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CoTi-SUBSTITUTED Ba-FERRITE FILMS PREPARED BY PULSED LASER DEPOSITION

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Abstract- Well oriented CoTi-substituted Ba-ferrite thin films, with c-axis perpendicular to the film plane, have been produced by pulsed laser deposition on (0001) sapphire substrates at 840 °C. These films were of good optical quality and exhibited optical and magneto-optical properties similar to the bulk single crystal material. Furthermore, angular thickness variations caused by narrow plume distributions were studied by depositing onto glass substrates and correlating variations in reflectance with thickness variations through optical interference effects. The surface morphology of the ceramic Ba-ferrite targets after progressive laser illumination was studied by scanning electron microscopy.
KEYWORDS: PULSED LASER DEPOSITION, CoTi-SUBSTITUTED BARIUM FERRITE, MAGNETOOPTICAL PROPERTIES, TARGET MODIFICATION, MATERIAL DISTRIBUTION.

INTRODUCTION

Barium ferrite is of some interest for both short and long wavelengths magneto-optical (MO) recording [1]. Principally, this is because it possesses strong intrinsic perpendicular anisotropy, is resistant to oxidation, and shows large MO effects in the near UV, associated with the Fe ions. In addition, partial substitution of Fe ions with Co gives increased MO effects in the near IR, which can be considerably enhanced by incorporating the material into a properly designed multilayer structure [2]. This is because of the low optical absorption of the material in this region.

We have applied pulsed laser deposition (PLD) to produce CoTi-substituted Ba-ferrite films. In recent years PLD has gained a well established reputation for producing high quality high-T_c superconductors. One of the key advantages of the technique is its ability to evaporate congruently, multi-element targets and subsequently reproduce their stoichiometry on the deposited films, under controlled deposition conditions [3]. However, a major disadvantage of PLD is that the angular distribution of the ablated material is strongly peaked in the forward direction giving rise to extremely non-uniform films. A secondary problem is the texturing of the target surface during the process [4, 5].

In this paper we report on the fabrication of c-axis oriented CoTi-doped Ba-ferrite by pulsed laser ablation onto sapphire substrates. The good quality of the films was confirmed by comparison of the measured complex Kerr rotation spectra with predictions based on the fundamental constants of the bulk single crystal material, determined earlier [6]. First, we give some complimentary results on the laser ablation process, concerning the material distribution in the plume as studied from thickness variations in the deposited film. In addition, the alteration of the surface morphology of the ablated Ba-ferrite targets during laser irradiation and the effect of this on deposition rate has also been studied.

EXPERIMENT

The targets were sintered ceramic discs of CoTi-doped Ba-hexaferrite ($BaFe_{12-2x}Co_xTi_xO_{19}$) with composition parameter x ranging from 0 to 1. The excimer laser beam (KrF λ=248 nm, 30 ns pulse width, 10 Hz repetition rate) was spatially filtered and focused at $\pi/4$ onto the rotating target to yield a laser fluence of 2 J/cm^2. An optical mirror scanning system was used to improve the uniformity of the deposited material and to minimise undesirable surface texturing on the target. The laser ablated material, ejected normal to the target surface, was collected onto 10 x 10 mm^2 single crystal (0001) sapphire (Al_2O_3), heated to a temperature of approximately 840 °C and located 3 cm from the target. The deposition was carried out at an oxygen pressure (P_{O_2}) of 0.1 mbar. Typically 20000 pulses yielded films 300 nm thick.

Angular thickness variations caused by a stationary plume were studied by depositing onto 75 x 25 mm^2 glass substrates at room temperature and correlating variations in reflectance with thickness variations through optical interference effects. The reflectance variations in the films were measured by scanning a laser diode beam over the surface of the film.

The surface morphology of stationary targets irradiated by fixed laser beams was examined by scanning electron microscopy.

A spectroscopic Kerr polarimeter with a rotating analyser was used to measure the complex Kerr rotation of the films.

MATERIAL DISTRIBUTION

Figure 1 represents a typical thickness variation with the angle of observation, parallel to the long axis of the focused laser spot (4 x 1 mm^2) of a Ba-ferrite film. Curves similar to these ones have been previously observed for other compounds [7, 8]. The shape of the curves has been classically explained by the existence of two separate components in the pulsed laser ablation process. At small angles there is a stoichiometric forward peaked component with a very strong angle dependence ($\cos^n\Theta$ with $n>>1$, the precise value depending upon the experimental conditions) representative of the ablation process; and at large angles, there is a non-stoichiometric cosine component ($\cos\Theta$) which can be explained by the thermalisation of the material due to collisions.

