Hybrid garnet crystal growth for thin-disc lasing applications by multi-beam Pulsed Laser Deposition

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Abstract Composite crystals are of great interest for side-pumped thin-disc laser applications. We present a novel technique for fabricating hybrid films for such applications based upon multi-beam, multi-target Pulsed Laser Deposition (PLD). A two-garnet example was grown as proof of concept, representing the first known demonstration of controlled, continuous horizontal grading in a PLD-grown crystal. The results of shadow masking experiments and the suitability of such a method for growth of the desired structures are also discussed.

1 Introduction

A thin-disc laser crystal is typically 100-300μm thick and millimetres in diameter, highly reflective on the back side and bonded to a heat-sink. The chief advantage over bulk crystal lasers is that heat dissipation is essentially one dimensional due to the high surface-to-volume ratio. High pump power densities can hence be achieved whilst thermal effects (e.g. lensing) remain much reduced, resulting in a laser with high output power but excellent beam quality [1]. Such discs have traditionally been quasi-end pumped, requiring a complicated parabolic mirror arrangement. Side or edge pumping, a recent variant, drastically simplifies the setup [2–4].

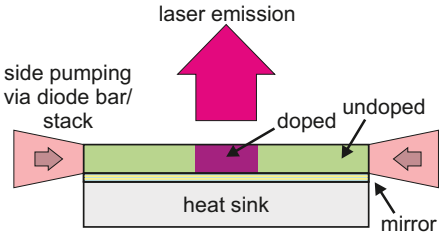

Fig. 1: Schematic of diode side-pumped composite thin-disc laser.

Composite crystals, with dopants confined to the centre of the disc only (such as that illustrated in Figure 1), may be preferred over single material samples, both to simplify the pumping setup by guiding pump light to the core and to reduce the effect of amplified spontaneous emission [2,3]. However, fabrication of such structures can be slow or laborious and structures may be

vulnerable to bending or breaking at high temperatures. A single all-crystal film should not be subject to such problems, particularly if growth could be achieved in one step. Quick, simple growth of appropriate materials (such as mixed garnets) via Pulsed Laser Deposition (PLD) has been demonstrated on a number of occasions [5,6], making this method ideal for further investigation.

In preparation for the growth of hybrid crystals, two approaches have been developed to confine materials horizontally: shadow masking and cone growth. The latter was found to be superior in the context of side-pumped laser crystals due to the lack of a distinct boundary between doped and undoped areas, resulting in the first known example of controlled, continuous horizontal grading in a PLD-grown crystal. However, physical masking may still be of interest for other applications.

2 Shadow masking

A mask or stencil is applied to the substrate. Doped material is deposited through this mask. The mask is then removed to leave a feature that may then be overgrown with undoped material. This method is simple but requires a masking material that can be both accurately machined and applied to the substrate without melting or indiffusing at the temperatures required for crystalline growth. A negative masking approach may also be taken: material is deposited through a mask without heating, forming an amorphous negative mask. Subsequently deposited material grows crystalline on the bare substrate only, and amorphous material can be removed by etching. In the context of garnets, however, this approach was found to be unsuitable due to the similar etch rates for amorphous and crystalline material. Growth of nanoscale epitaxial oxide structures by PLD through a mask has been demonstrated previously, with SiN or alumina membranes favoured as masking materials [7–9].

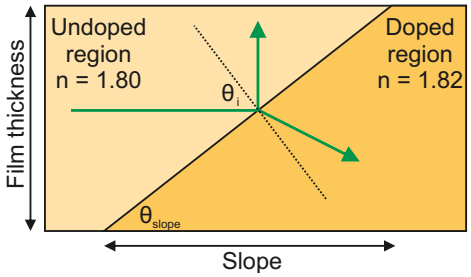

Fig. 2: Diagram of model used to estimate Fresnel reflection of side pumped light (green) due to the interface between areas of higher and lower refractive index.

Any sloping interface between doped and undoped material will result in Fresnel reflections of any side-pump beam (see Figure 2). Such an interface is hard to avoid due to the spreading of the plume beyond the mask, so close mask-substrate contact is crucial to increase the slope angle θ_{slope} and reduce reflections.

2.1 Experiment

Laser machined silicon was chosen as the most suitable masking material. Silicon's low thermal conductivity (desirable to prevent heat sinking) and a high melting point (~1400 °C) are desirable for masking. Wafers thin and flat enough to form a close mask, but strong enough to withstand machining, are readily available.

