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Reduced temperature and bias-voltage dependence of the magnetoresistance in magnetic tunnel junctions with Hf-inserted Al_2O_3 barrier

Byong Guk Park[a] and Taek Dong Lee
Department of Materials Science and Engineering, KAIST, 373-1, Kusong-dong, Yusong-gu, Taejeon, 305-701, Korea

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A modified tunnel barrier structure for the magnetic tunnel junction (MTJ) was fabricated by inserting a Hf layer in the middle of the Al_2O_3 tunnel barrier. MTJs with the Hf-inserted barrier show a higher tunnel mangnetoresistance (TMR) ratio and weaker temperature and bias-voltage dependence of TMR compared to the MTJs with a conventional Al_2O_3 barrier. The enhancement of the TMR ratio and the reduction of the temperature and bias-voltage dependence were attributed to the reduction of defects in the barrier. © *2002 American Institute of Physics.*

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Magnetic tunnel junctions (MTJs) have been extensively studied for technological applications in devices such as magnetic recording heads or nonvolatile magnetic random access memory[1,2] since the discovery of the large magnetoresistance effect at room temperature.[3,4] However, the tunneling magnetoresistance (TMR) ratio has been observed to decrease significantly with increasing temperature and bias voltage, which can hinder industrial application of the TMR junction. Zhang and White[5] proposed that the temperature and bias-voltage dependence of the TMR could be explained by spin independent two-step tunneling via defect states in the barrier. The key issue to improve the TMR ratio and to minimize the temperature and bias-voltage dependence is to fabricate a high quality tunnel barrier with low defect density. Shang *et al.*[6] also suggested that the temperature dependence of the TMR could be described by two current contribution; spin dependent and spin independent contribution. Spin dependent contribution with increasing temperature is based on the reduction of polarization and broadening of the Fermi distribution while spin independent contribution is due to a hopping process via localized defect states in the barrier.

In this letter, we have modified the oxide barrier by inserting a Hf layer in the middle of Al film prior to the oxidation. Since Hf oxide has a larger heat of formation (−268 Kcal/mol) than Al oxide (−202 Kcal/mol) does, the Hf layer could act as an oxide stabilizer at high temperature and decrease the defects in the tunnel barrier. MTJs with a Hf-oxide inserted barrier were fabricated and their properties such as the TMR ratio, junction resistance, and temperature and bias-voltage dependence were investigated and compared with those from the conventional Al_2O_3 barrier junctions.

MTJs consisting of Hf (5 nm)/$Ni_{81}Fe_{19}$ (8 nm)/$Ir_{25}Mn_{75}$ (20 nm)/$Co_{50}Fe_{50}$ (4 nm)/Al oxide or Al–Hf–Al oxide (Hf-inserted barrier)/$Co_{50}Fe_{50}$ (3 nm)/$Ni_{81}Fe_{19}$ (15 nm)/Hf (5 nm) were deposited on the thermally oxidized Si wafer by a magnetron sputtering system. The tunnel barriers were formed by depositing an Al layer or Al/Hf/Al layer and subsequently exposing to an ozone atmosphere for tens of minutes. The oxidation process involving ozone generation by dielectric the barrier discharge method was described in our previous work.[7,8] A Hf layer of 0.2 nm was inserted in the middle of the Al layer and the total thickness of the barrier was kept around 1.4 nm. These samples were annealed at temperatures from 250 to 350 °C in vacuum ($\sim 10^{-6}$ Torr) for 50 min. A magnetic field of 1 kOe was applied along the easy direction of magnetization during the annealing process. Magnetoresistance was measured using a four probe method at temperatures ranging form 30 to 300 K. $I-V$ characteristics and bias-voltage dependence were obtained by sweeping the voltage from 5 to 800 mV.

Figure 1 shows the TMR curves for junctions with Al oxide (a) and Hf-inserted (b) barriers after annealing at different temperatures. The TMR ratio of the as-deposited junctions was near 23% for both junctions and increased when annealed at 300 °C. The TMR ratio, however, dropped sharply above 300 °C for both junctions. The TMR ratio for the junctions with the Hf-inserted barrier was 38% after annealing at 300 °C whereas, without the Hf layer, the TMR ratio was only 30%. Although the TMR ratio decreased when the junctions were annealed above 300 °C, the Hf-inserted junction maintained a substantially high TMR ratio of 28% even after annealing at 350 °C.

