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Structural and magnetic properties of Gd/Fe multilayers grown by pulsed laser deposition

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This work investigates the structural and the magnetic properties of Gd/Fe multilayered thin films grown by pulsed laser deposition onto Si (001) substrates at room temperature. The Fe layer thickness is varied from 70 to 150 nm and its effect on the structural and magnetic properties of Fe/Gd/Fe sandwich multilayers has been explored. Gd films were found to change from amorphous to polycrystalline at a critical thickness of 20 nm.

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Keywords: Pulsed laser ablation, Thin films and magnetization

1. Introduction

Magnetic layered systems exhibit various fascinating physical properties including giant magnetoresistance (GMR). These properties not only arouse fundamental interest, but are also important because of their possible applications in magnetic storage devices and sensors. For instance, double-layer films consisting of two different magnetic layers i.e. two ferromagnetic materials, or a ferromagnetic and an antiferromagnetic layer are being used in magnetoresistive heads[1] and magneto-optical media[2-3], utilizing the exchange coupling acting between two layers. Phenomena such as giant magnetoresistance and interlayer magnetic coupling have stimulated a great theoretical interest and opened new pathways for the design of novel devices by correct choice of materials. The periodic stacking of two distinct ferromagnetic materials gives rise to a variety of magnetic exchange interactions. Magnetic multilayers composed of a rare-earth (RE) element, such as Gd, and a transition metal (TM) like Fe, Co and Ni are interesting example systems. Due to their very different ordering temperatures, magnetic configurations depending on the structural parameters, temperature, and magnetic field may occur. In the case of Gd/Fe multilayers the impact of the variation of the growth parameters and thickness of the individual constituting Fe and Gd layers upon the transport and magnetic properties has been investigated extensively. [4-7]. However, most of these investigations were only carried out in the thickness regime of few monolayers (MLs).[8-13] The present study is aimed at enhancing the magnetic properties of Gd layer by sandwiching it in-between the two ferromagnetic Fe thin films with Fe thickness varying from 70 nm to 150 nm.

2. Experimental

The Gd, Fe and Fe/Gd/Fe thin films have been deposited using the pulsed laser deposition (PLD) facility at Risø National Laboratory for Sustainable Energy at the Technical University of Denmark, DTU, from polycrystalline Gd and Fe targets. A focused excimer laser was used for ablation (wavelength: 248 nm and frequency: 40 Hz) with a fluence of 6 Jcm^{-2}. Thin films were grown at room temperature, on Si (001) substrates at a background base pressure of 10^{-7} mbar. Substrate pre-treatment consisted of rinsing in ethanol, and in a 10 mol% diluted HF solution to remove native SiO_2 layer. The target-substrate distance was maintained at 65 mm. An Ag film deposited for 5 min, under the above mentioned conditions was deposited on the top of the films to prevent the oxidation of the metallic films.

The structure of Gd, Fe thin films and Fe/Gd/Fe multilayers was characterized by X-ray diffraction (Philips PANalytical and STOE) using Cu-K_α radiation. The thickness of the films was either measured directly from the cross-section of films using a Zeiss Scanning Electron Microscope (SEM) or from the EDX data by using the program CASINO V2.42.[14] A vibrating sample magnetometer (LakeShore 7407) was used to measure magnetic properties of thin films at temperatures ranging from 80 - 300 K.

3. Results and discussion

The phase purity and crystalline quality of the Gd, Fe films and Fe/Gd/Fe multilayers grown on Si (001) substrates by pulsed laser deposition were verified by room temperature X-ray diffraction. The θ - 2θ range was varied between 20° – 90° at a scan rate of 0.05°/step and integration time of 20 s/step. Slow scans were carried out between 25° – 55° at a scan rate of 0.01°/step and with

integration time of 60 s/step. The broadening of the most intense XRD peaks of Gd and Fe was extracted by recording the XRD pattern of a well crystallized silicon standard and correcting for the instrumental broadening.

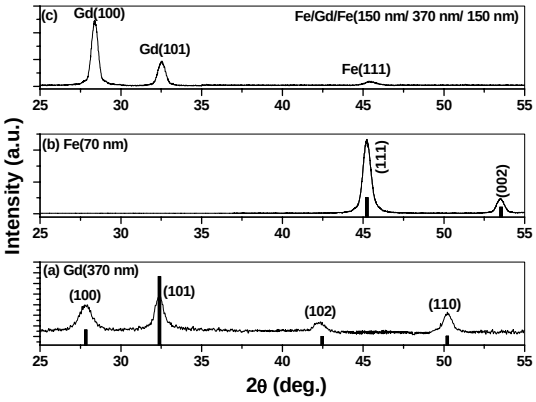

Fig.1. X-ray diffractograms of (a) Gd (370 nm), (b) Fe (70 nm) and (c) Fe/Gd/Fe (150 nm/370 nm/ 150 nm) films produced at 248 nm, 40 Hz and 6 J/cm^2.

