

MAGNETIC PROPERTIES OF Fe/Cu MULTILAYERS PREPARED USING PULSED-CURRENT ELECTRODEPOSITION

Conrad Rizal^{1*}, Yuji Ueda², and Bhisma R. Karki³

1 Department of Electrical and Computer Engineering, University of British Columbia, Vancouver, BC, V6T 1Z4, Canada

2 Department of Electrical and Electronic Engineering, Muroan Institute of Technology, Hokkaido, 050-8585, Japan

3 Department of Physics, Texas Tech University, Lubbock, TX, 79409, USA

ABSTRACT

[Fe (t_{Fe}) nm/Cu (t_{Cu}) nm]_N multilayer films were prepared using pulsed-current electrodeposition method. The role of the pulsed-current electrodeposition on microstructure and magnetic properties was investigated. The microstructure of the multilayer films is dependent on the thicknesses of both the Fe and Cu layers. The saturation magnetization of the multilayers strongly correlated with the crystalline structure of Fe at the interface of Fe and Cu layers.

Key words: pulsed-current electrodeposition, Fe/Cu, multilayers, saturation magnetization, microstructure

INTRODUCTION

Interest in artificially tailored nanostructures has greatly surged in recent times because of their novel magnetic properties and potential technological applications [1]. One of the excellent examples of artificially tailored nanostructure is Fe/Cu, which is composed of mutually insoluble metals such as iron and copper. In spite of its complex structure, Fe can be combined with Cu to prepare multilayers of fcc-Cu and bcc-Fe [2-4]. Extensive studies have been done on the crystalline structure and magnetization properties of the multilayers of Fe/Cu prepared by physical methods such as sputtering [5, 6], molecular beam epitaxy [7], and e-beam evaporation [8]. However, these methods require an ultra-high vacuum to control their film thicknesses. In contrast, the electrochemical method employs the liquid phase, which is relatively simple, inexpensive, and efficient for growing multilayers at the atomic level.

Pulse electrodeposition offers an easy adjustment of the current and time required in finding appropriate electrodeposition conditions. It also offers direct control over the composition and thickness, which can be achieved by adjusting the quantity of the pulsed-current through the electrolyte. Also, the

* e-mail: crizal@ece.ubc.ca, tel:+1(604)8226268, fax:+1(604)8225949

thickness and composition of the multilayer containing the layers of Fe and Cu can be conveniently controlled by changing pulse amplitude and width [9].

Earlier, we reported findings on the chemical compositions and magneto-resistance of compositionally modulated Fe-Cu-Ni alloys [9], and Fe-Ni [10] and Fe-Cu [11] multilayers and alloys.

In this study, we re-examine the relationship between saturation magnetization and the microstructure of the Fe/Cu multilayers. The saturation magnetization of the multilayer strongly correlated with the crystalline structure of Fe at the interface of Fe and Cu layers.

EXPERIMENTAL PROCEDURES

Fe/Cu multilayers were electrodeposited in a 250 cc glass beaker from a single sulphate electrolyte using pulsed-current electrodeposition, which utilized two electrodes. The pulsed-current (potential) was supplied and controlled by a programmable micro-computer and a digital current source as shown in [12].

The solution was composed of $\text{FeSO}_4 \cdot 6\text{H}_2\text{O}$ (25.01 gm/litre), $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ (2.5 gm/litre), and NaCl (2.5 gm/litre). Ammonium hydroxide (NH_4OH) was used as a complexing agent because it improves the quality of the deposit [13]. The pH was maintained at 3.5 and it was controlled by the addition of sulphuric acid (H_2SO_4). The total composition of the solution was 0.1 mol/litre. The solution was prepared in double distilled water. The temperature of the bath was maintained at room temperature. Further details are available in [11] and [14]. The distance between the cathode and anode was maintained at 2 cm. The current density was changed from 0.2 to 20 mA/cm^2 . Metal ions were collected at the cathode. Ferromagnetic Fe and non-magnetic Cu were alternately deposited to form very fine layered films. After deposition, the films were cleaned with double distilled water, dried, and immediately wrapped in paraffin paper.

The saturation magnetization of the Fe/Cu multilayers was measured in the range of ± 21 kOe, using a vibrating sample magnetometer. The microstructure was analyzed with $\text{CuK}\alpha$ radiation using an X-Ray diffractometer. The lattice spacing, d , was calculated from the diffraction curve using Bragg's law as:

$$2d \sin \theta_B = n\lambda \quad (1)$$

where n is the integer, λ is the wavelength of the X-rays, and θ_B is the Bragg's angle.

The multilayer periodicity was determined using a composition of Fe and Cu in the film, and the composition was estimated using micro-balance and energy dispersive X-ray analysis.

RESULTS AND DISCUSSIONS

Fig.1a shows pulsed-current waves with different widths and intervals

and (Fig 1b) the corresponding multilayer structures. The composition of each layer of Fe and Cu in (Fig 1b) corresponds to the height of the pulse wave whereas the thickness corresponds to the pulse width, which is also a deposition time in (Fig 1a). The beauty of this process is that by controlling both the pulse amplitude and widths one can produce various combinations of multilayers and alloys on an atomic scale via a single electrolyte [12].

