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**Mn diffusion and the thermal stability of tunneling spin polarization**


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We examine the role of Mn diffusion in the thermal stability of tunneling spin polarization $P$ by directly measuring $P$ of $\text{Al/AlO}_x/\text{Co}/\text{FeMn}$ and $\text{Al/AlO}_x/\text{Co}_{90}\text{Fe}_{10}/\text{FeMn}$ junctions using superconducting tunneling spectroscopy (STS). We confirm Mn diffusion in our junctions using x-ray photoelectron spectroscopy after a 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.

We demonstrate that $P$ in our $\text{Al/AlO}_x/\text{Co}/\text{FeMn}$ and $\text{Al/AlO}_x/\text{Co}_{90}\text{Fe}_{10}/\text{FeMn}$ 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 $\text{FeMn/Co/AlO}_x/\text{Co/Ta}$, 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 $\text{Al/AlO}_x/\text{Co}(200\text{ Å})/\text{FeMn}(100\text{ Å})/\text{Co}(200\text{ Å})$ 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$_\alpha$). Logically, we assert that if Mn diffuses to the surface of the 200 Å thick top Co layer, it should also diffuse towards the AlO$_x$/Co interface below the FeMn layer. Also, any significant Mn accumulation near the surface of 200 Å thick top Co layer should be detectable by XPS. The AlO$_x$ 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 (2$p_{3/2}$) and 650.05 eV (2$p_{1/2}$), respectively. 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 2$p_{3/2}$ Mn oxide peak is found to be around 641.4 eV. The literature values for the 2$p_{3/2}$ peaks of various manganese oxides are found to lie between 641–642 eV. 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 introduced by the degassing of adsorbed oxygen from the sample plate during the anneal. Although we do not com-

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No oxidation of Co is evident, since there is no oxidation of Co near the surface of the sample is much larger than when the sample has been annealed, confirming the decrease of Co.

These results are in agreement with the fact that the free energy of formation of Mn 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.

To measure the effect of Mn diffusion on P, cross-striped 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 10^5 kΩ μm^2. 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/AlO_x/Co and an Al/AlO_x/Co_{05}Fe_{10} 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 maxima and extracted by a fit of the model based on the Maki theory. This model accounts for the effect of orbital-depairing and spin-orbit scattering on the superconducting density of states. The extracted P (38% ± 1%) for Co and (48% ± 1%) Co_{05}Fe_{10} junctions are in fair agreement with earlier work.

Figure 4 shows P as a function of postdeposition anneal temperature for junctions which do (closed squares) and do not (open circles) contain an FeMn layer. Remarkably, P does not suffer any degradation in response to anneal up to 500 °C for both types of ferromagnets, independent of the presence of FeMn. Also, the absolute values of P for a particular ferromagnet does not change before and after the anneal, irrespective of Mn diffusion into the layers. This result is in qualitative agreement with the work of Kim and Moodera, who report that Mn concentrations as high as 30% in Al/AlO_x/Co_{05}Mn_{1−y} junctions have only a weak negative effect on P. In addition, the anneals do not affect other junction parameters such as junction resistance and the superconducting band gap of our Al electrode. The thermal robustness of P above 300 °C (evident in Fig. 4) is in sharp contrast with the effect of postdeposition annealing on the TMR of MTJs. In order to clarify this apparent contradiction further experiments are indispensable.

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.
FIG. 4. (Color online) $P$ measured after an in situ 30 min postdeposition UHV anneal in Al/AlO$_x$/Co and Al/AlO$_x$/Co$_{90}$Fe$_{10}$ junctions which do (closed squares) and do not (open circles) contain an FeMn layer. This data shows that $P$ is not affected by the presence of an FeMn layer on top of the ferromagnet.

We now turn our attention to another interesting observation. Typically, anneals below 300 °C enhances TMR. One explanation of this enhancement in TMR is an improvement of $P$ due to migration of oxygen from the bottom F electrode into the AlO$_x$ barrier. This stoichiometric redistribution of oxygen in the barrier results in a sharper interface and improved barrier properties. Consequently, the barrier height should increase, and spin-independent tunneling should decrease, both leading to higher TMR. Another explanation which concerns both electrodes is the possibility of a change in the ferromagnet structure at the interface after the anneal. However, our measurements do not show an increase in $P$ when the Al/I/F stack is annealed at 100–300 °C, which indicates that there is no change at the I/F interface or in the structure of the F electrode which contributes to enhancement of $P$, and subsequently, TMR. Therefore, the second explanation, i.e., change in the F electrode structure is not supported by our measurements.

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/AlO$_x$/Co and Al/AlO$_x$/Co$_{90}$Fe$_{10}$ junctions is thermally stable up to 500 °C, despite the likelihood of Mn diffusion towards the I/F interface.

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