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Yttrium iron garnet surface modification during pulsed laser ablation deposition

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This paper reports on the formation of cone-like structures on yttrium iron garnet (YIG) targets during the ablation of YIG thin films. Energy dipersive X-ray analysis (EDS) performed in the targets show that the cones are rich in yttrium.

Key words: *PLAD; surface modification; YIG*

1. Introduction

Pulsed laser ablation deposition (PLAD) has become a popular technique for the preparation of various kinds of thin films of metals, chemical compounds and organic materials [1]. This technique has many advantages: it can easily produce thin films from materials with high melting points and it can produce thin films with compositions similar to the target material compositions. Despite these advantages, one of the disadvantages is that the ablation rate can be influenced by cone-like structures, which form on the surface of an ablation target during ablation [2]. Dyer et al. [3] studied laser cone formation on polyimide films and showed how seeding with impurities affected their formation. The high-temperature superconductor – yttrium-barium-copper oxide (YBCO) has been extensively studied by Foltyn et al. [4, 5] who suggested that vaporization-resistant impurities are responsible for the cone formation. They demonstrated that the cone tips are rich in yttrium. With incongruent melting compounds like YBCO or Yittrium Iron Garnet (YIG) surface segregation can occur with the resolidification of higher melting components such as Y_2O_3 [6].

This paper reports the study of cone formation on YIG targets due to ablation.

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2. Experimental details

YIG ($Y_3Fe_5O_{12}$) targets were prepared using conventional ceramic methods as reported by Ibrahim et al. [7]. The laser used was a Lambda Physik Excimer (LPX100) XeCl laser (λ = 308 nm and repetition rate – 21 Hz). It hit the target at an incidence angle of 45°. Laser fluence was 1 J·cm^{-2}. During the process of ablation the laser was moved horizontally by wobbling lens.

An optical microscope and scanning electron microscope (SEM) were used to study the surface morphologies of the unablated and ablated YIG targets. X-ray analysis of the targets was done using energy dispersive X-ray analysis (EDS).

3. Results and discussion

Optical micrographs in Fig. 1 show three different stages of cone formation. Shallow ripple-like structures occur at the edge of the ablated area, which become deeper as the laser damage increases and eventually full cones develop.

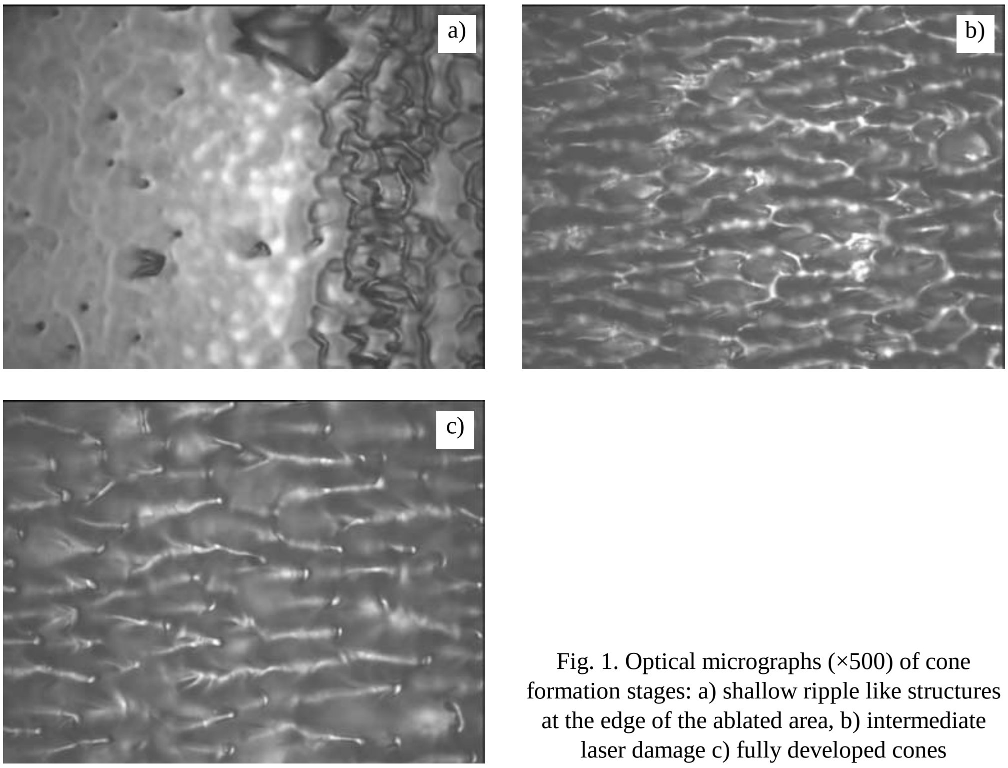
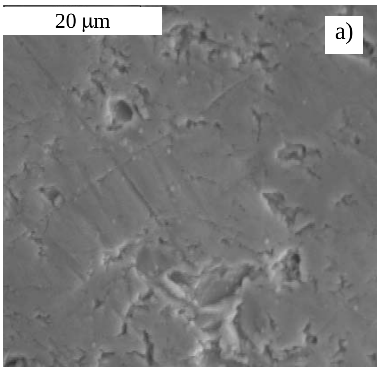