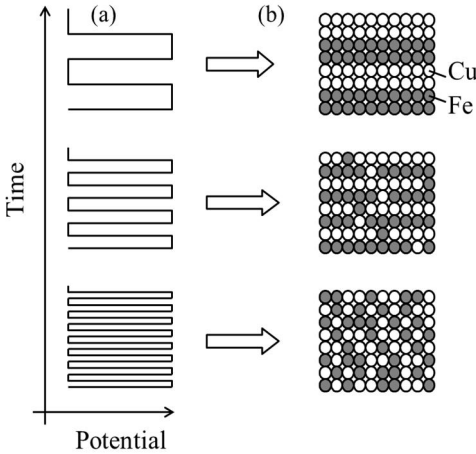


Fig.1 - (a) Deposition times versus potential corresponding to the pulsed-current and (b) Film structures corresponding to (a).

The reasons for the decrease in magnetization with the change in thickness of Cu layer are likely due to a change-over of fcc-Fe from the bcc-Fe crystalline structures [14]. The reasons for further decrease in magnetization due to heat treatment are likely due to the inter-diffusions of Fe and Cu atoms at the interface [15] [16].

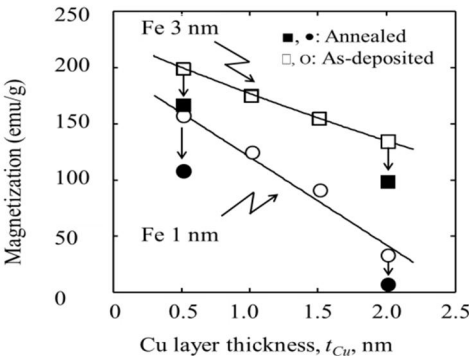


Fig.2 shows the saturation magnetization of [Fe 1 nm/Cu (t_{Cu})]₁₂₀ and [Fe 3 nm/Cu (t_{Cu})]₄₀ multilayers as the Cu layer thickness, t_{Cu} is changed from 0.5 to 2 nm. The total M_s tended to decrease when t_{Cu} is increased. However, decrease in magnetization for t_{Fe} = 1 nm is different from when t_{Fe} = 3 nm, and the gap between the two widens with the increase of t_{Cu} , i.e., the decrease of M_s with t_{Fe} = 1 nm is sharper than when t_{Fe} = 3 nm. Furthermore, M_s tends to decrease when subject to heat treatment.

Fig. 2 – Cu layer thickness, t_{Cu} dependence of the saturation magnetization of the [Fe1 nm/Cu (t_{Cu}) nm]₁₂₀ and [Fe3 nm/Cu (t_{Cu}) nm]₄₀ multilayers: Open rectangles (□): as-deposited and t_{Fe} = 3 nm. Closed rectangles (■): Annealed for 60 minutes at 300 °C and t_{Fe} = 3 nm. Open circles (○): as-deposited and t_{Fe} = 1 nm. Closed circles (●): Annealed for 60 minutes at 300°C and t_{Fe} = 1 nm.

The details about how the lattice constant changes with the layer thickness are given in *Fig. 3* and *Fig. 4*. It should be noted that total Fe content in the multilayers is always kept constant.

Fig. 3 shows the dependence of Cu layer thickness on the lattice constant of Fe, a_{Fe} , which increases as the Cu layer thickness, t_{Cu} , of $[Fe\ 1\ \text{nm}/Cu(t_{Cu})]_{120}$ multilayer is changed from 1 to 3 nm.

The inset shows the X-ray diffraction curves. A sharp diffraction peak of $[Fe\ 1\ \text{nm}/Cu(t_{Cu})]_{120}$ corresponding to bcc-Fe (110) is indicated by long dashed lines. When the fcc-Cu<111> layer thickness is increased, the peak shifts towards the lower side, and also results in a decrease in the diffraction peak height and an increase in the diffraction width, a sign of significant change in the crystalline structure of bcc-Fe (110) [15]. The broad peak of Fe (110) is believed to be originated from the intermixing of atoms of Fe and Cu in the interface as the thickness of Cu is increased [14].

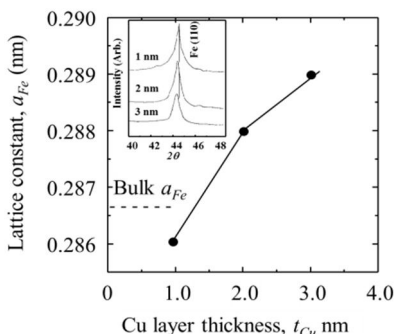


Fig. 3 – The lattice constant of Fe as the Cu layer is changed from 1 to 3 nm, obtained from the X-ray diffraction analysis.

The inset shows the X-ray diffraction pattern of $[Fe\ 1\ \text{nm}/Cu(t_{Cu})]_{120}$ multilayer films.

