Physical and magnetic microstructure of rapid thermally annealed thin film bismuth-doped garnets

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Bismuth-doped iron garnets are promising materials for blue light magneto-optic recording media. These materials have been deposited on glass substrates with bulk magnetic and magneto-optic properties suitable for magneto-optic recording. However, these films exhibit a very low carrier to noise ratio (CNR) when crystallized in conventional furnaces. The low CNR is due to the inability to write circular domains in these materials. Rapid thermal annealing (RTA) has been reported to produce films with a much finer grain size. We have confirmed by transmission electron microscopy that RTA does indeed reduce the grain size over an order of magnitude from films crystallized in a conventional furnace. Faraday microscopy has shown that films processed by RTA with smaller grain size (~ 30 nm) support circular magnetic domains of much greater regularity than films crystallized in conventional furnaces. The improved regularity of static thermomagnetically written domains, in films crystallized by RTA, lends promise that the noise in these materials can be reduced.

INTRODUCTION

Bismuth-substituted iron garnets are promising short wavelength magneto-optic recording materials since they have a strong magneto-optic effect at wavelengths less than 550 nm. Shono et al. demonstrated that bismuthsubstituted garnet films grown on single crystal GGG substrates exhibit 54 dB of CNR (30 kHz bandwidth, 5 MHz writing frequency, 14 m/s linear velocity, 514 nm wavlength, and 1.4 μ m bit length). The single crystal GGG substrates are expensive and limited in size so a more practical substrate is needed. Garnet films deposited on glass disks, crystallized in situ by deposition at elevated temperatures, or by post-deposition annealing have exhibited low CNR's due to high media noise. The media noise is due in part to the inability to write regular circular domains when the grain size is comparable to the magnetic domain size.³ The magnetic domain walls follow the grain boundaries thus the irregularity in domain shape is a result of the average grain size being of the order of the written domain size.

Much effort has been made to control the grain size and film morphology through different dopants, 4,5 underlayers, 6,7 and annealing schedules with only limited success. Suzuki et al. 10 reported that rapid thermal annealing (RTA), a process which employs are lamps or flash lamps as the thermal source and can provide heating rates as high as 200 °C/s, was successful in reducing the grain size to about 30 nm. In a transmission electron microscope (TEM) study Bechevet et al. 11 observed an inhomogeneous microstructure of large crystallites (450 nm) embedded in a matrix of small crystallites (35 nm) in cerium-substituted iron garnets crystallized by RTA. The purpose of this work was to confirm Suzuki's results for bismuth-substituted garnets and to determine if the reduced grain size enables regularly shaped magnetic domains to be written in these materials.

EXPERIMENTAL DETAILS

Thin films were deposited by rf magnetron sputtering onto Corning No. 7059 glass substrates with the sputtering

conditions listed in Table I. The as-deposited films were amorphous and were crystallized by either conventional annealing at 670 °C for 3 h in an air atmosphere or by RTA in an AG Associates 610i heat pulse system with a heating rate of 100 °C/s and a dwell time of 2 min at 710 °C in a nitrogen atmosphere.

Microstructural investigations were conducted on planar ion-milled specimens in a Philips 420 T TEM. Micromagnetic investigations were conducted using Faraday microscopy (50×0.5 or 125×0.8 NA) with an argon ion laser (λ =488 nm), modulated by an electro-optic modulator, as the writing source.

RESULTS AND DISCUSSION

A TEM image of a film annealed in a conventional furnace is shown in Fig. 1. This film has a polycrystalline microstructure with a grain size of the order of 1 μ m. A Faraday microscope image of thermomagnetically written domains in this film using a writing power of 20 mW, pulse width of 1 μ s, and a bias field of 70 Oe is shown in Fig. 2. The written domains are clearly irregularly shaped. In a previous paper³ it was shown that the domain walls are located along grain boundaries, thus the film microstructure plays a dominant role in determining the written domain shape. Further, it was shown that for films crystallized by post-deposition annealing the garnet phase nucleated at sites

TABLE I. rf magnetron sputtering conditions.

