

# Investigation of Giant Magnetoresistance in Vacuum-Annealed NiFe/Ag Discontinuous Multilayers

Chang-Min Park<sup>a)</sup>, Young Eok Kim<sup>b)</sup>, and Kyung Ho Shin

*Thin Film Technology Research Center,*

*Korea Institute of Science and Technology, P.O.Box 131, Cheongryang, Seoul, 130-650, Korea.*

(Received 13 May 1997)

The vacuum-annealed Ni<sub>80</sub>Fe<sub>20</sub>/Ag discontinuous multilayers were found to show giant magnetoresistive behaviors comparable to those of corresponding multilayers annealed at atmospheric pressure in a mixture of H<sub>2</sub> and Ar. This vacuum-annealing process will offer potential advantages, enabling a continuous batch process from the deposition to the annealing. Their giant magnetoresistive behaviors were attributed to the magnetostatic coupling that are induced at the edges of the discontinuous magnetic grains. We also present our results about the multilayer patterned into a basic device for the magnetic field sensor.

## I. Introduction

Since the discovery of giant magnetoresistance (GMR) in [Fe/Cr]<sub>n</sub> superlattice [1,2], various GMR materials have received a lot of attention from the magnetic recording industry as promising candidates for new magnetoresistive (MR) field sensor materials. Some of the important parameters for field-sensor applications are the maximum MR ratio, the field sensitivity, the saturation field, and the offset field of a transfer curve. NiFe/Ag discontinuous multilayers discovered by T. L. Hylton et al. [3] display a high magnetoresistance and sensitivity in relatively small magnetic fields. The high field-sensitivity was attributed to the weakness of the magnetostatic interlayer coupling [4] caused by discontinuities among magnetic grains. It is generally accepted that the magnetostatic coupling between adjacent NiFe layers is induced by edge moments located at discontinuities in the NiFe layers. Several studies [3,5] indicate that these discontinuities are produced by grain boundary diffusion of Ag into the NiFe layers during a post-annealing and thus these multilayers require post-deposition annealing to obtain a GMR response. Usually the annealing is performed in a rapid annealing furnace for about 10 min in a H<sub>2</sub>-Ar ambient at temperature of 300°C to 400°C.

In this article, we present an alternative *vacuum-annealing approach* for the annealing process, rather than the previous annealing process [3,5,6]. This new process offers many potential advantages as compared to the earlier technique, making it possible to complete the deposition and annealing process continuously. After annealing, the magnetoresistive behaviors of these vacuum-annealed discontinuous multilayers were examined and interpreted in terms of a interlayer coupling.

In addition, an annealed sample was patterned using standard photographic technique and its magnetoresistive behaviors were also investigated.

## II. Experiment

The multilayer structures consisted of the following stack: Ta (40Å, buffer layer)/Ag (20Å)/Ni<sub>80</sub>Fe<sub>20</sub> (15Å)/[Ag (t<sub>Ag</sub>)/Ni<sub>80</sub>Fe<sub>20</sub>(15Å)]<sub>7</sub>/Ag (20Å)/Ta (100Å, capping layer), where t<sub>Ag</sub> is 40Å (sample set A), 45Å (sample set B), and 50Å (sample set C), deposited on Si substrates. They were prepared by 3-gun DC magnetron sputtering system with a base pressure below 2 × 10<sup>-6</sup> Torr. The natural oxide layers on substrates were not eliminated. The films were deposited at room temperature and in 3 mTorr of Ar, and with a

This work was supported by the Ministry of Information and Communication.

a) Electronic mail: cm@kistmail.kist.re.kr

b) Permanent address; K-MAG Incorporated, 603-82, Chang 2-Dong, Do Bong-Gu, Seoul 132-042, Korea.

magnetic field of 300 Oe during the film growth. The samples were annealed in a vacuum chamber for 10 min with a base pressure below  $2 \times 10^{-6}$  Torr at various temperatures from 320°C to 420°C. Some samples with equal structures were prepared with and without a magnetic field and compared with each other, but they exhibited no difference in GMR behaviors before and after an annealing treatment.

