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Magnetic tunnel junctions with high magnetoresistance and small bias voltage dependence using epitaxial NiFe(111) ferromagnetic bottom electrodes

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Magnetic tunnel junctions (MTJs) were fabricated using an Al–O insulating layer prepared on an epitaxially grown $Ni_{80}Fe_{20}$ bottom electrode and on a polycrystalline $Ni_{80}Fe_{20}$ bottom electrode. Crystallographic orientations and surface morphology of the films were examined using x-ray diffraction and atomic force microscopy, respectively. The MTJ with an epitaxial bottom electrode showed a tunnel magnetoresistance (TMR) ratio of 51% after annealing at 250 °C. This value was about two times larger than that of the MTJ with a polycrystalline bottom electrode (27%). The applied bias voltage dependences of the TMR ratios were also much different. The V_{half} values of epitaxial and polycrystalline samples were about 750 and 400 mV, respectively. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1544458]

I. INTRODUCTION

Polycrystalline ferromagnetic (FM) films with an fcc(111) preferred orientation have been used often as bottom electrodes in magnetic tunnel junctions (MTJs). Previous works have shown that initial plasma oxidation of a thin Al film progresses through grain boundaries.[1,2] Inhomogeneity of the insulating layer can be one reason for the decrease of tunnel magnetoresistance (TMR) ratio with increasing bias voltage. This is fatal in development of magnetoresistive random access memory with large capacity of Gbit order. Insulating properties of Al–O thin film prepared on a single crystalline or epitaxial FM electrode probably differ from those of Al–O thin film prepared on a polycrystalline FM electrode. Recently, high quality MTJs with single crystalline or epitaxial FM electrodes were successfully fabricated.[3,4] In this study, MTJs are fabricated using Al–O insulating layers prepared on an epitaxial $Ni_{80}Fe_{20}$ (NiFe) bottom electrode and on a polycrystalline NiFe bottom electrode. Differences in TMR ratios and their bias dependences are discussed.

II. EXPERIMENTS

It was reported that NiFe(111) film on Si(111) substrate/Ag(111) 100 nm/Cu(111) 50 nm multilayer grew epitaxially by sputter at room temperature (RT).[5] We adopted and modified this film structure as a buffer layer for an epitaxial NiFe layer. A Si(111) substrate was etched by NH_4F solution to remove the native oxide layer for growth of an epitaxial bottom electrode. The multilayer of Si(111)/Ag 3 nm/Cu 50 nm/NiFe 50 nm/Al–O/$Co_{75}Fe_{25}$ 4 nm/IrMn 20 nm/NiFe 20 nm/Ta 5 nm was sputtered (Sample A). The Al–O was prepared by plasma oxidation of a 1.6-nm-thick Al layer. Sputtering power densities of Ag, Cu, NiFe were about 0.74, 1.48, and 1.48 W/cm^2, respectively. For a sample with a polycrystalline bottom electrode (Sample B), the same stacking and the same Al oxidation condition were used except for a thermally oxidized Si substrate with Ta 3 nm/Cu 20 nm buffer layers and higher sputtering power density for every material used. Both samples were patterned into several μm^2 using a microfabrication process including photolithography and Ar ion etching. Crystallographic orientation and surface morphology of films were examined using x-ray diffraction and atomic force microscopy (AFM), respectively. TMR curves were measured at room temperature. The junction area was 30×30 μm^2. Measurements were done by dc four-probe method at a bias voltage of 1 mV.

III. RESULTS AND DISCUSSION

The x-ray diffraction pattern for the bottom electrode of Sample A showed only fcc(111) peaks of NiFe and Cu, except for some peaks originated in the Si substrate. No peak was observed for Ag because it was very thin. The rocking curve measured for the NiFe(111) peak showed the full width at half maximum of 0.80°, inferring a very small dispersion. The ϕ-scans of the {111} planes of NiFe, Cu and Si revealed that all layers grew epitaxially and that twin epitaxy existed in these films. The x-ray diffraction pattern for the bottom electrode of Sample B showed that the bottom electrode grown on a thermally oxidized Si had a textured polycrystalline structure with a fcc(111) preferred orientation.

