Interface characterization and thermal stability of Co/Al–O/CoFe spin-dependent tunnel junctions

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A detailed study of the interface properties as well as the thermal stability has been done for the Co/Al–O/CoFe/NiFe magnetic tunnel junction, by using high resolution transmission electron microscopy equipped with energy dispersive x-ray spectrum. The Al behaves more stable against thermal annealing compared with the Fe, Co, Ni, and O elements. The reduction of the tunnel magnetoresistance ratio for the low annealing temperature (200 °C) may be caused by the spin flip scattering at oxide ion, rather than by the change in magnetic properties. The annealing at higher temperatures (300 °C and 400 °C) results in a strong interdiffusion, and in turn the disappearance of the magnetoresistance due to the shortcut of the junction.
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Spin-dependent tunnel junction or magnetic tunnel junction (MTJ)[1] has attracted much attention due to its huge application in advanced technologies, such as magnetic sensor and memory. The ultrathin insulator in nanoscale limit acts as a tunneling barrier between two ferromagnetic (FM) electrodes. The spin-dependent tunneling behavior or the magnetoresistance (TMR) is strongly effected by the interface properties, such as the effective interfacial spin polarization and roughness. In the simple Jullièr's model,[2] the TMR ratio was merely determined by the spin polarization of both FM electrodes. Slonczewski[3] indicated however that the discontinuous change in the potential at the electrode–barrier interface as well as the effective interfacial exchange coupling may play an important role in the spin-dependent tunneling process. Moreover, thermal stability is also one of the crucial issues while one combines the fabrication of the MTJ with the semiconductor processing, which requires an annealing temperatures up to 400 °C. The previous study has shown that the spin-dependent junctions may still maintain a significant value of the TMR ratio after thermal annealing at elevated temperatures around 300 °C.[4,5] A detailed understanding of the interdiffusion mechanism at the interface between FM electrodes and insulator layer and its effect on the magnetoresistance during thermal annealing however requires further investigation.

In this work, a detailed study of the effects of the thermal stability on the interface structure and magnetic as well as magnetoresistive properties has been done for the Co/Al–O/interlayer CoFe/NiFe MTJ. Junctions with the structure, Si/Co/Al–O/CoFe/NiFe,[6,7] were fabricated by high-vacuum magnetron sputtering system with the base pressure of 1×10^{-7} Torr.[7] These multilayers were deposited by dc power with a deposition voltage of about 300 V in a 5×10^{-3} Torr Ar atmosphere. To form the insulating layer, the Al layer was plasma-oxidized by rf glow discharge (64% Ar+36% O). Cross strip junctions with 1 mm×1 mm area were fabricated for the four-probes measurement of the tunnel resistance in a current perpendicular to the film plane geometry.[7] The MTJs were annealed at a pressure of 1×10^{-7} Torr with temperatures ranging from 200 °C to 400 °C for 1 h. For the study of interface properties, a field emission gun transmission electron microscopy JEOL 2010F equipped with energy dispersive x-ray spectrum (EDX) and gatan imaging filter systems was utilized to investigate the structure and composition profile of the ultrathin film in the MTJ annealed at different temperatures. The point-to-point spatial resolution of the 2010F microscope is near 0.2 nm with a minimum probe size of 0.5 nm.

Figure 1 shows the TMR curves for the as-deposited junctions and annealed at different temperatures (200 °C and 300 °C). The junction as deposited has a TMR ratio around 16%. After 200 °C annealing, the TMR ratio decreases sig-

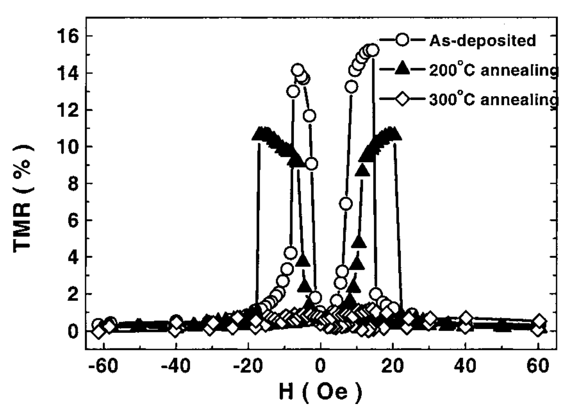

FIG. 1. TMR curves for various annealing temperatures.