MR transfer curves were measured using the four-point-probe method with maximum magnetic fields of 120 Oe in the film plane. From the transfer curves, two MR characteristics, i. e., the maximum MR ratio ( $MR_{max}$ ) and the offset field ( $H_{off}$ ) were calculated. X-ray diffraction (XRD) scans were obtained in the conventional  $\theta$ - $2\theta$  reflection geometry.

An annealed sample among sample set B ( $t_{Ag} = 45\text{\AA}$ ) was patterned into device using standard photolithographic techniques and then etched by ion beam. The patterned device was then overcoated with Au current pads to eliminate the lead resistance during the MR measurement.

### III. Results and Discussion

Figure 1 shows the variation of magnetoresistance transfer curves with the annealing temperature for three

samples with different Ag thickness of a) 40Å, b) 45Å, and c) 50Å. The magnetoresistance curves of corresponding unannealed films were also shown. No significant GMR response was observed before the annealing treatment, i. e., in the as-deposited stage. After the vacuum-annealing, samples manifested GMR responses and their MR characteristics were found to vary as a function of the annealing temperature. As the annealing temperature is increased, the  $MR_{max}$  first increases then decreases and the  $H_{off}$  increases, regardless of the thickness of Ag layer. For convenience,  $MR_{max}$ 's and  $H_{off}$ 's calculated from transfer curves for three sample sets are summarized in Figure 2. Sample set C ( $t_{Ag}=50\text{\AA}$ ) achieved the largest  $MR_{max}$  of 4 % for an annealing temperature of 350°C. For all sample sets,  $H_{off}$ 's fall into a range from 0 Oe to 25 Oe. It can be found that there exists temperature regions where the  $MR_{max}$ 's remain quite high with slight decreases while the  $H_{off}$ 's experience significant increases. For example, in sample set A, as the annealing temperature is increased from 350°C to 380°C the  $MR_{max}$  decreases from 4 % to 3.5 % while the  $H_{off}$  increases from 1.81 Oe to 5.2 Oe. The  $H_{off}$  often correlates with the field-sensitivity for a given MR curve. In general, a GMR multilayer with a large  $H_{off}$  is likely to have a small field-sensitivity. From the results shown in Figure 2, it is also found that the samples with the thicker Ag layers have

