

## Excellent Magnetic Properties of $\text{Co}_{53}\text{Fe}_{22}\text{Hf}_{10}\text{O}_{15}$ Thin Films

L. V. Tho<sup>1</sup>, K. E. Lee<sup>1</sup>, C. G. Kim<sup>1,2</sup>, C. O. Kim<sup>1,2</sup>, and W. S. Cho<sup>2,3\*</sup>

<sup>1</sup>Department of Materials Science and Engineering, Chungnam National University, Daejeon, Korea

<sup>2</sup>ReCAMM, Chungnam National University, Daejeon, Korea

<sup>3</sup>Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 0ME United Kingdom

(Received 28 November 2006)

Nanocrystalline CoFeHfO thin films have been fabricated by *RF* sputtering method. It is shown that the CoFeHfO thin films possess not only high electrical resistivity but also large saturation magnetization and anisotropy field. Among the composition investigated,  $\text{Co}_{53}\text{Fe}_{22}\text{Hf}_{10}\text{O}_{15}$  thin film is observed to exhibit good soft magnetic properties: coercivity ( $H_c$ ) of 0.18 Oe; anisotropy field ( $H_k$ ) of 49.92 Oe; saturation magnetization ( $4\pi M_s$ ) of 15.5 kG. The frequency response of permeability of the film is excellent. The excellent magnetic properties of this film in addition of an extremely high electrical resistivity ( $r$ ) of 185  $\mu\text{cm}$  make it ideal for uses in high-frequency applications of micromagnetic devices. It is the formation of a peculiar microstructure that resulted in the superior properties of this film.

**Keywords :** CoFeHfO thin film, soft magnetic thin film, anisotropy field

### 1. Introduction

During the last decades, searches for suitable magnetically soft, low coercivity, high saturation magnetization and anisotropy and resistivity materials, have been carried out to fulfill the requirements of writing heads and high-frequency inductors.

The growing interest in micromagnetic integrated devices such as inductors and transformers for power delivery and signal conditioning, has renewed interest in highly resistive granular magnetic films [1, 2]. Among these materials, soft magnetic films based on Co-Fe permalloy and ion-rich Fe-Zr, Fe-Ti, Fe-Ta have been investigated [3-5]. However, desirable soft magnetic properties are typically obtained only after postdeposition heat treatment, for nanocrystallization, of the deposited films at relatively high temperature of 300°-600°C such a high temperature post heat treatment is an added complication in the processing of devices which contain other components and materials, some sensitive to the high-temperature post heat treatment [3]. In the present work, we have successfully achieved the desirable to avoid the high-temperature post heat treatment.

### 2. Experiment

Co-Fe-Hf-O thin films were fabricated on Si wafer by *RF* reactive sputtering under Ar+O<sub>2</sub> atmosphere, at the O<sub>2</sub>/(Ar+O<sub>2</sub>) gas flow ratio is 2 to 10%. Magnetic field of 100 Oe is applied during sample preparation to induce an in-plane uniaxial anisotropy. Other condition in this study were as follows: base vacuum of less than  $2 \times 10^{-7}$  Torr; working pressure of  $2 \times 10^{-3}$  Torr; AC sputtering power of 300 W; the time sputtering is controlled from 1 min to 5 min to obtain thickness of the film is about 300 nm.

The exactly composition of Co-Fe-Hf-O films was measured by auger electron spectra (AES). The electrical resistivity was measured with a conventional four-point probe. The magnetic properties were measured with a DMS vibrating sample magnetometer (VSM) and a SHB-109 BH loop tracer. The effective permeability of the thin films was measured in the range of 1 to 3000 MHz by network analyzer HP4191A. The crystal structure was observed by x-ray diffraction (XRD) and high resolution transmission electron microscopy (HRTEM).

