Atomic-scale characterization of a Co/AlO\textsubscript{x}/Co magnetic tunnel junction by scanning transmission electron microscopy

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Analytical electron microscopy has been employed to characterize the localized physical and electronic structure of a Co/AlO\textsubscript{x}/Co magnetic tunnel junction. The tunnel barrier is amorphous alumina with an extensive conduction band tail due to disorder. Both barrier edges are Al terminated and an Al-rich layer exists at the bottom Co/AlO\textsubscript{x} interface. \textit{sp}-\textit{d} hybridization between interfacial Co and Al atoms is observed and it is likely that the interfacial Al is metallic. All of these features are expected to be important to the magnetoresistance behavior of the junction. © 2001 American Institute of Physics.

Magnetic tunnel junctions which consist of two ferromagnetic electrodes separated by a thin (~10 Å) tunnel barrier can exhibit a magnetoresistance of greater than 30\% at 295 K.\textsuperscript{1} Such structures hold substantial potential for application as computer memory and magnetic field sensors. Much about the spin-dependent tunneling process is still not understood. The Juliere model\textsuperscript{2} predicts magnetoresistance based on the bulk spin polarization of the density of states in the two ferromagnetic electrodes. However, experimental values often do not match this model indicating that the detailed nature of the tunnel barrier and/or its interfaces with the ferromagnetic electrodes can play a dominant role in determining the spin polarization of the tunneling current. To investigate this issue we have used scanning transmission electron microscopy (STEM) to characterize the physical and electronic structure of a magnetic tunnel junction system.

The magnetic tunnel junction for this study was grown by dc magnetron sputter deposition on Si, with the layer sequence Si/Co 60 Å/Al 12 Å/O\textsubscript{2} exposure/Co 100 Å. The Si(100) substrate was spin etched with HF to remove the native oxide layer. Co and Al deposition rates were 1.8 and 0.4 Å/s, respectively. During oxygen exposure 99.998\% O\textsubscript{2} was maintained at a pressure of 100 mTorr for 15 min.

Cross-section samples for STEM analysis were prepared with a mechanical wedge-polish method.\textsuperscript{3} They were analyzed using a 100 keV VG HB501A STEM equipped with a McMullan-style parallel electron energy loss spectrometer and a computer-controlled data acquisition system. This STEM has a demonstrated resolution of better than 2 Å.\textsuperscript{4}

Figures 1(a) and 1(b) show bright field (BF) and annular dark field (ADF) images, respectively, of a 200-Å-thick cross-sectioned magnetic tunnel junction sample. Images were taken with a 10.4 mrad objective aperture and 0.4 mrad collector aperture. The growth direction is from left to right, and the thin bright (dark) vertical region in the bright (dark) field image is the AlO\textsubscript{2} barrier. Note the Si lattice visible on the left of the ADF image. Insets show line scans across the tunnel junction for different focal values with roughly equal changes in defocus.

The ADF intensity is due to high-angle Rutherford scattering and varies approximately as $Z^{1.3}$ (Z is the atomic number).\textsuperscript{5} The AlO\textsubscript{2} barrier layer appears fairly uniform in thickness, about 15 Å full width at half maximum. The position of the AlO\textsubscript{2} layer varies with a root-mean-square...
roughness of 1.7 Å. Note that even the best-focused image has a somewhat diffuse barrier profile. This is partly an effect of the layer roughness and does not necessarily indicate substantial intermixing of atomic species.

BF intensity is more difficult to interpret due to its coherent nature. Changing focus results in very different junction profiles due to the appearance of Fresnel fringes. The under-focused image in Fig. 1(a) has deceptively sharp-looking Co/AlO$_x$ interfaces, and the line scan from this image shows the presence of Fresnel fringes. Even the “best” focus (defocus with least Fresnel contrast) shows some fringes. The reciprocity theorem guarantees that these effects will also appear in conventional transmission electron microscopy (TEM) images. Thus it is very difficult to determine junction width and profile from a single such BF image.

Figure 2 shows an element map of O, Co, and Al across the magnetic tunnel junction. Each signal was measured by integrating the area under the background-subtracted O K, Co L$_{3,2}$ and Al K edges in the electron energy loss spectrum (EELS). The profiles are somewhat blurred due to roughness of the AlO$_x$ layer; however, some information can be gained from them. The ratio of O to Al atoms in the barrier center was determined to be 1.4±0.1 by comparison with sapphire. The profiles are somewhat affected by the O content of the barrier, which was determined to be 1.4±0.1 by comparison with sapphire. The center of the tunnel barrier contains fully or near-fully oxidized Al, (defined as O:Al of 1.5). At the bottom Co/AlO$_x$ interface, the O:Al ratio decreases indicating an Al-rich region. The top interface may be slightly Al rich as well. The Al-rich bottom interface is consistent with x-ray photoemission spectroscopy studies, which suggest that the relatively mild oxidation conditions used to form the tunnel barrier can be expected to leave some metallic Al.