25 μm thick silicon wafers were laser machined into 10×11 mm rectangles, with central holes of 3 mm diameter. To allow substrate heating and to prevent contamination from adhesives, methanol bonding [10] was the chosen as the method of applying the mask to the substrate. The (100) orientated single-crystal YAG substrate was cleaned, placed in a dish and covered in methanol. The mask was applied and the dish left on a hotplate at 50 °C until the methanol had evaporated (2-3 days). Any residue on the substrate was ignored as it was unlikely to survive heating to deposition temperatures. The mask was easily removed post-deposition using a solvent.

Films were grown using the setup illustrated in Figure 3. A frequency-quadrupled Nd:YAG laser of wavelength 266 nm and fluence ∼1 J/cm^2 ablated a rotating single-crystal $Gd_3Ga_5O_{12}$ (GGG) target. Laser repetition rate was 10 Hz and ambient oxygen pressure during deposition was 4×10^{-2} mbar. The plume-substrate angle of incidence was varied over a range of 8° in order to obtain a flat thickness profile and increase target lifetime. Substrates were heated to ∼650 °C via a raster-scanned 10.6 μm wavelength CO_2 laser, and the substrate was rotated to aid uniform temperature growth. Target-substrate distance was ∼ 50 mm. A Zemetrics ZeScope optical profiler and a KLA Tencor P-16 stylus profiler were used for 2D shape and 1D height profiling respectively. X-ray diffraction (XRD) was carried out via a Siemens D5000 powder x-ray diffractometer.

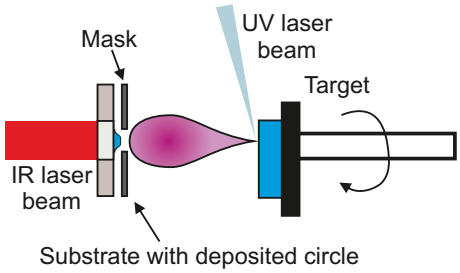

Fig. 3: Schematic of single-plume masked growth.

2.2 Results and discussion

Single-phase crystalline growth of a GGG circle ∼3 mm diameter was achieved; XRD data is shown in Figure 4. Stylus profiling (Figure 5), however, shows that θ_{slope} is small, leading to a potential pump reflection of ∼80% and implying that mask contact was poor. These estimates, calculated using the Fresnel equations, assume a change in refractive index Δn of 0.02 (50% Yb:YAG surrounded by undoped YAG) and a straight edge profile. Mask attrition rates were high: even with careful handling, masks frequently cracked and/or snapped during bonding or substrate loading. Because only small amounts of force could be applied, bonding was also not as strong as was required, with masks coming away from the substrate when in the deposition position.

3 Cone growth

Concerns with mask adhesion led to the move away from shadow masking towards dual-plume cone growth. This technique has many advantages over shadow masking in terms of speed and simplicity: extra equipment is minimal and the number of fabrication steps is reduced.

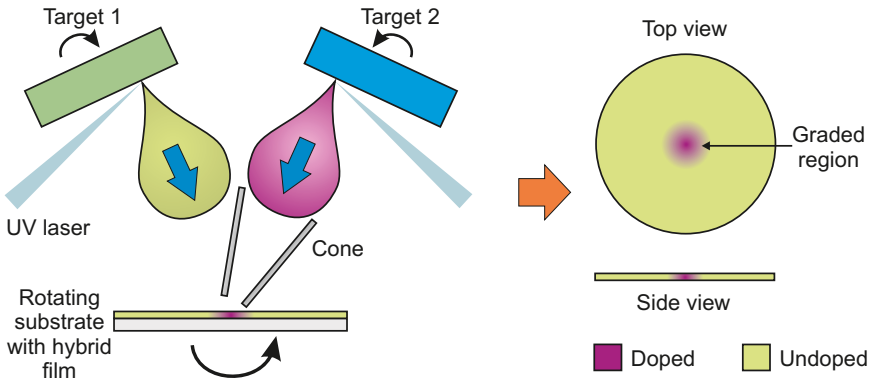

Fig. 6: Schematic illustrating cone growth concept (left) and resulting hybrid structure (right)