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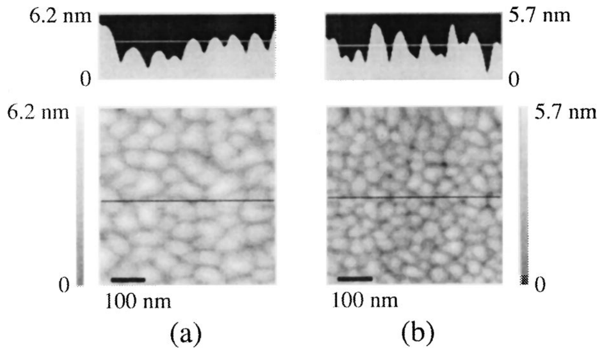

FIG. 1. AFM images of the surfaces of NiFe bottom electrodes in (a) Sample A (epitaxial) and (b) Sample B (textured).

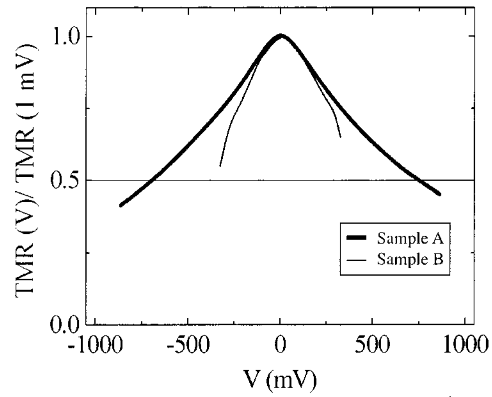

FIG. 3. Normalized TMR ratio vs dc bias voltage curves for Samples A and B measured at RT after annealing at 250 °C.

Surface morphology of the bottom electrodes of Samples A and B are shown in Figs. 1(a) and 1(b), respectively. Image size of each sample is 500 nm$\times$500 nm. Upper figures show cross sectional profiles along the lines in the corresponding images. Surface roughness of the epitaxial film (R_a $\cong$0.9 nm) was slightly larger than that of the textured films ($R_a$$\cong$0.7 nm). However, the cross section of the epitaxial film shows a more rounded and smooth surface in contrast to the textured film surface.

TMR ratios measured at as-deposited state were 32% for Sample A and 8% for Sample B. Figure 2 shows TMR curves measured after annealing at 250 °C. The resistance-area product of Samples A and B measured about 5.5 $\times 10^5$ $\Omega\,\mu\mathrm{m}^2$ and 3.2×10^5 $\Omega\,\mu\mathrm{m}^2$, respectively. The TMR ratio of Sample A was 51%, about two times larger than that of Sample B (27%).

It is also remarkable that the bias dependences of TMR ratio in the two samples differed greatly. Figure 3 shows the normalized TMR ratio versus dc bias voltage curves measured at RT after annealing. Bold and solid lines represent results of Samples A and B, respectively. Both curves were obtained from the current (I)-dc bias voltage (V) curves measured at antiparallel alignment and parallel alignment of the magnetization of top and bottom electrodes. Positive bias was defined as the direction of current flow from the bottom electrode to the upper electrode. The V_{half}, the bias voltage at which the normalized TMR ratio becomes 0.5, of Sample A was about +750 and −700 mV for positive and negative biases, respectively. These good properties shown in Figs. 2 and 3 were observed at all junctions in the same substrate, which means that they exhibited high reproducibility. On the other hand, the V_{half} of Sample B was estimated about 400 mV at both sides, which were much smaller than those of Sample A. The large difference of V_{half} between the two samples would be due to different interfacial structures. In Sample B, the bottom FM electrode was composed of fcc(111) oriented grains that were randomly rotated in the film plane. Thus, there existed many high angle grain boundaries in the polycrystalline NiFe layer. The metallic Al layer on the polycrystalline NiFe also had the same grain structure and included a number of grain boundaries as well. Due to the high angle grain boundaries of the metallic Al precursor layer, the Al–O layer would have decreased density after oxidation. Considering our previous work, oxygen atoms would penetrate into the NiFe bottom electrode along grain boundaries and create defects near the interface between the insulating layer and the NiFe layer.[1] Therefore, the grain boundaries themselves would act as defects in spin-polarized electron tunneling. These defects must be one origin of strong bias dependence of TMR.[6] On the contrary, in the case of Sample A, layer structure was much denser than that of Sample B. It was reported that Al grows epitaxially on epitaxial NiFe, despite the large lattice parameter misfit of 12%.[7] Therefore, the absence of high angle grain boundaries in the Al metallic layer would lead to better uniformity of oxygen distribution in the insulating layer. Furthermore, over oxidation along the grain boundaries of the NiFe would be suppressed, resulting in more homogeneous oxidation near the interface between the NiFe and Al–O layer. Consequently, in Sample A, there existed few defect states through which spin-independent tunneling occurs.[6]

