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**The effect of particulate density on performance of Nd:Gd<sub>3</sub>Ga<sub>5</sub>O<sub>12</sub> waveguide lasers grown by Pulsed Laser Deposition.**

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**Abstract:** We have successfully grown a range of waveguiding layers of Nd:Gd<sub>3</sub>Ga<sub>5</sub>O<sub>12</sub> (Nd:GGG) on Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> (YAG) by pulsed laser deposition for purposes of studying the effects of particulates on waveguiding and lasing performance. We have found that particulates have a detrimental effect on lasing threshold for the range of particulate densities studied, and can increase lasing thresholds from as low as 2.5mW up to 167mW. We have also shown that the detrimental effect of particulates in waveguides becomes less significant with increasing waveguide thickness.

## Introduction.

Pulsed laser deposition (PLD) has always been an attractive method of fabricating waveguides. The ability to quickly and easily grow stoichiometric, epitaxial films of a large variety of laser media makes it an almost ideal fabrication process<sup>[1]</sup>. PLD has been avoided as a *preferred* method, however, due to the large number of particulates that can occur on the waveguide surface during growth. These particulates, which originate from the target, are believed to occur, in the growth of dielectric films, through two main processes<sup>[1]</sup>.

The first is due to incomplete vaporisation of the target area by the ablation process itself. The area directly surrounding the vaporised material melts and is subsequently expelled due to the recoil pressure of the plasma plume shock wave. Liquid droplets of target material consequently form particulates on the growing film. Increasing the density of the target material can minimise this process of particulate production. Single crystal targets therefore are the optimum material for good film fabrication compared to alternatives such as compressed ceramic targets<sup>[2]</sup>. Alternatively, use of a femtosecond ablation source has also been considered as a potential way of avoiding particulates by this route<sup>[3]</sup>. The ablation process in femtosecond lasers is a non-thermal process, and therefore problems associated with melt expulsion do not occur. To date no multi-component materials (beyond simple oxides and nitrides<sup>[4]</sup>) have been deposited using femtosecond PLD and it is uncertain whether the stoichiometry of a complex target material can be reproduced in the film. Additionally, deposition rates appear to be very low, so growth of films of typical waveguide dimensions (few  $\mu\text{m}$ ) within an acceptable time scale may be difficult.

The second process, termed exfoliation is due to poor surface quality of the target. As the ablation laser pulse continually hits the target area, thin structures of  $\mu\text{m}$  dimensions can develop on the target, pointing towards the direction of the laser radiation, due to a shadowing effect<sup>[5]</sup>. These can eventually break off and form particulates on the film surface. Rotating the target and constantly changing the direction of rotation or re-polishing of the target after use can reduce, but not eliminate, this effect. There have also been many other inventive attempts to remove particulates from the plasma plume<sup>[6-10]</sup> however these have often involved complicated set-ups or sensitive triggering equipment and may be considered to be generally impractical.

There has been no direct correlation however, between the number of particulates seen on the films and the losses generated when used as a waveguide. In this paper we report on the systematic comparison between lasing threshold observed in waveguides of differing thickness and varying particulate density.

### **Waveguide fabrication**

Using conditions similar to those reported earlier<sup>[2]</sup>, we have grown pulsed laser deposited films from a 1 at % Nd doped GGG target on a (100) YAG substrate. The XRD data shown in fig.1 confirms the crystallinity of the films and shows growth in the (400) Nd:GGG direction parallel to the (400) YAG peak.

Films have been grown at thicknesses of order  $2\mu\text{m}$ ,  $4\mu\text{m}$  and  $8\mu\text{m}$ . A piezoelectric gas valve was used to modify the particulate density in the film during growth<sup>[9,10]</sup> as shown in figure 2. Opening of the gas valve (backed with  $\text{O}_2$ ) ejects a beam of

molecular oxygen in a direction perpendicular to the expanding plasma plume. The particulates contained within the plasma plume have a significantly lower velocity than the fast neutral and ionic species assumed to contribute to good stoichiometric film growth<sup>[9]</sup>. The firing of the ablation laser is triggered from the valve such that the beam of molecular oxygen is coincident with the particulates in the ablation plume. The particulates are deflected, by collision with the oxygen, away from the substrate.