By fitting of $I-V$ curves to the Simmons theory,[9] the barrier width and height for the junctions were estimated.[10] The junctions with the Hf-inserted tunnel barrier had the barrier height of 2.2 eV, which is higher compared to the junctions with a normal Al oxide tunnel barrier. The barrier parameters of the junctions with the Hf-inserted barrier did not change after annealing at 350 °C at which the TMR ratio was observed to drop significantly, whereas the barrier height decreases and the barrier width increases after annealing at 350 °C for the junctions without the Hf layer. The results suggest that the Hf-inserted barrier is more stable than the conventional Al oxide barrier and that the enhancement of the TMR ratio is attributable to the improved tunnel barrier quality through insertion of the Hf layer.

Figure 2 shows the temperature dependence of the TMR

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[a] Electronic mail: bgpark@kaist.ac.kr

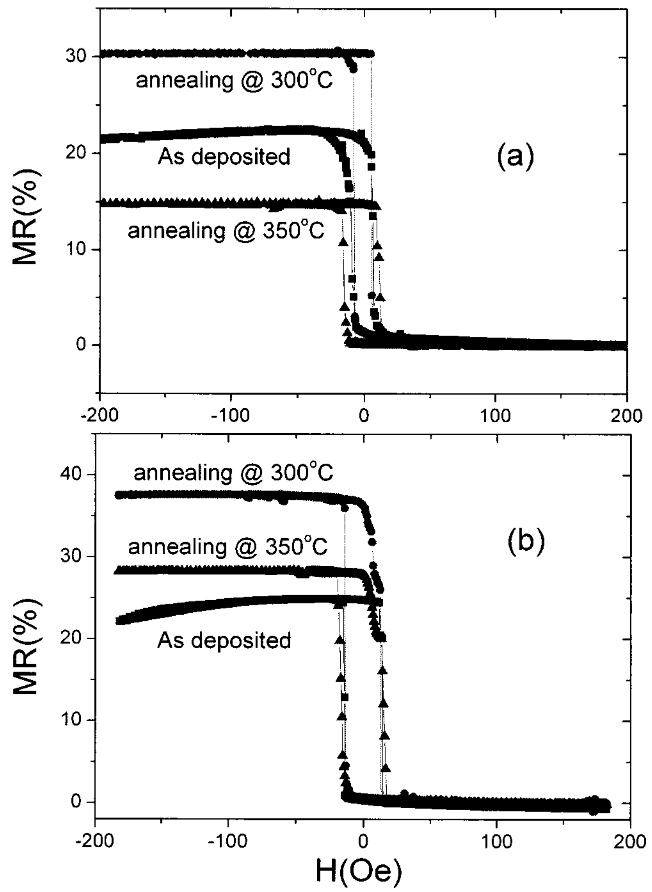

FIG. 1. TMR curves for the as-deposited junctions and the same junctions after annealing at 300 and 350 C. The curves correspond to junctions with Al oxide (a) and those with a Hf-inserted barrier (b).

