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# Fabrication of high-magnetoresistance tunnel junctions using $Co_{75}Fe_{25}$ ferromagnetic electrodes

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Temperature dependence of tunnel magnetoresistance (TMR) ratio, resistance, and coercivity from 4.2 K to room temperature and applied voltage dependence of the TMR ratio and resistance at room temperature for a tunnel junction, Ta (5 nm)/$Ni_{79}Fe_{21}$ (3 nm)/Cu (20 nm)/$Ni_{79}Fe_{21}$ (3 nm)/$Ir_{22}Mn_{78}$ (10 nm)/$Co_{75}Fe_{25}$ (4 nm)/Al (0.8 nm)-oxide/$Co_{75}Fe_{25}$ (4 nm)/$Ni_{79}Fe_{21}$ (20 nm)/Ta(5 nm), were investigated. TMR ratio, effective barrier height and width, and breakdown voltage of the junction can be remarkably enhanced after annealing at 300 °C for an hour. High TMR ratio of 49.7% at room temperature and 69.1% at 4.2 K were observed. The value of spin polarization of $Co_{75}Fe_{25}$, $P = 50.7\%$, deduced from the TMR ratio at 4.2 K was corresponding well to the experimental data measured at 0.2 K in a spin polarized tunneling experiment using a superconductor/insulator/ferromagnet tunneling junction. © *2000 American Institute of Physics.* [S0003-6951(00)05028-2]

Since the spin tunneling magnetoresistive effect in ferromagnet/insulator/ferromagnet (FM/I/FM) junctions was discovered in 1975,[1] much attention has been paid to the tunneling magnetoresistance (TMR) studies.[2–6] These earlier research works have made the tunneling magnetoresistive effect more interesting and stimulated further experimental and theoretical investigations of spin polarized tunneling properties in FM/I/FM junctions.[7–11] The TMR junction devices and TMR random access memories have been designed and fabricated in the laboratory using the optical and electron beam lithograph methods on micron and submicron sizes.[12,13] Many studies on TMR effect and the tunnel junction in recent years have opened a potential application field in developing high density magnetic storage and magnetic sensors, etc.

Fabricating high TMR junction with low resistance is very important, not only because it is essential for magnetic read head and memory device applications, but also because it can help us to understand well the intrinsic properties and physical mechanism of TMR phenomenon. In this letter, magnetoresistance, effective barrier height and width of the tunnel junction using $Co_{75}Fe_{25}$ as the ferromagnetic electrodes, and Cu as bottom conduction electrode were investigated before and after annealing. Temperature dependence of the TMR ratio, resistance, and coercivity from 4.2 K to room temperature (RT) and applied voltage dependence of the TMR ratio and resistance at RT for the tunnel junction were measured. A high TMR ratio of 49.7% at RT and 69.1% at 4.2 K was achieved.

The multilayer, Ta (5 nm)/$Ni_{79}Fe_{21}$ (3 nm)/Cu (20 nm)/$Ni_{79}Fe_{21}$ (3 nm)/$Ir_{22}Mn_{78}$ (10 nm)/$Co_{75}Fe_{25}$ (4 nm)/Al (0.8 nm)-oxide/$Co_{75}Fe_{25}$ (4 nm)/$Ni_{79}Fe_{21}$ (20 nm)/Ta (5 nm), was first prepared using radio frequency (rf) magnetron sputtering on Si(100)/$SiO_2$ substrate. All the deposition processes were done at a base pressure of about $3\times10^{-6}$ Pa without breaking vacuum in any process. The Al–O insulating layer was formed by inductively coupled plasma oxidation with an oxidation time of 40 s in a mixture of oxygen and argon at a pressure of 1.0 Pa. Tunnel junctions with small active area from 100×100 down to 3×3 $\mu m^2$ were fabricated using a microfabrication technique, i.e., lithographic technique combined with Ar ion-beam etching and $CF_4$ reactive etching. The resistance decreases with increasing junction area from 3×3 to 100×100 $\mu m^2$. Although TMR ratio, in principle, is independent from junction area on micron and submicron sizes, high TMR ratio can be usually abtained in small size junctions due to the reduction of defects in the Al–O barrier. The optimum annealing temperature and time for the highest TMR obtained were inferred to be around 300 °C for an hour, beyond which the TMR ratio of the junctions began to decrease quickly with increasing annealing temperature, which is due to diffusion of the metal atoms into the Al–O barrier and degeneration of the interfaces between FM/I/FM layers. The barrier heights $\phi_1$ and $\phi_2$ and barrier width $d$ were obtained by fitting the current $I$ vs dc bias voltage $V$ curves to Simmons' equation with an asymmetric potential barrier in the insulating layer between the top and bottom magnetic electrodes.[14,15]