A fast rise time of the oxygen pulse and, consequently, the gas valve opening time are critical for the molecular beam to interact with only the slower, detrimental, particulates and not interfere with the faster neutral and ionic species. Gas pulse rise times on the order of  $\sim 200\mu\text{s}$  are required, and for this reason we chose a piezoelectric gas valve (highly modified Maxtek MV-112). Solenoid gas valves can achieve equally fast rise times, but at far greater expense. Furthermore, driving circuits for piezoelectric gas valves are trivial to build compared to those required for solenoid devices.

The gas valve nozzle was placed 2 mm below the ablation area. The close proximity of the nozzle and the ablation area is to maximise the probability of collision as the density of both the oxygen pulse and the plasma plume are at a maximum. Typical delays between gas valve and laser pulse triggering were around 1ms. The backing pressure of oxygen on the valve was 2 atm. The piezoelectric valve was also used as the oxygen source for reactive PLD, therefore the valve was kept open for 3ms to achieve an oxygen pressure in the chamber of  $\sim 5$  Pa during ablation.

Fig.3 shows the surface of two films grown with and without the gas-jet interaction with the plasma plume. Fig.3a shows high particulate density when the film is grown with no gas-jet interaction with the plume, while fig.3b shows appreciably lower particulate density when the gas jet is directed into the plume.

We have grown films with particulate densities that vary between  $10^4$  and  $10^7$  particulates/cm<sup>2</sup>. This large difference in particulate densities may not be entirely due to the gas jet and can be attributed in part to the increased degradation of the target and increased exfoliation. XRD spectra, shows a slight broadening of the (400) GGG peak with increased particulate density, implying a slight degradation in crystallinity. Figure 4 shows the increase in the FWHM of the (400) GGG peak in 4 $\mu$ m thick waveguides with increased particulate density. Figure 5 shows typical particle size distributions for films containing the high and low particulate densities. All films show an approximately normal distribution of particle sizes.

The films were cut back to 2mm in length, corresponding approximately to a 1/e absorption length for the pumping wavelength of 808nm, and polished to optical flatness on both ends. Horizontal and vertical parallelism was maintained between the end faces by a laser collimator.

#### **Threshold results for waveguide lasing.**

To confirm spectroscopic properties, the waveguides were pumped using an Ar<sup>+</sup> pumped Ti:sapphire laser capable of delivering 600mW at 808nm, matching the strongest absorption band of Nd:GGG<sup>[11]</sup>. Light was coupled into the active layer using x25 objectives for the 2 $\mu$ m and 4 $\mu$ m guides and x16 for the 8 $\mu$ m guide. Figure

6 shows the fluorescence spectra of the Nd doped waveguides. This agrees with previous results<sup>[2]</sup>, showing peak fluorescence around 1060nm. Fluorescence lifetime was also measured to be 260μs, in close agreement with previous results and the bulk crystal value.

Two plane dielectric mirrors were carefully attached to the end faces using Fluorinert FC-70 fluorinated liquid. Both mirrors were high reflectivity (>99.5%) mirrors at the signal wavelength. The typical lasing spectra of waveguides examined are shown in figure 7, and consists of a broad emission over ~3nm. Multiple peaks (splitting) can be seen due to etalon effects within the dielectric mirrors. The output profile at the lasing wavelength was single mode for all waveguides, regardless of how many spatial modes could be supported within the waveguide.

The lasing threshold was measured for films of varying thickness and particulate density. Figure 8 shows the threshold recorded for the three different thickness films. It can be seen that the high particulate density has a detrimental effect on the lasing threshold of the film. This is largely due to the particulates acting as scattering sites for light within the guide, which consequently increases the loss in a waveguide. As can be seen from the equation.<sup>[12]</sup>

$$P_{th} = \left( \frac{\pi \hbar \nu_p}{2 \sigma_e \eta_p \tau_f} \right) \sqrt{(W_{lx}^2 + W_{px}^2)} \sqrt{(W_{ly}^2 + W_{py}^2)} L$$

$\hbar$  = Plank's constant,  $\nu_p$  is the pump laser frequency  $\sigma_e$  is the stimulated emission cross section,  $\eta_p$  is the pump quantum efficiency (assumed as 1),  $\tau_f$  is the fluorescence lifetime,  $W_l$  and  $W_p$  are the average spot sizes for the laser and pump beam

respectively in the guided (x) and unguided (y) direction, and L is the single pass exponential loss in the guide.

