Philip W. T. Pong, Moshe Schmoueli, and William F. Egelhoff Jr.

Magnetic Materials Group, Materials Science and Engineering Laboratory, National Institute of Standards and Technology, Gaithersburg, MD 20899 USA

The technique of pre-oxidation of the bottom CoFe electrode prior to deposition of Al_2O_3 was previously shown to be capable of suppressing intermixing at the ferromagnet/Al interface and removing partial shorts near Al grain boundaries. In this paper, we studied the influence of the plasma oxidation on the pre-oxidized magnetic tunnel junctions (MTJs). In general, the tunneling magnetoresistance (TMR) is independent of the plasma oxidation time whereas the resistance-area product (RA) increases with it. However, for the pre-oxidized MTJs with very thin Al_2O_3 (Al thickness < 0.7 nm prior to plasma oxidation), the TMR decreases with plasma oxidation while the RA increases with it.

Index Terms-Intermixing, magnetic tunnel junction, orange-peel coupling, plasma oxidation, pre-oxidation.

I. INTRODUCTION

AGNETIC TUNNEL JUNCTIONS (MTJs) are very useful in ultra-low magnetic field sensors. In order to realize this potential application, the MTJs must exhibit small orange-peel coupling. Therefore, a technique which can reduce the orange-peel coupling would be of paramount importance for building ultrasensitive magnetic field sensors with MTJs.

The ferromagnet/oxide interface plays a critical role in determining the tunneling magnetoresistance (TMR), the junction resistance, and the magnetic properties. Surface oxidation achieved by exposing the bottom ferromagnetic layer to oxygen before depositing the Al layer helps to suppress intermixing at the ferromagnet/Al interface and reduce orange-peel coupling [1]-[6]. For a discussion of pre-oxidation and orange-peel coupling in MTJs, see [4] and [5]. Initially, the deposited Al takes oxygen away from the ferromagnetic oxide to form Al₂O₃. After the oxygen is consumed and Al is deposited, the Al layer is oxidized in plasma to form Al_2O_3 barrier [1], [7]. This approach was previously demonstrated on spin valves [3] and it provides a very promising means of suppressing orange-peel coupling in MTJs [8], [9]. In this paper we studied the influence of the plasma oxidation on the pre-oxidized MTJs to obtain deeper understanding of the phenomenon.

II. EXPERIMENT

The thin films were deposited on thermally oxidized silicon wafers by dc magnetron sputtering in an ultrahigh vacuum chamber with a base pressure of 2×10^{-8} Pa. The metal films were deposited at room temperature in 0.2 Pa argon. Pre-oxidation of CoFe layer was carried out with 0.1 Pa oxygen for 30 s prior to the Al metal deposition. The oxide barrier layer was made by first depositing a thin Al metal and then oxidizing it in oxygen plasma (0.4 Pa argon, 0.2 Pa oxygen). The plasma oxidation time and the thicknesses of the Al metal were varied. The sample structure is substrate 2.5 Ta\5 Au\10 IrMn\4 CoFe\X Al: oxidized \5 CoFe\5 Ta\7 Ru. (X was varied,



Fig. 1. Plot of TMR versus RA for pre-oxidized samples with different plasma oxidation time.

all units in nm). Electrical characterization was performed with current-in-plane-tunneling (CIPT) technique.

III. RESULTS AND DISCUSSION

We have explored the influence of the plasma oxidation duration on the pre-oxidized MTJs. These samples consist of a wedge of Al₂O₃ barrier with Al thicknesses of around 0.3-2.3 nm. Five different samples of this structure with the plasma oxidation time of 15, 30, 60, 120, and 240 s, respectively, were made. Fig. 1 displays the plot of the TMR versus resistance-area product (RA) of these samples. The approximate Al thickness $(\pm 30\%)$ of each data point is specified in Table I. The highest TMR were achieved with similar Al thicknesses of around 1.25 nm (these thicknesses are asterisked in Table I). Comparing the data points with the highest TMR, the sample with 240 s plasma oxidation time exhibits the largest RA on the order of $10^5 \Omega \mu m^2$ while the sample with 15 s shows the smallest RA on the order of $10^4 \Omega \mu m^2$. In general, the RA increases with the plasma oxidation time. This implies that longer plasma oxidation time increases the thickness of the tunneling barrier probably because the underlying CoFe, even though being pre-oxidized, was further oxidized by the oxygen plasma. Previously we proposed that the pre-oxidation should passivate the CoFe surface and prevent the further oxidation

