

Magnetic Properties of Fe-Ni-N/Cu Multilayered Films by DC Magnetron Sputtering Method

Jung Gi Kim, Hyun Joong Kim, Ji Young Jang and Kyung Hunn Han^{1*}

Department of Physics, Hanyang University, 17 Haengdang-Dong, Seongdong Gu, Seoul 133-791, Korea

¹Quantum Photonic Science Research Center, Hanyang University, Seoul 133-791, Korea

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The structure and magnetic properties of Fe-Ni-N/Cu multilayered films, prepared by the DC magnetron sputter, as a function of different thicknesses of Fe-Ni-N (t_{FeNiN}) and Cu (t_{Cu}) layers have been studied by the methods of x-ray diffraction and measurement of magnetic moment. It has been found that the enhancement of (200) orientation in Fe-Ni-N layers is observed at the ratio of layer thickness with about $t_{\text{FeNiN}}/t_{\text{Cu}} \cong 3.75$. The reduction of magnetization due to the formation of interdiffusion near the interface is explained by means of the dead layer model. The temperature dependence of magnetization exhibits the feature of Bloch's $T^{3/2}$ law. The layer thickness dependence of Curie temperature has been discussed by critical temperature theory of Heisenberg model.

Key words : multilayered film, Fe₃NiN film, Curie temperature, Magnetization, Spin wave theory

1. Introduction

Artificially composition-modulated materials whose microstructure and magnetic properties depend strongly on the thickness of the individual elemental layers have attracted considerable attention in recent years. In particular, such multilayered structures based on magnetic/non-magnetic bilayers can exhibit noble and interesting physical effects with technologically important applications like magnetic recording media, devices, and sensors [1]. In a search for the new material, the crystal structure of Fe₃NiN alloy is very similar to that of copper (Cu) which is composed of the face-centered-cubic cell. The lattice mismatch of Fe₃NiN ($a_0 = 3.79 \text{ \AA}$) and Cu ($a_0 = 3.61 \text{ \AA}$) is about 0.5% [2]. Also the thermal expansion and forced magnetostriction of Fe₃NiN alloy showed similarities with the Fe-Ni Invar alloys [3]. In this study, a new series of Fe-Ni-N/Cu multilayers were prepared by dc magnetron sputter in ultrahigh vacuum and we have studied their structure, magnetic properties, and interlayer exchange interaction.

2. Experimental

Fe-Ni-N/Cu multilayered films were synthesized using

*Corresponding author: Tel: +82-2-2290-0919, e-mail: ilywamh@ihanyang.ac.kr

dc magnetron sputter under a basic pressure 3×10^{-7} Torr or better and prepared on Si(111) substrate at 250 °C. The Fe-Ni composite target consisted of Ni (the purity of 99.9%) chips placed on a Fe (the purity of 99.9%) target covering 10% of the disc surface. The Fe-Ni-N layer was deposited in a mixture of argon and nitrogen at a total gas pressure of 2 mTorr. The deposition rate was approximately 1 and 4 nm/min for Cu and Fe-Ni-N, respectively. Two series of samples were prepared; one where the Cu layer thickness (t_{Cu}) was varied from 0.5 to 8 nm and Fe-Ni-N layer thickness (t_{FeNiN}) was fixed at 5 nm, and another with $t_{\text{Cu}} = 2$ nm and t_{FeNiN} was varied from 1 to 15 nm. The number of bilayers (n) was in the range 4~60. The total thickness of the Fe-Ni-N for multilayered films was fixed at approximately 60 nm. The crystal structure was investigated using an X-ray diffraction (XRD) in θ - 2θ geometry. Magnetic hysteresis loops were obtained by vibrating sample magnetometer (VSM). The magnetizations within the temperature range from 20 to 150 K were measured by SQUID magnetometer under the external field ($H = 1$ kG) parallel to the film plane.