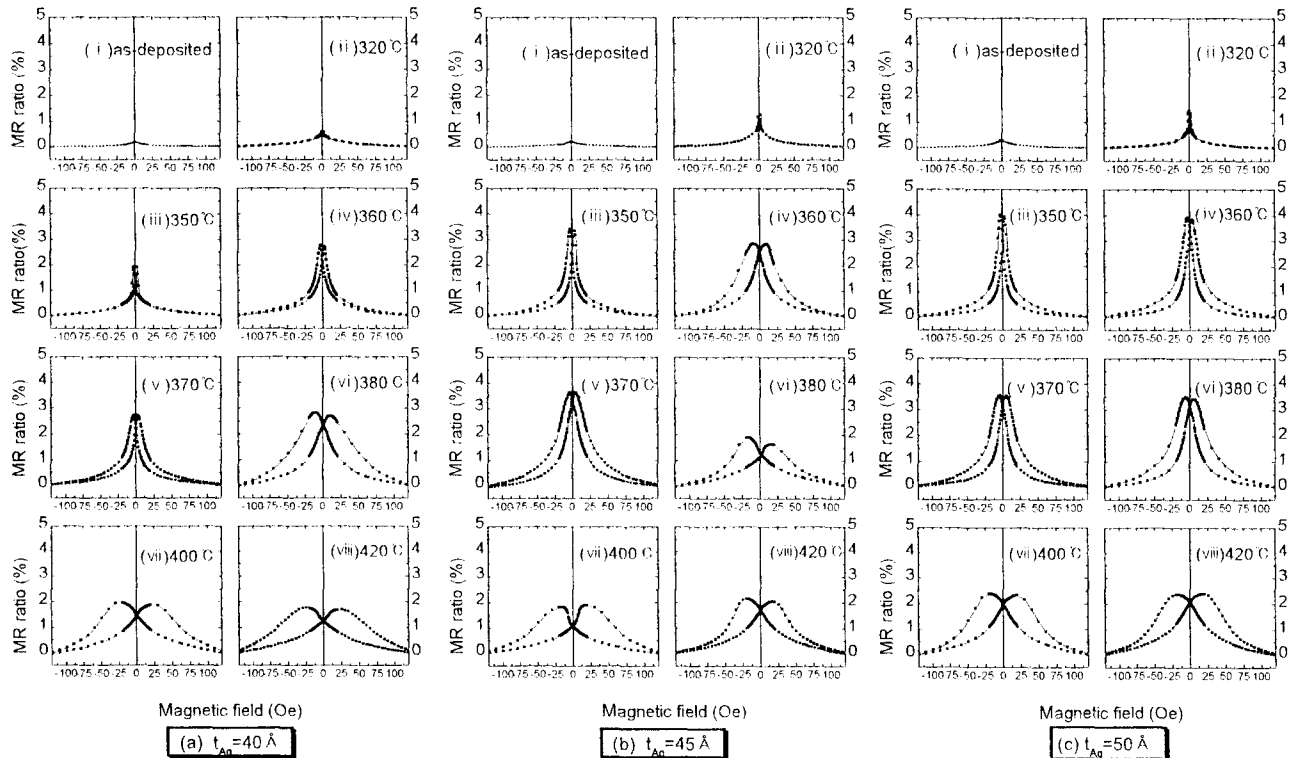


Fig. 1. Magnetoresistance curves for Si/Ta(40Å)/Ag(20Å)/NiFe(15Å)/[Ag( $t_{Ag}$ )/NiFe(15Å)]<sub>7</sub>/Ag(20Å)/Ta(100Å) with (a)  $t_{Ag}=40\text{\AA}$ , (b)  $t_{Ag}=45\text{\AA}$ , (c)  $t_{Ag}=50\text{\AA}$ . Annealing temperatures were (i) as-deposited, (ii) 320°C, (iii) 350°C, (iv) 360°C, (v) 370°C, (vi) 380°C, (vii) 400°C, (viii) 420°C.

