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Abnormal Temperature Dependence of Tunneling Magnetoresistance for Magnetic Tunnel Junctions

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Magnetic tunnel junctions (MTJs) were fabricated with high bias for plasma oxidation and the effects of annealing on the temperature dependence of tunneling magnetoresistance (TMR) were investigated experimentally. As-grown, TMR increases, peaks around 160 K, and decreases with increasing temperature from 80 K to 300 K. When MTJs are annealed, T_{max}, the temperature at which maximum TMR is obtained, decreases as annealing temperature increases to the optimal point. In order to explain this abnormal temperature dependence of TMR, the difference of conductance between parallel and antiparallel alignments of magnetizations as a function of temperature is also analyzed. The shifts of T_{max} due to annealing process are described phenomenologically with spin-dependent transfer rates of electrons tunnel through the barrier.

Key words : Mangetic Tunnel Junction, Tunneling Magnetoresistance

1. Introduction

Sizable room temperature tunneling magnetoresistance (TMR) in magnetic tunnel junctions (MTJs) [1, 2], composed of two ferromagnetic electrodes seperated by a thin insulating barrier, has ignited the intensive research both from scientific and technological points of view [3, 4]. A simple model proposed by Julliere has explained the observed TMR relatively well, where the TMR is expressed in terms of the spin polarization of the ferromagnetic electrodes [5]. After that report, theoretical and experimental studies of the temperature dependence of TMR have been conducted to investigate the transport mechanism of spin-dependent tunneling in MTJ. It is well known that the appropriate annealing enhances TMR at room temperature [6], but the effects of annealing on the temperature dependence of TMR are not much studied. In this work, the effects of annealing on the temperature dependence of TMR are investigated experimentally, and it is found that the T_{max}, the temperature where the maximum TMR value is obtained, decrease with annealing. Analysis on the temperature dependence of conductance in parallel and antiparallel configurations of ferromagnetic layers suggests the presence of spin-dependent current via impurities, which diminish with the thermal treatment.

2. Experiment

Bottom exchange-biased magnetic tunnel junctions with the layer stacks of 5 nm Ta/6 nm $Ni_{81}Fe_{19}$/8 nm $Fe_{50}Mn_{50}$/4 nm $Co_{84}Fe_{16}$/1.6 nm Al_2O_3/2 nm $Co_{84}Fe_{16}$/10 nm $Ni_{81}Fe_{19}$/5 nm Ta (from bottom to top) were fabricated *in-situ* on thermally oxidized Si(100) wafers by 6-target DC magnetron sputter system with a base pressure better than 4×10^{-8} Torr. Tunneling barrier (Al_2O_3) was formed by plasma oxidation of 1.6 nm thick Al in a seperated load-lock chamber, by applying relatively high DC bias (-150W) to Al target in pure O_2 of 20 mTorr. The cross geometry junctions were fabricated by photolithography and ion beam etching process and the junction size is 50 μm × 50 μm. Four point probe method was used for characterization in the temperature range of 77 K~300 K and magnetic field up to 1 Tesla in Quantum Design PPMS (Physical property measurement system). A series of samples were heat treated by the rapid thermal annealing (RTA) process for 10 sec at the each annealing temper-

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*Corresponding author: Tel: +82-2-958-5418, Fax: +82-2-958-6851, e-mail: kshin@kist.re.kr atures (T_A) of 250 °C, 300 °C, and 400 °C.

