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Intrinsic thermal robustness of tunneling spin polarization in Al/Al₂O₃/Co junctions

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Through a direct observation, based on the spin-polarized tunneling technique, we explore the thermal stability of tunneling spin polarization in Al/Al₂O₃/Co junctions. Thermal robustness of this parameter, which is of key importance for magnetic tunnel junction performance, is established for *in situ* postdeposition anneal temperatures up to 500 °C. This stability is consistent with detailed *in situ* x-ray photoelectron spectroscopy measurements on the Al₂O₃/Co system which show no structural changes during the anneal. Our results imply that, for comparable magnetic tunnel junction devices, thermal stability is not limited by intrinsic processes in the Al₂O₃ barrier and its interfaces. With *ex situ* postdeposition annealing in an Ar-atmosphere, which leads to severe degradation of the spin polarization above 250 °C, we demonstrate that the spin polarization is extremely vulnerable to diffusion of impurities. © 2004 American Institute of Physics.

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Magnetic tunnel junctions (MTJs) have a high potential for industrial applications¹ because of their large room temperature tunnel magnetoresistance (TMR) effect.² MTJs are under development to utilize the TMR effect in magnetic random access memories.³ Postdeposition annealing of MTJs typically leads to a severe degradation of the TMR effect above 200 °C. Since incorporation of MTJs into existing semiconductor technology requires thermal stability of TMR up to 400 °C, the influence of a postdeposition annealing process is currently under investigation.^{4–6}

TMR originates from the tunneling spin polarization determined by the atomic and electronic structure at both barrier/ferromagnetic interfaces. This crucial role of the interface structure has been suggested in several experimental papers, e.g., using dusting layers,^{7–9} alternative barrier/electrode combinations,¹⁰ and by theoretical calculations.^{11,12} The tunneling spin polarization from a single barrier/ferromagnet interface can be measured directly with the spin-polarized tunneling (SPT) technique,¹³ in which the Zeeman-split superconducting density of states in a thin Al counter electrode is used as a spin detector.

In this article, we present an investigation of the thermal stability of tunneling spin polarization based on SPT. We show that the spin polarization in Al/Al₂O₃/Co tunnel junctions is stable up to 500 °C. Additionally, with *in situ* x-ray photoelectron spectroscopy (XPS) measurements we can exclude structural changes of the Al₂O₃/Co system during the postdeposition anneal. These results imply that the thermal stability of TMR in comparable MTJs is not limited by the intrinsic stability of the Al₂O₃ barrier and its interfaces. Furthermore, we demonstrate that annealing under high-vacuum conditions is of crucial importance.

Our tunnel junctions are prepared by dc magnetron sputtering (base pressure <10^{−9} mbar) through metal shadow masks on glass substrates at room temperature. The Al₂O₃

tunnel barrier is obtained by partially oxidizing a 40 Å Al bottom electrode with an *in situ* oxygen plasma (10^{−1} mbar, 5 W) for 200 s. The plasma-oxidation is followed by deposition of 200 Å Co top-electrodes in a cross-stripe configuration which are finally capped with 60 Å Ta. The resulting junctions have an area of 400 μm×400 μm and a resistance-area product of roughly 10⁵ kΩ μm².

For the spin-polarized tunneling measurements the junctions are cooled to 0.3 K in a sorption-pumped ³He-cryostat. Current–voltage (*I*–*V*), and conductance (*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 and have critical fields of 4.5 T. Figure 1(a) shows a representative conductance measurement in zero field and a field of 3.0 T performed on a junction which is not annealed. The spin polarization is extracted by fitting the model based on the Maki theory^{14–16} to the conductance curves. This model accounts for the effect of orbital-depairing and spin-orbit scattering on the superconducting density of states.¹³ We find a spin polarization of 39±1%, with the error margin determined by sample-to-sample variation, which is in fair agreement with earlier work.^{17,18}

To investigate the thermal stability of the spin polarization, junctions are postdeposition *in situ* annealed for 30 min while the pressure was in the low 10^{−9} mbar range. The junctions start to show metallic shorts for anneal temperatures exceeding 500 °C, well above the thermal stability requirement for device implementation. Probably, this is related to thermal stresses in the Al₂O₃ barrier, or other processes, which are, however, outside the scope of this article. For temperatures below 500 °C the junctions are stable without significant changes in the junction resistance. However, this does not exclude delicate structural or chemical changes at the barrier interfaces, which, as mentioned before, are key to the spin polarization. Several thermal processes, possibly affecting the spin polarization, have been proposed.

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FIG. 1. Conductance measurements of Al/Al₂O₃/Co junctions at 0.3 K in zero-field and an applied field of 3 T performed before (a) and after a 500 °C *in situ* postdeposition anneal (b). The solid lines are theoretical fits.

Monsma and Parkin¹⁹ consider oxidation of the ferromagnet at the interface. Closely related, Sousa *et al.*²⁰ investigated the Al₂O₃ barrier in their MTJ structure with Rutherford backscattering and observed a redistribution of oxygen.

