

# The Properties of Spin Valves with a Partially Oxidized Fe or CoFe Ultra-Thin Layer Inserted in the Magnetic Layers

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We successfully enhanced the performance of a spin valve by inserting an ultra-thin layer of partially oxidized Fe in the pinned and free layers. With the exchange bias field kept large, the spin valve reached a GMR of 12%, which corresponded to a 55% increase in GMR when we compared it with that of spin valves without any inserted layer. The layer of partially oxidized Fe was more effective for improving the properties of the spin valve than the layer of partially oxidized  $\text{Co}_{90}\text{Fe}_{10}$ . Considering all the results, we can contribute the significant improvement to the combined effect of the modified local electronic structures at the Fe impurities and the enhanced spin-dependent reflections at the  $\alpha\text{-Fe}_2\text{O}_3$  phase in the magnetic layer.

**Key words** : spin valve, giant magnetoresistance, ultra-thin layer, oxidation

## 1. Introduction

Giant magnetoresistance (GMR) spin valves have been used as read elements in recording heads for high storage capacity due to their high sensitivity and small noise in reading bits of hard disk drives [1]. The effort to achieve the high performance spin valves has been extensive and has over the past decade resulted in a rate of increase of over 60% per year in the areal density of magnetic recording. Because the GMR value (or equivalently the sheet resistance change,  $\Delta R_s$ ) is directly proportional to the performance of a spin valve, the GMR value has been the most important parameter determining the spin-valve head performance, and the aim in the development of the head has been to increase the GMR value. A GMR value of ~10% is typical in spin valves used in the drive. The ever-increasing need for higher areal density of magnetic disk drives will demand a spin-valve read head with a GMR value (or  $\Delta R_s$ ) higher than ever before.

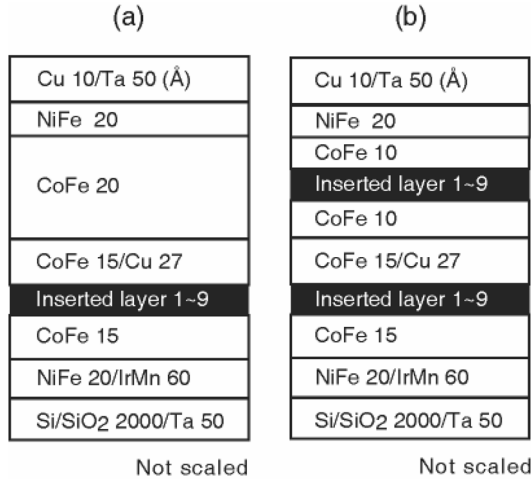
One attempt in the quest to improve a spin valve's performance has been to apply an ultra-thin layer of impurities to pinned and free layers of a spin valve [2, 3]. The ultra-thin layer is made by inserting impurities inside the magnetic layers of a spin valve, and the impurities can act as spin-dependent scatterers, depending on their

localized density of states. Our previous experiments showed that an ultra-thin layer of Ni, Cu, or Fe in a spin valve with Co magnetic layers increased the GMR due to enhanced spin-dependent scattering at the impurities [4]. Another attempt has been to increase specular reflections at magnetic layers by inserting the ultra-thin oxide layer into them, which results in a high probability of spin-dependent scattering at the interfaces [5]. Our recent experiments have revealed that an ultra-thin oxide layer in the  $\text{Co}_{90}\text{Fe}_{10}$  ferromagnetic layer indeed increased the GMR. In particular, an  $\alpha\text{-Fe}_2\text{O}_3$  phase played an important role in enhancing the properties of the spin valves [6]. Those two encouraging results were our motivation to study the effect of an ultra-thin layer of partially oxidized Fe inside the magnetic layer with the aim of further enhancing the GMR performance. In this paper, we compare and discuss the effect of an ultra-thin layer of partially oxidized Fe or CoFe inserted in a  $\text{Co}_{90}\text{Fe}_{10}$  pinned and/or free layer on GMR in a spin valve.

## 2. Experimental Procedures

We sputter-deposited three kinds of spin valves on a thermally oxidized silicon wafer in an ultra-high-vacuum DC magnetron sputtering system. They were 1) a spin valve without any inserted layer (we will call this spin valve Conventional Spin Valve, CSV, hereafter), 2) a spin valve with an ultra-thin layer in the pinned layer, and 3) a

