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Citation for published version (APA):

DOI:
10.1063/1.1856291

Document status and date:
Published: 01/01/2005

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:
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Download date: 08. Oct. 2023
Mn diffusion and the thermal stability of tunneling spin polarization


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(Received on 10 November 2004; published online 5 May 2005)

We examine the role of Mn diffusion in the thermal stability of tunneling spin polarization $P$ by directly measuring $P$ of Al/AIO$_x$/Co/FeMn and Al/AIO$_x$/Co$_{90}$Fe$_{10}$/FeMn junctions using superconducting tunneling spectroscopy (STS). We confirm Mn diffusion in our junctions using x-ray photoelectron spectroscopy after an ultrahigh vacuum 500 °C anneal. Surprisingly, and in contrast to the current belief, no drop in $P$ is observed using STS. Therefore, though Mn diffuses significantly, it cannot be solely responsible for the drop in tunneling magnetoresistance observed after postdeposition anneals above 300 °C. © 2005 American Institute of Physics.

Magnetic tunnel junctions (MTJs) demonstrate large room temperature tunneling magnetoresistance1 (TMR) which makes them suitable for application in magnetoresistive random access memories (MRAMS).2 A standard TMR stack generally consists of an antiferromagnet/ferromagnet/insulator/ferromagnet multilayer, where the antiferromagnetic (AF) layer is used to pin the direction of the magnetic moment of the adjacent ferromagnetic layer, and the ferromagnet-insulator-ferromagnet (F/I/F) sandwich is responsible for the TMR effect. The AF layer generally contains a Mn alloy (e.g., Fe$_{50}$Mn$_{50}$, Pt$_{50}$Mn$_{50}$) to allow device operation at elevated temperatures (above 150 °C).3 Presently, one of the major areas of research in MTJs is the miniaturization of these elements for application in MRAM and their integration with CMOS (complementary metal oxide semiconductor) processing.4 Successful integration of the MTJ in MRAM requires the device to be thermally resistant against standard high temperature CMOS processing steps (400–450 °C).5 However, postdeposition anneals of tunnel junctions below 300 °C enhances TMR, and those above 300 °C lead to its severe degradation.6 The physical mechanism behind this drop in TMR after anneals above 300 °C is not yet completely understood. Several causes have been suggested for this drop, among which Mn diffusion from the AF layer into the F electrode and towards the F/I interface is believed to play the principle role.6,7

In this paper, we combine a study of the thermal stability of tunneling spin polarization ($P$) based on the superconducting tunneling spectroscopy (STS) technique8 with an x-ray photoelectron spectroscopy (XPS) analysis of Mn diffusion. The spin polarization of the tunneling electrons in the F/I/F junction is the fundamental parameter responsible for the TMR effect9 and is very sensitive to the interfacial density of states at the F/I/F interfaces.10 Any change in $P$ should ensue from chemical and/or morphological changes at or near the F/I interface. We demonstrate that $P$ in our Al/AIO$_x$/Co/FeMn and Al/AIO$_x$/Co$_{90}$Fe$_{10}$/FeMn junctions is thermally stable up to 500 °C, even when Mn diffuses towards the I/F interface.

Detection of the influence of Mn diffusion in a conventional tunnel junction stack, for example, one consisting of FeMn/Co/AIO$_x$/Co/Ta, is difficult to probe experimentally with XPS, since the escape depth of the photoelectrons is much less than the standard thickness of the top Co and Ta layers. Therefore, we deposited Al/AIO$_x$/Co(200 Å)/FeMn(100 Å)/Co(200 Å) layers on silicon substrates using dc magnetron sputtering (base pressure <10$^{-8}$ mbar), in situ annealed them at 500 °C in ultra high vacuum (UHV, pressure <10$^{-8}$ mbar during anneal) for 30 min, and then studied them with in situ XPS (Al $K_{α}$). Logically, we assert that if Mn diffuses to the surface of the 200 Å thick top Co layer, it should also diffuse towards the AIO$_x$/Co interface below the FeMn layer. Also, any significant Mn accumulation near the surface of 200 Å thick Co layer should be detectable by XPS. The AIO$_x$ barrier layer (which is 10–22 Å thick) was formed by partially plasma oxidizing the 40 Å Al bottom electrode for 200 seconds.