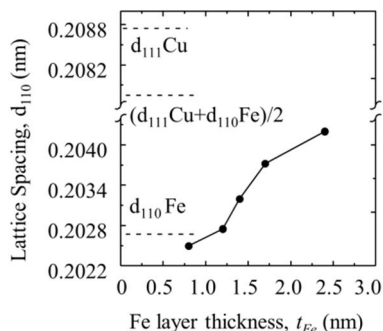


Fig. 4 – The lattice spacing, d_{110} of Fe/Cu multilayers as the Fe layer thickness, t_{Fe} is increased in thickness from 1 to 3 nm.

The bulk values of $d_{110}Fe$, $d_{111}Cu$ and $d_{110}Fe + d_{111}Cu$ are indicated by straight dashed lines.

Fig. 4 shows the Fe layer thickness dependence on lattice spacing. The lattice spacing of $[Fe(t_{Fe})\ \text{nm}/Cu\ 1\ \text{nm}]_N$ multilayers, where N is varied from 120 to 40, increases as the thickness of the Fe layer in the multilayer is increased from 1 to 3 nm. The lattice spacing is lower than the bulk value of $d_{110}Fe$ for $t_{Fe} < 1$ nm, followed by an increase that approaches the bulk value of $d_{110}Fe + d_{111}Cu$ for $t_{Fe} = 3$ nm. These multilayers display a combination of properties whose atomic structures and properties vary strongly with the individual layer thickness of Fe and Cu, indicating how the electronic structure might be engineered to enhance saturation magnetization [17].

CONCLUSIONS

We have observed unusual changes in the atomic structures of the Fe/Cu multilayers depending on the layer thickness of Fe and Cu. The change in the electronic properties of Fe is believed to arise from the intermixing of atoms at the interface between Fe and Cu layers. The thickness dependence of the magnetization shows that the decrease in magnetization is caused by changes in the crystalline structures from bcc-Fe to fcc-Fe, which is consistent with the results of X-ray diffraction. Research on the Fe/Cu multilayer is growing rapidly and has many technological applications including in the biomedical sectors.

Acknowledgements

The authors are grateful to the Natural Science and Engineering Research Council scholarship, Government of Canada, for partial funding for this work.

REFERENCES

- [1] P. J. Sadashivaiah, T. Sankarappa, T. Sujatha et al. *Vacuum*, 2010, vol. 85, pp. 466-473
- [2] C. L. Foiles, *Ultra-microscopy*, 1989, vol. 29, pp. 80-87.
- [3] S. El Khiraoui, M. Sajieddine, M. Hehn, et al. *Physica B: Condensed Matter*, 2008, vol. 403, pp. 2509-2514.
- [4] X. Jin, Y. Zhou, C. O. Kim, et al. *J. Appl. Phys.*, 2002, vol. 91, pp. 6071-6076.
- [5] A. P. Kuprin, L. Cheng, D. W. Lee, et al. *J. Appl. Phys.*, 1999, vol. 85, pp. 5738-5740.
- [6] J. M. Florczak, E. D. Dahlberg, E. E. Fullerton and I. K. Schuller, *J. Appl. Phys.*, 1991, vol. 70, pp. 5836-5836.
- [7] Y. Harada, Y. Nakanishi, N. Yoshimoto et al. *J. Magn. Magn. Mater.*, 2004, vol. 272-276, pp. e969-e970.
- [8] J. F. M. Borges, G. Tosin, L. F. Schelp, et al. *J. Magn. Magn. Mater.*, 1993, vol. 121, pp. 53-56.
- [9] C. Rizal and Y. Ueda, *International Conference on Magnetic Films and Surfaces (ICMFS)*, Kyoto, Japan, 2001, pp. 27-28.
- [10] A. Yamada, M. Shirota, T. Houga, C. Rizal, Y. Ueda, *Japan Inst. of Metals*, 2002, vol. 66, pp. 869-872.
- [11] H. Zaman, A. Yamada, H. Fukuda, Y. Ueda, *J. Electrochem. Soc.*, 1998, vol. 145, pp. 565-568.
- [12] A. Yamada, T. Houga and Y. Ueda, *J. Magn. Magn. Mater.*, 2002, vol. 239, pp. 272-275, 2.
- [13] I. W. Wolf, *Appl. Phys.*, 1962, vol. 33, pp. 1152-1159.
- [14] Y. Ueda, S. Ikeda, Y. Mori, H. Zaman, *Materials Science and Engineering (A)*, 1996, vol. 217, pp. 371-375.
- [15] Y. Ueda and M. Takahashi, *J. Magn. Magn. Mater.*, 1988, vol. 71, pp. 212-218.
- [16] R. M. Bozorth, *Ferromagnetism*. New York: Van Nostrand, 1947., pp.48
- [17] P. Ohresser, J. Shen, J. Barthel, et al. *Phys. Rev. B*, 1999, vol. 55 (5), pp. 3696-3706