Fig.1(a) shows the X-ray diffractogram of a Gd film with a thickness (t) of ~ 370 nm . The film thickness of Gd (370 nm) and Fe (70 nm) films (Fig. 1a and b) was obtained from scanning electron microscopy and CASINO V2.42 respectively. The peaks in the XRD pattern (Fig.1a) are in agreement with the hexagonal close packed (*hcp*) structure of Gd. The Gd films with thickness greater than 20 nm showed a polycrystalline nature and the reflections corresponding to (100), (101), (102) and (110) planes were observed at 2θ values of 28.3^o, 32.4^o, 42.4^o and 50.2^o (JCPDS: 89-2924). However, for thickness values of t < 20 nm the films tend to become amorphous with evolution of broad XRD peaks (not shown in this figure). Figure.1 (b) shows X-ray diffractogram of Fe film with a thickness of ~70 nm. The reflections corresponding to Fe(111) and Fe(002) were observed at 2θ values of 45.8^o and 53.4^o for Fe thin film (JCPDS: 88-2324).

The full width at half maximum (FWHM) of the Fe(111) peak found to be 0.5^o, corresponds to crystallite size of ~ 55 nm. The FWHM of Fe thin films remain unaltered for 70 nm and 150 nm thickness. Fig. 1(c) shows the X-ray diffractogram of Fe/Gd/Fe (150 nm/370 nm/150 nm) multilayer film. Reflections corresponding to Gd (101), Gd (102) and Fe(111) were observed. The reflections from Gd (102) and (110) disappeared and the FWHM of Gd peaks were reduced compared to the Gd films grown on Si (001) (Fig. 1. a). Additionally, the (002) of the Fe seems to disappear as well while the intensity of the (111) peak reduces drastically. Similar trends were observed in Fe/Gd/Fe (70 nm/740 nm/ 70 nm) multilayers.

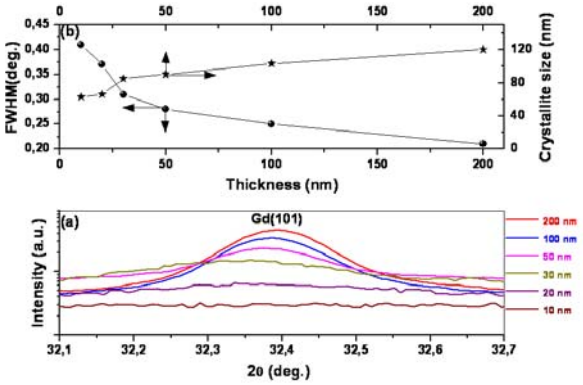

Fig. 2. (a) Slow X-ray diffraction scans of Gd(101) peak and (b) Full width at half maxima and crystallite size vs. thickness for Gd thin films.

It was found from X-ray diffraction data that all the Gd films with thicknesses ranging from 20 - 370 nm were predominantly (101) orientated (not shown here). Slow XRD scans of (101) peak for Gd films with different thicknesses were obtained, and it was observed that the Gd(101) peak clearly shifts toward lower angle suggesting an increase in lattice parameter (Fig. 2a). The shift in Gd(101) peak position towards lower angle could be due to the strain in the lattice (lattice mismatch of ~6%). This result shows that the Si (001) substrate induces tensile strain in the in-plane of the Gd thin film. The peak position of Gd(101) shifts towards a higher angle (~32.39^o) with an increase in thickness indicating the lattice parameters tend towards bulk value of Gd (a = b = 0.363 nm and c = 0.577 nm). The FWHM decreases from 0.41^o to 0.21^o with increase in thickness for the Gd film indicating the possible formation of larger grains in thicker films (Fig. 2b). The lower thickness of thin films assists in the growth of finer grains resulting in an increase in FWHM to 0.41^o. X-ray diffraction patterns corresponding to Gd thin films with thickness 10 nm, 20 nm and 30 nm show a broad reflection compared to bulk polycrystalline Gd due to the formation of small crystallite size in these films. The crystallite size of Gd thin films was evaluated by Debye-Scherrer formula by obtaining the full width at half maximum of the Gd(101) peak. The crystallite size is found to increase from 66 nm to 120 nm with an increase in thickness values from 20 nm to 200 nm (Fig. 2b).