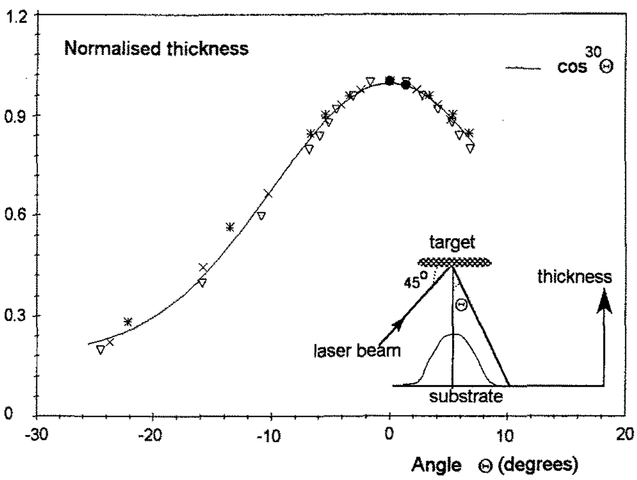

Fig. 1. Normalised angular thickness distributions of Ba-ferrite films deposited from a single irradiated spot of size 4 mm^2 at laser fluence of ~2 J/cm^2.

It was characteristic of our measurements, that an approximate distribution $\cos^{30}(\Theta)$ was observed parallel to the laser spot long axis while a lower order dependence ($\sim\cos^{18}(\Theta)$) was observed perpendicular to it. These narrow distributions have their origin in the profile of the excimer laser beam. Owing to the nature of the beam profile it is not possible to obtain a homogeneous laser spot with constant intensity. The best solution to the problem is the use of a beam homogeniser combined with appropriate scanning of the laser beam.

TARGET MODIFICATION

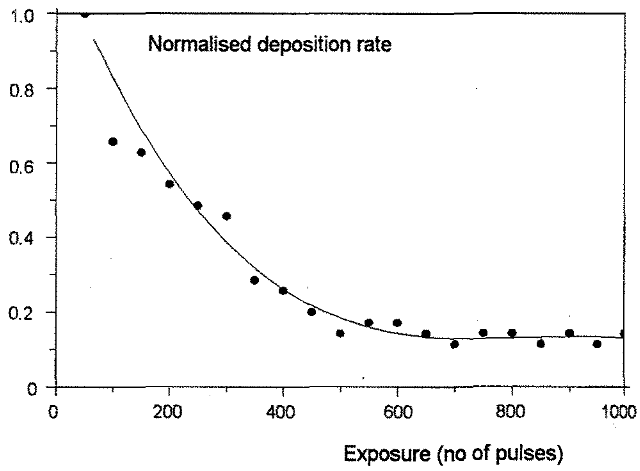

Fig. 2. Deposition rate as a function of laser exposure.

Under conditions used for pulsed laser deposition of Ba-ferrite films, it was observed that there was a noticeable decrease in the size of the plume with increased shot number when the laser beam was hitting the target at the same position. This was confirmed by a quartz crystal monitor which showed that the deposition rate dropped exponentially and attained a steady state only after prolonged exposure (fig. 2). The reason for such behaviour can be found in the modification of the surface by accumulated laser irradiation. Figure 3 shows electron micrographs of the target surface after sequential exposure.

The initial granular surface of the unexposed target has been smoothed by melting after 50 pulses. The observed cracks have been generated by the laser induced shock waves. For 200 pulses the surface has developed small protrusions. At 600 pulses the illuminated area is populated by columnar structures, though the morphology is not the same throughout the whole irradiated site. As a consequence of the Gaussian beam profile the laser energy deposited at the periphery of the crater was less, resulting in columnar surface features which point in the direction of the laser beam. At the centre of the crater, where the deposited energy was larger, the target material was easily ablated, leaving a wavy morphology. At 2000 pulses the above surface effects are more pronounced.

These columns are responsible for the pronounced decrease in film deposition rate. Since the laser beam irradiating a textured surface is spread over a larger area than on a smooth surface, the laser fluence is therefore decreased as is the ablation rate.

It is worth emphasising that the deposition profiles were not symmetric about the target normal. This phenomenon can be explained by the deviation of

