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Electron transport properties in magnetic tunnel junctions with epitaxial NiFe (111) ferromagnetic bottom electrodes

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By employing epitaxial NiFe (111) films as ferromagnetic bottom electrodes, magnetic tunnel junctions with layer sequence of Si (111)/epitaxial Ag/epitaxial Cu/epitaxial NiFe/Al-oxide/CoFe/IrMn/NiFe/Ta were prepared. High tunneling magnetoresistance (TMR) ratios were obtained and the bias dependence of TMR was remarkably reduced. The reason for the small bias dependence of TMR was explained by inelastic electron tunneling spectroscopy. It was clearly elucidated that a well-defined sharp interface formed between the tunnel barrier and the ferromagnetic electrode that is nearly free of crystalline defects. This magnetic tunnel junction has a large capability in engineering aspects if we can reduce the barrier thickness further by decreasing the interface roughness. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1587271]

The high-quality magnetic tunnel junction (MTJ) with single-crystalline or epitaxial ferromagnetic (FM) electrodes has been successfully made[1,2] and the authors were also able to fabricate MTJs using Al–O insulating layers prepared on an epitaxial $Ni_{80}Fe_{20}$ (NiFe) bottom electrode.[3] It showed a tunneling magnetoresistance (TMR) ratio of 50.7% after annealing at 250 °C. This value was about two times larger than that of the MTJ with a polycrystalline NiFe bottom electrode (ratio of 27%). The applied bias voltage dependence of the TMR ratio was also so small that the V_{half} value was about 750 mV. However, the question of the possibility of the enhanced interfacial structure and the asymmetric nature of the bias dependence remained and it will be studied in this work. As in a previous work of ours,[3] we focused on the engineering aspects of MTJs with epitaxial FM electrodes and thus fabricated MTJs using an Al–O insulating layer that was prepared on epitaxially grown NiFe bottom electrodes.

Si (111) substrates were first cleaned in $H_2SO_4:H_2O_2 = 4:1$ solution for 20 min to remove organic impurities, then rinsed in deionized water. Subsequently, they were etched in NH_4F solution for 10 min to remove the native oxide layer and to obtain hydrogen-terminated flat surfaces. The epitaxial NiFe (111) film was grown successfully by Gong *et al.*[4] on the Si (111) substrate/Ag (111) 100 nm/Cu (111) 50-nm multilayer by sputtering at RT. We used this film structure as a buffer layer to grow the epitaxial NiFe bottom electrode and fabricated MTJs with the following stacking sequence: Si (111)/epitaxial Ag 3 nm/epitaxial Cu (50 and 100 nm)/epitaxial $Ni_{80}Fe_{20}$ 50 nm/Al–O 1.6 nm/$Co_{75}Fe_{25}$ 4 nm/$Ir_{22}Mn_{78}$ 20 nm/$Ni_{80}Fe_{20}$ 20 nm/Ta 5 nm. We prepared two types of samples—Sample A (with $d_{Cu} = 100$ nm) and B (with $d_{Cu} = 50$ nm)—and compared them. All the layers in the junction were prepared using inductively coupled plasma (ICP)-assisted magnetron sputtering with base pressure below 1×10^{-6} Pa without breaking vacuum. The insulating barrier was formed by depositing Al, using ICP-assisted rf magnetron sputtering at the rate of about 0.07 nm/s and oxidizing by ICP oxidation under 1 Pa of $Ar/O_2 = 1:3$ gas mixture. The buffer layers of Ag, Cu, and NiFe were sequentially deposited at RT with rates of about 0.06, 0.06, and 0.03 nm/s, respectively, and the corresponding Ar pressure at deposition was 0.15, 0.12, and 0.15 Pa, respectively.

Crystallographic structures and surface morphology of the films were investigated using x-ray diffraction (XRD) and atomic force microscopy (AFM). Nine junction areas were patterned using a microfabrication method including photolithography, and the area patterned was in the range of 3×3 through 100×100 μm^2. All the samples were annealed at 250 °C for 1 h under vacuum and external magnetic field. The interface between the FM electrode and the insulating layer was investigated by inelastic electron tunneling spectroscopy (IETS).

The epitaxial degree of FM bottom electrodes in all the samples was checked by XRD. The θ–2θ scans of the following layer structure—Si(111)/Ag (3 nm)/Cu (100 nm)/NiFe (50 nm)—shows only the {111} peaks of NiFe and Cu, indicating 111-orientation [Fig. 1(a)]. The rocking curve of the NiFe peak had a full width at half-maximum of 0.77°, inferring a very small dispersion [Fig. 1(b)]. The ϕ-scan of {111} planes of NiFe revealed three peaks at the same ϕ positions with those of Si, verifying epitaxial growth [Fig. 1(c)]. The other three peaks were also observed at angles of 180° translated from the former three peaks, ascribing to the existence of twin epitaxy.