Target Composition:	$Gd_2Bi_1Fe_{4.6}Al_{0.2}Ga_{0.2}O_{12}$
rf power	0.57 W/cm^2
Bleed gas	Argon
Bleed pressure	5 mTorr
Base pressure	5×10^{-6} Torr
Deposition rate	150 nm/h
Deposition time	2 h



FIG. 1. Bright field TEM micrograph of garnet film annealed in a conventional furnace at $670~^{\circ}\text{C}$ for 3 h in an air atmosphere.

spaced approximately 1 μ m apart which grew rapidly until they impinged upon one another forming the grain boundaries. Therefore, the grain size is controlled by the density of garnet nuclei obtained before significant grain growth could occur.

A TEM image of an identical film crystallized by RTA is shown in Fig. 3. The average grain size is approximately 30 nm in agreement with Suzuki's results measured by scanning tunneling microscopy and x-ray diffraction. No evidence was found of the inhomogeneous microstructure, of large crystallites imbedded within a matrix of smaller crystallites, observed by Bechevet. A Faraday microscope image of domains written in this film using identical writing conditions as the conventionally annealed sample is shown in Fig. 4. These domains are much more circular than the domains written in the conventionally annealed sample, which lends promise that the noise in these materials can be reduced.

These results can be qualitatively understood in terms of the effects of different heat treatments on the nucleation and growth kinetics of the garnet phase. When the heating rate is

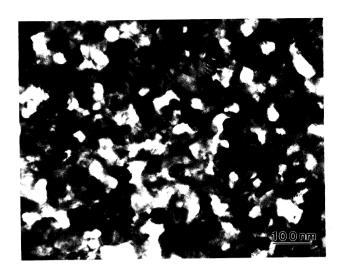


FIG. 3. Bright field TEM micrograph of a garnet film crystallized by RTA with a ramp rate of $100\,^{\circ}\text{C/s}$ and a dwell of 2 min at $710\,^{\circ}\text{C}$ in a nitrogen atmosphere.

slow (\sim 0.1 °C/s) nucleation of the garnet phase will commence at a relatively low temperature. This means that the nucleation proceeds under a small driving force, thus leading to a low nucleation rate (number of garnet phase nuclei per unit volume of untransformed region per unit time). The small number of nuclei, which occur at the lowest temperatures, are given time to grow with the rising temperature and consume other possible nucleation sites. This produces a low effective nucleation density and therefore the large crystallite size observed of approximately 1 μ m.

In the case of RTA, the rapid temperature rise will enable the nucleation to start at a relatively higher temperature (i.e., under a larger driving force). Further, the rapid temperature rise does not provide sufficient time for those nuclei which occur at the lowest temperatures to grow and consume the other possible nucleation sites. This creates a high effective nucleation density and the fine grained microstructure observed.

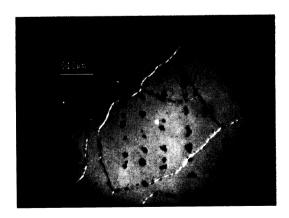


FIG. 2. Faraday micrograph of thermomagnetically written domains in post-deposition annealed garnet film (20 mW writing power, 1 μ s pulse width, and 70 Oe bias field).

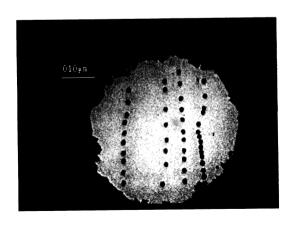


FIG. 4. Faraday micrograph of thermomagnetically written domains in RTA crystallized film (20 mW writing power, 1 μ m pulse width, and 70 Oe bias field).

CONCLUSIONS

We have confirmed Suzuki's results that RTA significantly reduces the crystallite size of bismuth-doped iron garnet films on glass substrates. Further, we have shown that films crystallized by RTA with the smaller grain size support circular magnetic domains of much greater regularity than films crystallized in conventional furnaces. The improved regularity of static thermomagnetic written domains in these films lends promise that the media noise problems of these materials can be solved. We believe the reduced grain size in the RTA processed films is a result of improved nucleation of the garnet phase and suppression of grain growth from the rapid temperature rise. A quantitative understanding of the kinetics of the phase change in these materials would be helpful in determining the optimal heating rate and crystalli-

zation temperature and possibly point to better methods of controlling the crystallite size in these materials.

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