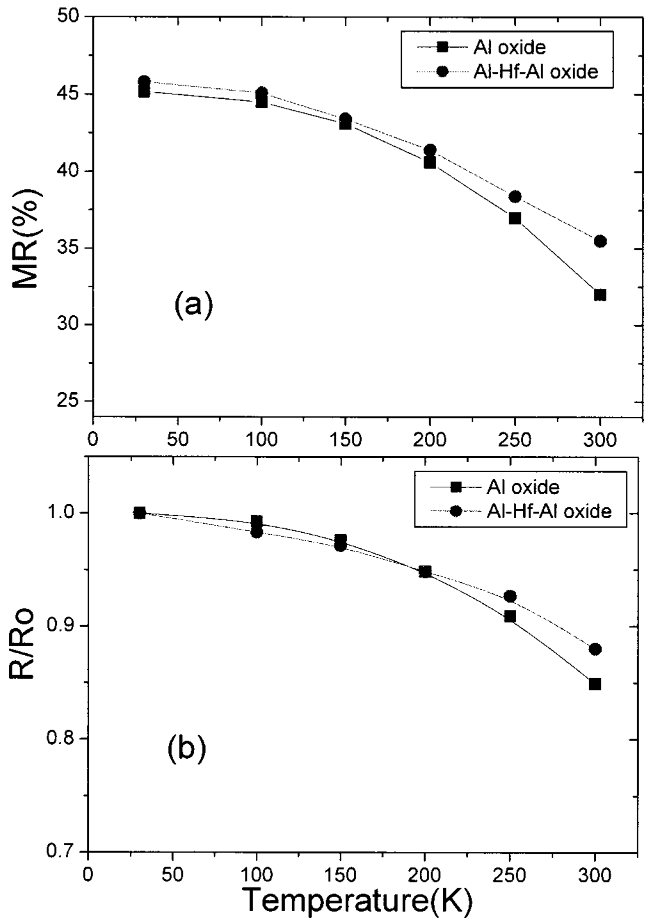

FIG. 2. Temperature dependence of the TMR (a) and junction resistance (b) for junctions with different tunnel barriers: Al oxide barrier (square) and Al–Hf–Al oxide barrier (circle).

ratio (a) and the junction resistance (b) for the junctions with Al oxide (square) and Al–Hf–Al oxide (circle) tunnel barriers. As the samples were cooled from room temperature, the TMR and junction resistance increased for both samples. The TMR ratio of the junction with Al oxide increased from 31% to 45% while the TMR ratio of the junction with the Hf-inserted barrier improved from 36% to 45.5% as the measuring temperature was lowered from 300 to 30 K. The difference in the TMR ratio between the two types of junctions at room temperature is 5% in absolute value; however, the difference becomes nearly negligible as the temperature was decreased to 30 K. The junction resistance also increased by 17.5% for the junction with the normal Al oxide barrier and by 14% for the junction with the Hf-inserted barrier. The junctions with the normal Al oxide barrier appear to have a stronger dependence of the TMR ratio and the junction resistance on the temperature compared to the junctions with the Hf-inserted tunnel barrier.

According to the model proposed by Shang *et al.*,[6] the temperature dependence of the tunnel junction is due to the spin dependent and spin independent tunneling. In their model, the total conductance was expressed as

$$G(\theta)=G_T(1+P_1P_2\cos\theta)+G_{\mathrm{SI}}, \tag{1}$$

where θ is the angle between the magnetization directions of the two electrodes, P is the effective spin polarization, G_T is the conductance for direct elastic tunneling through the junction, and G_{SI} is additional conductance due to spin independent contribution. It was suggested that hopping conductance via localized state is responsible for the spin independent contribution.[6]

Figure 3(a) shows the $\Delta G(T)/\Delta G(30\ \mathrm{K})$ as a function of temperature for the junction with the two different types of barriers. The change of conductance $[\Delta G(T)=G(\theta=0)-G(\theta=180)]$ does not contain spin independent contribution, but contains contribution from the reduction of spin polarization with temperature. In Fig. 3, $\Delta G(T)$ decreased approximately by 6% as the temperature increased from 30 to 300 K for both junctions, which indicates an equal magnitude of spin polarization reduction of the magnetic electrodes for both junctions. This result is expected because the same electrodes were used in both junctions and the electrode material was in contact with Al oxide.

Figure 3(b) shows the dependence of $G_{\mathrm{SI}}(T)/G_{\mathrm{SI}}(30\ \mathrm{K})$ on the temperature for the junctions with an Al oxide barrier and Al–Hf–Al barrier. The term G_{SI} represents the spin independent contribution of the MTJ, which increases total conductance but reduces the TMR ratio. From Eq. (1), G_{SI} is given by

$$G_{\mathrm{SI}}=\langle G\rangle-G_T, \tag{2}$$

where $\langle G\rangle$ is the average conductance over parallel and antiparallel magnetization. By fitting the experimental data to $G_{\mathrm{SI}}(T)\propto T^{\gamma}$, we obtained $\gamma\ (\mathrm{Al\ oxide})=2.16$ and $\gamma\ (\mathrm{Al{-}Hf{-}Al\ oxide})=1.7$. This result suggests that the in-