3. Results and Discussion

Figure 1 shows the temperature dependence of TMR for samples annealed at different temperatures. The solid circles represent the TMR of as-grown junction, and the triangles, the open circles, and the squares are for MTJs annealed at 250 °C, 300 °C, and 400 °C, respectively. The TMR is improved significantly by annealing at 250 °C and 300 °C, but deteriorates severely when annealed at 400 °C. The deterioration at high annealing temperature may be attributed to reaction between the oxide barrier and adjacent ferromagnetic layers [7] or the interdiffusion of Mn at the interface of CoFe and FeMn [8, 9]. The optimal temperature for the RTA process is about 300 °C. At this annealing temperature, the TMR value reaches to 48% measured at room temperature and 59% at 80 K. It is known that the TMR usually decreases monotonically with increasing temperature mainly due to the decrease of the polarization [10, 11]. However, our samples show an abnormal temperature dependence of TMR. It is surprising that the TMR increases as a function of temperature in a certain range. This is in contrast to the temperature dependence of TMR observed by others [10, 12, 13]. As grown, the TMR increases with increasing temperature from 80 K to 160 K and decreases thereafter. For the junction annealed at 250 °C, the highest TMR is observed at 140 K. When annealed at 300 °C, the temperature dependence is similar to those observed by others, but still the TMR measured at 100 K is slightly larger than that measured at 80 K. This novel temperature dependence of TMR seems to be related to the high plasma oxidation power (-150 W). This high bias for oxygen plasma is a prerequisite to observe the novel features of our MTJs and may cause to form impurity states in the tunneling barrier [14], as well as, MTJs fabricated with low plasma power exhibit usual decrease of TMR as a function of temperature [13, 15].

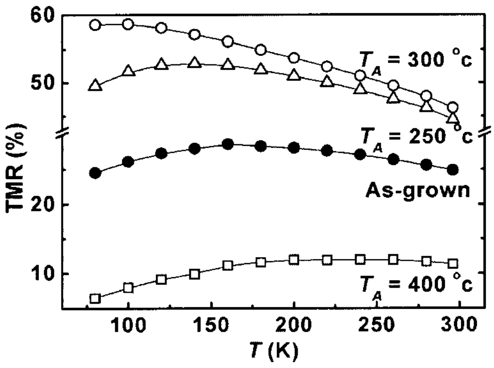

Fig. 1. Temperature dependence of TMR of MTJs annealed at different temperatures. (T_A represents the annealing temperature).

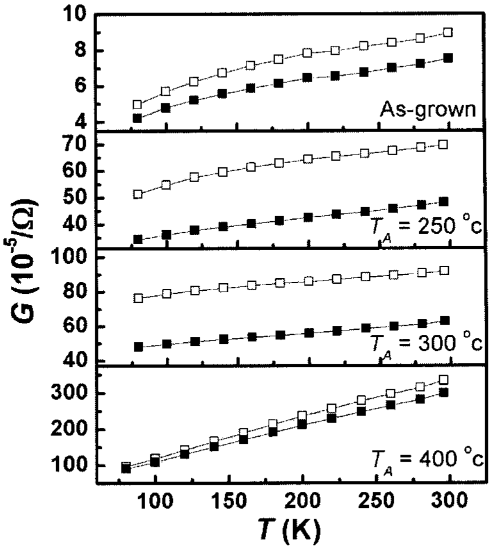

Fig. 2. Temperature dependence of junction conductance for MTJs annealed at different temperatures. The top panel if for as-grown and the followed are for MTJs annealed at 250 °C, 300 °C, and 400 °C. The solid and open symbols represent the parallel and antiparallel configurations of magnetization, respectively.

In Fig. 2, the junction conductance G is plotted as a function of temperature. From the top to the bottom panel, shown is the conductance for MTJs as grown, annealed at 250 °C, 300 °C, and 400 °C, respectively. The solid (open) squares represent the parallel (antiparallel) magnetization configurations of the ferromagnetic electrodes. Annealing process affects the behavior of the junction conductance especially in two aspects. First, the magnitude of the conductance is enhanced significantly after the annealing process. Second, the temperature dependence of conductance becomes weaker for junctions annealed at 250 °C or 300 °C. Again, the MTJ annealed at 400 °C shows different behavior from other junctions. As grown, the conductance increases by about 80% as temperature changes from 80 K to 300 K. This is rather strong temperature dependence compared with results by Shang *et al.* [10]. In the same temperature range, 35% increase of conductance is observed for the junction annealed at 250 °C and 20% increase for the MTJ annealed at 300 °C. When annealed at 400 °C, the conductance increases much more rapidly with temperature than the as-grown sample.