Digital Object Identifier 10.1109/TMAG.2008.2001329

Color versions of one or more of the figures in this paper are available online at http://ieeexplore.ieee.org.

Sample 1: 15 sec	6.2 8.5 9 9.3 9.8 10.8* 11.4 12
plasma oxidation	12.3 12.6 13.2 13.8 15.4
Sample 2: 30 sec	8.7 12.3 12.7 13.5 14.3 15.1*
plasma oxidation	15.9 16.7 17.5 18.3 19.1 19.5 23.1
Sample 3: 60 sec	3.4 4.5 5.7 6.8 7.9 10.2* 11.3
plasma oxidation	12.5 13.5 14.7 15.8 17 19.2
Sample 4: 120 sec	2.8 3.4 3.9 4.5 5.1 5.7 6.2 6.8
plasma oxidation	7.4 7.9 8.5 9.1 11.3* 13 13.5
-	14.7 15.3 16.4 17 17.5 18.1
Sample 5: 240 sec	5.1 5.7 6.2 6.8 7.4 7.9 9.1 10.8
plasma oxidation	12.5* 15.3 17 18.1
*data points with the high	ghest TMR

in the CoFe [4]. However, apparently the oxygen plasma is so reactive that it can attack the underneath of the pre-oxidized CoFe and oxidize the CoFe deeper. The maximum TMR for these samples are all around 35% which means TMR is more or less independent of the plasma oxidation time and the RA. This further evidences that the increase of the RA is due to the formation of CeFe oxide instead of the complete oxidization of the underoxidized Al because otherwise, the TMR would have improved due to the elimination of the partial shorts in the Al layer. The thickness of the Al oxide barrier plays an important role in TMR. The TMR attains its highest value at an optimum Al thickness. Therefore, the plasma oxidization time must be optimized together with the Al thickness in order to obtain the highest TMR.

Next we investigated the influence of the plasma oxidation on the pre-oxidized MTJs with very thin Al₂O₃ tunnel barrier (Al < 0.7 nm prior to plasma oxidation). Thin Al₂ O₃ MTJs are of particular interest because low RA is necessary for MTJs working as a hard-disk readhead. A pre-oxidized MTJ sample with the same structure was made. However, there are double wedges in this sample: on one edge, the Al deposited varies from 0.4 nm to 0.7 nm; on the other edge, the plasma oxidation duration was varied from 25 s to 175 s. This was controlled by placing an adjustable shutter very close to the sample to separate it from the oxygen plasma and gradually removing the shutter and exposing the sample to the plasma. As we can observe from Fig. 2, the TMR in general increases with the thickness of Al which is because thicker Al₂O₃ barrier contains less pinholes. Interestingly, rather than being unaffected, the TMR of this thin Al₂O₃ MTJ sample decreases with the plasma oxidation duration. This is probably because the CoFe underneath the Al oxide barrier was excessively oxidized by the oxygen plasma and the quality of the barrier interface was severely deteriorated. On the other hand, as shown in Fig. 3, the RA increases with the thickness of Al and the plasma oxidation duration as expected. The thicker Al made the resulting Al₂O₃ barrier thicker and the longer plasma oxidation further oxidized the underneath CoFe, both leading to thicker oxide barrier and thus larger RA.



Fig. 2. Plot of TMR versus plasma oxidation time and thickness of Al for the pre-oxidized sample with double wedges. The TMR values are specified on top of each bar.