### 3. Results and Discussion

Fig. 1 shows the result of  $\text{Co}_{70}\text{Fe}_{30}\text{Hf}_{10}\text{O}_6$  thin film that was prepared with 300 W power, 6% partial of oxygen

\*Corresponding author: Tel: +82-42-821-6227,  
Fax: +82-42-822-6272, e-mail: wscho77@yahoo.com

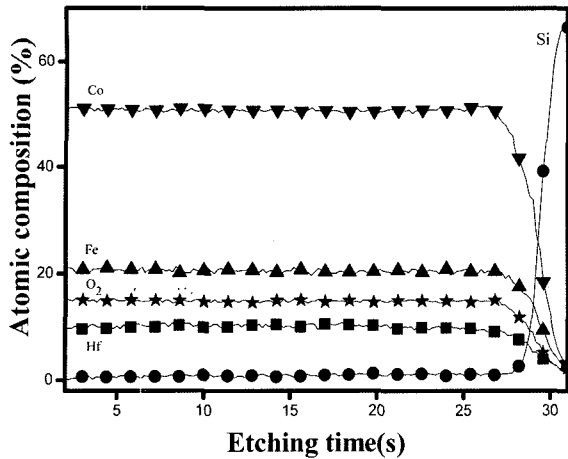


Fig. 1. AES-depth profile of CoFeHfO thin film as deposited with 6% oxygen.

during 3 minutes. This result shows that, the composition is almost constant along to thickness of thin film and indicated that our sputtering technique is good method to make thin film. The exact composition is  $\text{Co}_{53}\text{Fe}_{22}\text{Hf}_{10}\text{O}_{15}$ .

Fig. 2 shows XRD patterns of  $\text{Co}_{53}\text{Fe}_{22}\text{Hf}_{10}\text{O}_{15}$  film, the diffraction strong peaks is distributed around  $2\theta$  of ( $44^\circ$ - $46^\circ$ ), corresponding to the diffraction from CoFe (110). It is the main crystalline in our thin films and plays important role in making excellent magnetic properties of thin films. So we concentrated to fabricate this crystal. The other broad and low peak at  $2\theta$  of ( $41^\circ$ - $43^\circ$ ), ( $65^\circ$ - $67^\circ$ ) due to the diffraction from monoclinic, CoO (200), CoFe (200), respectively. These crystals are not good for magnetic properties, so we try to decrease amount of CoO by controlled  $\text{O}_2$  gas flow [6] and CoFe (200) by controlled thickness of film [4].

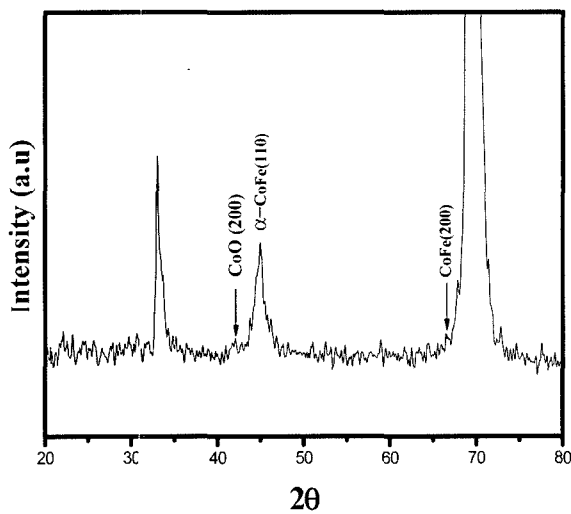


Fig. 2. XRD diffraction patterns of  $\text{Co}_{53}\text{Fe}_{22}\text{Hf}_{10}\text{O}_{15}$  reactive sputter-deposited thin films.

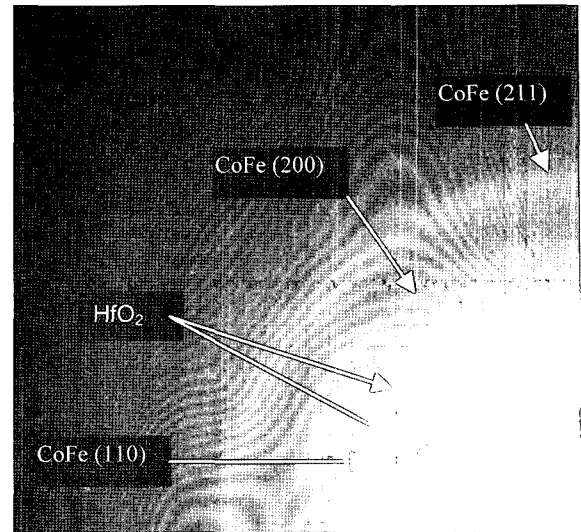


Fig. 3. SAD patterns of  $\text{Co}_{53}\text{Fe}_{22}\text{Hf}_{10}\text{O}_{15}$  thin film.