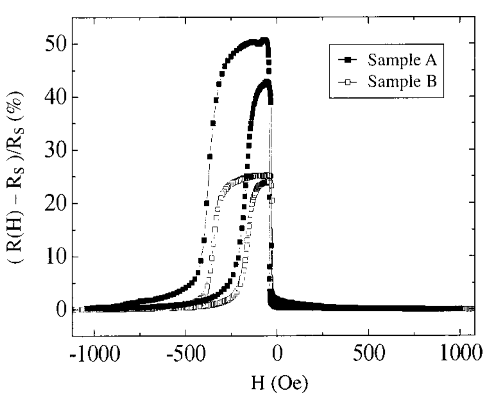

FIG. 2. TMR curves measured at RT for Samples A and B after annealing at 250 °C.

It should be noted that V_{half} values of Sample A were slightly asymmetric. This small asymmetry would be due to different interfacial structures between the upper and lower interfaces. However, analysis of an I–V curve for Sample A revealed nearly symmetric barrier heights at both interfaces. Therefore, further investigations with precise analysis, for example, inelastic electron tunnel spectroscopy, are necessary.

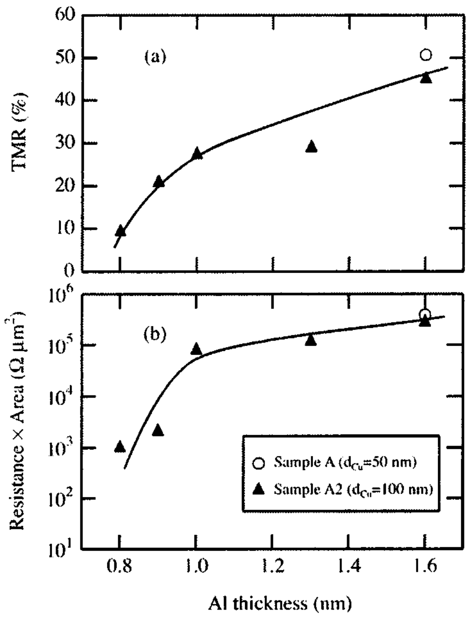

FIG. 4. Al thickness dependences of (a) TMR and (b) R×A for samples with an epitaxially grown NiFe bottom electrode.

Figure 4 shows Al-thickness dependences of TMR and resistance $(R)\times$junction area (A). Triangular dots represent the data of Sample A with a thicker Cu buffer layer (Sample A2, $d_{\mathrm{Cu}}=100$ nm) in which NiFe bottom electrodes showed more highly oriented structure. However, it also had larger roughness ($R_{\mathrm{a}}\cong 1.3$ nm) than that of Sample A. When Al thickness exceeded 1 nm, the TMR ratio and RA slightly decreased with decreasing Al thickness. However, both values decreased rapidly at Al thickness of less than 1 nm. Nonuniform current flow due to rough interface and nonuniform thickness of the insulating layer would be responsible for these rapid decreases. Buffer layer stacking should be examined in order to suppress that roughness.

In summary, a great improvement in bias dependence of the TMR ratio was obtained in MTJs with an epitaxial FM bottom electrode. An insulator grown on the epitaxial bottom electrode would have fewer defects than the one on the textured bottom electrode. The result suggests that output signals of the devices based on MTJs, e.g., MRAM, can be enhanced very much by improving insulating layer quality.

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