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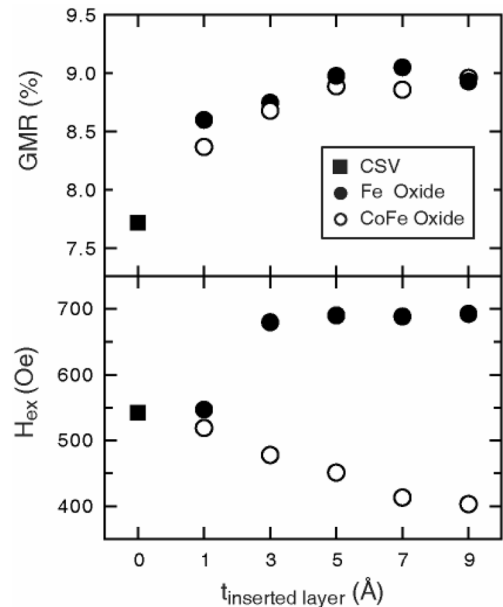
**Fig. 1.** Structures of the spin valve (a) with an ultra-thin layer only in the pinned layer and (b) with ultra-thin layers in both pinned and free layers.

spin valve with ultra-thin layers in both pinned and free layers (shown in Fig. 1). The basic structure of the CSV was Si/SiO<sub>2</sub> 2000/Ta 50/Ni<sub>80</sub>Fe<sub>20</sub> 20/Ir<sub>81</sub>Mn<sub>19</sub> 60/pinned layer (Co<sub>90</sub>Fe<sub>10</sub> 30)/Cu 27/free layer (Co<sub>90</sub>Fe<sub>10</sub> 20/Ni<sub>80</sub>Fe<sub>20</sub> 20)/Cu 10/Ta 50 (Å). The ultra-thin layer of partially oxidized Fe or Co<sub>90</sub>Fe<sub>10</sub> was inserted into the pinned layer (Fig. 1a) or into both pinned and free layers (Fig. 1b). The Co<sub>90</sub>Fe<sub>10</sub> pinned layer was cut in half and three steps were taken to intermittently deposit the ultra-thin layer on it: Co<sub>90</sub>Fe<sub>10</sub> 15/Fe or Co<sub>90</sub>Fe<sub>10</sub> layer/exposure of purified oxygen/Co<sub>90</sub>Fe<sub>10</sub> 15 (Å). The thickness of the inserted layer was varied from 1 to 9 Å by 2 Å increments. We varied the flow rate and the duration of exposure to oxygen to optimize the properties of spin valves. The condition for the oxidation was optimized at an oxygen exposure of 0.002 Pa·s for the CoFe inserted layer, which resulted in partial oxidation of the layer, and we applied the same oxidation condition to the Fe ultra-thin layer. The base pressure was less than 4 × 10<sup>-6</sup> Pa and a magnetic field of ~90 Oe was applied during the deposition in order to induce uniaxial anisotropy. After the deposition, we field-annealed the spin valves at 250 °C for 1 h in vacuum under a magnetic field of 5 kOe at a pressure of less than 7 × 10<sup>-5</sup> Pa. The magnetic and electrical properties of the spin valves were measured by a DC 4-point probe method in the field range of -5 kOe to 5 kOe at room temperature. We used vibrating sample magnetometry (VSM) to measure the saturation magnetic flux density B<sub>s</sub> and carried out near-edge x-ray absorption fine structure (NEXAFS) spectroscopy to obtain the chemical states of the ultra-thin layers.

## Results and Discussion

The GMR and ΔR<sub>s</sub> of the CSV with a 30 Å-thick Co<sub>90</sub>Fe<sub>10</sub> pinned layer were 7.7% and 1.37 Ω, respectively. The exchange bias field H<sub>ex</sub> was 542 Oe. Here, we describe the properties of the spin valves after field-annealing unless we stated otherwise. As we inserted the ultra-thin layer of Fe or Co<sub>90</sub>Fe<sub>10</sub> in the pinned layer and then exposed the layer to oxygen, the GMR increased to over 8.5%. However, the H<sub>ex</sub> showed contrary behavior for the two inserted layers. The H<sub>ex</sub> of the spin valve with the layer of partially oxidized Fe increased to almost 700 Oe (more than 25%) for a layer of as little as 3 Å in thickness, while that with the layer of partially oxidized CoFe decreased as a function of thickness, as shown in Fig. 2. We will discuss the oxidation states of the inserted layer below. The typical properties of the spin valves with each layer are listed in Table 1.

The increase in GMR could be attributed to the enhanced spin-dependent scattering due to 1) modified local electronic structures at the Fe impurities and 2) specular reflections at the α-Fe<sub>2</sub>O<sub>3</sub> impurities in the ultra-thin layer [7]. We think that the large H<sub>ex</sub> additionally contributed to the increase in GMR for the case of an ultra-thin layer of partially oxidized Fe. However, the



**Fig. 2.** Variations in the GMR and H<sub>ex</sub> with the ultra-thin layer inserted only in the pinned layer of the spin valves: The closed squares (■) indicate the CSV with 30 Å-thick Co<sub>90</sub>Fe<sub>10</sub>, the closed circles (●) spin valves with the ultra-thin layer of partially oxidized Fe, and the open circles (○) spin valves with the ultra-thin layer of partially oxidized Co<sub>90</sub>Fe<sub>10</sub>.

