Oxidation Processes in Magnetic Tunnel Junctions

L. Seve,¹ W. Zhu,¹ B. Sinkovic,¹ J. W. Freeland,² I. Coulthard,² W. J. Antel Jr.,² S. S. P. Parkin³

¹ Department of Physics, University of Connecticut, Storrs, CT, U.S.A.

² Experimental Facilities Division, Argonne National Laboratory, Argonne, IL, U.S.A.

³ IBM Research Division, Almaden Center, San Jose, CA, U.S.A.

Introduction

Recent success in fabrication of magnetic tunneling junctions (MTJ) with magnetoresistance (MR) above 40% at room temperature has generated intense interest because of their potential application as efficient magnetic random access memory (MRAM) elements.¹ MTJs are composed of two ferromagnetic (FM) electrodes separated by an insulating (I) layer with typical structures being $Co_{1-x}Fe_x/Al_2O_3/Co_{1-x}Fe_x$. It has been reported by several authors¹⁻⁵ that these record high MR values are obtained only after annealing the MTJ to 200-300°C, which leads to increase in MR by ~30% when compared to as-grown structures. This is suggestive of partial oxidation of the ferromagnetic electrode in the as-grown sample resulting in degradation of the MR, which is subsequently removed upon annealing.

We use x-ray absorption spectroscopy (XAS) to address this issue by directly monitoring the chemical states of FM in a $Co_{84}Fe_{16}/Al_2O_3$ bilayer before and after annealing. XAS spectra at the L₃-edge of 3d transition metal elements and their oxides exhibit different, easily distinguishable features, which makes XAS an ideal tool to investigate the oxidation states of FM electrodes in MTJ. We employ a bilayer consisting of an Al wedge in order to perform systematic study of the effect of annealing as a function of Al_2O_3 thickness. This is of particular importance since the Al_2O_3 barriers of MTJ are fabricated by oxidation of Al layers and an optimal FM/I interface should have all of the Al and none of the FM oxidized Our results demonstrate clearly that the Co and Fe oxide observed in the as grown structures can be completely removed by annealing to 250°C for a certain range of Al thicknesses.

Methods and Materials

The samples for the XAS experiments were fabricated at the IBM Almaden Research Center by magnetron sputtering. A 200 Å layer of Co₈₄Fe₁₆ was first deposited on a silicon wafer followed by an aluminum wedge of 5-20 Å thickness deposited over a distance of 23 mm using computer-controlled mask. The Al wedge was then plasma oxidized in an oxygen atmosphere to form optimally a 13 Å oxide layer, and was subsequently capped by 40 Å of Ti to protect it from further oxidation in the air. XAS measurements were performed at the Advanced Photon Source high-resolution intermediate x-ray spectroscopy facility (beamline 2-ID-C) located at Argonne National Laboratory. Co and Fe L-edges and Al K-edge absorption spectra were collected in total yield mode at locations of different Al thickness along the wedge. The photon energy resolution was 100 meV for Co and Fe measurements and 250 meV for Al XAS with an x-ray spot size of 0.25 mm x 2 mm. The sample was annealed in situ with e-beam bombardment to 250°C.

Results and Discussion

Figure 1 shows the Co L edge absorption as a function of position along the Al wedge. For thin Al thicknesses there are clear signs of oxidation. For this study we will track the pre-edge





FIG. 2. Co oxide feature intensity vs. Al thickness.

oxide feature as a function of both Al thickness and annealing temperature. The data for Fe are not shown as they track with the behavior of the Co.

Figure 2 shows the intensity of the pre-edge peak (776.3 eV) of the Co L₃ spectra before (black circles) and after (red squares) annealing, respectively. A large reduction of Co oxide after annealing is clearly seen. For as-grown films and for a given oxidation time, the Co oxide is expected to be present only if the Al layer is thinner than the corresponding oxidation depth. The annealing to 250°C reduces the amount of Co oxide throughout the wedge (red squares). Most notably the Co oxide completely disappears at the FM/I interface for Al thicknesses larger than ~9 Å. Since an optimal MTJ uses a barrier made of 10-15 Å Al, this figure nicely demonstrates how annealing completely removes the interfacial Co oxide present after the fabrication of these junctions. Upon subsequent annealing to 350°C for another 1/2 hour, no further change occurred in the XAS spectra of the Co or Fe L edge. This is in agreement with recent experiment⁵ in which the degrading of MTJ performance after high-temperature annealing was attributed to the interfacial diffusion at an exchange-biased antiferromagnetic/ferromagnetic interface rather than a change at FM/I interfaces.

We now turn to study of the insulator layer (Al₂O₃) of the sample before and after annealing (250°C) by examining a set of Al K-edge absorption spectra shown in Fig. 3. Similar to previous analysis of Co data, the spectra are recorded at different positions of the wedge corresponding to different Al layer thickness. Only the pre-anneal data are shown as there is little change due to annealling. Included for comparison in Fig. 3 are data for an oxidazed Al foil and sapphire. A comparison of the spectra as a function of thickness shows several features worth noting. First, the edge position of the oxide features moves to higher energy at thinner Al layer thickness, which can be attributed to excess oxide in the layer (O/Al ratio >1.5). Second, the oxide is very disordered as noted by comparing with the single-crystal phase (sapphire) and that of the polycrystalline Al foil. Last, note that there is very little signal for the Al metal, which might indicate lateral motion of the excess oxygen in the layer.

Acknowledgments

Use of the Advanced Photon Source was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. W-31-109-Eng-38.



FIG. 3. Al K edge absorption spectra.

References

- ¹S.S.P. Parkin et. al., J. Appl. Lett. 85, 5828 (1999).
- ² R.C. Sousa et. al., Appl. Phys. Lett. 73, 3288 (1998).
- ³ M. Sato, H. Kikuchi, and K. Kobayashi, J. Appl. Phys. **83**, 6691 (1998).
- ⁴ R.C. Sousa et. al., J. Appl. Lett. 85, 5258 (1999).
- ⁵ S. Cardoso et. al., Appl. Phys. Lett. 76, 610 (2000).