In order to analyze the temperature dependence of TMR, it is useful to investigate the difference of conductance between parallel and antiparallel alignments of magneti-

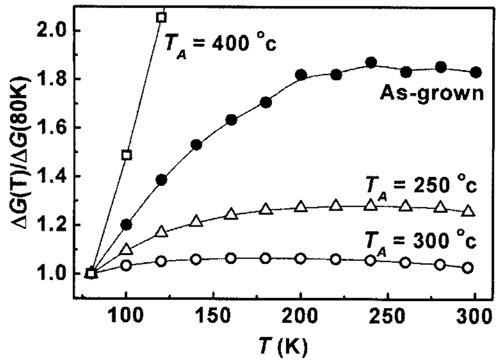

Fig. 3. Temperature dependence of the normalized ΔG for MTJs annealed at different temperatures: as-grown (solid circles), annealed at 250 °C (triangles), 300 °C (open circles), and 400 °C (squares).

zations. The difference of conductance is denoted as $\Delta G = G_P - G_{AP}$, where G_P (G_{AP}) is the conductance for the parallel (antiparallel) alignment of magnetizations. In Fig. 3, the normalized ΔG is displayed for MTJs annealed in different conditions. The solid circles are for as-grown sample, and the open triangles, the open circles, and the open rectangles are for junctions annealed at 250 °C, 300 °C, and 400 °C, respectively. As-grown, the normalized ΔG is observed to increase and saturate with increasing temperature. When the junction is annealed at 250 °C, the normalized ΔG decreases slightly at higher temperatures. For the junction annealed at 300 °C the decrease of normalized ΔG at higher temperatures is seen more clearly.

Shang *et al.* [10] observed that normalized ΔG decreases with increasing temperature. They assumed that the junction conductance consists of spin-dependent and spin-independent parts. They further argued that the spin-dependent conductance decreases due to that of spin polarization in the magnetic layer and, consequently, the normalized ΔG decreases. Zhang and White [12] suggested that currents through vacancies in the Al_2O_3 barrier contribute to the spin-independent conductance. Regardless of the origin, the presence of spin-independent conductance diminishes TMR. Thus, the increase of ΔG in our results strongly suggests that there exists another source of spin-dependent conductance which increases as a function of temperature.

The aforementioned effects of annealing on the junction conductance can be explained in terms of the change in the oxide barrier. It is considered that the oxygen ions with high energy may penetrate through Al and reach to the bottom ferromagnetic electrode and form magnetic impurities during the plasma oxidation process. In Fig. 4, study of transmission electron microscopy on these junctions reveals that the interface of Al_2O_3 and pinned layer is obscure before annealing, and becomes clear after annealing. One may safely assume that the initially oxidized ferromagnetic electrode is deoxidized during the annealing process. Thus, the effective barrier thickness is lager than expected as grown and decreases after annealing, which results in the increase of conductance when the junction is annealed. Since the Al layer is oxidized more easily, it is suspected that the ferromagnetic electrode is not evenly oxidized, and unoxidized magnetic particles can be considered as impurities. Conductance *via* impurity states is strongly dependent on temperature [12]. When annealed, oxygen migrates from the oxidized magnetic layer to Al_2O_3 layer and the impurities decrease as the ferromagnetic layer is recovered. This qualitatively explains the temperature dependence of the conductance after annealing. Furthermore, conductance *via* magnetic impurity states is spin-dependent and the increase of ΔG as a function of temperature can be explained as well. By annealing, magnetic oxide layer becomes thinner and number of magnetic impurities decreases and the spin-dependent impurity effect diminishes until optimum annealing temperature. This explains the temperature dependence of G and normalized ΔG for junctions as grown, and annealed at 250 °C and 300 °C but it cannot be applied to the junction annealed at 400 °C since the junction is literally destroyed during the annealing process [7-9].