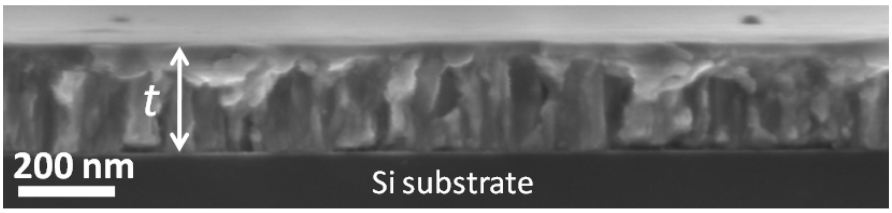

Fig. 3. Scanning electron micrograph of a cleaved Fe thin film cross-section. The bar indicates 200 nm.

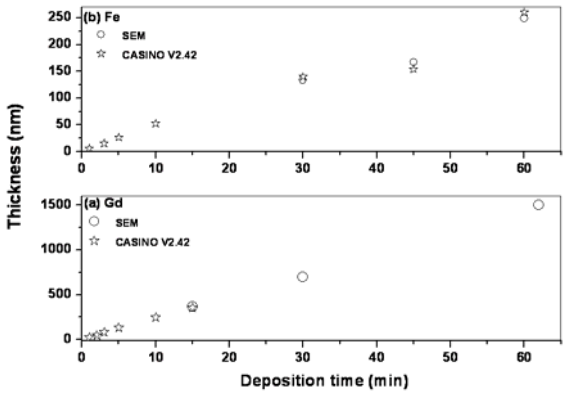

Fig. 4. Film thickness vs. deposition time of (a) Gd and (b) Fe thin films deposited by pulsed laser deposition.

Fig. 3 shows SEM image of a 260 nm thick Fe film deposited on a Si (001) substrate with columnar grains. The thickness measurements were carried out on Gd and Fe films with different deposition times. The growth rate is plotted as thickness vs. deposition time graphs for Gd and Fe thin films (Figures 4a and b). Films with lower deposition time (< 10 min) were evaluated for thickness by CASINO V2.42 while for some of the thicker films these values were measured directly from the film's cross section, and indeed a good agreement is observed between the two. The growth rate for Gd (24.5 nm/min) is larger than that of Fe (4.1 nm/min) films. The growth rate is found to decrease with increase in deposition time. Films deposited for less than about 10 min have a higher growth rate compared to the films deposited for a longer duration. This trend is observed both in Gd and Fe thin films. Since Gd and Fe atoms have nearly equal binding energy values (Gd: 4.14 eV/atom and Fe: 4.29 eV/atom) they are expected to yield similar growth rates. The lower growth rate at higher deposition time could be attributed to a change in energy absorbed by metal surfaces with increase in ablation time.

Gd, Fe and Fe/Gd/Fe thin films were studied for their magnetic properties by applying a magnetic field in parallel and perpendicular directions (with reference to the surface normal of the substrate). Gd and Fe have similar bulk saturation magnetization values at room temperature (2020 and 1750 emu/cm^3).[15] Fe saturates at low magnetic field compared to Gd and may be expected to enhance the magnetic properties of Gd. Figures 5(a) and (b) show temperature dependent magnetic hysteresis of Gd and Fe thin films in parallel configuration. The Gd thin film has a Curie temperature of 293 K, unaltered from bulk Gd. The Fe thin films (t ~ 260 nm) exhibited a low coercivity (H_C ~ 10 Oe) and its magnetic properties were found to be independent of the applied direction of the field.

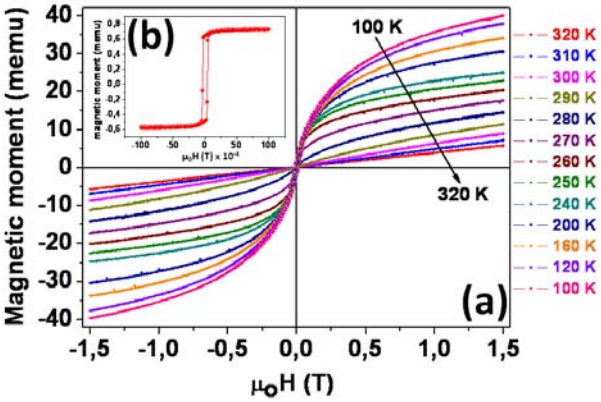

Fig. 5. Field dependence of magnetization curves for (a) Gd (1.5 μm) film and the inset (b) shows magnetization vs field curve of Fe (260 nm) film at room temperature in parallel configuration.