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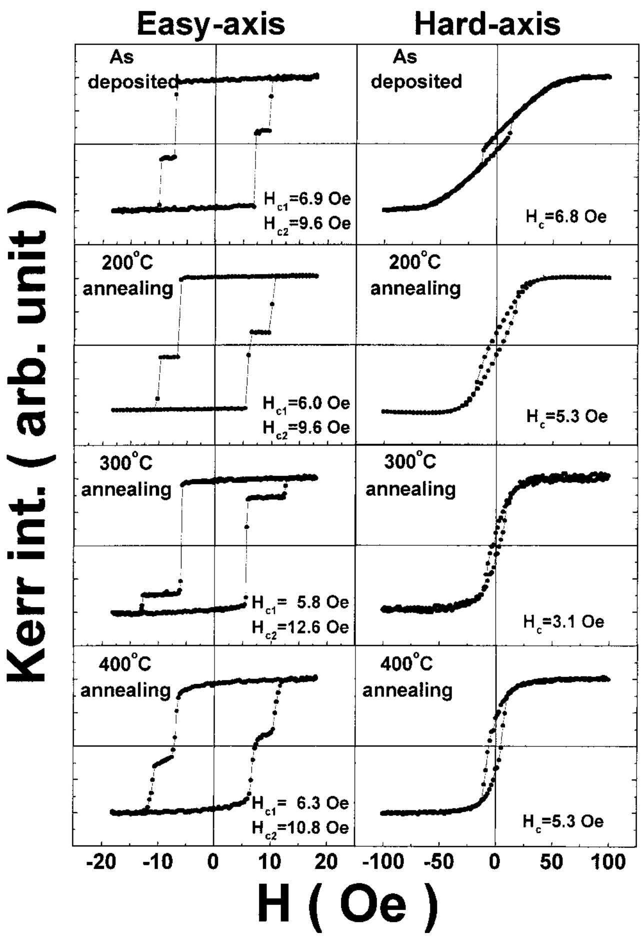

FIG. 2. Magnetic hysteresis loops by MOKE for the MTJs as deposited and annealed at different temperatures in both easy and hard axes. The H_{c1} and H_{c2} indicate the different coercivity field of the FM electrodes in a pseudospin valve structure. Note that the difference between H_{c1} and H_{c2} is even larger for the MTJs after annealing up to 300 °C.

nificantly down to 10%. However, the width of the plateau or the field range, in which the relative magnetization orientation of both FM electrodes is antiparallel (AP), increases (~4 Oe for as deposited junction and ~10 Oe for the 200 °C annealing one). The TMR ratio of the junction annealed at 300 °C drops drastically to around 1%. Obviously, the thermal annealing strongly changes the spin-dependent transport properties.

As mentioned, the final value of the TMR ratio should be determined *intrinsically* by the spin polarization or a well-defined magnetic configuration contrast (parallel and antiparallel alignments of FM electrode magnetizations), and may also be influenced *extrinsically* by the interface properties. The effect of the thermal annealing on the former one may be checked by the magnetic measurement. Figure 2 compiles the magnetic hysteresis loops in both easy and hard axes by using magneto-optical Kerr effect (MOKE). The hysteresis loops of the MTJs before annealing and after 200 °C annealing reveal a very similar feature. The AP field range for the one after 200 °C annealing is even larger than that before annealing. This is consistent with the TMR loops shown in Fig. 1. There is no significant decrease of the magnetic signal observed. The drastic TMR decrease, as shown in Fig. 1, is not due to the change of the magnetic configuration of both FM electrodes. The significant change of the TMR ratio could be thus attributed to the effect of the thermal annealing on the interface properties. After 400 °C annealing, the hysteresis loop along the easy axis becomes rounder and has no more a perfect plateau of the AP field range.

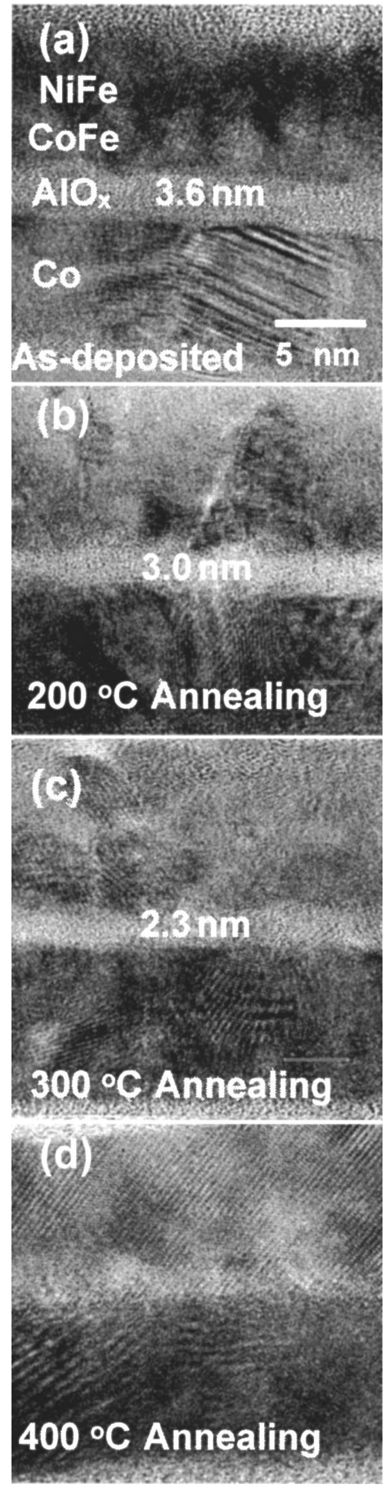

FIG. 3. TEM images for junctions annealed at different temperatures. The observed Al–O thickness is 3.6 nm, 3.0 nm, and 2.3 nm for as deposited, 200 °C and 300 °C annealing, respectively. The Al–O junction area becomes obscure after 400 °C annealing.