Fig. 3. Plot of RA versus plasma oxidation time and thickness of Al for the pre-oxidized sample with double wedges. The RA values are specified on top of each bar.

IV. CONCLUSION

It was previously shown that pre-oxidation could suppress the orange-peel coupling and it is a very useful technique for fabricating high-quality MTJs. In this work, the influence of the plasma oxidation on pre-oxidized MTJs was investigated. We found that in general the longer plasma oxidation time will increase the RA but it does not affect the TMR. Pre-oxidation of CoFe and an optimum Al oxide barrier thickness should be used to achieve the highest possible TMR. However, when the thickness of Al_2O_3 is very thin (Al < 0.7 nm prior to plasma oxidation), the TMR decreases with the plasma oxidation time instead of increases with it. Therefore, for thin Al oxide barrier MTJs, pre-oxidation of CoFe and minimum plasma oxidation should be used.

TABLE I Approximate AL Thickness (nm) for the Plot of TMR Versus RA in Fig. 1

REFERENCES

- W. F. Egelhoff, Jr., P. J. Chen, R. D. McMichael, C. J. Powell, R. D. Deslattes, F. G. Serpa, and R. D. Gomez, "Surface oxidation as a diffusion barrier for Al deposited on ferromagnetic metals," *J. Appl. Phys.*, vol. 89, p. 5209, 2001.
- [2] J. Wolfman, D. Mauri, T. Lin, J. Yang, and T. Chen, "Low resistance Al₂O₃ magnetic tunnel junctions optimized through in situ conductance measurements," *J. Appl. Phys.*, vol. 97, p. 123713, 2005.
- [3] K. Sin, S. Funada, M. R. Gibbons, W. Jensen, C. Hiner, X. Shi, and H. C. Tong, "Effects of CoFe surface oxidation on tunneling magnetore-sistance," in *IEEE Int. Magnetics Conf. (INTERMAG) Eur. 2002. Dig. Tech. Papers*, p. ER10.
- [4] W. F. Egelhoff, Jr., R. D. McMichael, C. L. Dennis, M. D. Stiles, A. J. Shapiro, B. B. Maranville, and C. J. Powell, "Suppression of orange-peel coupling in magnetic tunnel junctions by preoxidation," *Appl. Phys. Lett.*, vol. 88, p. 162508, 2006.
- [5] W. F. Egelhoff, Jr., R. D. McMichael, C. L. Dennis, M. D. Stiles, A. J. Shapiro, B. B. Maranville, and C. J. Powell, "Preoxidation as a general approach to suppressing orange-peel coupling in magnetic tunnel junctions," *IEEE Trans. Magn.*, vol. 42, p. 2664, 2006.

- [6] 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, "Oxygen as a surfactant in the growth of giant magnetoresistance spin valves," *J. Appl. Phys.*, vol. 82, p. 6142, 1997.
- [7] J. D. R. Buchanan, T. P. A. Hase, B. K. Tanner, P. J. Chen, L. Gan, C. J. Powell, and W. F. Egelhoff, Jr., "Intermixing of aluminum-magnetic transition-metal bilayers," *J. Appl. Phys.*, vol. 93, p. 8044, 2003.
- [8] J. D. R. Buchanan, T. P. A. Hase, B. K. Tanner, C. J. Powell, and W. F. Egelhoff, Jr., "Interface intermixing and in-plane grain size in aluminum transition-metal bilayers," *J. Appl. Phys.*, vol. 96, p. 7278, 2003.
- [9] J. D. R. Buchanan, T. P. A. Hase, B. K. Tanner, P. J. Chen, L. Gan, C. J. Powell, and W. F. Egelhoff, Jr., "Anomalously large intermixing in aluminum-transition-metal bilayers," *Phys. Rev. B*, vol. 66, p. 104427, 2003.

Manuscript received March 03, 2008. Current version published December 17, 2008. Corresponding author: P. W. T. Pong (e-mail: ppong@nist.gov).