In the center of the tunnel barrier EELS fingerprinting analysis shows that the AlO$_x$ is physically disordered. Figures 3(a) and 3(b) show that the O K and Al L$_{3,2}$ edges from the barrier center match very well at all energies with those from an evaporated alumina thin film and are very different from those from sapphire. The selected area diffraction pattern (not shown) from the evaporated alumina shows very diffuse rings confirming an amorphous structure. EELS near-edge fine structure is very sensitive to local atomic structure, so the positive fingerprint identification indicates that the barrier oxide is amorphous, not crystalline. Furthermore, it confirms that there is no substantial amount of Co in the central region of the tunnel barrier as the concentration profile in Fig. 2 might seem to imply.

Intimately related to physical disorder is electronic disorder in the tunnel barrier AlO$_x$ layer. The inset in Fig. 3(a) shows that the O K edge from amorphous alumina has a much more gradual onset than the edge from crystalline alumina. Since the O K edge shows the shape of the unfilled density of states with $p$ symmetry, this indicates the conduction band in the tunnel barrier has a significant band tail. This single-particle interpretation of the O K edge is likely valid since the O K edge calculated for other forms of alumina shows no significant core-hole effects, and core-hole effects tend to sharpen the edge onset rather than blur it. This extensive band tail is consistent with ballistic electron emission microscopy measurements of a relatively low tunnel barrier height for Co/AlO$_x$/Co junctions. Tsymbal and Pettifor predict for tunneling through a disordered barrier that the spin polarization decreases with increasing disorder and barrier thickness.

The nature of the ferromagnet/oxide interface is of great interest since tunneling is expected to occur from the few atomic layers of the metal nearest the insulator. Figure 4(a) shows the averaged Al L$_3$ edge measured at both Co/AlO$_x$ interfaces. The edge onset is much more sloped and lower in energy than the Al L$_3$ edge taken at the center of the AlO$_x$ layer. This is in contrast to the O K edge, which has no measurable change at either Co/AlO$_x$ interface. Since the atomic layer at a Co/AlO$_x$ interface is expected to have a significantly different local density of states than those away from an interface, these results indicate that the barrier is Al terminated and not O terminated. Furthermore, the much lower edge onset, comparable to our measurements for the Al L$_3$ edge at a Co/Al interface, indicates the interfacial Al probably has states at the Fermi energy and is metallized by its contact with Co.
In summary, we have observed some important physical and electronic features in magnetic tunnel junctions that can significantly affect the spin-polarized tunneling current. The tunnel barrier oxide was identified as amorphous alumina, which is physically and electronically disordered. This disorder strongly influences the tunneling process by lowering the height of the tunnel barrier and is predicted to affect the spin polarization of the tunneling current. Fully or near-finely oxidized Al is found at the barrier center, but a region of under-oxidized Al is found at the bottom interface. Both oxide edges are Al terminated, and the interfacial Co–Al bonds likely result in (i) metallization of the interfacial Al and (ii) a reduction in the interfacial Co moment. Furthermore, the observed $sp$-$d$ hybridization of these interfacial bonds is predicted to reverse the polarization of the tunneling current through the barrier. There is no evidence of Co–O bonds, which may cause spin-flip scattering at either interface. Finally, through-focal measurements demonstrate that due to the appearance of Fresnel fringes it is very difficult to determine oxide layer width and interface sharpness from a single coherent STEM or TEM image.

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The Al $L_3$ edge shows the interfacial Co $L_3$ edge spectrum. Compared to the bulk Co signal, the interfacial Co has a shorter and wider $L_3$ peak. This matches the change we observe for Co at a Co/Al interface, indicating that the interfacial Co has some Al nearest neighbors. This may result in a reduction of the magnetic moment of the Co at the Co/AlO$_x$ interfaces, which implies a reduced spin polarization at these interfaces. There appears to be no significant CoO formation at either interface since Co with O nearest neighbors has an $L_3$ peak that is taller and narrower with a slightly lower onset, none of which is observed in the interfacial Co $L_3$ peak. This is consistent with the interfacial Al and O signals discussed above.

The Co $L_3$ edge changes indicate that the Co/AlO$_x$ interfacial bond has $sp$-$d$ hybridization, which also has important implications for the spin polarization of the tunneling current. As a result of the $sp$-$d$ hybridization the Co $d$ band spreads in energy, and the $L_3$ edge which reflects the empty $d$-band density of states (DOS) becomes broader as well. A strong $sp$-$d$ Al–Co bond has been used to explain experimental observations of a positive spin-polarized tunneling current from Co through an aluminum-oxide barrier, despite a negative spin polarization of the DOS at the Fermi energy in Co.

**Figure 4.** (a) The Al $L_3$ edge at Co/AlO$_x$ interfaces has a lower onset than that of the AlO$_x$ layer, similar to a Co/Al interface. This indicates interfacial Al is likely metallized by its contact with Co. (b) The Co $L_3$ edge at Co/AlO$_x$ interfaces matches that of a Co/Al interface and is dissimilar to CoO. This indicates interfacial Co bonds with Al, and there is no substantial formation of CoO.