**Table 1.** Typical properties of spin valves with different ultra-thin layers

Property	CSV	Partially oxidized CoFe layer (5 Å)	Partially oxidized Fe layer (5 Å)
GMR (%)	7.7	8.9	9.0
$\Delta R_s$ ( $\Omega$ )	1.37	1.50	1.45
$R_s$ ( $\Omega$ )	17.71	17.37	16.10
$H_{ex}$ (Oe)	542	451	691

contribution of  $H_{ex}$  should not be dominant because the insertion of the 1 Å-thick layer increased the GMR but did not change the  $H_{ex}$ . For reference, as we increased the thickness of the CoFe pinned layer without oxygen exposure, the properties were not significantly enhanced but the tendency was similar to that of the spin valve with the ultra-thin layer of partially oxidized CoFe (not shown). Therefore, we focus our discussion on the spin valves with the partially oxidized ultra-thin layers.

Of particular interest is the contrary behavior of the  $H_{ex}$  of the spin valves with the different layers. As the thickness  $t$  or saturation magnetization  $M_s$  of a pinned layer increases, the  $H_{ex}$  is expected to be decreased since the relation can be expressed by

$$H_{ex} = J_K / (M_s \cdot t)_{pinned}, \quad (1)$$

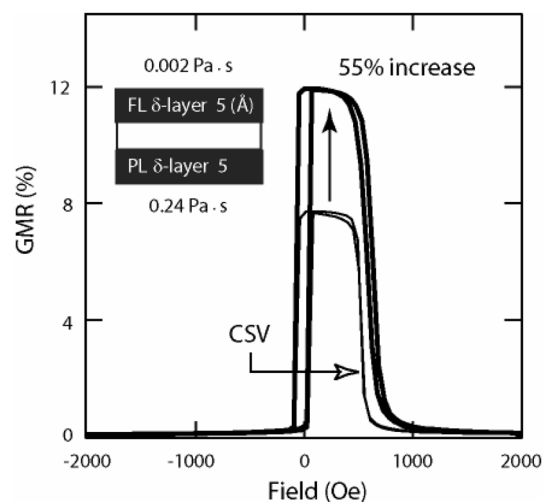
where  $J_K$  is the exchange coupling strength. Equation (1) indeed describes well the behavior of the  $H_{ex}$  of a spin valve with the layer of partially oxidized  $\text{Co}_{90}\text{Fe}_{10}$ . However, in the case of the layer of partially oxidized Fe, the behavior was the opposite: the  $H_{ex}$  increased with the insertion of the layer even though the insertion increased the value of  $(M_s \cdot t)_{pinned}$ . Other researchers, too, have observed such an increase in the  $H_{ex}$  [8]. As they increased the Fe content in a  $\text{Co}_{1-x}\text{Fe}_x$  alloy pinned layer, the  $H_{ex}$  increased. However, in their case there was a tradeoff in the form of a lower GMR, which is different from our result. Our spin valves showed a significant increase in both GMR and  $H_{ex}$ .

Note that before the field-annealing, the as-deposited spin valves with the layer of partially oxidized Fe had shown the  $H_{ex}$  that decreased with the thickness as is the case for partially oxidized CoFe, which is consistent with (1). Therefore, it is conceivable that the increase in the  $H_{ex}$  could be caused by a structural modification due to intermixing of small amounts of Fe into  $\text{Co}_{90}\text{Fe}_{10}$  through the field-annealing. An increase in Fe content in CoFe alloys is known to increase the magnetic anisotropy [9], and this can lead to an increase in the  $J_K$  and thereby an increase in the  $H_{ex}$ .

To obtain structural information in other ways, we

compared the saturation magnetic flux density  $B_s$  of the pinned layers by VSM, and carried out NEXAFS spectroscopy as we had done before [6]. The  $B_s$  of the pinned layer with the 5 Å-thick layer of partially oxidized Fe was nearly the same (~21 kG) before and after thermal treatment, and the  $B_s$  without the layer was ~19 kG. This indicates that the layer inserted in the pinned layer did not exist as a continuous layer but rather existed as impurities inside the  $\text{Co}_{90}\text{Fe}_{10}$  matrix. We believe that local enrichment of Fe in the pinned layer may be responsible for the increase in  $B_s$ . The increased  $B_s$  also indicates that the inserted ultra-thin layer was not fully oxidized, which was also confirmed by NEXAFS [7]. We found by NEXAFS spectroscopy that the Fe was in part oxidized to  $\alpha\text{-Fe}_2\text{O}_3$  (The phase was evidenced by the difference between the energy of the main peak and that of the peak in the shoulder of the  $L_{III}$ -edge,  $\Delta E$ ) while most Co in the pinned layer did not react with oxygen and was left as a metallic phase, which is consistent with the experiment reported [6].