3. Results and Discussion

Figure 1(a) and (b) show XRD patterns of multilayered films with various t_{Cu} and t_{FeNiN} , respectively. These

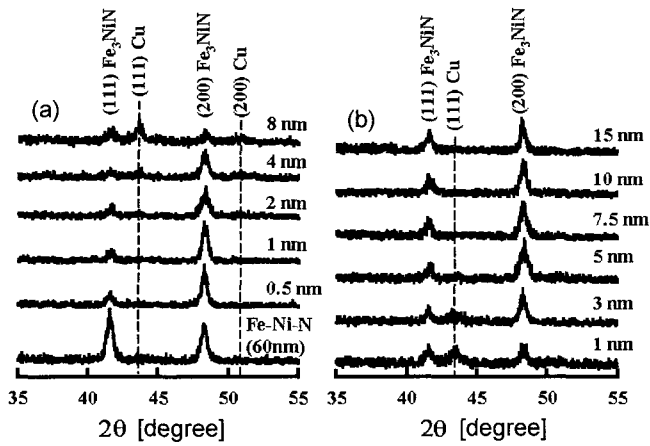


Fig. 1. XRD patterns of multilayered films with various (a) t_{Cu} with a fixed $t_{FeNiN} = 5$ nm (“Fe-Ni-N” in figure means single-layered Fe-Ni-N film) and (b) t_{FeNiN} with a fixed $t_{Cu} = 2$ nm. The dash lines correspond to the positions of the (111) and (200) diffraction peaks of bulk Cu.

patterns show that the Fe_3NiN phases in the multi- and single-layered films having the total thickness of 60 nm are composed of (200) and (111) planes and the Cu crystal phases appear in the ratio of layer thickness, $t_{FeNiN}/t_{Cu} \leq 2.5$. The area ratio, $A_{(200)}/[A_{(111)} + A_{(200)}]$, and lattice constant of the Fe_3NiN obtained using the XRD patterns are plotted in Fig. 2(a) and (b), respectively. As shown in Fig. 2(a) and (b), the area ratio for the multilayered films decreases rapidly with increase of t_{Cu} when the Fe-Ni-N layer thickness is fixed, but the ratio increases up to $t_{FeNiN}/t_{Cu} = 3.75$ (t_{FeNiN} of 7.5 nm and t_{Cu} of 2 nm) and then decreases with increase of t_{FeNiN} when the Cu layer

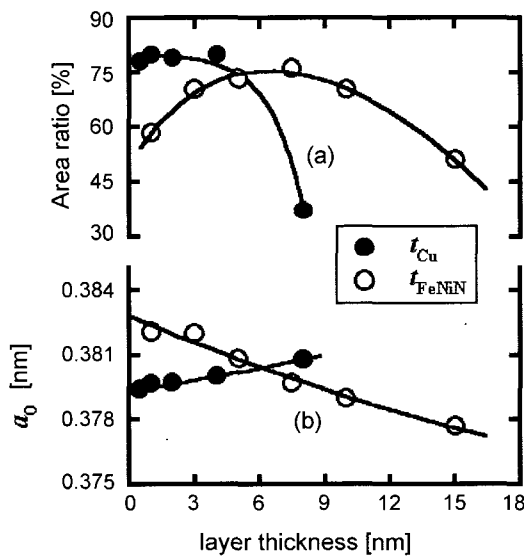


Fig. 2. (a) the area ratio, $A_{(200)}/[A_{(111)} + A_{(200)}]$, and (b) lattice constant as a function of t_{Cu} and t_{FeNiN} , respectively.

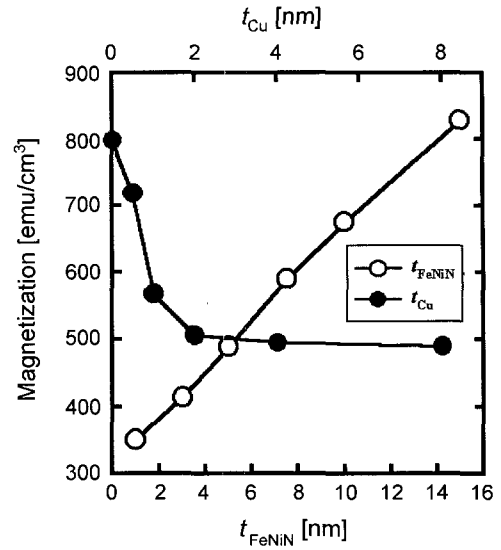


Fig. 3. The magnetization of Fe-Ni-N/Cu multilayered films with a fixed $t_{FeNiN} = 5$ nm as a function of Cu layer thickness (\bullet) and with a fixed $t_{Cu} = 2$ nm as a function of Fe-Ni-N layer thickness (\circ).

thickness is fixed. Also, the lattice constants for both cases vary linearly with increase of the layer thickness.