To confirm these phases, SAD of thin film was determined. Fig. 3 shows a halo ring is observed at the first diffraction ring suggesting the presence of remaining amorphous and granular phases in the films, which can be indexed using the  $\alpha\text{-Co(Fe)}$  (110),  $\alpha\text{-CoFe}$  (110), amorphous oxide phases a-HfO phases. The intensity of this ring is so high indicated that amount of these phases is high also. The second diffraction ring could be due to a distribution of CoFe (200) in the film. The third diffraction ring can be indexed using a combination of the  $\alpha\text{-Co(Fe)}$  (211),  $\alpha\text{-CoFe}$  (211). The second and third ring is weak indicated that amount of CoFe (200) and CoFe (211) are very low. The outer diffraction rings correspond to reflections from monoclinic Co phases.

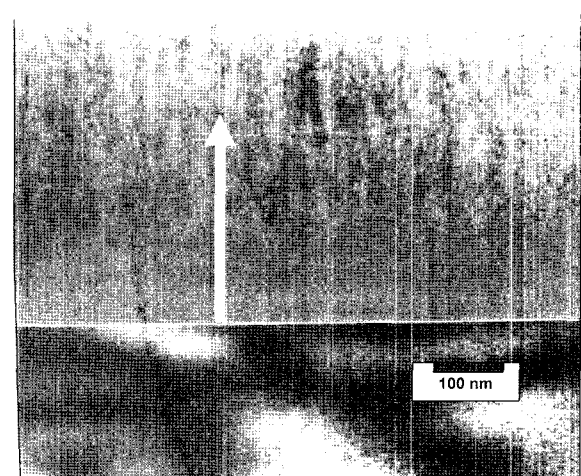


Fig. 4. HRTEM view of film with 6%  $\text{O}_2$ . Arrows point to the direction of nanocrystalline CoFe-rich layers (black) and amorphous HfO-rich layers (white), respectively.

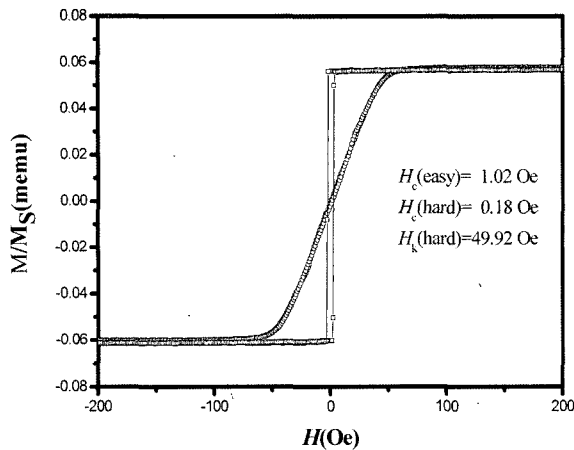


Fig. 5. Magnetization curves of nanocrystalline  $\text{Co}_{53}\text{Fe}_{22}\text{Hf}_{10}\text{O}_{15}$  thin film as deposited with 300 nm and 6% Oxygen.

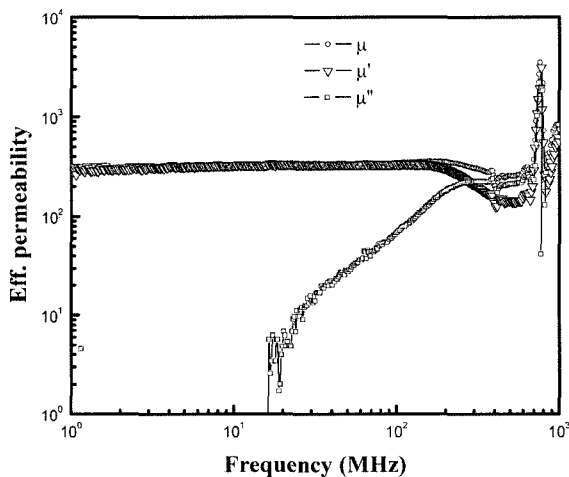


Fig. 6. Frequency dependence of effective permeability of  $\text{Co}_{53}\text{Fe}_{22}\text{Hf}_{10}\text{O}_{15}$  film measured along easy axis.

