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Journal of Magnetism and Magnetic Materials 239 (2002) 116–119

Journal of
magnetism
and
magnetic
materials

www.elsevier.com/locate/jmmm

Thermal stability study of the insulator layer in NiFe/CoFe/ Al₂O₃/Co spin-dependent tunnel junction

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Abstract

Spin-dependent tunnel junction, NiFe/CoFe/Al₂O₃/Co//Si, was fabricated to investigate the thermal stability induced diffusion behaviors. The interfacial diffusion causes the degradation of the ratio of the TMR, the enhancement of the switching field of the two magnetic electrodes, the thickness decrease of the insulator layer, and the increase of the interfacial roughness. The outward diffusion of oxygen from the insulator layer is faster than that of aluminum for samples annealed below 400°C. The degradation of the ratio of TMR is attributed to the disturbance of the spin polarization in the magnetic layers, and the increase of the pinholes and spin-flip effect in the insulator layer. The relative roughness between the two interfaces of the insulator induces the surface magnetic dipoles, and hence, increases the switching field of the ferromagnetic electrodes. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Tunneling magnetoresistance; Thermal stability; Interfacial roughness; Spin polarization

1. Introduction

Since the successful development of the large tunneling magnetoresistance (TMR) [1], there has been a great interest in the potential application of this structure due to the highly field-sensitive tunnel resistance. The TMR tunnel junctions with the structure of two ferromagnetic (FM) layers separated by a thin insulating layer (I) are known to change the tunnel resistance according to the relative magnetic orientations within the two FM layers [2]. However, the tunnel junction should be able to withstand thermal treatment up to 400°C for complementary metal oxide semiconductor compatibility, which is encountered in baked microelectronics integration processing with on-chip IC [3]. However, the success of FM/I/FM tunneling behaviors depends critically on the quality of the insulating layer. Extensive

researches on the thermal stability in the tunnel junction have been carried out in the recent years. Ando et al. have studied the influence of the interlayer roughness using the atomic force microscopy on the TMR ratio [4]. They reported that the corresponding TMR ratio is very small (2%) for the junction with a rough interface. Parkin et al. [5] have shown that the tunnel resistance and the TMR ratio are slightly affected by thermal annealing at temperatures up to ~300°C and that the Al₂O₃ thickness is slightly decreased under the annealing temperature (T_A) of 350°C for CoPt/Al₂O₃/CoPtCr tunnel junction. In this study, the diffusion mechanism between layers was investigated in order to understand the effect of the thermal annealing on the TMR ratio.

2. Experiments

Spin-dependent tunnel (SDT) junctions were prepared in a high vacuum magnetron sputtering system with a base pressure lower than 1×10^{-7} Torr. The junctions

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had the following sequence: NiFe(10 nm)/CoFe(1 nm)/Al₂O₃(3.6 nm)/Co(10 nm)//Si. These multilayers were deposited by DC power with the deposition voltage of about 300 V in a 5×10^{-3} Torr Ar ambient. In order to form the insulating layer, the Al surface was plasma oxidized by RF glow discharge with the bias voltage of -350 V. Samples were annealed at the ambient of 1×10^{-7} Torr at T_A ranging from 200°C to 400°C for 1 h. Cross strip junctions with 1 mm $\times$ 1 mm dimension were designed to measure junction resistance by current perpendicular to plane and 4-probe methods. A field emission gun high resolution transmission electron microscopy (HRTEM) JEOL 2010F equipped with the energy dispersive X-ray spectrometer (EDX) system was utilized to investigate the structure and composition of the ultrathin film in the TMR junctions. The point-to-point spatial resolutions of the 2010F microscope are close to 0.2 nm with a minimum probe size of 0.5 nm.

3. Results and discussion

The HRTEM images associated with different T_A are shown in Fig. 1 for TMR junctions (a) before annealing, (b) annealing at 200°C for 1 h, (c) annealing at 300°C for 1 h, and (d) annealing at 400°C for 1 h. The insulator layer shows a lighter contrast in the image due to the amorphous characterization of AlO_x. The value of x , evaluated using nanobeam EDX, is about 2.5–2.6 for the

as-deposited junction. Apparently, the AlO_x is an oxygen-rich aluminum oxide compared with Al₂O₃. This indicates that the enhancement of TMR after annealing will not occur in our system. The strong dark contrast implies that they are columnar, similar in nature to the metallic layers corresponding to the regions of metallic grains. As shown in Fig. 1(a)–(c), the horizontal white contrast lines with slight waviness are the insulating layer. The average thicknesses of the insulating layer (which is 3.6, 3.0 and 2.3 nm) decreases with increasing T_A . Meanwhile, the interfaces of FM/I or I/FM were obscured after annealing at 400°C, as shown in Fig. 1(d), and thus cannot evaluate the average insulating layer thickness of the junction by HRTEM image. It is apparent that the diffusion or interfacial reaction leads to the interfacial mixing of the NiFe/CoFe/AlO_x/Co structure during the annealing treatment at 400°C. To further assess the interfacial mixing quantitatively, the insulating layer thickness and interfacial roughness of the FM/I were analyzed from the HRTEM images. The average insulating layer thickness and NiFe/CoFe/AlO_x/Co roughness for each annealing temperature are sequentially listed in Table 1.