From the equation above it can be seen that lasing threshold is expected to increase linearly with loss. Using measured spot sizes and threshold values we have estimated the loss in each guide. This is plotted against particulate density in fig 8. It can also be seen that the effect of the particulates has a far more dramatic effect on those guides with increased confinement. An increase in particulate density from  $3.5 \times 10^4$  to  $9.7 \times 10^5$  particulates/cm<sup>2</sup> led to an increase in absorbed power threshold from 2.5mW to 167mW. A 4μm thick guide with a particulate density of  $3.2 \times 10^6$  particulates/cm<sup>2</sup>, however, lased easily at 19mW where a 2μm thick waveguide would be unable to lase. Even an increase up to  $1 \times 10^7$  particulates/cm<sup>2</sup> (a very poor quality film) would still lase still at around 60mW. The 8μm thick waveguide showed good waveguide performance regardless of particulate density, lasing at 2.2mw and 12.2mw with particulate densities of  $5.5 \times 10^4$  cm<sup>-2</sup> and  $6.6 \times 10^6$  cm<sup>-2</sup> respectively, a change in particulate density of over 2 orders of magnitude.

The loss mechanism due to scattering in waveguides from particulates is believed to be largely due to surface scattering, as opposed to scattering from within the guide, and the increased effect of particulates on the more confined waveguides (2μm thick) is possibly an example of this effect. As the particulates in these guides are of order of the same size as the guide itself then there is a higher probability of surface scattering, as most particulates will protrude from the waveguide surface. In the larger guides, however there is more chance that the particulates will be enclosed within the guide and therefore, not contribute as significantly to losses.

The slight decrease in crystal degradation with increasing particulate density will also contribute to the increase in threshold. This may account for the observed non-linear increase of threshold power with particulate density.

It must be noted that the particulate density counts given in this paper are taken from an optical microscope using particulate analysis software and so represent the particulate count throughout the entire guide per  $\text{cm}^2$ . Many reports take particulate counts from SEM data, which can only yield particulate counts for particulates that extend beyond the surface.

## **Discussion**

These results suggest that were particulates to remain an intrinsic problem in PLD fabrication of waveguide devices, then the solution is to move to thicker waveguides where the particulate density is not such a critical factor in the performance of the resultant laser. Although this seems a relatively intuitive deduction it is in contrast to the accepted notion that lower thresholds can be attained with more confined waveguides structures.

The fabrication of thicker waveguides, however, is easily compatible with PLD as deposition rates are achievable of up to  $1\mu\text{m}/\text{minute}$ , and there need be no significant increase in fabrication times. High Numerical aperture waveguides are also readily achievable with PLD. Coupling this with larger thickness waveguides make PLD fabricated waveguides an attractive prospect for coupling with high power, high divergence sources. Finally it should be noted that proximity coupling is also readily achievable with these devices<sup>[13]</sup>.



## **Summary**

We have shown that the presence of particulates in PLD waveguides increases the losses and lasing thresholds and have analysed this quantitatively. We have also shown that this effect is less pronounced in thicker waveguides. This is believed to be due to surface scattering of light from the particulates being the dominant cause of loss within the guide.

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## **Figure Captions.**

**Figure 1.** X-ray diffraction data for the Nd:GGG films. The YAG (400) line is weak due to the thickness of the sample. No other peaks are observed.

**Figure 2.** Experimental set-up showing piezoelectric gas valve mounted in chamber.

**Figure 3a.** Optical microscopy image of film surface under normal growth conditions.

**Figure 3b.** Optical microscopy image of film surface with gas-jet directed into plume during growth.

**Figure 4.** Increase of FWHM of the (400) GGG peak with particulate density

**Figure 5.** Typical particle size distributions for films with high and low particulate distributions.

**Figure 6.** Typical fluorescence spectrum of a waveguide.

**Figure 7.** Typical lasing spectrum of a waveguide.

**Figure 8** Lasing threshold versus particulate density for films of different thickness.

**Figure 9** Estimated loss versus particulate density for films of different thickness

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