Fig. 1. Optical micrographs (×500) of cone formation stages: a) shallow ripple like structures at the edge of the ablated area, b) intermediate laser damage c) fully developed cones

Figures 2a and b show SEM micrographs of the YIG target before (×1700 magnification) and after ablation (×200 magnification). The surface has cone-like structures approximately 70 μm long and 30 μm wide, and the surfaces of these structures are smooth (the facetted structure is caused by the resolution of the digital photograph and printer). The ablated target was hit with ~30 000 shots per site and heated to 500 °C during ablation. The number of laser shots per site was calculated by multiplying the total number of laser pulses by the ratio of beam area to total exposed area. The cone axes are not normal to the surface; they appear to be lying at an angle. Similar cone structures have been observed by several researchers on YBCO targets [4, 5, 8] and on aluminium [9]. They observed that the cones point towards the laser beam. As the beam in this study is incident at 45° to the target, they would be expected to form at 45° to the surface of the YIG targets. Figure 3 shows a cone under higher magnification. It has a rounded surface with some evidence of flowing, which indicates that the surface has been in a liquid state.

To study the nature of the cones on YIG, EDS analysis was done on the unablated and ablated targets. Figures 4 a and b show the results. They indicate that the ablated target contains more yttrium and less iron (49%Fe_2O_3:51%Y_2O_3) as compared to the unablated target (67%Fe_2O_3:33%Y_2O_3). This yttrium enrichment may be due to the incongruent melting of YIG.



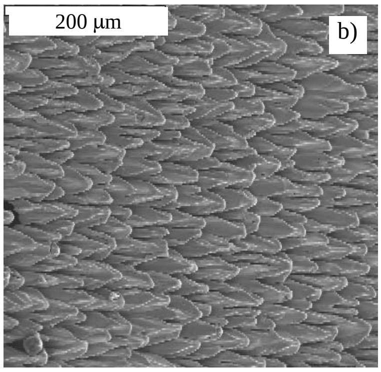


Fig. 2. SEM micrographs of YIG target: a) unablated target, b) ablated target showing cone structures

