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# Magnetism of $Co_{1-x}Fe_x$ -NOL in Specular Spin-Valves

Masaaki Doi, Masato Izumi, Hiromi Niu Fuke, Hitoshi Iwasaki, and Masashi Sahashi

Abstract-We investigated the magnetic properties of the Co1-xFex-natural oxidized nano-oxide layer (NOL) in the specular spin-valve system by precise measurement of magnetization at 77 K and M-T curves after field cooling at  $\pm 5$  kOe. The result suggests the antiferromagnetic component behavior of the NOL and its Néel temperature  $(T_N)$  shifts to the higher temperature with increasing Fe composition. Below the  $T_N$ , the antiparallel spin configuration at the pinned layers is stabilized by the exchange bias field induced from antiferromagnetic component of NOL by the field cooling in negative field direction.

Index Terms-Exchange coupling, giant magnetoresistance, nano-oxide layer (NOL), specular reflection, spin-valves.

#### I. INTRODUCTION

• O CONTINUE to apply the giant magnetoresistive (GMR) spin valves sensor as an element of reading heads for the advanced recording densities in hard-disk drive, the increment of head sensitivity is required. To realize high sensitivity in CIP (current into the plane) configuration, the specular GMR spin-valve (SPSV) multilaver is attractive [1]. It is now well known that the introduction of a nano-oxide layer (NOL) in a SPSV multilayer is fairly effective to enhance the MR ratio [2]–[4]. However, the structural and magnetic properties of the NOL itself are still not clear because of its very thin thickness. Recently, Co<sub>90</sub>Fe<sub>10</sub>-NOL was reported as antiferromagnetic and the Néel temperature  $(T_N)$  is around 175 K [5], [6]. Understanding the NOL in an SPSV multilayer is of great interesting from the view point of the specularity of the electron and the magnetic exchange coupling in the spin-valve type multilayers. Beside the enhancement of the MR ratio, strong pinning field is also required for the application. Fig. 1 shows the typical structure of our synthetic SPSV with NOL. In order to keep the superior pinning effect  $(J_{ex})$  from the antiferromagnetic (AFM) PtMn layer to CoFe  $(m_{p4})$  layer, the NOL  $(m_{p3})$  layer should provide strong ferromagnetic coupling between ferromagnetic  $m_{p4}$  and  $m_{p2}$  layers. It is necessary to clarify the magnetic properties of NOL to realize strong ferromagnetic coupling through NOL.

In this paper, we focus on investigating the magnetic properties of the NOL ( $Co_{1-x}Fe_x$ -natural oxidation) in the SPSV

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Fig. 1. Schematic diagram of a synthetic spin valve with specular NOL structure.

system by precise measurement of magnetization from 77 to 293 K (R.T.).

#### **II. EXPERIMENTAL PROCEDURE**

The sequential sputtering method was used for the sample preparation. The typical design of the synthetic SPSV sample was underlayer (NiFeCr/NiFe)/PtMn/CoFe /Ru  $/Co_{1-x}Fe_x(x = 0.08, 0.17, 0.26)$  /Cr /natural oxidation /CoFe /Cu /CoFe /NiFe /Cu /Ta on Si substrate (Fig. 1). The conventional natural oxidation process was carried out in an extra chamber [7]. The thickness of Co<sub>1-x</sub>Fe<sub>x</sub> was 0.6 nm. All of the film were annealed at 270 °C for 1 h under a magnetic field of 7 kOe. Details of the sample preparation were given in [3] and [7]. It should be mentioned that the 0.08 nm of Cr layer was deposited on the surface of  $Co_{1-x}Fe_x(m_{p2})$  for the improvement of the stability of NOL layer [8] while the without oxidation sample was prepared without Cr. The MR ratio and the flat area with high resistance of the R-H curve (Hflat) estimated from the extrapolated cross point of the flat line and decreasing slope line of R-H curve for each sample are listed in Table I. The flat area of the R-H curve corresponse to the antipallalell configration of the magnetization for the free layer  $(m_{f1})$  and  $m_{p4}$ , which is stabilized by the total exchange coupling of  $J_{ex}$ ,  $J_{Ru}$ ,  $J_{NOL}$ , and  $J_{Cu}$ . MR ratio of the without NOL sample was 13%. In comparison, NOL inserted samples show constant

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M. Doi, M. Izumi, and M. Sahashi are with the Department of Electronic Engineering, Tohoku University, Sendai 980-8579, Japan (e-mail: doi@ecei.tohoku.ac.jp).