Figure 2(a) shows TMR curves that were measured for samples A and B at RT. Measurement was performed by dc four-probe method at a bias voltage of 1 mV. The TMR ratios were obtained 45.5% and 50.7% for Samples A and B, respectively. Sample B exhibited a similar but slightly larger value of TMR than that of sample A. The resistance–area

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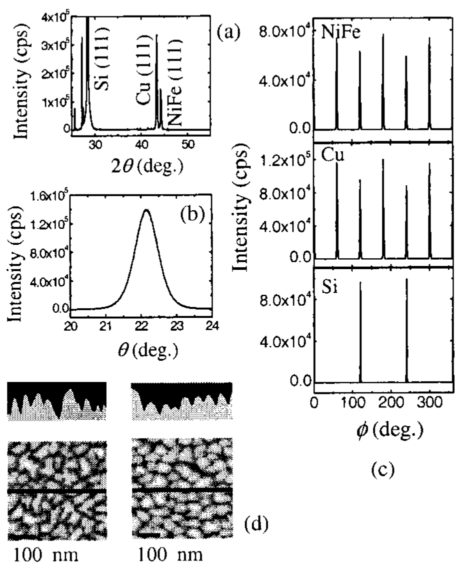

FIG. 1. (a) The θ–2θ scans of the layer, Si(111)/Ag(3 nm)/Cu (100 nm)/NiFe (50 nm). (b) The rocking curve of the NiFe peak. (c) The ϕ-scan of {111} planes of NiFe, Cu, and Si. (d) AFM images of NiFe FM bottom electrodes of samples A (left side) and B (right side).

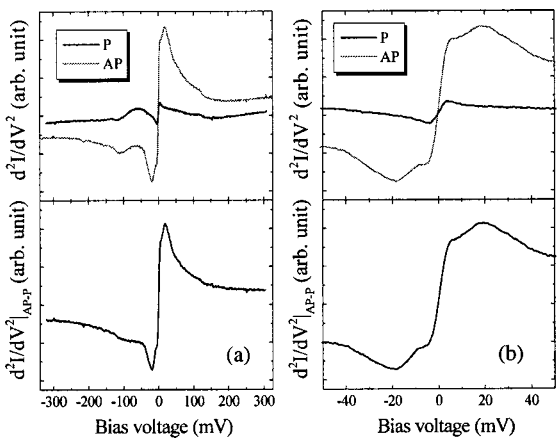

FIG. 3. (a) IETS of P and AP magnetization states (upper) and subtraction IETS of sample A (bottom) measured at 10 K. (b) Expanded spectra of (a).

product was 3.1×10^5 and 5.5×10^5 $\Omega\ \mu m^2$, respectively, for all the junction areas in both sample types. Barrier height ϕ and barrier width d were obtained by fitting the current (I) versus dc bias voltage (V) curves to Brinkman's relation, which can explain the tunneling with an asymmetric barrier. They were determined about 3.0 eV (nearly symmetric) and 0.94 nm, respectively, irrespective of sample types.

The bias dependence of TMR is significant especially from an engineering standpoint and has attracted much interest both in experiment and theory.[5,6] The normalized TMR–V curves measured for samples A and B at RT are described in Fig. 2(b). The curves were obtained from $I-V$ curves measured for antiparallel (AP) and parallel (P) alignment states of the magnetization of top and bottom electrodes. The curves for samples A and B are almost identical. The positive bias is defined as the direction of electron tunneling from top to bottom electrode. The V_{half}, the bias voltages at which the TMR ratio is reduced to half near the zero bias, were measured about $+750$ and -700 mV, much higher values than those of conventional MTJs. Previously, a high V_{half} in MTJs with single-crystalline electrodes was obtained,[1] however, there existed a large asymmetry and the TMR was not so high. These good properties shown in Fig. 2 were observed at all the junctions, which means that they exhibited high reproducibility.

The AFM images of NiFe FM bottom electrodes are shown in Fig. 1(d). The surface roughness R_a, was determined about 1.3 and 0.9 nm for samples A (left) and B (right), respectively. Although the interface between the FM bottom electrode and the insulating layer is rather rough in both samples, it did not exert any significant effect to the bias dependence itself, except that V_{half} and TMR ratios decreased with a reduction in insulating layer thickness.[3]

The difference in the normalized TMR–V curves between samples A and B did not exist in spite of different TMR ratios. As a result, the interface roughness did not seem to have an intrinsic effect on the bias dependence. It is likely that the TMR ratios were affected by dipole coupling. In depositing NiFe bottom electrodes with a higher rate, it was observed that V_{half} in the positive bias became smaller, although it was still larger than those of conventional MTJs, and there was a slight decrease of V_{half} in the negative bias. As the effect of the Cu buffer layer thickness was found not so significant on the performance of TMR and the bias dependence, only sample A was continued for further analysis, keeping in mind that sample B would behave similarly to sample A.