Figure 1 shows the TMR curves measured at RT for the tunnel junction at the as-deposited state and after annealing. The junction area is 8×8 $\mu m^2$. The value of the TMR ratio increases more than two times from 22.5% (as-deposited state) to 49.7% after annealing due to the improvements in the junction properties of the interface between FM/I/FM layers and barrier homogenization and due to the reduction in the defect density of the $Al_2O_3$ barrier upon annealing. The barrier heights $\phi_1$ and $\phi_2$, barrier width $d$, and resistance-area product were 1.38 and 1.56 eV, 0.94 nm, and 2947 $\Omega\ \mu m^2$, respectively, at the as-deposited state, and 2.28 and 2.33 eV, 0.78 nm, and 4011 $\Omega\ \mu m^2$, respectively, after annealing. It can be seen that the average barrier height $\phi$ increases about 57% and the barrier shape becomes near to a rectangular potential barrier in the insulating layer after annealing. The effective barrier width $d$ decreased from 0.94

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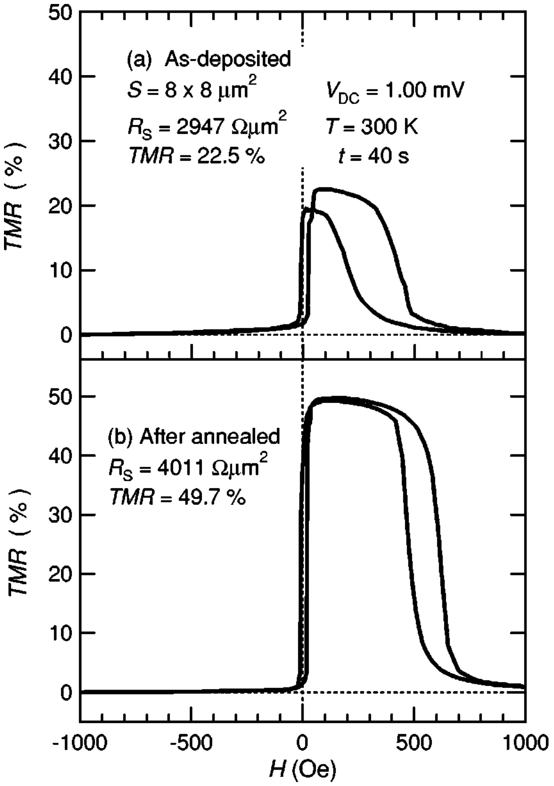

FIG. 1. TMR curves measured at RT for the tunnel junction Ta (5 nm)/$Ni_{79}Fe_{21}$ (3 nm)/Cu (20 nm)/$Ni_{79}Fe_{21}$ (3 nm)/$Ir_{22}Mn_{78}$ (10 nm)/$Co_{75}Fe_{25}$ (4 nm)/Al (0.8 nm)-oxide/$Co_{75}Fe_{25}$ (4 nm)/$Ni_{79}Fe_{21}$ (20 nm)/Ta (5 nm) at the as-deposited state (a) and after annealing (b).

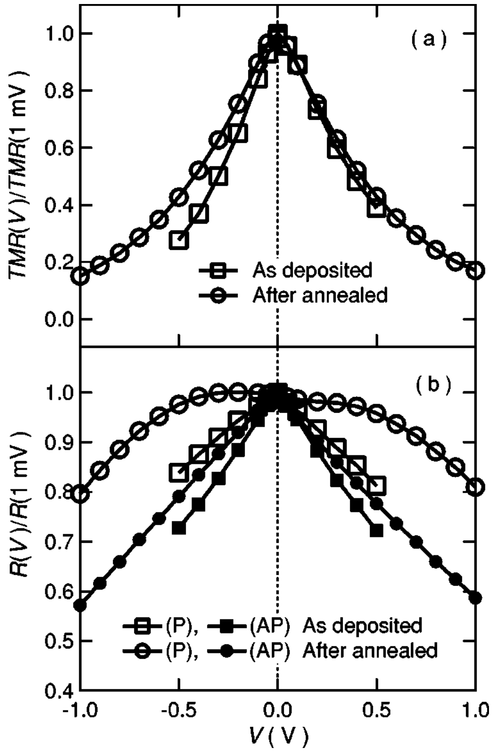

FIG. 2. Normalized TMR ratio and resistance *R* vs dc bias voltage curves measured at RT for the same junction with the size of $8\times8\ \mu m^2$ after anealing as that showing in Fig. 1(b) and for another same stacking junction with the size of $5\times5\ \mu m^2$ at the as-deposited state as a reference.

nm near to the deposited Al thickness of 0.80 nm after annealing, which suggested that the diffusion of oxygen and metal atoms Al, Co, and Fe between the FM/I interfaces originated from the deposited and plasma-oxidation procedures was reduced and sharpening of the FM/I interfaces was achieved upon annealing.