Figure 3 shows the t_{Cu} and t_{FeNiN} dependence of the magnetization M_s at room temperature for the multilayered films. It is shown that $M_s(t_{Cu})$ decreases rapidly up to about $t_{Cu} = 2$ nm and keeps approximately unchanged when $t_{Cu} > 2$ nm, whereas $M_s(t_{FeNiN})$ increases linearly with increase of t_{FeNiN} when t_{Cu} is fixed. Generally, these behaviors of magnetization have been attributed to the formation of interdiffusion layers near the interface,

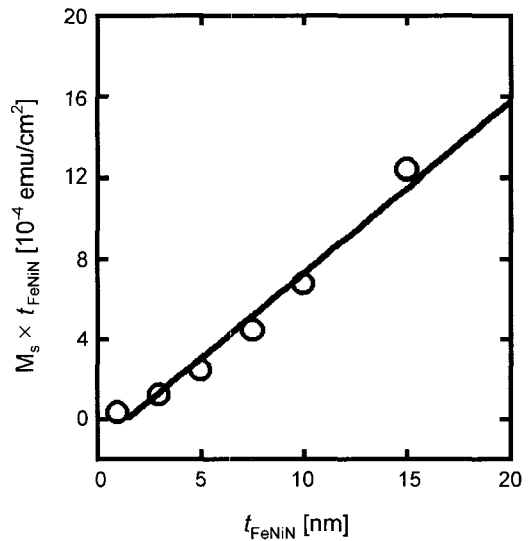


Fig. 4. The variation of the product $M_s \times t_{FeNiN}$ as a function of Fe-Ni-N layer thickness.

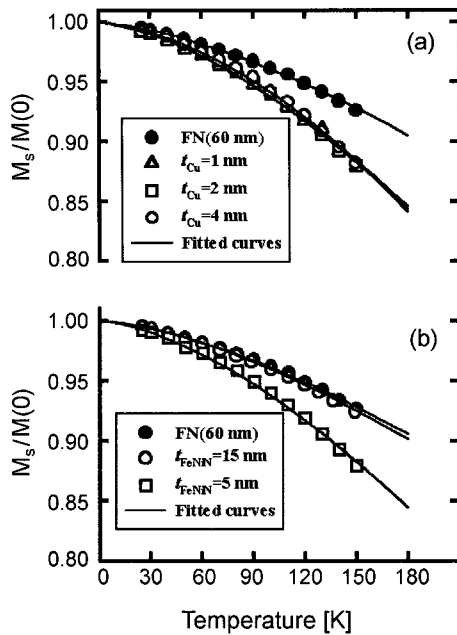


Fig. 5. The temperature dependence of the reduced magnetization for Fe-Ni-N/Cu multilayered films: (a) $t_{\text{Cu}} = 1, 2, 4$ nm with a fixed $t_{\text{FeNiN}} = 5$ nm and (b) $t_{\text{FeNiN}} = 5, 15$ nm with a fixed $t_{\text{Cu}} = 2$ nm.

which leads to a reduction of magnetic moment [4]. The layer thickness dependence of magnetization is well described in terms of the dead layer model by equation $M_s = M_0(1 - 2\delta/t_{\text{FeNiN}})$ [5]. Here M_0 is the magnetization for a bulk Fe-Ni-N alloy and δ the dead layer thickness. Magnetization per unit area, $M_s \times t_{\text{FeNiN}}$, for multilayered films with a fixed $t_{\text{Cu}} = 2$ nm is plotted in Fig. 4 as a function of Fe-Ni-N layer thickness. From linear fits to data, the dead layer thickness d and M_0 are determined to be about 0.7 nm, which is compared with the value of 0.2 nm for the $[\text{Co/V}]_n$ [5], and 850 emu/cm^3 for single-layered films having the total thickness of 60 nm, respectively.

Figure 5(a) and (b) show the reduced magnetization $M(T)/M(0)$ for the some Fe-Ni-N/Cu multilayered films as a function of temperature. Here $M(0)$ is the magnetization at 0 K. It is evident that the values of $M(T)/M(0)$ decrease rapidly with increasing the temperature. The temperature dependence of reduced magnetization is well explained by the Bloch's law, $M(T)/M(0) = 1 - \delta T^{3/2}$ [6]. Here β is a best-fit constant. From the fitting result of the experimental data, the values of $M(0)$ and β are determined within the temperature range of 20~150 K and listed in Table 1 using the law. The spin wave stiffness constant D and the coefficient β are related by $D = 5.79[g/(M(0)\beta)]^{2/3}$ [8]. Here g is the spectroscopic splitting factor referred to the value [7]. In the Fe-Ni-N/Cu multilayered films, β is

