Magnetotransport properties of CoFeB/MgO/CoFe/MgO/CoFeB double barrier magnetic tunnel junctions with large negative magnetoresistance at room temperature

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Abstract. CoFeB/MgO/CoFe/MgO/CoFeB double-barrier magnetic-tunnel junctions were fabricated using an ultrahigh vacuum magnetron sputtering system, and their magnetotransport properties were characterized at room temperature. After post-deposition annealing, the polarity of TMR changed from negative to positive with increasing bias voltage. A relatively high negative TMR ratio of 30% was obtained at a negative bias voltage. Furthermore, a unique bias voltage dependence of conductance was observed at room temperature. This behavior may be attributable to the large minority density of states caused by the interfacial oxidation of the middle CoFe layer.

1. Introduction
Ever since the discovery of tunnel magnetoresistance (TMR) at room temperature [1], [2], magnetic tunnel junctions (MTJs) have become essential components in spintronics applications. The improvement in the quality of fabrication processes has led to the development of single MTJs with high TMR ratios; this has been achieved by employing a MgO barrier along with Heusler alloy or CoFeB electrodes. MTJs with high TMR ratios can be used to develop novel spintronics applications such as magnetic random access memories using spin-transfer torque switching (spin-RAMS) and spin-transistors. Double-barrier magnetic tunnel junctions (DBMTJs) have certain desirable features because of which they are considered to be one of the promising next-generation spintronics applications. For example, the decay of the TMR with bias voltage is significantly slower in DBMTJs than in single MTJs [3]. Then DBMTJs exhibit quantum well states [4] and resonant tunnelling at the bias voltages matches with the energy of the quantum well states; because of this feature, DBMTJs can be used as spontaneous oscillation devices and/or three terminals devices. Finally, DBMTJs can reduce the spin transfer switching current [5], which in turn can reduce the electrical consumption of the spin-RAM. In this study, we fabricated DBMTJs with a MgO barrier and systematically...
investigated the effects due to varying middle-layer thickness and the annealing temperature dependence of magnetotransport properties at room temperature.

2. Experimental procedure
CoFeB/MgO/CoFe/MgO/CoFeB DBMTJs with a thin middle layer were fabricated using an ultrahigh vacuum (UHV) magnetron sputtering system with a base pressure of less than $2 \times 10^{-6}$ Pa. The stacking structure was as follows: Si/SiO$_2$/Ta (10)/Ru (10)/IrMn (10)/Co$_{25}$Fe$_{75}$ (2)/Ru (0.8)/Co$_{08}$Fe$_{40}$B$_{20}$ (5)/MgO (2.5)/Co$_{08}$Fe$_{40}$ (t)/MgO (2.5)/Co$_{08}$Fe$_{40}$B$_{20}$ (5)/Ru (0.8)/Co$_{25}$Fe$_{75}$ (2)/IrMn (10)/Ta (10) (thickness in nm). The thickness of the Co$_{50}$Fe$_{50}$ ($t_{FeCo}$) middle layer was changed between 0.8 and 2.3 nm. After microfabrication, these DBMTJs were annealed at temperatures ranging from 523 to 623 K by applying a magnetic field of 10 kOe. The interfacial structure was confirmed by cross-sectional high resolution transmission electron microscopy (HRTEM) observation using an FEI Titan 80-300 TEM, operating at 300 kV with a CEOS image corrector. The magnetotransport properties were measured at room temperature using a standard four-probe method.

3. Results and discussion
3.1 Structure of middle Co$_{50}$Fe$_{50}$ layer
The structure of the middle Co$_{50}$Fe$_{50}$ layer was investigated by cross-sectional HRTEM analyses. Figure 1 shows the HRTEM images of DBMTJs with a middle layer having a thickness $t$ of 1.7 nm before and after annealing. As-deposited DBMTJs had continuous middle layers, and the exterior Fe$_{40}$Co$_{40}$B$_{20}$ layers had an amorphous structure. The HRTEM images of the interfaces between the MgO, Fe$_{40}$Co$_{40}$B$_{20}$, and Co$_{50}$Fe$_{50}$ layers were obscure. After annealing at 623 K, Fe$_{40}$Co$_{40}$B$_{20}$ layers were crystallized, and the interface between Co$_{50}$Fe$_{50}$ and MgO layers became clearer than that of the as-deposited DBMTJs. However, the lattice contrast was still not continuous, and an obscure area was observed near the interface between MgO and the Co$_{50}$Fe$_{50}$ middle layer. This suggests a possible interfacial oxidation of the middle Co$_{50}$Fe$_{50}$ layer; however, the details of such a process have not yet been clarified.