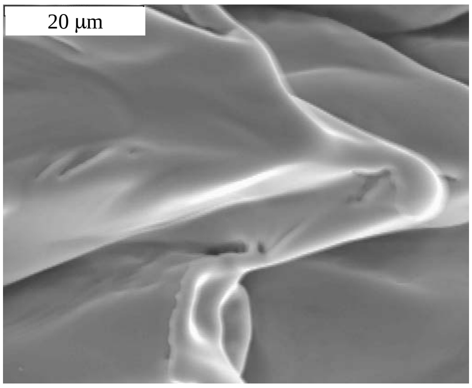


Fig. 3. Close-up SEM micrograph of ablated YIG target

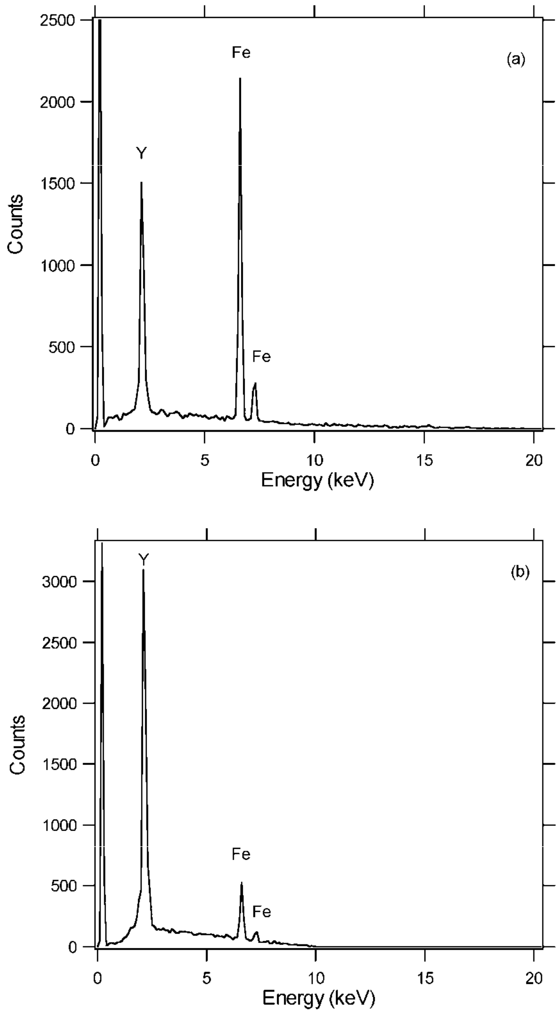


Fig. 4. SEM X-ray analysis of YIG target:
a) before ablation, b) after ablation (~30 000 shots/site)

A similar yttrium enrichment process has also been observed on YBCO targets, which also melt incongruently [4, 5]. The laser rapidly heats the target to form a very high-temperature plasma. Some liquid will be present on the target’s surface, and this will cool between laser pulses and resolidify. As the resolidification process on the YBCO target begins, Y_2O_3, the solid phase with the higher melting point will tend to freeze first, leading to yttrium enrichment [6]. This segregation does not occur significantly in a single ablation pulse, but when the process is repeated thousands of times during an ablation. As Y enrichment proceeds, the laser will begin to interact with the transparent material (Y_2O_3 is used for UV optics) [2]. The Y rich material then shields the underlying material, leading to cone formation and an decrease in the ablation rate.

YIG melts incongruently to form liquid and yttrium orthoferrite ($YFeO_3$). In this case, $YFeO_3$ is the solid phase with the higher melting point, and will tend to freeze first, leading to yttrium enrichment and cone formation. The cones have larger surface area than the original target; the laser's fluence is therefore lowered, leading to a reduction in the ablation rate. This effect was visible during ablation, as there was a noticeable decrease in the size of the ablation plume after approximately half an hour. Foltyn et al. [4] reported a drop in the deposition rate by a factor of 4 after 1000 laser shots per site for YBCO targets. Krajnovich and Vasquez [10] reported that the reduction trend stops when cones have completely formed on excimer irradiated polymers.

As the formation of these cones reduces the film deposition rate, we suggest using an adjustable mirror to move the laser beam to a fresh area of the target whenever there is a noticeable decrease in the size of ablation plume.

4. Conclusions

Cone-like structures and yttrium enrichment processes have been observed on YIG ablation pellets. Cone formation reduces the ablation rate. This is believed to be the first report of cone structures on ablated YIG targets.

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