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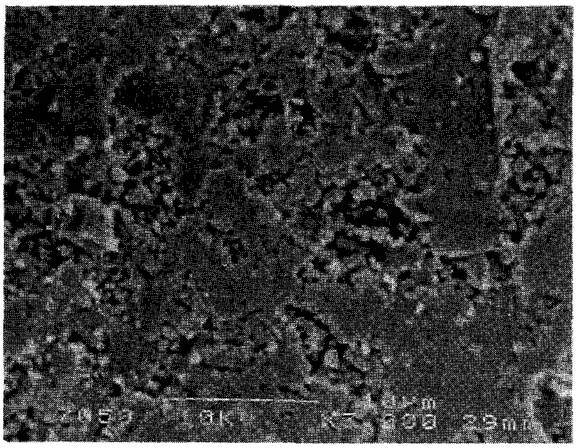
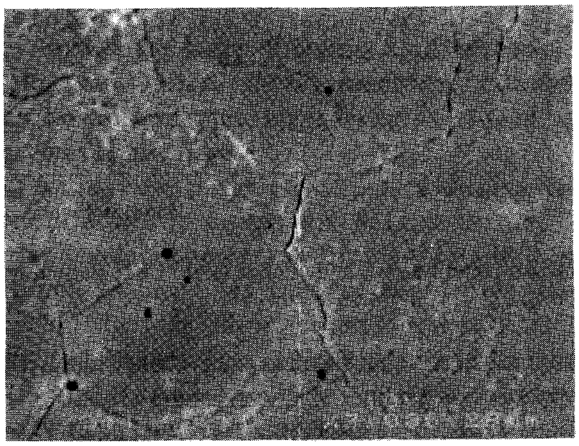
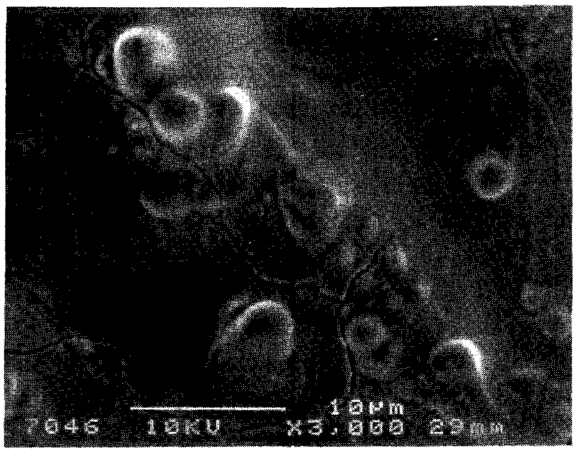


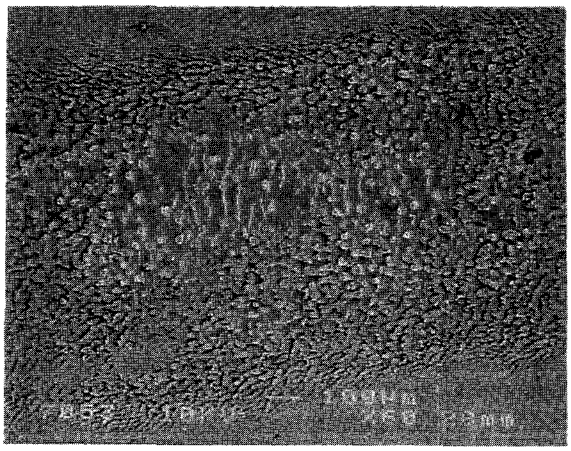
0 pulses (⊢——⊣ 10 µm)



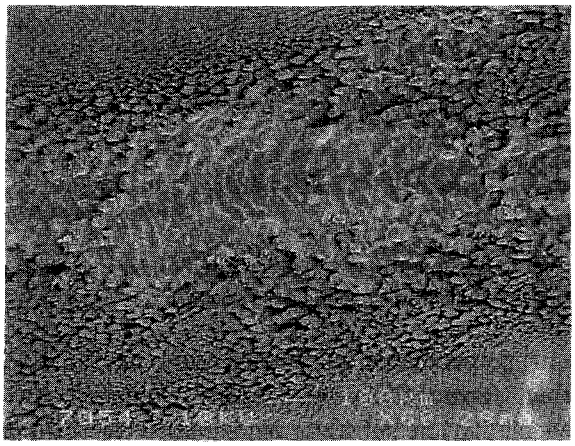
50 pulses (⊢——⊣ 10 µm)



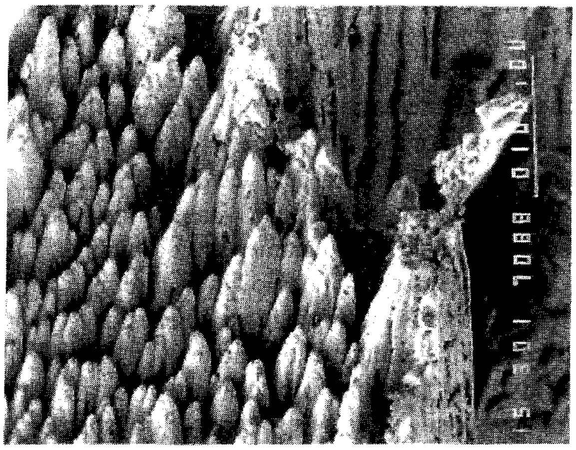
200 pulses (⊢——⊣ 10 µm)



600 pulses (⊢⊣ 100 µm)



2000 pulses (⊢⊣ 100 µm)



100000 pulses (⊢——⊣ 100 µm)

Fig. 3. Modification of the target surface after 0, 50, 200, 600, 2000 and 10000 pulses.

the ablated plume away from the target normal caused by the evolution of the columnar features on the target.