The upper figure of Fig. 3(a) shows the IET spectra of sample A measured at 10 K for P (dark line) and AP (light line) states. The peaks by excitation of magnon and Al–O phonon were observed around ±20 and ±120 mV, similar to previous studies of conventional MTJs.[7–9] The subtraction spectrum defined by the difference between the spectra of both magnetization states was obtained in order to eliminate the contribution from spin-independent excitation [the bottom figure of Fig. 3(a)].

Our result reveals remarkable features. First, the intensity of spectra near the zero bias is small. The peak observed at small bias of several millivolts is induced by inelastic tunneling due to impurities at the interface and in the tunnel barrier, and clearly separated with the peak of magnon excitation in case of conventional MTJs.[9] On the other hand, the intensity of the peak near the zero bias is smaller compared with magnon excitation peak in our epitaxial MTJs [Fig. 3(b)], signifying that the density of impurities is small, and thus, we could say that a clear interface structure was obtained for epitaxial MTJs in this study, compared with conventional MTJs. Second, the spectrum of P state reveals remarkably small intensity, suggesting that inelastic excitation

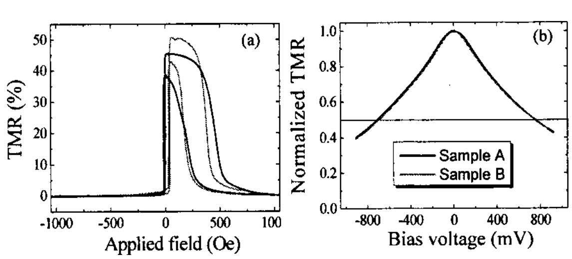

FIG. 2. (a) TMR—applied magnetic field curves and (b) normalized TMR—dc bias voltage curves measured at RT for samples A and B.

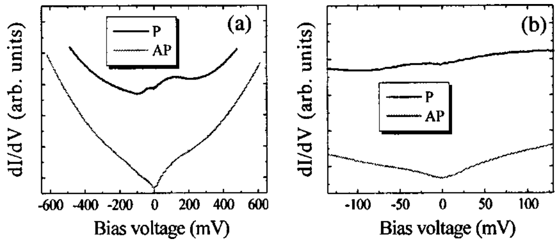

FIG. 4. (a) Dynamic conductance of P and AP magnetization states of sample A measured at 10 K. (b) Expanded spectra of (a).

due to spin scattering is small. Finally, the spectral intensity of positive bias is larger than that of negative bias and a hillock appears around −60 mV, exhibiting asymmetry in the bias voltage. The electronic structure and the spin-dependent density of states (DOS) of the FM electrodes must be responsible for this. The band structure and DOS of FM electrodes should also be considered to explain the bias dependence of TMR.[10]

Dynamic conductance measured simultaneously with IETS is shown in Fig. 4. It reveals an obvious asymmetry and the curve for P state has local minima at about −100 and +230 mV. The similar shape of the spectrum was reported for the Al/Al–O/Ni junction.[11] Thus, we may suggest that the conductance caused by the DOS of NiFe is superimposed and reveals a large asymmetry. This elastic component must have affected the shape of IETS. In fact, the subtraction spectrum in Fig. 3 canceling the spin-independent elastic component exhibited an improved symmetrical behavior.

According to the results of XRD ϕ-scan and IETS, we could suggest the reason for the improved bias dependence of TMR in epitaxial samples. In FM electrodes of conventional MTJs, there existed many high-angle grain boundaries. They served as sites of defects or impurities both at the Al–O/FM interface and the insulating layer. These localized defects in the insulating layer increase the inelastic tunnel process, resulting in a strong bias dependence of TMR according to the two-step tunneling model.[12] On the other hand, the grains in the epitaxial NiFe layer do not have high-angle grain boundaries, but only twin boundaries. It was reported that Al grew epitaxially on epitaxial NiFe, even if the lattice parameter difference between Al and NiFe is as large as 12%.[13] The absence of high-angle grain boundaries in the Al precursor metallic layer and the NiFe layer would lead to better uniformity of the insulating layer and the rough but well-defined sharp interface of Al–O/FM. They could reduce the number of trap sites through which spin-independent tunneling occurs in the insulating layer and could affect the interfacial DOS, resulting in a small bias dependence of TMR in this epitaxial work in spite of large interface roughness.

In summary, the MTJs were fabricated by sputter deposition at RT using an epitaxial NiFe as the bottom FM electrode. High TMR ratios were obtained and the bias dependence of TMR was remarkably reduced. It was clearly elucidated that it was induced by the formation of a well-defined sharp interface between the tunnel barrier and the FM electrode that is nearly free of crystalline defects. This MTJ has a large capability in engineering aspects if we can reduce the barrier thickness further by decreasing the interface roughness.

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