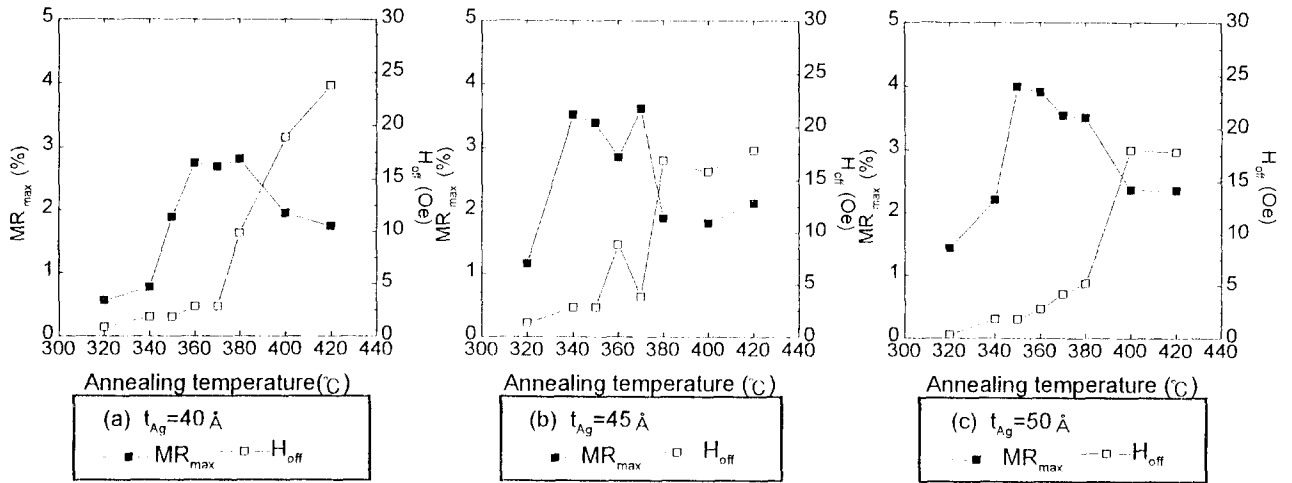


Fig. 2. Maximum magnetoresistance (MR<sub>max</sub>) and offset field (H<sub>off</sub>) versus annealing temperature for Si/Ta(40Å)/Ag(20Å)/NiFe(15Å)/[Ag(t<sub>Ag</sub>)/NiFe(15Å)]<sub>7</sub>/Ag(20Å)/Ta(100Å), where (a) t<sub>Ag</sub>=40Å, (b) t<sub>Ag</sub>=45Å, (c) t<sub>Ag</sub>=50Å.

somewhat larger MR<sub>max</sub>. This result will be discussed in detail below.

The most prominent features of the MR characteristics data are (i) that the vacuum-annealing process yielded a GMR performance comparable to the earlier Ar-H<sub>2</sub> annealing process [3,5,6] and (ii) that the combination of a large MR<sub>max</sub> and small H<sub>off</sub>, which is desirable for the high-performance field sensors application, can be realized through the appropriate control of the vacuum-annealing temperature.

As the physical basis of the magnetotransport properties

of GMR multilayers is the spin dependent scattering [1], the antiparallel alignment of magnetic moments at small field region is essential to the development of GMR in discontinuous multilayers. T. L. Hylton et al. [3] and J. C. Slonczewski [4] proposed that the annealing treatment of NiFe/Ag multilayers causes layer discontinuities, thus creating a magnetostatically induced antiferromagnetic order which results from the magnetostatic energy of adjacent-but-separate magnetic moments. The coupling whose origin is ferromagnetic is reduced dramatically as the NiFe grains are physically separated by the annealing treatment. The relative

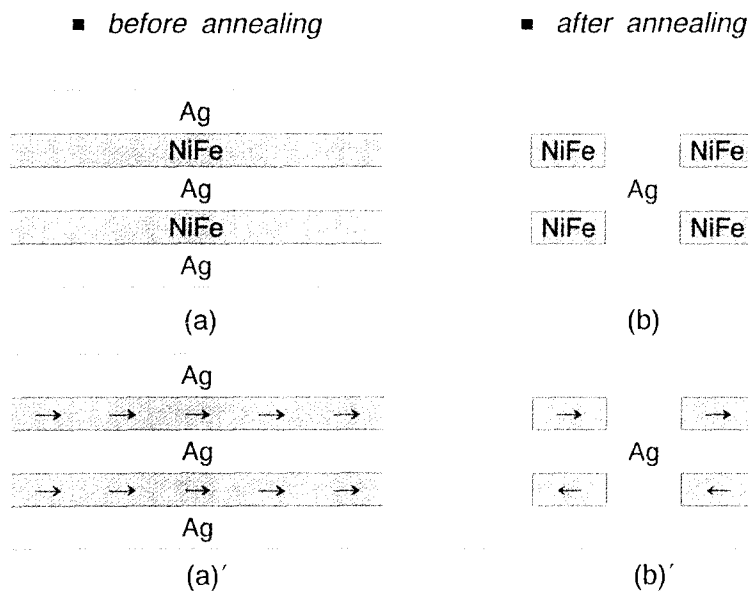


Fig. 3. Idealized diagrams illustrating the development of discontinuity in NiFe/Ag multilayers upon annealing. (a) and (b) represent the simplified cross-section of an as-deposited and annealed multilayer, respectively. Corresponding arrangements of magnetic moments for two cases are shown in (a') and (b').

strength of the two types of couplings dominantly affect the MR characteristics. To overcome any ferromagnetic exchange coupling and destructive effects like the anisotropy dispersion, the strength of the magnetostatic coupling must be strong enough. At the same time, however, it is desirable to make the coupling strength as small as possible for the field sensitivity is inversely proportional with it. Earlier reports suggested that the high field-sensitivity of annealed NiFe/Ag multilayers can be attributed to the weakness of the magnetostatic interlayer coupling.