Figure 2 displays the normalized TMR ratio and resistance *R* versus dc bias voltage curves, which are slightly asymmetric with respect to zero dc bias voltage, measured at RT for the same junction with the size of $8\times8\ \mu m^2$ after anealing as that showing in Fig. 1(b) and for another same stacking junction with the size of $5\times5\ \mu m^2$ at the as-deposited state as a reference. A smaller size junction with $5\times5\ \mu m^2$ was selected to measure the normalized TMR ratio versus dc bias voltage curves before annealing is due to its relative large resistance of 100 Ω compared to that of 46 Ω for the junction with a size of $8\times8\ \mu m^2$. The breakdown voltage of the junction with a small size and a large resistance is higher than that with a relative large size and a small resistance. After annealing the resistance stability and breakdown voltage of the junctions increase due to barrier homogenization and reduction of the defect density of the Al–O barrier. We note that the drop of the resistance for the antiparallel (AP) alignment of the magnetization of the two electrodes is larger than that for the parallel (P) alignment with increasing applied dc bias voltage. The TMR ratio decreases with increasing dc bias voltage. The half-peak widths in the normalized TMR ratio versus dc bias voltage curves before and after annealing were about 390 and 430 mV, respectively, when a positive dc bias voltage was applied and they were about 300 and 400 mV, respectively, when a negative dc bias voltage was applied.

As an example, Fig. 3 shows the TMR curves measured at 4.2 and 77 K for the same junction after annealing as that

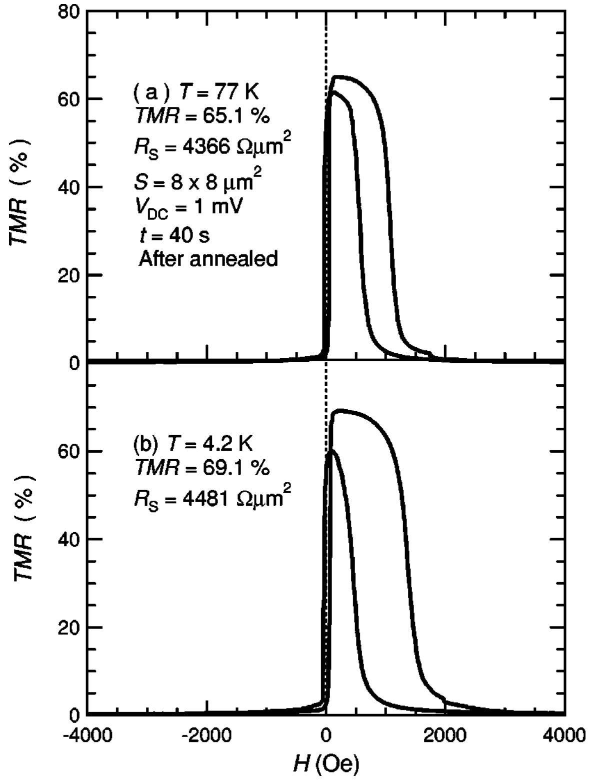

FIG. 3. TMR curves measured at 77 (a) and 4.2 (b) K for the same junction after annealing as that shown in Fig. 1(b).