Fig. 4 shows the TEM image of  $\text{Co}_{53}\text{Fe}_{22}\text{Hf}_{10}\text{O}_{15}$  thin film with microstructure consists of Co(Fe) nanograins randomly arranged within an amorphous HfO matrix. It has a peculiar microstructure (arrow pointing) which consist of nanocrystalline Co(Fe) -rich laminates of  $\sim 2$  nm thickness indicated by black color separated one by one by amorphous HfO-rich laminates of  $\sim 0.5$  nm thickness indicated by white color. This structure can be considering column structure, thereby providing a preferentially uniaxial anisotropy.

Fig. 5 shows typical normalized magnetization curves of nanocrystalline  $\text{Co}_{53}\text{Fe}_{22}\text{Hf}_{10}\text{O}_{15}$  film. The film exhibits excellent magnetic Properties, with  $H_c$  of 0.18 Oe, anisotropy field,  $H_k$  of 49.92 Oe and saturation magnetization of 15.5 kG. The large  $H_k$  of this film may be connected with

the anisotropic form of nanocrystalline in the film. The large saturation magnetization is relation of a lot of amount crystalline CoFe (110).

Fig. 6 illustrates the effective permeability ( $\mu_{eff}$ ) spectra in the 0 to 2 GHz band. The effective permeability of thin film is about 300, which is maintained up to 650 MHz.

The behavior of  $\mu'$  (in this case corresponding with  $\mu_{eff}$ ) agrees with the Stoner and Wolfarth theory, which explains the permeability at low frequency the value  $\mu' = M_s/H_k$ . The excellent  $\mu$ -f characteristics are due to the high ferromagnetic resonance frequency,  $f_{FMR}$  originated from the factors of thin film as mentioned above. It can be said that the high resistivity soft magnetic films with excellent characteristics perform well in the very high frequency range.

#### 4. Conclusion

In summary of this report, the optimal sputtering conditions have been obtained with 300 nm at 6%  $\text{O}_2$ . The optimized film composition is  $\text{Co}_{53}\text{Fe}_{22}\text{Hf}_{10}\text{O}_{15}$  exhibit very low  $H_c$  0.18 Oe and high resistivity 185  $\mu\text{cm}$  compared with 5 Oe and 60  $\mu\text{cm}$  observed by Jeon *et al.* [4], it also have higher  $H_k$  and saturation magnetization compared with Crawford *et al.* It indicated that  $\text{Co}_{53}\text{Fe}_{22}\text{Hf}_{10}\text{O}_{15}$  film exhibit excellence magnetic properties and it is very promising for high frequency applications (GHz) was proved in Fig. 6.

#### Acknowledgements

This work was supported by the Korean Science and Engineering Foundation through the Research Center for Advanced Magnetic Materials at Chungnam National University and BK 21 program.

#### References

- [1] A. M. Crawford, D. Gardner, and S. X. Wang, IEEE Trans. Magn. **38**, 3168 (2002).
- [2] M. Xu, T. M. Liakopoulos, and C. H. Ahn, IEEE Trans. Magn. **34**, 1369 (1998).
- [3] N. Kataoka, M. Hosokawa, A. Inoue, and T. Masumoto, Jpn. J. Appl. Phys. **28**, L462 (1989).
- [4] N. Kataoka, K. Sumiyama, and Y. Nakamura, Trans. Jpn. Inst. Met. **27**, 823 (1986).
- [5] Xiaomin Liu and Giovanni Zangari, Liyong Shen, J. Appl. Phys. **87**, 9, 2000.
- [6] Liangliang Li, Ankur M. Crawford, Shan X. Wang, and Ann F. Marshall, J. Appl. Phys. **97**, 10F907, 2005.