Dependence of Giant Tunnel Magnetoresistance of Sputtered CoFeB/MgO/CoFeB Magnetic Tunnel Junctions on MgO Barrier Thickness and Annealing Temperature

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We investigated the dependence of giant tunnel magnetoresistance (TMR) on the thickness of an MgO barrier and on the annealing temperature of sputtered CoFeB/MgO/CoFeB magnetic tunnel junctions deposited on SiO$_2$/Si wafers. The resistance-area product exponentially increases with MgO thickness, indicating that the quality of MgO barriers is high in the investigated thickness range of 1.15–2.4 nm. High-resolution transmission electron microscope images show that annealing at the annealing temperature and can be as high as 260% at RT and 403% at 5 K. [DOI: 10.1143/JJAP.44.L587]

KEYWORDS: tunnel magnetoresistance effect, magnetic tunnel junction, MgO barrier, CoFeB, crystallization

MTJ films with synthetic pin layers were formed on SiO$_2$/Si substrates. The order of the film layers was as follows, starting from the substrate side: Ta(5)/Ru(50)/Ta(5)/NiFe(5) / MnIr(10) / CoFe(2) / Ru(0.8) / CoFeB(3) / MgO / CoFeB(3)/Ta(5)/Ru(5). The numbers in parentheses indicate the thickness in nm of the layers, and the thickness of the MgO barrier was varied from 1.15 to 2.4 nm. We use CoFe to represent a Co$_{90}$Fe$_{10}$ alloy and CoFeB to represent a Co$_{90}$Fe$_{10}$B$_{20}$ alloy. The MgO layer was formed using an MgO target at a pressure of 1 mTorr in a Ar atmosphere. The MTJs were annealed at 270 to 375°C for 1 h in a vacuum of 10$^{-6}$ Torr under a magnetic field of 4 kOe. All junctions, sized from 0.8 × 0.8 μm$^2$ to 0.8 × 5.6 μm$^2$, were fabricated using a conventional photolithography process. An atomic force microscopy (AFM) image of the surface of Ta/Ru/Ta underlayer (ρ = 7.5 μΩ·cm) indicating that surface is smooth with $R_a$ of 0.17 nm.

MTJs were annealed at 375°C for 1 h with a 2.0 nm MgO barrier, typical TMR ratios (ΔR/R) versus magnetic field (H) loops measured at RT (dashed line) and
5 K (solid line) are shown in Fig. 2; the resistance-area product (RA) is 3.4 kΩm² at RT and 3.5 kΩm² at 5 K. We found that the TMR ratio is as high as 260% at RT, and reaches 403% at 5 K; these are the highest yet reported TMR ratios of MTJs using conventional 3d ferromagnetic metals and oxide barriers. By using Julliere’s formula, these TMR ratios are found to correspond to tunneling spin-polarizations of 0.75 and 0.82. The exchange bias field, TMR ratios are found to correspond to tunneling spin-polarizations of 0.75 and 0.82. The exchange bias field, 

\[ T = 375 \text{ C} \]

The exponential increase of RA with increasing MgO barrier thickness (tMgO), shown in Fig. 3(a) indicates that pinhole-free barriers with constant barrier height (φ) are formed and maintained at elevated T. The φ which is estimated using the Wenzel-Kramer-Brillouin approximation is in the range of 0.34–0.38 eV, indicating that it varies little upon annealing. In the ranges from 1.35 to 2.2 nm, the TMR ratio increases dramatically with increasing T. For example, in a MTJ with a 2.0 nm MgO barrier, the TMR ratio is 100% when \( T = 270 \text{ C} \), 200% when \( T = 325 \text{ C} \), and 260% when \( T = 375 \text{ C} \). However, the ratio does not significantly increase with increasing MgO barrier thickness when tMgO = 2.4 and 1.15 nm. The 110% TMR ratio at tMgO = 1.15 nm, however, is still four times greater than the ratios of aluminum oxide barrier MTJs with similar RAs of 30 Ωm².