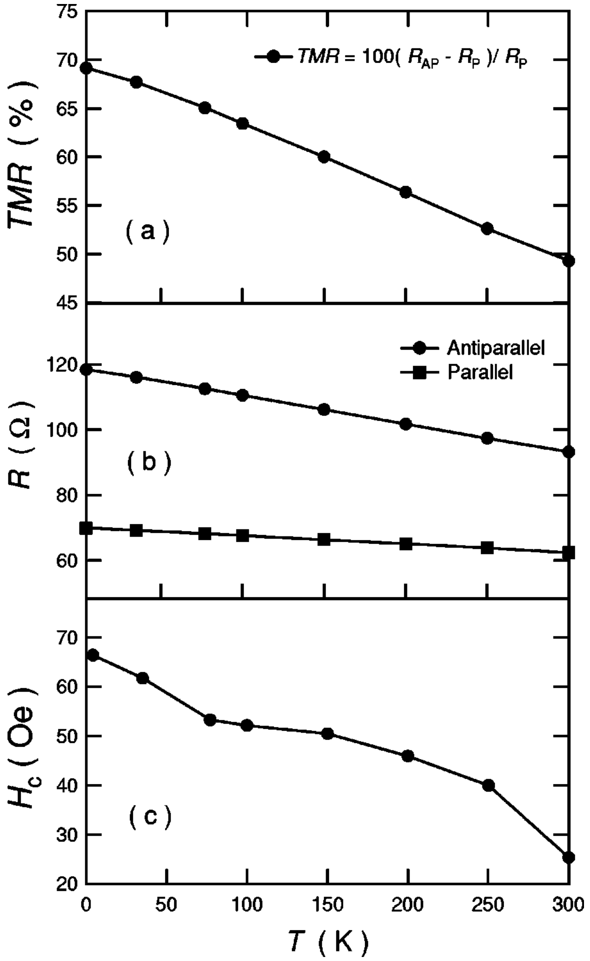

FIG. 4. Temperature dependence of the TMR ratio (a), resistances (b), and coercivity (c) from 4.2 K to RT for the same junction after annealing as that shown in Fig. 1(b).

shown in Fig. 1(b). A high TMR ratio of 69.1% was observed at 4.2 K, which increased 19.4% compared with that of 49.7% at RT due to the decrease of magnon excitations and the increase of spin-polarization of the magnetic electrodes at 4.2 K. It can be supposed that the spin polarization is identical, i.e., $P=P_1\simeq P_2$, for top and bottom $Co_{75}Fe_{25}$ (4 nm) magnetic electrodes based on the facts that an approximate rectangular potential barrier is formed in the insulating layer and the improvements of barrier homogenization and both FM/I interfaces are achieved after annealing. The value of spin polarization of $Co_{75}Fe_{25}$ at 4.2 K, $P=50.7\%$, can be deduced from the Jullière's formula, i.e., $\mathrm{TMR}=69.15\% =2P_1P_2/(1-P_1P_2)\simeq 2P^2/(1-P^2)$. It corresponds well to the experimental data of 50%–52% for $Co_{50}Fe_{50}$, $Co_{60}Fe_{40}$, and $Co_{84}Fe_{16}$, measured at 0.2 K by Parkin[16] with a spin polarized tunneling experiment using a S/I/FM tunneling junction, wherein Al is used as superconductor (S). This experimental value of TMR ratio at 4.2 K is very close to the expected value of the junction using $Co_{75}Fe_{25}$ ferromagnetic electrodes, which means that defects in the Al–O barrier and at the interfaces between FM/I/FM layers are very few after annealing and good quality junctions, as theoretically expected, can be achieved by the optimum fabricating processes and heat treatment. This experimental result also satisfied the Jullière's model.

Figure 4 shows the temperature dependence of the TMR ratio, resistances, and coercivity from 4.2 K to RT for the same junction after annealing as that shown in Fig. 1(b). It can be seen that the TMR ratio, the resistances which are corresponding to the P and AP magnetization configurations of the $Co_{75}Fe_{25}$ electrodes, and the coercivity of the free layer which is corresponding to the critical field and at which the step change of TMR ratio occurs when the magnetic field increases from −100 to 100 Oe, decrease with increasing temperature from 4.2 K to RT. The temperature dependence of the TMR ratio and resistances can be calculated[17] by fitting two parameters of the spin polarization of the density of the state for $Co_{75}Fe_{25}$ $(\rho^M/\rho^m)$ and the matrix element ratio $(T^d/T^J)$ using the theory and formulas developed by Zhang *et al.*[8]

In conclusions, TMR ratio, effective barrier height and width, and breakdown voltage of the junction can be remarkably enhanced after annealed at 300 °C for an hour using $Co_{75}Fe_{25}$ as ferromagnetic electrodes and Cu as bottom conduction electrodes. TMR ratio and resistance decrease with increasing dc bias voltage from 0 to 1000 mV or with increasing temperature from 4.2 K to RT. High TMR ratio of 69.1% at 4.2 K, which corresponds to the high tunneling electron spin polarization of $Co_{75}Fe_{25}$, 50.7%, was observed. This experimental value of TMR ratio at 4.2 K is very close to the expected value of the junction using $Co_{75}Fe_{25}$ ferromagnetic electrodes, and this is the first result observed of such a high TMR ratio at 4.2 K.

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