The anneal-induced magnetic discontinuities can be attributed to the diffusion of Ag into the NiFe layers along the vertical columnar-grain boundaries which normally exists in sputtered structures. Before annealing, a multilayer consisting of immiscible magnetic and non-magnetic layers can be simplified like the idealized diagram shown in Figure 3 (a). When it is annealed at some temperature of 300°C to 400°C, the Ag diffusion along grain boundaries occurs and it causes structural discontinuities between NiFe grains, as shown in Figure 3 (b). Previous transmission electron microscopy (TEM) and X-ray diffraction work [5] has illustrated the separation of the NiFe grains with increasing annealing temperature. The breakup of NiFe layers due to the annealing might be also supported by our XRD results. Although no x-ray diffraction models have been published

for discontinuous multilayers, some researchers could carry out some helpful analyses qualitatively. Figure 4 shows the results of the  $\theta$ - $2\theta$  diffraction scans for selected samples in set A. It is apparent from Figure 4 that as the annealing temperature is increased the two dominant Ag peaks go through gradual changes. A increase in the diffraction intensity was observed, for which a reduction of scattering defects in the film seems to be responsible [3]. As the annealing temperature increases from 340°C to 400°C, the satellite peak adjacent to the Ag (111) main peak of the multilayer tends to weaken relatively, suggesting a degradation of the original layer-by-layer structure of the as-deposited superlattice. Unexpectedly, the satellite peak of the as-deposited film is less distinct than the one annealed at 340°C.

The observed results about the MR characteristics can be explained as follows. The initial increase in the  $MR_{max}$  with the annealing temperature is attributed to the enhanced antiparallel alignment of the zero-field magnetization as the annealing procedure causes the discontinuities of the NiFe grains [3,4]. The decrease in the  $MR_{max}$  for higher annealing temperatures has been understood in terms of a possible bridging of the NiFe grains across the Ag layers, which would increase the effective ferromagnetic coupling between layers [3,5]. The small  $MR_{max}$  for samples with small  $t_{Ag}$  was mentioned in preceding paragraph and can be also understood. Previous studies [3,8] ascribed this Ag thickness effect to the combinations of the changes in the ferromagnetic coupling and the magnetostatic coupling. For thicker  $t_{Ag}$ , the ferromagnetic interlayer coupling would significantly weaken and the magnetostatic coupling is expected to become dominant. For thinner  $t_{Ag}$ , the ferromagnetic coupling is still so strong that the magnetostatic coupling is not dominant enough to provide the moments with a strong antiparallel order. In addition, a

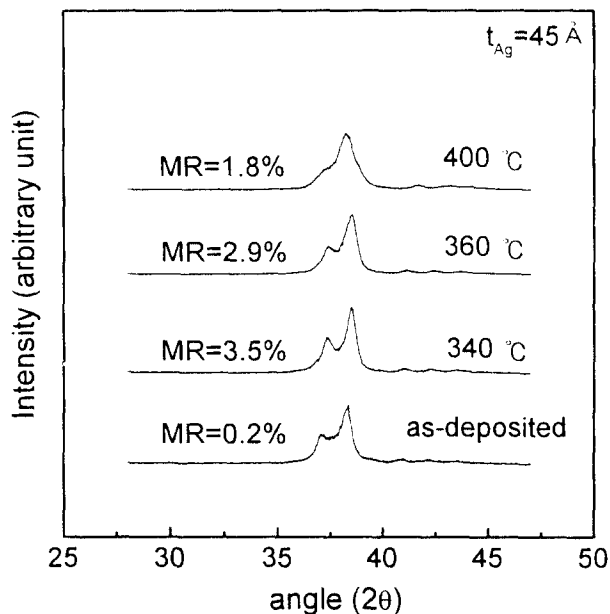


Fig. 4. Normalized high-angle XRD patterns and magnetoresistance values for Si/Ta(40Å)/Ag(20Å)/NiFe(15Å)/[Ag(45Å)/NiFe(15Å)]<sub>n</sub>/Ag(20Å)/Ta(100Å) in the as-deposited condition and after annealing for 10 min at different temperatures. The diffraction intensity (in counts) tends to increase as the annealing temperature increases. Their values are 1150, 2430, 2400, and 3650.