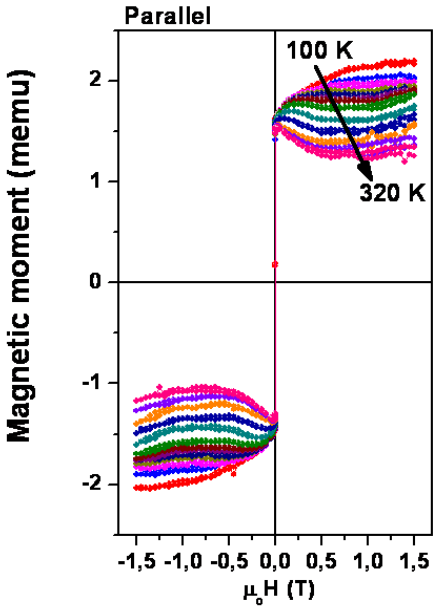

Fig. 6. Field dependence of magnetization curves for Fe/Gd/Fe (150 nm/ 370 nm/ 150 nm) multilayers in

parallel configuration at various temperatures.

Fig. 6 shows the field dependence of the magnetization of Fe/Gd/Fe (150 / 370 / 150 nm) multilayers in parallel configuration obtained at different temperatures in the range of 100 K to 320 K. Fe saturates at a low magnetic field and yields spontaneous magnetization. Similar trends of Fe saturation are observed in Fe/Gd/Fe (70 / 740 / 70 nm) multilayers. Below the Gd transition temperature, Gd appears to magnetize only after complete saturation of the Fe layers present on either side. In parallel configuration, the paramagnetic state of Gd is observed to reduce the magnetization in the Fe/Gd/Fe (150 / 370 / 150 nm). In perpendicular configuration, the magnetization of Gd appears to be unsaturated, however it shows a continuous increase with increase in magnetic field. The absolute magnetization values at low magnetic fields (< 100 Oe) roughly correspond to the magnetization of Fe. The Curie temperature of Gd remained unaltered in Fe/Gd/Fe (70 / 740 / 70 nm) multilayers.

4. Conclusions

Gd/Fe multilayer films have been prepared by pulsed laser deposition, and their magnetic properties have been investigated. The following results were obtained.

(i) When the Gd layer thickness is about 20 nm, the film structure changes from amorphous to polycrystalline.

(ii) The transition temperature of Gd thin films remains unaltered in Fe/Gd/Fe multilayers, and is found to be independent of Fe thin film thickness.

References

[1] H. H. Bertschat, J. Magn. Magn. Mater. **198-199**, 636 (1999).
[2] T. Shinjo, W. Keune, J. Magn. Magn. Mater. **200**, 598 (1999).
[3] H. Awano, S. Ohnuki, H. Shirai, N. Ohta, IEEE Trans. Magn. **33**, 3214 (1997).
[4] N. Nishimura, T. Hiroki, T. Okada, S. Tsunashima, Jpn. J. Appl. Phys., Part **1**(35), 403 (1996).
[5] A. A. Kusov, S. Jaswal, Z. Shan, Phys. Rev. B **46**, 3123 (1992).
[6] K. Takanashi, Y. Kamiguchi, H. Fujimori, M. Motokawa, J. Phys. Soc. Japan **61**, 3721 (1992).
[7] M. Vaezzadeh, B George, G Marchal Phys. Rev. B **50**, 6113 (1994).
[8] J. P. Andrés, L. Chico, J. Colino, J. M. Riveiro, Phys. Rev. B, **66**, 094424 (2002).
[9] R.E. Camley, R.L. Stamps, J. Phys. Condens. Matter **5**, 3727 (1993).
[10] R. E. Camley, Phys. Rev. B **39**, 12316 (1989).
[11] T. Morishita, Y. Togami, K. Tsushima, J. Phys. Soc. Jpn. **54**, 37 (1985).
[12] K. Takanashi, H. Kurokawa, H. Fujimori, Appl. Phys. Lett. **63**, 1585 (1993).
[13] S. Honda, M. Nawate, I. Sakamoto, J. Appl. Phys. **79**, 365 (1996).
[14] N. Pryds, B. Toftmann, J. B. Bilde-Sørensen, J. Schou, S. Linderoth, Appl. Surf. Sci. **252,** 4882 (2006).
[15] D. Haskel, G. Srajer, J. C.Lang, J. Pollmann, C. S. Nelson, J. S. Jiang, S. Bader, Phys. Rev. Lett, **87,** 207201 (2001).

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