H. N. Fuke and H. Iwasaki are with the Storage Material and Devices Laboratory, Corporate Research and Development Center, Toshiba Corporation, Kawasaki 212-8582, Japan (e-mail: hiromi.fuke@toshiba.co.jp; hitosi.iwasaki@toshiba.co.jp).

TABLE I MR RATIO AND THE H<sup>flat</sup> ESTIMATED FROM R-H CURVE FOR WITHOUT NOL SV AND  $Co_{1-x}Fe_x$ -NOL SPSV SAMPLE

| Co <sub>1-x</sub> Fe <sub>x</sub> | MR ratio(%) | H <sup>flat</sup> (Oe) |
|-----------------------------------|-------------|------------------------|
| X=0.08<br>Without NOL             | 13.0        | <600                   |
| 0.08                              | 14.3        | 410                    |
| 0.17                              | 14.1        | 390                    |
| 0.26                              | 14.4        | 280                    |



Fig. 2. Magnetization curve of SPSV with  $Co_{1-x}Fe_x$ -NOL at R.T. within the magnetic field of 2.5 kOe. Inset shows an enlargement at the low magnetic field.

value around 14%. Increment of MR ratio is caused by the specular effect of the NOL. However, the H<sup>flat</sup> decreases with increasing of Fe composition. Taking into account that the variation of  $J_{Cu}$ ,  $J_{Ru}$ , and  $J_{ex}$  with increasing Fe composition are negligible, the decrease of H<sup>flat</sup> suggests the ferromagnetic coupling between ferromagnetic  $m_{p4}$  and  $m_{p2}$  layers through NOL ( $m_{p3}$ ) layer is weakened for Fe-rich-NOL. Accordingly, magnetization process through  $J_{NOL}$  should be discussed within relatively low magnetic field. The magnetization was measured by conventional vibrating sample magnetometer (VSM) within the sample size of 10 × 10 mm. The temperature dependence of magnetization was measured in the magnetic field at 3 kOe and 300 Oe with heating rate of 1.2 C°/min. The field cooling effect (FC:H =  $\pm 5$  kOe) was measured from room temperature to 77 K.

### **III. RESULT AND DISCUSSION**

Fig. 2 shows the magnetization curve of SPSV with  $Co_{1-x}Fe_x$ -NOL (x = 0.08, 0.17, 0.26) at R.T. within the magnetic field of 2.5 kOe. The value of the magnetization in this figure is described per volume of the magnetic layer (m). The decrease of the coercive force and the increase of the exchange coupling through Cu ( $J_{Cu}$ ) in the negative sign were observed by the increment of Fe composition (see inset of Fig. 2). These results suggest that the roughness of CoFe( $m_{f1}$ )/Cu/CoFe( $m_{p4}$ ) interfaces are improved by the Fe-rich NOL. This result gives no explanation of the decreasing of H<sup>flat</sup> with the increase of Fe composition. Further, the magnetization was measured at 77 K after the field cooling (FC) for the characterization of



Fig. 3. Magnetization curves at 77 K for the Co<sub>1-x</sub>Fe<sub>x</sub>-NOL and without NOL SPSV sample after FC at H =  $\pm 5$  kOe.

the magnetic properties of NOL and  $J_{NOL}$ . We defined the positive field direction parallel to the pinned field direction of AFM PtMn layer. Fig. 3 shows magnetization curves at 77 K for the Co<sub>1-x</sub>Fe<sub>x</sub>-NOL SPSV sample and the sample without oxidation after FC at  $H = \pm 5$  kOe. No difference of the magnetization curve is observed for the sample without oxidation (Fig. 3). In contrast, the magnetization for the NOL introduced samples show a difference between field cooled at +5 kOe and -5 kOe in the positive applied field area. The smaller values of the magnetization are observed for the FC at -5 kOe in the positive applied field, compared to FC at +5kOe. These results suggest that the  $m_{p2}, m_{p3}$ , and  $m_{p4}$  layers are stabilized in the negative direction by the negative FC process. This result can be explained as follows. If we assume NOL as antiferromagnetic, the high negative field cooling through  $T_{\rm N}$  of the NOL should induce exchange bias effect in negative field direction. This effect leads to higher exchange coupling between  $m_{p2}$  and  $m_{p4}$  in negative field direction, which stabilizes antiparallel spin configuration at the pinned layers. In contrast, the positive field cooling induces exchange bias in the positive direction, which leads to unfavored parallel spin configuration of pinned layer. To confirm the AFM behavior of the NOL, FC M-T curves were measured (Fig. 4). The branch of the M-T curves is observed for Co<sub>1-x</sub> Fe<sub>x</sub>-NOL with x = 0.08 and 0.17, compare to the well agreement for the sample without NOL. It is considered that the branch point (shown by an arrow in the figure) corresponds to the



Fig. 4. M-T curves for the without NOL and  $Co_{1-x}Fe_x$  -NOL SPSV sample after FC at H  $\,=\,\pm5\,$  kOe.