Fig. 5. Optical micrograph for the MR element part of the patterned discontinuous multilayer with the structure of Si/Ta(40Å)/Ag(20Å)/NiFe(15Å)/[Ag(45Å)/NiFe(15Å)]<sub>n</sub>/Ag(20Å)/Ta(100Å).

stronger interlayer ferromagnetic coupling and more probable bridging between NiFe layers resulting from thinner Ag layers are expected.

A vacuum-annealed film with  $t_{Ag}=45\text{\AA}$  (sample set B) was patterned with Au current leads and the track width of 2 mm. In Figure 5, the optical micrograph of the selected area on the patterned device is shown. The element height was about 100  $\mu\text{m}$ . The electrical resistance of the patterned sample was 57  $\Omega$ , while the unpatterned sample has a very small resistance of 0.4  $\Omega$ . Figure 6 shows the magnetoresistance of the patterned device and corresponding unpatterned film. Both samples display the  $MR_{max}$  of  $\sim 3\%$  and  $H_{0.5}$  of  $\sim 2$  Oe. There is no observable difference in GMR responses between two cases and their MR characteristics are nearly identical. These characteristics of the patterned vacuum-annealed NiFe discontinuous multilayers are sufficient for the magnetic field sensor with a large readout signal.

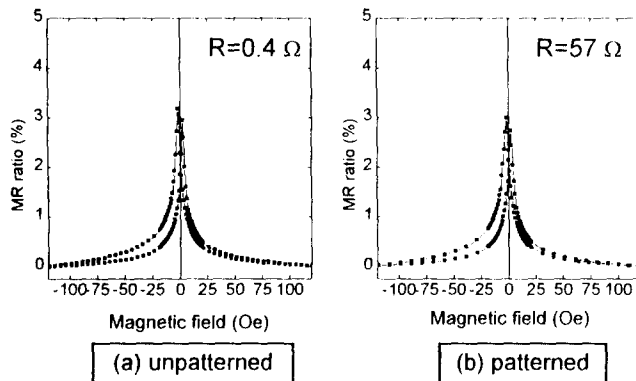


Fig. 6. Magnetoresistance curves for an annealed Si/Ta(40Å)/Ag(20Å)/NiFe(15Å)/[Ag(45Å)/NiFe(15Å)]<sub>7</sub>/Ag(20Å)/Ta(100Å) of a sample (a) before patterning and (b) after patterning. The annealing temperature is 345 °C.

## IV. Conclusion

We have produced vacuum-annealed discontinuous NiFe/Ag multilayers. The vacuum annealing process yielded a GMR response comparable to the conventional Ar-H<sub>2</sub> annealing process. Our XRD analyses support the earlier theoretical model in which the development of GMR is attributed to an antiparallel alignment of moments due to magnetostatics. The MR behavior of the patterned multilayers indicates that these structures may be useful as magnetic field sensors.

## References

- [1] M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, and F. Petroff, Phys. Rev. Lett. **61**, 2472 (1988).
- [2] G. Binasch, P. Grunberg, F. Saurenbach, and W. Zinn, Phys. Rev. B **39**, 4828 (1989).
- [3] T. L. Hylton, K. R. Coffey, M. A. Parker, and J. K. Howard, Science **261**, 1021 (1993).
- [4] J. C. Slonczewski, J. Magn. Magn. Mater. **129**, L123 (1994).
- [5] M. A. Parker, T. L. Hylton, K. R. Coffey, and J. K. Howard, J. Appl. Phys. **75**, 6382 (1994).
- [6] Y. K. Kim and S. C. Sanders, Appl. Phys. Lett. **66**, 1009 (1995).
- [7] A. Yelon, "Interactions in multilayer magnetic films", in Physics of Thin Films, vol. 6, New York; Academic, 1971, pp. 205-300.
- [8] T. L. Hylton, K. R. Coffey, M. A. Parker, and J. K. Howard, J. Appl. Phys. **75**, 7058 (1994).