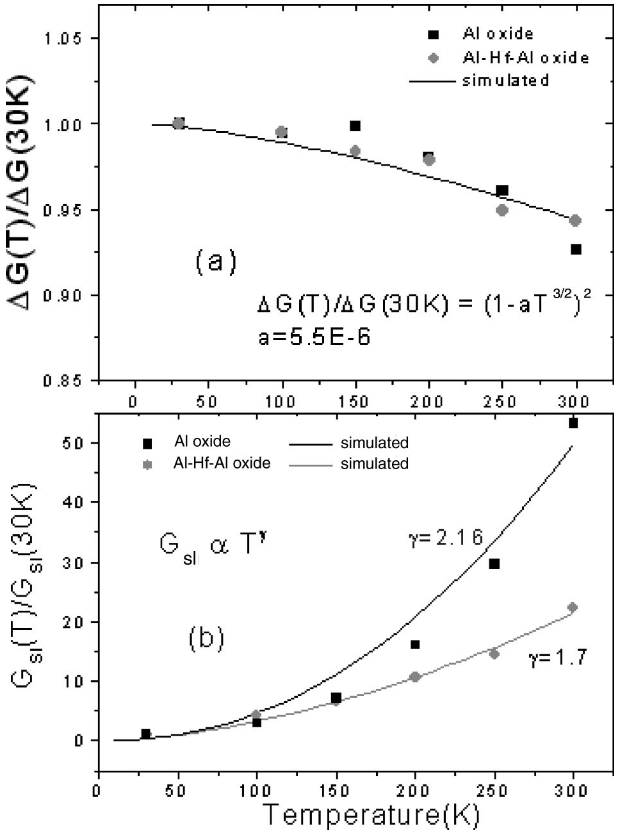

FIG. 3. Temperature dependence of the normalized ΔG (a) and spin independent conductance G_{SI} (b) for junctions with the Al oxide barrier (square) and Al–Hf–Al oxide barrier (circle). The lines represent the theoretical fits.

sertion of Hf would reduce the defects in the barrier as γ mainly depends on the defect concentration.[11]

Figure 4 shows the bias-voltage dependence of the TMR ratio for the junctions with Al oxide (square) and Hf-inserted barrier (circle) after annealing at 300 °C. The TMR ratio decreased gradually with increasing bias voltage. The voltage $V_{1/2}$, at which the TMR is reduced by 50%, was around 625 mV for the junctions with the Hf-inserted barrier and 530 mV for the junctions with the normal Al oxide barrier. This indicates that modifying the barrier structure by inserting the Hf layer reduces the bias-voltage dependence of the TMR. This reduced bias-voltage dependence could be explained by the same reason used for the reduced temperature dependence. As disorder or defects in the tunnel barrier are known to increase the bias-voltage dependence through increased contribution from the spin independent parts of the two step tunneling or other spin flip processes.[5]

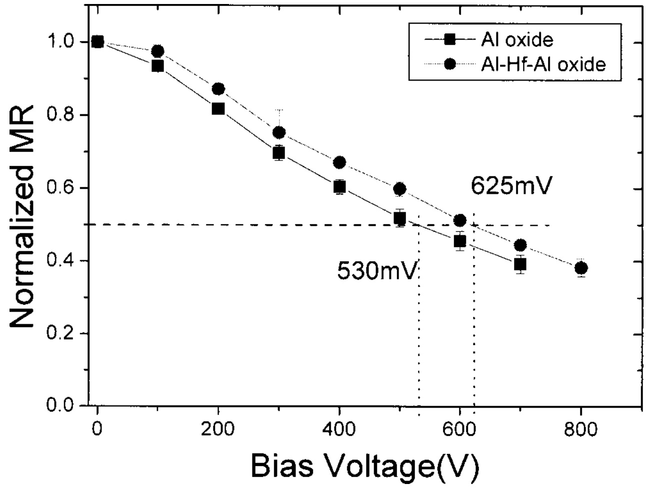

FIG. 4. Bias-voltage dependence of the TMR for junctions with Al oxide (square) and Hf-inserted oxide (circle) after 300 °C annealing. The voltage $V_{1/2}$, at which the TMR is reduced by 50%, is around 625 mV for junctions with Hf-inserted oxide and 530 mV for junctions with Al oxide.

In conclusion, we introduced the artificial modification of the oxide barrier by inserting the Hf layer into the Al oxide tunnel barrier. The Hf-inserted barrier enhances the TMR effect and reduces the temperature and bias-voltage dependence of TMR ratio by decreasing the defect concentration in the tunnel barrier.

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