicates the 2θ database value of the (400) YAG peak. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)