3.2 Magnetotransport properties of fabricated DBMTJs
Figure 2 shows the bias voltage dependence of the TMR ratio at various annealing temperatures and the typical TMR curves of a DBMTJ with a 1.7-nm-thick middle layer that was annealed at 573 K. The TMR ratio is defined as $(R_{ap} - R_{p})/R_{p}$, where $R_{ap}$ and $R_{p}$ are resistances of the antiparallel magnetization configurations between the middle Co$_{50}$Fe$_{50}$ layer and the exterior ferromagnetic Co$_{08}$Fe$_{40}$B$_{20}$ layers, respectively. Electrons flow from the bottom to the top electrode, at negative bias.
The as-deposited DBMTJs varied positive, normal TMR curves (not shown). However, after annealing, the polarity of TMR changed to negative at negative bias voltage. In this study, the largest negative TMR ratio obtained at room temperature was approximately 30%, which was obtained after annealing at 623 K; this value is comparable to the values reported in other studies. There are some reports of negative TMR caused by negative spin polarization due to a magnetic oxide layer [6], [7], which imply that the oxidization of the magnetic layer might cause negative TMR. The oxidation layer in DBMTJs can be elucidated by studying the relationships between the spin configurations and the magnetotransport properties using typical TMR curves. Figure 2 shows that the resistances were large at high magnetic fields with parallel spin configuration of the three magnetic layers and the resistance was the least at a small positive magnetic field. When the annealing temperature was increased to 623 K, the top junction broke, because the step near the zero field disappeared. This indicates that the negative polarization was caused by the oxidation of the bottom side of interface in the Co_{50}Fe_{50} layer due to the oxidation from MgO barrier during annealing.

Figure 2 Bias voltage dependence of TMR ratio at various annealing temperature, and the typical TMR curves of DBMTJs with middle layer thickness of 1.7 nm.

Figure 3 (a) Bias voltage dependence of conductance (G-V), (b) the electron transmission in parallel state, (c) and (d) electron transmission in antiparallel (AP) state at positive voltage and negative voltage respectively.
Figure 3(a) shows the bias voltage dependence of conductance ($G-V$). Although the bias voltage dependence of conductance was symmetric in the parallel (P) state, it exhibited a unique asymmetric behavior for negative bias with antiparallel (AP) state. At negative bias voltage, the conductance of AP state increased to more than that in the P state, and a small step was observed at -0.25 V in the AP state. DBMTJs that were annealed at different temperatures and that had middle Co$_{50}$Fe$_{50}$ layers of varying thickness exhibited similar tendencies with respect to the bias voltage dependence of the TMR ratio. Taking these results into consideration, we imaged the band structures of FeCo-O and FeCoB to illustrate the voltage dependence of conductance. The imaged band structures of FeCoO and FeCoB are shown in Fig. 3(b), (c), and (d). In the P state, majority tunneling is dominant, and therefore $dl/dV$ is symmetric. In the AP state, when a positive voltage was applied to the junction, the electrons traversed from the minority of FeCoO to majority of FeCoB; this led to the conductance normal in positive voltage. However, when a negative voltage was applied to the junction, the electrons traversed from FeCoB to FeCoO. Application of a suitable voltage would lead to the minority density of state of FeCo-O shift to largest position, thus enhancing the conductance. Here, the negative TMR can be explained by assuming a large minority density of state (DOS) of FeCo-O above $E_F$. However, it will be necessary to carry out further investigations using HRTEM before it can be confirmed that the oxidation of only the bottom side of the middle Co$_{50}$Fe$_{50}$ layer induced the large negative TMR ratio of 30% and the related unique bias voltage dependence of conductance.

4. Summary
We fabricated DBMTJs with a MgO barrier using a UHV sputtering system, and we investigated their magnetotransport properties at room temperature. The polarity of TMR became negative on increasing the bias voltage, and a large negative TMR of 30% was obtained at -0.4 V at room temperature. The bias voltage dependence of the conductance curves indicates that the negative TMR observed at negative bias voltage is attributable to the large Fermi level in the minority band structure. The cross-sectional HRTRM images showed that the interface between MgO and the Co$_{50}$Fe$_{50}$ middle layer became clear after heat treatment. However, an obscure area was observed near the interface between the MgO and Co$_{50}$Fe$_{50}$ middle layer; this obscure area was identified as a magnetic oxide layer.

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