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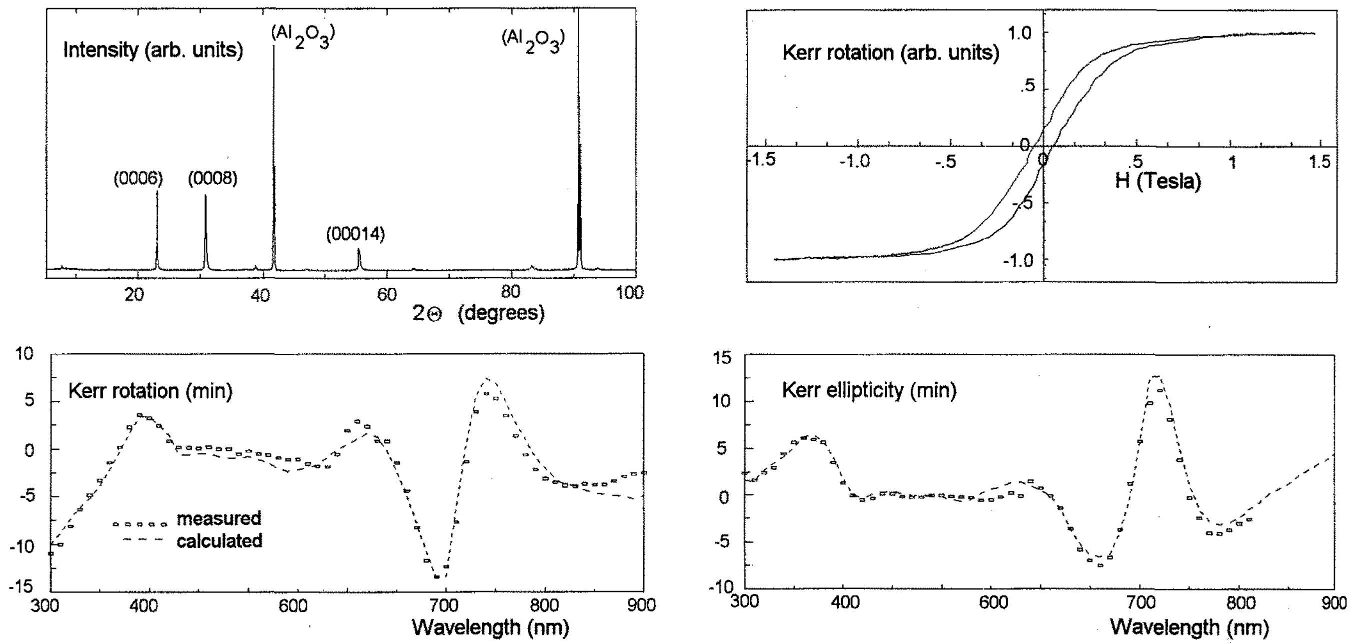

Fig. 4 Complex Kerr rotation , Kerr hysteresis loop and X-ray diffraction pattern of a heavily doped (x=1) CoTi-Barium ferrite film produced by pulsed laser deposition.

C-axis perpendicularly oriented pure and CoTi-doped Barium ferrite films were easily produced by PLD provided that the substrate temperature was kept higher than 830 °C and the laser fluence at about 2 J/cm^2, at the substrate target distance of 3 cm and at the oxygen pressure of 0.1 mbar. A typical XRD pattern of heavily doped film is shown in fig. 4. The coercivity measured from Kerr hysteresis loops was found to range from 0.18 Tesla for pure Ba-ferrite to only ~0.05 T for the heavily doped material. The films were of good optical quality possessing excellent mechanical hardness. The measured MO spectra of the films showed good agreement with predictions based on theoretical calculations using the fundamental constants of the bulk single crystal material [6]. However, a refined reduction of the absorption coefficient was necessary to predict the absolute magnitude of the major Kerr rotation peaks indicating that the film properties are similar to those of the bulk with slightly higher degree of transparency.

CONCLUSIONS

Narrow angular distributions have been observed in the pulsed laser deposition of Ba-ferrite films. The development of columnar features on the target surface due to laser irradiation, is responsible for the pronounced decrease of deposition rate. Oriented CoTi -substituted Ba-ferrite films with c-axis perpendicular to the film plane were produced on single crystal sapphire (0001) substrates by PLD. The optical and MO properties of these films are consistent with the properties of the bulk single crystal material.

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