Figure 1(a) shows the XPS spectra for Mn electrons measured before and after a 500 °C anneal. As expected, as-deposited samples show no evidence of Mn peaks in the intensity scan, confirming the absence of Mn at or near the surface of the 200 Å thick top Co layer. However, after the anneal, two explicit peaks appear near the energies of known Mn p-level peaks, which for pure Mn, are expected to be at 638.8 eV (2p$_{3/2}$) and 650.05 eV (2p$_{1/2}$), respectively.12 This result is a proof of Mn diffusion from the FeMn layer towards the surface of the 200 Å thick top Co layer. Careful examination of the spectra show that the Mn peaks are shifted to higher binding energies, evincive of an oxidized state of Mn in the Co layer. In our sample, the 2p$_{3/2}$ Mn oxide peak is found to be around 641.4 eV. The literature values for the 2p$_{3/2}$ peaks of various manganese oxides are found to lie between 641–642 eV.12,13 The formation of Mn oxide near the surface of the top Co layer is purely due to the background partial pressure of oxygen in the chamber, which is induced by the degassing of adsorbed oxygen from the sample plate during the anneal. Although we do not com-

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completely exclude the possibility of oxygen driven Mn diffusion (reported for a 30 Å thick CoFe layer by Ref. 14) towards the surface of the top Co layer, we strongly believe that our 200 Å thick top and bottom Co layers should inhibit such a process.

Figure 1(b) indicates the corresponding XPS spectra for Co 2p_{3/2} (778.1 eV) and 2p_{1/2} (793.0 eV) peaks. It can be seen that the spectral intensity of peaks for the as-deposited sample is much larger than when the sample has been annealed, confirming the decrease of Co near the surface of the layer. However, no oxidation of Co is evident, since there is no distinguishable shift in the peaks. Co 2p_{3/2} peaks are expected at 778.1 eV, and those for its oxides are expected between 779.8–780.2 eV. These results are in accordance with the fact that the (negative) free energy of formation is lowest for cobalt oxides, intermediate for manganese oxides, and highest for aluminum oxides, making it difficult for Co to oxidize in the presence of Al and Mn. Figure 2 shows the Mn to Co spectral intensity ratio in our XPS measurements as a function of anneal temperature. The ratio is calculated by removing the background in the measurements, fitting the peaks to expected Mn and Co peaks, and then deriving the area under the curve. It is notable that the Mn/Co ratio at or near the surface of the 200 Å top Co layer starts increasing around 300 °C, which has been reported as the onset temperature for TMR collapse.¹⁵

To measure the effect of Mn diffusion on P, cross-stripped tunnel junctions with and without FeMn were prepared similar to the XPS samples. A 60 Å Ta capping layer was added on top. The junctions have an area of 400 μm × 400 μm and a resistance-area product of roughly 10⁸ kΩ μm². Current-voltage (I–V), and conductance-voltage (dI/dV-V) characteristics are measured in a four-terminal configuration using a standard lock-in technique. Our Al bottom electrodes become superconducting at about 2.2 K (Ref. 8) and have critical fields of 4.5 T. Figure 3 shows representative measurements of an Al/AlOₓ/Co and an Al/AlOₓ/Co₀ₓFe₁₀ junction at 0.3 K. In zero field the conductance reflects the superconducting density of states with its sharp peaks at the band-gap edge. In a magnetic field the conductance shows four maxima due to the Zeeman splitting of the density of states. The P is directly revealed by the asymmetry in the maxima² and extracted by a fit of the model based on the Maki theory.¹⁶ This model accounts for the effect of orbital-depairing and spin-orbit scattering on the superconducting density of states.³ The extracted P for Co (38% ± 1%) and Co₀₉Fe₁₀ junctions are in fair agreement with earlier work.¹⁸

FIG. 1. (Color online) In situ XPS (Al Kα) intensity spectra for (a) Mn peaks observed on the 200 Å top Co layer in the Al/AlOₓ/Co/FeMn(100 Å)/Co(200 Å) stack before and after in situ postdeposition UHV anneal at 500 °C. The dash-dotted (Mn) and solid (Mn₂O₃) lines indicate the expected 2p_{3/2} and 2p_{1/2} peak locations. (b) Co peaks for the same sample, the intensity of the Co 2p_{3/2} and 2p_{1/2} peaks decreases after the anneal.

FIG. 2. Mn/Co intensity ratio measured in our XPS spectra as a function of postdeposition anneal temperature.

FIG. 3. (Color online) Conductance of Al/AlOₓ/Co (a) and Al/AlOₓ/Co₀₉Fe₁₀ junction (b) at 0.3 K in zero field and an applied field of several Tesla. The solid lines are theoretical fits.


In summary, we investigated Mn diffusion in Al/I/F junctions and its effect on the P of the electrons tunneling from the ferromagnet. Contrary to the current belief, we have shown that P in Al/AlOx/Co and Al/AlOx/Co90Fe10 junctions is thermally stable up to 500 °C, despite the likelihood of Mn diffusion towards the I/F interface.

This research was supported by NanoNed, a nanotechnology program of the Ministry of Economic Affairs, and by the Dutch Foundation of Fundamental Research on Matter (FOM).

3For example, with respect to industrial and automobile sensors, see the German BMBF project “Magnetoelectronic” specifications led by Robert Bosch GmbH.