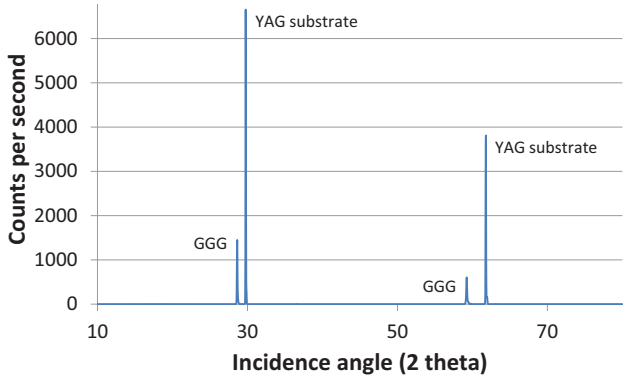

Fig. 4: X-ray diffraction spectrum of GGG circle. (400) and (800) peaks can be observed for film and substrate.

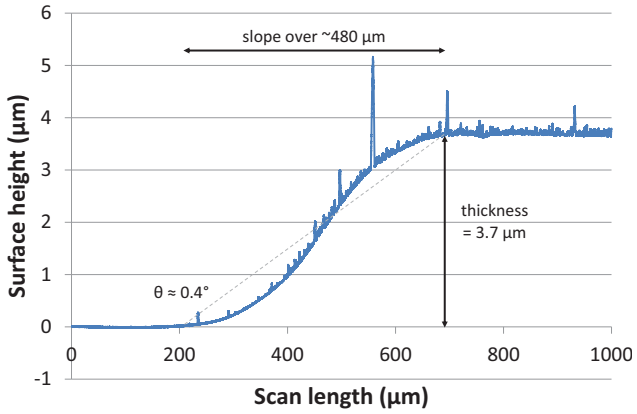

Fig. 5: Stylus profile of the edge of a GGG circle grown via a machined silicon mask showing a θ_{slope} of ~0.4°.

Cone growth also has the advantage of allowing horizontal grading between materials, removing the sloping boundary that leads to significant side pump reflections.

Figure 6 shows a schematic of the concept. A cone is placed in front of a target of doped material and aligned to the centre of a crystal substrate. When the target is ablated the plume is funnelled through the cone and deposition is confined to the centre of the substrate. If a second undoped target is ablated simultaneously and the substrate is rotated, a hybrid crystal will be formed consisting of doped material in the centre, undoped material around the outside and a graded interface between.

3.1 Experiment

Stainless steel cones (such as those used for cake frosting) were used for the proof of principle experiments detailed in this paper. They are cheap, easy to obtain and available with apertures of a number of sizes and shapes. A cone of 2.3 mm internal diameter aperture was used for these experiments.

An excimer laser was used to ablate a YAG target, with the resulting plume funnelled through the cone, while a Nd:YAG laser simultaneously ablated an undoped GGG target. Fluences in both cases were ~2.5

Jcm^{-2} and the ambient background gas pressure was 1 $\times$ 10^{-2} mbar. Targets were rotated but kept at a constant angle of 24°; the cone was held perpendicular to the target and hence was not perpendicular the substrate, as can be seen in Figure 6. In order to include the cone without blocking either ablating beam, the minimum target-substrate distance was found to be 50 mm, including a ~1 mm gap between cone edge and substrate to prevent heat sinking. While not appropriate materials for a functioning thin-disc laser, YAG and GGG were chosen for this proof of principle experiment as they were known to grow well as separate and mixed garnets but were dissimilar enough for relative concentrations to be distinguished via energy dispersive X-ray analysis (EDX). EDX was carried out using an Oxford Inca PentaFet-x3 EDX analyser and a Zeiss EVO-50 scanning electron microscope (SEM).

3.2 Results and discussion

The resulting structure consisted of a GGG outer region, mixed GGG-YAG centre and graded interface. This represents, to our knowledge, the first instance of smooth horizontal grading via PLD. Both plumes were directed towards the centre of the substrate; the combination of this and plume spreading make some mixing in the centre unavoidable. However, in the context of a doped-undoped hybrid crystal, such mixing would not present a problem; doping levels in the cone-funnelled plume should merely be increased to compensate.