When we inserted an additional ultra-thin layer of partially oxidized Fe into the free layer, the GMR was also increased, and it was optimized at the 5 Å-thick layer. The GMR value was 9.6% and the  $\Delta R_s$  was 1.56  $\Omega$ . On the other hand, the additional ultra-thin layer of partially oxidized CoFe in the free layer did not enhance the properties of the spin valve significantly. To increase the GMR further, we increased the oxidation exposure of the ultra-thin layer of Fe in the pinned layer while



**Fig. 3.** Large GMR increase for the spin valve with the ultra-thin layers of partially oxidized Fe in both pinned and free layers (thick line). The thin line indicates the GMR loop of the CSV, which does not have any inserted layers. The inset illustrates the ultra-thin layers in both pinned and free layers with the optimized oxygen exposure.

keeping it at 0.002 Pa·s for the inserted layer of Fe in the free layer. Fig. 3 shows our optimized spin valve with the ultra-thin layers (thick line). The GMR reached its maximum at an oxidation exposure of 0.24 Pa·s for the pinned layer. The GMR,  $\Delta R_s$ ,  $R_s$ , and  $H_{ex}$  of the spin valve were 12%, 2.0  $\Omega$ , 16.5  $\Omega$ , and 632 Oe, respectively. The  $H_{ex}$  was still larger than that of the CSV by 17%. In the case of the ultra-thin CoFe layer, the over-exposure of oxygen to the layer could not improve the properties of the spin valve at all: Rather both GMR and  $H_{ex}$  were significantly decreased at large oxygen dose, which makes the spin valve unusable for applications. The decreases were consistent with our previous experiment [6]. The spin valve with the insertion of partially oxidized Fe is very robust and attractive for applications.

### Conclusion

We successfully enhanced the performance of a spin valve by inserting an ultra-thin layer of partially oxidized Fe in the pinned and free layers. With the  $H_{ex}$  kept large, the spin valve reached a GMR of 12% and a  $\Delta R_s$  of 2.0  $\Omega$  which correspond to a 55% increase in GMR and a 44% increase in  $\Delta R_s$  compared to the GMR and  $\Delta R_s$  of the spin valve without the layer. The ultra-thin layer of partially oxidized Fe was far more effective for improving the properties of the spin valve than that of partially oxidized  $\text{Co}_{90}\text{Fe}_{10}$ . Considering all the results, we can contribute such substantial improvement to the combined effect of the modified local electronic structures at the Fe impurities and the enhanced spin-dependent reflections at the  $\alpha\text{-Fe}_2\text{O}_3$  phase in the ultra-thin layer.

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### References

- [1] D. E. Heim, R. E. Fontana, Jr., C. Tsang, V. S. Speriosu, B. A. Gurney, and M. L. Williams, IEEE Trans. Magn. **30**, 316 (1994).
- [2] P. Zhan, J. Binder, I. Mertig, R. Zeller, and P. H. Dederichs, Phys. Rev. Lett. **80**, 4309 (1998).
- [3] C. H. Marrows and B. J. Hickey, Phys. Rev. B **63**, 220405(R)1 (2001).
- [4] J. Hong and Hitoshi Kanai, Jpn. J. Appl. Phys. **42**, 7296 (2003).
- [5] (a) W. F. Egelhoff, Jr., P. J. Chen, C. J. Powell, M. D. Stiles, R. D. McMichael, J. H. Judy, K. Takano, and A. E. Berkowitz, J. Appl. Phys. **82**, 6142 (1997); (b) J. Hong, K. Noma, E. Kanda, and H. Kanai, Appl. Phys. Lett. **83**, 960 (2003).
- [6] J. Hong, Y. Lee, M. K. Lee, H. J. Song, H. Shin, Y. Yoo, and J. Suh, Appl. Phys. Lett. **83**, 4803 (2003).
- [7] J. In, Y. Han, S. Kim, J. Shim, and J. Hong, submitted for publication (2006).
- [8] J. R. Childress, M. J. Carey, R. J. Wilson, N. Smith, C. Tsang, M. K. Ho, K. Carey, S. A. MacDonald, L. M. Ingall, and B. A. Gurney, IEEE Trans. Magn. **37**, 1745 (2001).
- [9] T. Burkert, L. Nordstrom, O. Eriksson, and O. Heinonen, Phys. Rev. Lett. **93**, 0272031 (2004).