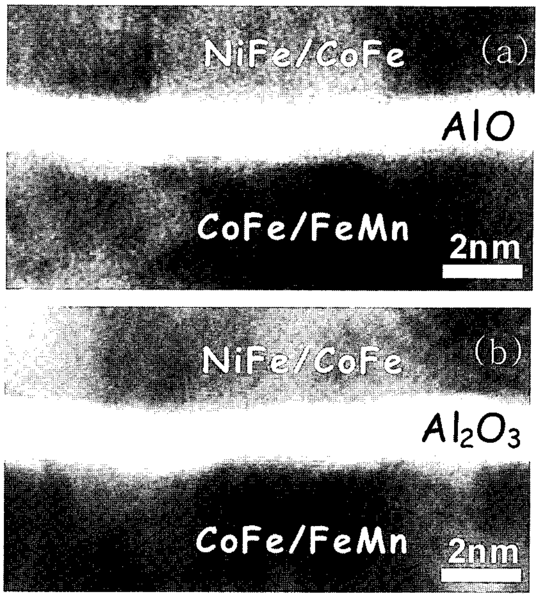

Fig. 4. Cross-sectional TEM micrographs (a) for as-grown MTJ and (b) for MTJ annealed at 300 °C.

Based on these arguments and the Julliere model [5, 11], the conductance of each spin-dependent conductance can be phenomenologically expressed as

$$G_P^{sd} \propto t_\uparrow N_{B\uparrow}(\varepsilon_F)N_{T\uparrow}(\varepsilon_F)+t_\downarrow N_{B\downarrow}(\varepsilon_F)N_{T\downarrow}(\varepsilon_F), \qquad (1)$$

$$G_{AP}^{sd} \propto t_\uparrow N_{B\uparrow}(\varepsilon_F)N_{T\downarrow}(\varepsilon_F)+t_\downarrow N_{B\downarrow}(\varepsilon_F)N_{T\uparrow}(\varepsilon_F), \qquad (2)$$

where t is the transfer rate of electrons, $N(\varepsilon_F)$ is the density of states of electrons at the Fermi energy ε_F, subscript $\uparrow$ ($\downarrow$) denotes the majority (minority) spin, and subscript $B(T)$ stands for the bottom (top) ferromagnetic layer. Since the magnetization of the magnetic impurity is in the same direction as that of the bottom magnetic layer, the transfer rate of the electrons with the majority spin in the bottom magnetic layer can be taken as $t_\uparrow$. The TMR is given as

$$\frac{G_P - G_{AP}}{G_{AP}} = \frac{2(P_B P_T + \tau P_T)}{1 - P_B P_T + \tau(P_B - P_T)}, \qquad (3)$$

where P is the spin polarization of $N(\varepsilon_F)$ with $P = (N_\uparrow - N_\downarrow)/(N_\uparrow + N_\downarrow)$ and τ is $\tau = (t_\uparrow - t_\downarrow)/(t_\uparrow + t_\downarrow)$. When there are no magnetic impurities, we have $t_\uparrow = t_\downarrow$ and Eq. (3) becomes the Julliere model. In our case, the same material is used for both ferromagnetic layers ($Co_{84}Fe_{16}$) and we have $P_B = P_T = P$. We assume that the energy of impurity states is spin-dependent and the conductance via magnetic impurity states is dominated by the majority spin. Introduction of magnetic impurities changes mainly $t_\uparrow$ and increases τ. Annealing decreases the number of impurities and, consequently, the magnitude of τ. For a given number of magnetic impurities, τ increases within a certain temperature range, which causes the increase of TMR in spite of the decrease of P. Eventually τ has a maximum value at a certain temparature and TMR begins to decrease thereafter. Thus, the temperature dependence of TMR and effects of annealing shown in Fig. 1 can be explained in terms of the phenomenological parameter τ.

4. Conclusion

In this study, we investigated the effects of annealing on the temperature dependence of TMR. The highest TMR value was observed at 160 K for the as-grown sample. When junctions are annealed at 250 °C and 300 °C, the temperature where the maximum TMR is observed shifts to 140 K and 100 K, respectively. This behavior is attributed to the oxidation of the bottom magnetic layer. The increase of TMR as a function of temperature in a certain range is explained phenomenologically with spin-dependent transfer rates of electrons through the barrier.

Acknowledgement

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