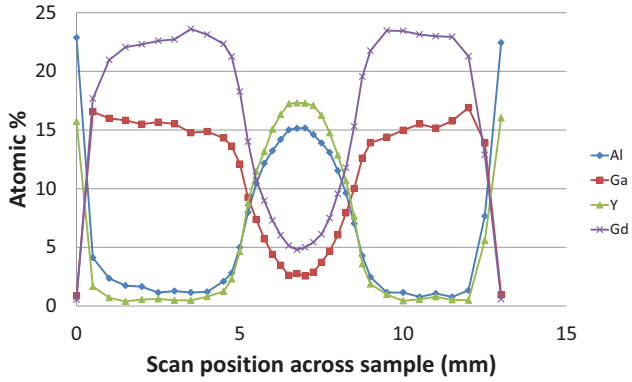

Fig. 7: EDX scan across dual-plume cone grown sample, showing the grading from GGG to mixed YAG-GGG.

The smooth grading from GGG into mixed YAG-GGG can be seen in Figure 7. Gadolinium and gallium atomic percentages drop from ~23 and ~15% to ~5 and ~3% respectively, while yttrium and aluminium atomic concentrations increase correspondingly. GGG growth is far from stoichiometric, a consequence of the compromised growing conditions required to grow GGG and YAG simultaneously. The grading can also be observed via XRD, as shown in Figure 8: a contribution from the graded and central areas can be observed between 27.8 and 28.7°, as can a sharper peak corresponding to the GGG-only outer region at ~27.6°. The total diameter of the mixed area including the graded region is ~5 mm, demonstrating the wide spread of the funnelled plume after exciting the cone. The small amount of yttrium and aluminium detected outside the central region is likely a contribution from the substrate.

The mixing of plumes in the centre resulted in a circular raised feature, which can be removed by polishing. Alternatively, the size and shape of such features could

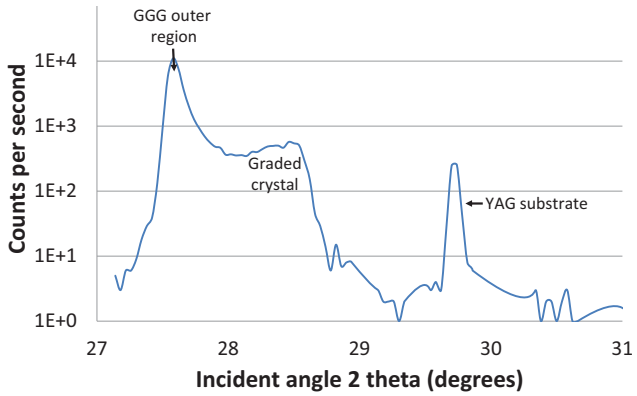

Fig. 8: XRD spectrum of cone grown hybrid crystal. Peaks corresponding to the non-stoichiometric GGG outer region and YAG substrate can be observed, along with a contribution from the central and graded areas.

be controlled by choice of cone size, shape and tip angle, as well as by changing relative growth rates.This presents an intriguing possibility in the field of micro- and integrated optics: cone growth has the potential to allow fabrication of crystalline microlenses, similar to amorphous examples demonstrated by CW laser effusion [11].

4 Next steps

Yb:GGG should prove a more useful cone growth material as, unlike Yb:YAG, GGG has been proven to ablate well under both excimer and Nd:YAG radiation [5, 6]. Ambient pressure can be increased for GGG-only growth, which is expected to result in stoichiometry closer to that of bulk as well as the higher growth rates required for laser crystal growth. Outer region growth rates could also be improved by introduction of a third target and laser to deposit additional undoped GGG. The effect of the various stress states in the crystal (due to the hybrid nature of the crystal, the uneven profile and any differences in stoichiometry compared to bulk), and any subsequent restrictions on film thickness or growth rate, must also be investigated before a practical crystal can be grown and tested.

5 Conclusions

Two methods of horizontal growth confinement in PLD have been investigated. Shadow masking was somewhat successful, with crystalline GGG features grown on YAG. However, plume spreading after passing through the mask resulted in a sloping edge, which would lead to significant pump reflection in a laser crystal.Consequently, a second approach was taken, utilising a stainless steel cone in a multi-beam, multi-target PLD setup. A hybrid crystal was grown using this technique as a proof of principle, consisting of a mixed YAG-GGG core with GGG surrounding and a graded interface between, the first example of such a hybrid structure grown by PLD.

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