Fig. 2 shows the ratio of TMR as a function of the applied magnetic field for samples of as-deposited (dots), annealed at 200°C (open triangles), and annealed at 300°C (open diamonds). For clarity, data of the sample annealed at 300°C are five times the original one. The ratio of TMR decreases from $\sim 15\%$ for the as-deposited sample to $\sim 11\%$ for samples annealed at 200°C for 1 h, and decreases further to $\sim 1.6\%$ for samples annealed at 300°C for 1 h. The pinning characteristics increased with increasing annealing temperature. This shows that the plateau region between the two coercivities of the two electrodes increased from ~ 6 Oe to 12 and 18 Oe for samples annealed at 200°C and 300°C, respectively.

In order to quantitatively determine the intermixing effect at the interface of CoFe/AlO_x and AlO_x/Co, a nanobeam EDX experiment was carried out in a full junction stack. The diffusion of the constituent element (Ni, Fe, Co, Al, and O) increases slightly with increasing T_A . A scanning focusing probe with a beam size of 0.5 nm was also applied to acquire nanobeam EDX spectra across the interfaces to investigate the compositional distribution of Ni, Co, Fe, Al, and O in the tunnel junction structure. The sampling spacing of the scanning focused electron beam is about 0.8 nm. The compositional profiles were interpolated using a polynomial fit. Nevertheless, the spatial resolution of the composition profile deduced from nanobeam EDX is degraded due to the electron beam broadening effect, which is due to the interaction of electron beam with the junction. The information of the diffusion among the tunnel junction structures may be obscured due to this broadening effect from the nanobeam EDX analysis. A Wiener filter

Fig. 1. High resolution transmission electron micrographs for TMR junction samples (a) as-deposited, (b) annealed at 200°C for 1 h, (c) annealed at 300°C for 1 h, and (d) annealed at 400°C for 1 h.

Table 1
The TMR ratio, the evaluated average insulating layer thickness, and the interfacial roughness of Co/AlO_x and AlO_x/CoFe/NiFe for a TMR junction before annealing, annealed at 200°C and 300°C, respectively

		As-deposited	200°C	300°C
TMR		15%	11%	1.6%
AlO _x thickness (nm)		3.6±0.1	3.0±0.1	2.3±0.1
Interfacial roughness (nm)	Co/AlO _x	0.24±0.01	0.70±0.03	0.86±0.03
	AlO _x /CoFe/NiFe	0.32±0.01	0.42±0.03	0.51±0.03

Fig. 2. The ratio of TMR as a function of the applied magnetic field for samples of as-deposited (dots), annealed at 200°C (open triangles), and annealed at 300°C (open diamonds).

deconvolution technique [6] was adopted to remove the beam broadening effect from nanobeam EDX. Fig. 3(a) shows the HRTEM image of a TMR junction before annealing. The deconvoluted results of nanobeam EDX counts for Co, O, and Ni as a function of its corresponding position are shown in Fig. 3(b) for individual T_A . It indicates that the FM elements, for example Co and Ni, diffuse into the insulating layer and that the O diffuses outward. In order to deduce the reasons for the deteriorated cause of TMR, the compositional variation of Al and O was further studied. Fig. 3(c) shows the diagram of the normalized Al/O ratio vs. its corresponding position. It is clear that the Al/O ratio increases with increasing T_A in the central area of the insulating layer. This suggests that the oxygen diffuses outward faster than that of aluminum from the central area of the insulating layer. The oxygen-rich aluminum oxide (AlO_x) seems to be unstable for the annealing process at an elevated temperature. This annealing result is different from that in Refs. [3,7], in which the TMR ratio increases with T_A due to the thermal improvement of the interfacial Al/O ratio.

The well-known bulk Al₂O₃ is a rather stable and passive phase, when the melting point is up to 1600°C. However, from the underlying viewpoint of the material, the transition metals could react with Al₂O₃ and form the spinel product at an elevated temperature [8].

Fig. 3. (a) The HRTEM image of a TMR junction before annealing, (b) the deconvoluted composition profile of Co, O, and Ni vs. its corresponding position, and (c) the diagram of the normalized Al/O ratio vs. its corresponding position.

Therefore, it is possible for the unstable AlO_x that the inward diffusion of Fe, Co and Ni interacts with AlO_x and forms the spinel materials, such as FeAl₂O₄, CoAl₂O₄ or NiAl₂O₄ etc. Meanwhile, the excess oxygen diffuses outward. The spinel resistivity is much lower than that of Al₂O₃ [9]. Accordingly, the formations of the spinel may cause pinholes in the insulating layer of the magnetic tunnel junctions, which are local shortcuts and cause the malfunctioning of such devices.

In summary, the diffusion effect of the insulator layer in NiFe/CoFe/Al-O/Co//Si spin dependent tunnel junctions has been studied experimentally. The interfacial diffusion causes the degradation of the ratio of the TMR, the enhancement of the switching field of the two magnetic electrodes, the thickness decrease of the insulator layer, and the increase of the interfacial roughness. The outward diffusion of oxygen from the insulator layer is much faster than that of aluminum for samples annealed below 400°C. The degradation of the ratio of TMR is attributed to the disturbance of the spin polarization in the magnetic layers and the increase of

the pinholes and spin-flip effect in the insulator layer. The relative roughness between the two interfaces of the insulator induces the surface magnetic dipoles, and hence, increases the switching field of the ferromagnetic electrodes.

Acknowledgements

The authors profusely thank the National Science Council of R.O.C (NSC 89-2218-E-007-056) for the financial support of this work.

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