Table 1. The layer thickness t_{FeNiN} and t_{Cu} , bilayer number n , the coefficient β of Bloch $T^{3/2}$, and spin wave stiffness constant D for the typical Fe-Ni-N/Cu multilayered films

t_{FeNiN} [nm]	t_{Cu} [nm]	n	$M(0)$ [emu/cm ³]	β [10 ⁻⁵ K ^{2/3}]	D [meVÅ ²]
60	0	0	859.5	3.92	91.1
15	2	4	915.1	4.13	84.4
5	2	12	708.2	6.28	75.7
5	1	12	745.6	6.22	73.6
5	2	12	708.2	6.28	75.7
5	4	12	693.9	6.25	77.6

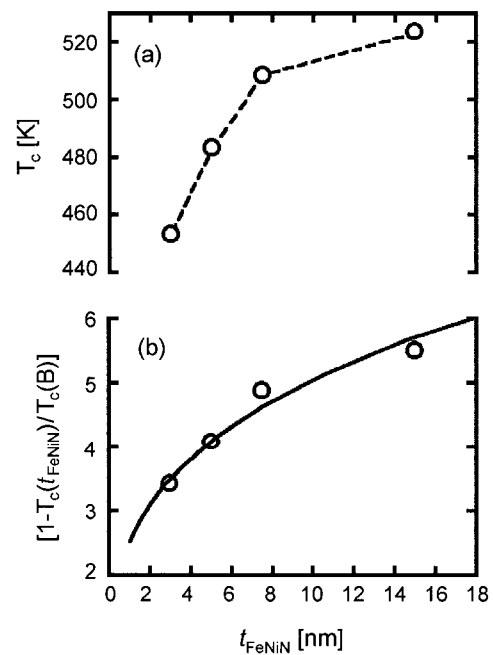


Fig. 6. (a) The Curie temperature T_c as a function of Fe-Ni-N layer thickness for the multilayered films with fixed $t_{\text{Cu}} = 2$ nm and (b) $[1 - T_c(t_{\text{FeNiN}})/T_c(B)]$ vs t_{FeNiN} for the multilayered films with a fixed Cu layer thickness of 2 nm.

large for thinner Fe-Ni-N layers when t_{Cu} is fixed, whereas for thinner Cu layers β is kept at approximately $6.25 \times 10^{-5} \text{ K}^{-3/2}$ independent of t_{Cu} when t_{FeNiN} is fixed. The t_{FeNiN} dependence of Curie temperature T_c for multilayered films with $t_{\text{Cu}} = 2$ nm is shown in Fig. 6(a). As shown in Fig. 6(b), T_c increases with increase of Fe-Ni-N layer thickness. The Fe-Ni-N layer thickness dependence of the Curie temperature is well described by high-temperature series expansion of the Heisenberg model resulted in a power law, $T_c = T_c(B)[1 - \alpha(t_{\text{FeNiN}})^{-\lambda}]$ [9]. Here $T_c(B)$ (=640 K) is the Curie temperature of the bulk Fe_3NiN magnet [10] and α , λ best-fit constants. The λ and α for the films are determined to be 0.31 and 0.32, respectively. The λ value of the films is lower than the

expected theoretical value of $\lambda = 2.00 \pm 0.05$ for cyclic lattice [9].

4. Conclusion

Multilayered $[\text{Fe-Ni-N/Cu}]_n$ films deposited on Si(111) substrate were prepared by the dc magnetron sputtering in order to investigate their structure and magnetic properties. The enhancement of (200) orientation of Fe-Ni-N layers occurred at about $t_{\text{FeNiN}}/t_{\text{Cu}} = 3.75$. The crystallites in Cu layers are observed at the ratio of layer thickness, $t_{\text{FeNiN}}/t_{\text{Cu}}, \leq 2.5$. The magnetization for the films decreases greatly with the Cu thickness within the range of $t_{\text{Cu}} \leq 2$, and increases with increase of t_{FeNiN} . This behavior of magnetization is well described by the dead layer model. Using the model, the dead layer thickness δ is determined to be about 0.7 nm. The temperature dependence of reduced magnetization is well described in terms of the Bloch's $T^{3/2}$ law. The coefficient β for multilayered films is larger than that of single layered film and keeps at approximately $6.25 \times 10^{-5} \text{ K}^{-3/2}$ independent of t_{Cu} . The small λ value obtained by the high-temperature series expansion of the Heisenberg model may mean the decrease of the exchange interaction between the magnetic layers due to the effect of intermixing of Cu. However, the additional studies still must be done.

Acknowledgements

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