The structure of similarly prepared samples without Ru in their underlayers with 240% TMR ratios was explored using high-resolution cross-sectional transmission electron microscopy (HRTEM). The HRTEM images in Fig. 4 show the regions containing the CoFeB(3)/MgO(2)/CoFeB(3) interfaces for samples as-deposited, (b) after annealing at 270 °C, and (c) after annealing at 375 °C. We can see three features in these images. First, as-deposited CoFeB electrodes have amorphous structures (Fig. 4(a)), and this structure remains amorphous after annealing at 270 °C (Fig. 4(b)). Second, the MgO barrier has NaCl structure that is highly (001) oriented and has been uniformly deposited on the amorphous CoFeB bottom ferromagnetic layer (Figs. 4(a) and 4(b)). Both of these observations are in accordance with a previous report. Third, by annealing at 375 °C, where the highest TMR ratio is obtained, full crystallization of the CoFeB ferromagnetic electrodes in body centered cubic structure is observed (Fig. 4(c)).

The highest TMR ratio together with the crystalline structure observed in the samples annealed at 375 °C strongly suggest that the origin of the giant TMR ratios in MgO-based MTJs with amorphous ferromagnetic electrodes resides in the fact that, in addition to the MgO barrier, the ferromagnetic electrodes, at least in the vicinity of the MgO barrier, is of crystalline nature, as Djavaprawire et al. speculated. This may resolve the apparent inconsistency with theory; theoretical studies have indicated that the giant TMR is an effect of a particular band structure combination between the crystalline barrier and the electrode material resulting in an effective half-metallic electrode. This combination is not possible when amorphous CoFeB ferromagnetic electrodes are crystallized by post annealing at 375 °C.
electrodes are used.

We believe that the crystallization process of CoFeB electrodes is initiated at both MgO/CoFeB interfaces and proceeds to the entire CoFeB layers. This is supported by Fig. 4(c), which shows smooth MgO/CoFeB interfaces, contrasting with the amorphous Ta/CoFeB upper interface and the rough CoFeB/Ru lower interface.

Current ($I$) is plotted in Fig. 5(a) as a function of bias voltage ($V$) in an MTJ that has a TMR ratio of 260% at RT. The solid line shows the data in a parallel (low-resistance) configuration and the dashed line shows data in an anti-parallel (high-resistance) configuration. Simmons’ equation\(^{13}\) yields $\phi = 0.4\, \text{eV}$ for a barrier thickness $d = 2.18\, \text{nm}$ using the anti-parallel data, which is in good agreement with values reported previously.\(^{5,9}\) The $I$–$V$ curve in the parallel configuration indicates virtually ohmic transport, while the anti-parallel configuration shows typical non-linear tunnel characteristics; note that for MTJs using amorphous aluminum oxide barriers, non-linear $I$–$V$ curves are generally observed in both parallel and anti-parallel configurations. The $I$–$V$ characteristics in the parallel configuration may be due to the matching of the symmetry of the tunneling electronic states predicted by theoretical studies.\(^{4–6}\)

The normalized TMR ratio (solid line) and output voltage ($\Delta V \equiv V_p \times (R_{ap} - R_p) / R_p$, dotted line) are plotted in Fig. 5(b) as functions of bias voltage. The TMR ratio was found to decrease with increasing bias voltage, and only a slight asymmetry was observed for positive and negative bias voltages compared with that in fully epitaxial Fe/MgO/Fe MTJs.\(^5\) The TMR ratios dropped to half at voltages of about 600 mV at positive bias and about 700 mV at negative bias. This asymmetry may have been caused by the differences in the electronic states at the MgO/CoFeB interfaces. Note that $\Delta V$ at negative bias reaches approximately 450 mV, which is approximately three times that of MTJs with amorphous aluminum oxide barriers.\(^{14}\)

In conclusion, we reported the dependence of MgO-based MTJs prepared using conventional sputtering on the thickness of MgO barriers and on the annealing temperature. By annealing at 375°C, CoFeB ferromagnetic electrodes separated by highly (001) oriented MgO are fully crystallized in body centered cubic structure. The increase in the TMR ratio due to annealing can be attributed to the formation of highly oriented crystalline CoFeB/MgO/CoFeB MTJs. The obtained TMR ratio in our experiment was as high as 260% at RT and 403% at 5 K. We observed an output voltage of up to 450 mV. The MTJs have characteristic $I$–$V$ responses in parallel configurations, which has not been seen in MTJs with amorphous aluminum oxide barriers.

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