 $T_N$  of the AFM NOL because of the appearance of induced exchange bias field below  $T_N$ . It is clear that the  $T_N$  shifts to the higher temperature with increasing Fe composition.  $T_N$ of x = 0.26 sample is suggested to be above R.T. The reason of the increment of  $T_N$  with increasing Fe composition is not clear from this experiment.  $T_N$  might vary with the thickness of NOL or by the composition of Fe, Co, Cr, and O in the oxide compound. The composition of the oxide compound is likely a CrO<sub>x</sub>, CoO, or Fe<sub>2</sub>O<sub>3</sub>, which may be a disordered phase or a phase substituted with other elements present in the film structure. In addition to the oxide compound, nonoxidized metallic area is partially observed by using our relatively soft oxidation process, which is confirmed by TEM observation [8]. The remained metallic ferromagnetic component maintains ferromagnetic coupling through  $J_{\rm NOL}$ . On the contrary, the paramagnetic component above  $T_{\rm N}$  weakens  $J_{\rm NOL}$  which leads the decrease of the stability of antipallalell configration of the magetization between  $m_{\rm f1}$  and  $m_{\rm p4}$  layer. The existence of the AFM component at R.T. might be one possibility for the explanation of the rapid decrease of  $H^{\rm flat}$  for Fe-rich sample. Further systematic measurements which include Mössbauer spectroscopy are now in progress to confirm more details of the structural and magnetic properties of the Co<sub>1-x</sub>Fe<sub>x</sub>-NOL.

## IV. SUMMARY

The magnetic properties of the  $Co_{1-x}Fe_x$ -natural oxidized NOL in the specular spin-valve (SPSV) system was investigated by precise measurement of magnetization at 77 K and M-T curves after field cooling at  $\pm 5$  kOe. The field cooling effect is observed. This result suggests that the antiferromagnetic component exists in the NOL and its Néel temperature ( $T_N$ ) shifts to the higher temperature with increasing Fe composition. To realize both high specularity and stable ferromagnetic coupling through  $J_{NOL}$  in the synthetic SPSV, the NOL should be uniformly ferromagnetic without paramagnetic or antiferromagnetic component.

#### REFERENCES

- [1] W. F. Egeloff Jr., P. J. Chen, C. J. Powell, M. D. Stiles, R. D. McMichael, J. H. Judy, K. Takano, and A. E. Berkowitz, "Oxygen as a surfactant in the growth of giant magnetoresistance spin valves," *J. Appl. Phys.*, vol. 82, p. 6142, 1997.
- [2] Y. Kamiguchi, H. Yuasa, H. Fukuzawa, K. Koi, H. Iwasaki, and M. Sahashi, *Dig. INTERMAG'99*, Seoul, Korea, May 1999, pp. 18–21.
- [3] H. Fukuzawa, K. Koi, H. Tomita, H. N. Fuke, H. Iwasaki, and M. Sahashi, "Specular spin-valve films with an FeCo nano-oxide layer by ion-assisted oxidation," *J. Appl. Phys.*, vol. 91, p. 6684, 2002.
- [4] N. Hasegawa, F. Koike, K. Ikarashi, M. Ishizone, M. Kawamura, Y. Nakazawa, A. Takahashi, H. Tomita, H. Iwasaki, and M. Sahashi, "Nano-oxide-layer specular spin valve heads with synthetic pinned layer:Head performance and reliability," *J. Appl. Phys.*, vol. 91, p. 8774, 2002.
- [5] J. O. Ventura, J. B. Sousa, M. A. Salgueiro da Silva, P. P. Freitas, and A. Veloso, "Anomalous magnetoresistance behavior of CoFe nano-oxide spin valves at low temperatures," *J. Appl. Phys.*, vol. 93, p. 7690, 2003.
- [6] J. O. Ventura, J. B. Sousa, P. P. Freitas, and A. Veloso, "Impact of the magnetism of nano-oxide layers on the GMR effect in specular spin valves," *J. Magn. Magn. Mater.*, 2004, to be published.
- [7] H. Fukuzawa, K. Koi, H. Tomita, H. N. Fuke, Y. Kamiguchi, H. Iwasaki, and M. Sahashi, "NOL specular spin-valve heads using an ultrathin CoFe free layer," *J. Magn. Magn. Mater.*, vol. 235, p. 208, 2001.
- [8] H. N. Fuke, H. Iwasaki, and M. Sahashi, "working paper,", 2004, to be published.