To address these issues, we have performed *in situ* XPS on the Al₂O₃/Co system before and after a 500 °C anneal. For careful investigation of the oxidation state of Co at the interface, the Co thickness in this case is only 5 Å. Figure 2(a) shows the XPS intensity in the Al 2s and the Co 3p region measured before annealing. Due to the oxidation state of the Al, the Al lines are chemically shifted with respect to the binding energies measured before the plasma oxidation (indicated by the vertical lines), in accordance with earlier reports which focus on the plasma oxidation of Al.^{21,22} The binding energies of the Co lines coincide with those documented for clean metallic Co. Figure 2(b) shows the spectrum measured after the 500 °C anneal. The Co lines are still located at their original positions indicating that the Co is not oxidized during the anneal. We have also verified that the Co 2p lines, which give more XPS signal and are more sensitive to chemical changes, are not affected by the anneal as well. In order to check the sensitivity for oxidation, the Co is oxidized deliberately revealing clear chemical shifts as shown in Fig. 2(c). These results show that Co at the Al₂O₃/Co interface in our junctions does not reduce to an oxide either during deposition or a 500 °C postdeposition anneal.

To investigate the issue of oxygen redistribution, as suggested by Sousa *et al.*, we have monitored XPS spectra of the O 1s line. In a careful comparison of normal and grazing

FIG. 2. *In situ* XPS study on the chemical stability of the Al₂O₃/Co interface during a 500 °C anneal. The binding energies of the Co lines measured before the anneal (a), after the anneal (b), and after deliberate oxidation (c), indicate that Co is not oxidized during the anneal.

emission spectra (which provides depth resolution), measured before and after the anneal, we could not identify oxygen in different chemical environments, significant changes in the O–Al ratio or a shift of the O 1s binding energy. To gain extra sensitivity for the O 1s line, we have repeated the experiment on another sample without the Co layer. Also in this case we could not observe significant changes. Thus, in contrast with the observation of Sousa *et al.*, we do not observe migration of oxygen in our junctions. This discrepancy may be explained by the fact that our barrier is formed by partial oxidation of a single Al bottom electrode, whereas in the work of Sousa *et al.* the barrier is formed by oxidation of an Al film deposited on top of a CoFe bottom electrode.

Figure 1(b) shows representative conductance measurements of a junction which is postdeposition annealed at 500 °C for 30 min. The spin polarization measured after the anneal is 37 ± 1%, essentially the same as before the anneal which is consistent with the absence of major structural and chemical changes as reported above. It could be argued that the anneal procedure affects the superconducting properties of the ultrathin Al electrode by which the obtained spin polarization is no longer accurate. This is, however, not the case. The superconducting transition temperature and the spin-orbit parameter, which is extracted from the conductivity curves measured in a magnetic field,¹³ are not significantly changed. This thermal stability of the Al superconducting electrode implies that the SPT technique is unambiguously applicable. The present results of an intrinsically stable Al₂O₃/Co system can be regarded as a test case and provides a basis for future research. We suggest that meta-stable systems, such as Al₂O₃/Ni (Ref. 19) or possibly Al₂O₃/Gd, studied with SPT in combination with XPS (and/or ultraviolet photo-electron spectroscopy), provide the opportunity for observing interesting correlations between polarization and barrier/ferromagnet interface structure.

The thermal stability reported here is in sharp contrast

FIG. 3. Spin polarization after *in situ* and *ex situ* postdeposition annealing.

with the severe degradation of the TMR effect above 200 °C observed in comparable MTJ structures (see, for example, Ref. 23). The suggested explanation for this degradation is the diffusion of Mn atoms from exchange-bias layers to the barrier/ferromagnet interfaces. Our results prove that indeed the stability of TMR in Co-based MTJs is limited by extrinsic processes (e.g., diffusion of Mn atoms) and not by intrinsic processes in the barrier and its interfaces.

To demonstrate the influence of impurity diffusion, we have annealed the same Al/Al₂O₃/Co junctions *ex situ* in an Ar-atmosphere instead of a high-vacuum chamber. Despite the fact that the Co electrode is capped with 60 Å Ta, impurities present in the Ar-atmosphere diffuse into the junction structure, which is, probably, occurring predominantly along grain boundaries. As in the case of the high-vacuum anneal, both the junction resistance and superconducting properties of the Al electrode remain unchanged. However, as shown in Fig. 3, the spin polarization starts to degrade severely above 250 °C, reminiscent of the degradation of TMR.²³ The degradation can be explained by contamination of the barrier/ferromagnet interface by foreign atoms,^{7–9} or by local oxidation of the ferromagnet as considered by Monsma and Parkin,¹⁹ and earlier in this article. This direct comparison between the high-vacuum and the Ar-atmosphere anneal confirms that the polarization is vulnerable to diffusion of impu-

rities. Extended anneal studies, based on SPT and Co electrodes containing well-defined impurity layers, further establishing these diffusion processes, will be the topic of future publications.

In summary, we have shown that the tunneling spin polarization in Al/Al₂O₃/Co junctions is intrinsically stable up to 500 °C. *Ex situ* postdeposition annealing in an Ar-atmosphere demonstrates that the polarization is extremely vulnerable to diffusion of impurities.

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