Figure 3 reveals the transmission electron microscopy

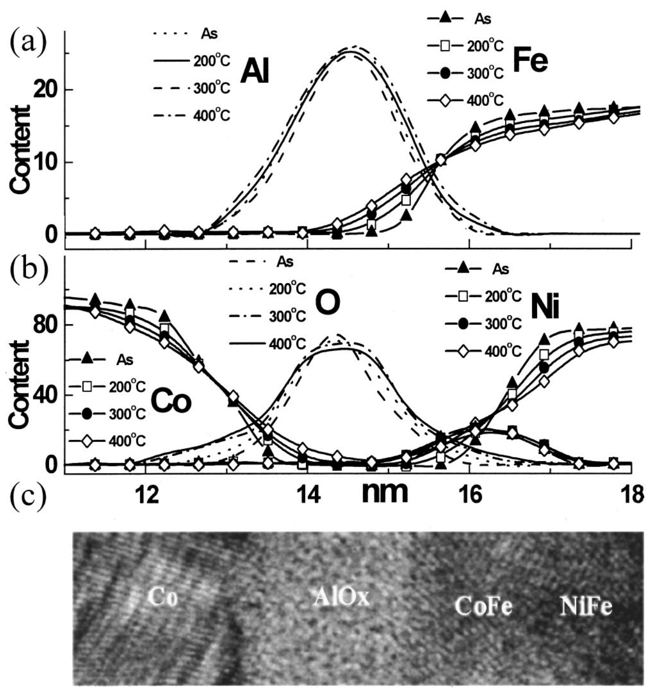

FIG. 4. Deconvoluted composition profiles for (a) Al and Fe and (b) O, Co, and Ni for different annealing temperatures. (c) The corresponding TEM imaging.

(TEM) images for junctions as deposited and annealed at different temperatures (200 °C, 300 °C, and 400 °C). The thickness of Al–O observed in the TEM images decreases with increasing annealing temperature (3.6 nm, 3.0 nm, and 2.3 nm for as deposited, 200 °C and 300 °C annealing, respectively). The Al–O junction area becomes obscure after 400 °C annealing, indicating a significant interdiffusion at junction interfaces. On the other hand, the interfacial roughness increases with increasing temperature. (0.25 nm, 0.41 nm, and 0.51 nm of Co/Al–O interface and 0.29 nm, 0.70 nm, and 0.86 nm for as deposited, 200 °C and 300 °C annealing, respectively.) From the TEM images, it shows that both Al–O thickness and roughness at the barrier–electrode interface are strongly changed by the thermal annealing, as compared to the magnetic properties.

The composition-resolved depth profiles in Fig. 4 were obtained by using the technique of the nanobeam EDX analysis supported by a Wiener filter deconvolution method.[8] Figures 4(a) and 4(b) indicate that all of the FM elements (Fe, Co, and Ni) diffuse into the Al–O layer and oxygen diffuses outward into both FM electrodes after annealing. The oxygen-rich AlO_x of the as deposited MTJ is extremely unstable against thermal annealing. The interesting finding is that the Al seems to be less effected as compared to other elements, such that Al/O ratio in the junction is enhanced upon thermal annealing.

As indicated, the thermal annealing has more of an effect on the electrode–barrier interface structure than on the magnetic properties of both FM electrodes. The interdiffusion of the Fe, Co, Ni, and O elements occurs already after annealing at low temperature 200 °C, and probably results in the formation of related oxides at interface, which may lead to the spin flip through the inelastic scattering, and in turn to the drastic reduction in the TMR ratio. After higher annealing temperature (300 °C and 400 °C), the total junction resistance is reduced from the initial value of several hundred ohms to few ohms only. A shortcut in the junction induced by the interdiffusion seems to be the main reason for the disappearance of the TMR ratio. The final structure after the interdiffusion may become a mixing of different phases, such as Al_2O_3 and X–O oxides (X=Fe, Co, and Ni). The spinel-like XAl_2O_4 (X=Fe, Co, and Ni), which have low resistivity and may cause the local shortcut, could be also one of the possible products. A detailed nanobeam diffraction study is however still required to provide further information on the final structure after annealing.

In summary, the TMR ratio is strongly reduced upon thermal annealing. At low annealing temperature of 200 °C, the reduction may be mainly due to the spin flip through the formation of X oxides rather than the change in magnetic configuration. At higher annealing temperatures (300 °C and 400 °C), the resistances of the junctions are reduced drastically to few ohms due to the shortcut of the junction through important interdiffusion.

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