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Abstract

Using the contact-shadow-mask method, magnetic tunnel junctions (MTJs) with the layer structures of Ta (5 nm)/Cu(25 nm)/Ni$_{79}$Fe$_{21}$(5 nm)/Ir$_{22}$Mn$_{78}$(10 nm)/Co$_{75}$Fe$_{25}$(4 nm)/Al(1.0 nm)-oxide/Co$_{75}$Fe$_{25}$(4 nm)/Ni$_{79}$Fe$_{21}$(20 nm)/Ta(5 nm) were fabricated. High magnetoresistance ratio of 41% and low resistance-area product $R_S$ of $1.9 \times 10^4 \ \Omega \mu m^2$ at as-deposited state were achieved at room temperature (RT). Coercivity of the free layer was 6.2 Oe and magnetic field sensitivity reached at 20.6%/Oe when an external magnetic field increases from -4.6 to 6.2 Oe. Additionally, using lithography method, MTJs with the layer structures of 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 fabricated. High magnetoresistance ratio of 22% and 50% and low resistance-area product $R_S$ of $3.0 \times 10^3$ and $4.4 \times 10^3 \ \Omega \mu m^2$ at as-deposited state and after annealing were attained at RT, respectively. After annealing, coercivity of the free layer was 25.7 Oe and magnetic field sensitivity of the free layer arrived at 9.7%/Oe when an external magnetic field increases from 21.8 to 25.7 Oe. Such MTJs can be used to fabricate the cell of MRAM and other magnetic field sensors.
after further optimizing. Concise explicit function expression for the bias voltage or/and temperature dependences of TMR ratio and resistances were shown with an unique set of intrinsic parameters for an MTJ.

**Key words:** Magnetic tunnel junction, TMR, MRAM, Magnetic field sensor, Magnon excitation.

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1. INTRODUCTION

Tunnel magnetoresistance (TMR) effect in magnetic tunnel junction (MTJ) with structure of ferromagnet/insulator/ferromagnet (FM/I/FM) is a very interesting and useful research topic in both fundamental and applied physics. Investigating the TMR effect is very important for developing the spin-electronics. In the contrary, fabricating the MTJs with high magnetoresistance and low resistance processes a very high application potential in magnetoresistive random access memory (MRAM), magnetic read head technology, and other highly sensitive magnetic field sensors. As we know that, in general case, fabricating MTJs with TMR ≥ 30% and resistance-area product $R_S \sim 10^3 \Omega \mu m^2$ or with TMR ≥ 20% and resistance-area product $R_S \sim 10 \Omega \mu m^2$ at room temperature (RT) are necessary for designing the MRAM with high integration of cell defined by one MTJ and one transistor or the magnetic read head with high magnetic recording density of 100 Gbit/inch$^2$ or more high, respectively. Therefore, how to fabricate the MTJs with high magnetoresistance and low resistance is a key technique for the MTJ application.

In addition, the investigation of intrinsic magnetoelectric properties of the MTJs with high magnetoresistance and low resistance can help us to understand accurately the TMR effect well for developing spin-electron polarization tunneling theory.

In this work, the MTJs with high magnetoresistance and low resistance were fabricated with the layer structures of Ta (5 nm)/Cu(25 nm)/Ni$_{79}$Fe$_{21}$(5 nm)/Ir$_{22}$Mn$_{78}$(10 nm)/Co$_{75}$Fe$_{25}$(4 nm)/Al(1.0 nm)-oxide/Co$_{75}$Fe$_{25}$(4 nm)/Ni$_{79}$Fe$_{21}$(20 nm)/Ta(5 nm) by using the contact-shadow-mask method and 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) by using lithography method, separately. A series of experimental data was measured for these MTJs. The high magnetoresistance ratio of 41% and low resistance-area product $R_S$ of $1.9 \times 10^4 \Omega \mu m^2$ as the deposited state were achieved at RT for the former. The high magnetoresistance ratio TMR of 22% and 50% and low resistance-area product $R_S$ of $3.0 \times 10^3$ and $4.4 \times 10^3 \Omega \mu m^2$ at the as-deposited state and after annealing, respectively,
were obtained at RT for the later. TMR ratio, effective barrier height and width of the MTJs can be remarkably enhanced after annealing at 300 °C for an hour.

II. METHOD OF ANALYSIS

Recently, a spin-electron polarization tunneling model developed by Zhang et al.\textsuperscript{15}, based on magnon excitations by the tunneling electrons during the tunnel process, was extended by defining an anisotropic-wavelength-cutoff energy $E_{\gamma}^{c}$ of spin-wave spectrum in MTJs.\textsuperscript{19} The difference in $E_{\gamma}^{c}$ can be interpreted as the difference in the energy gap between the ground and first excited energy levels of the s-d electron system in the MTJs for antiparallel (AP) and parallel (P) magnetic configurations. Such difference in the energy gap is mainly contributed from the s-d exchange interaction and the Zeeman interaction in an external magnetic field or/and a demagnetization field. Here, concise explicit function expression for the bias voltage or/and temperature dependences of TMR ratio and resistances are shown with an unique set of intrinsic parameters.

Let us consider a simple case for two identical FM electrodes in MTJs. The conductance $G = \frac{I}{V}$ is denoted as $G^{\gamma}(0, 0)$ at 0 K and zero voltage or $G^{\gamma}(V, T)$ at finite bias and temperature, where $\gamma = (P, AP)$ represents the parallel (P) and antiparallel (AP) alignment of the magnetization of the two FM electrodes. The normalized conductance or resistance can be written simply as follows:

$$\frac{G^{\gamma}(V, T)}{G^{\gamma}(0, 0)} = 1 + QC^{\gamma} \nu(V) + QC^{\gamma} \frac{2Sk_{B}T}{E_{m}} \tau(T),$$

where

$$\nu(V) = \begin{cases} 
S \frac{eV}{E_{m}}, & \text{for } eV < E_{m}, \\
S(2 - \frac{E_{m}}{eV}), & \text{for } eV > E_{m}.
\end{cases}$$

$$\tau(T) = -\ln[1 - e^{-\frac{E_{\gamma}^{c}k_{B}T}{k_{B}T}}]$$

$$= \ln\left(\frac{k_{B}T}{E_{\gamma}^{c}}\right), \text{ for } k_{B}T > E_{\gamma}^{c}.$$
\[ Q = \frac{1}{|T'_d|^2/|T'_j|^2 + 25^2}, \]  

(4)

\[ C^\gamma = \begin{cases} 
\xi, & \text{when } \gamma = P, \\
1/\xi, & \text{when } \gamma = AP, \text{ and} 
\end{cases} \]  

(5)

\[ \xi = \frac{2\rho_M\rho_m}{\rho_M^2 + \rho_m^2} = \frac{2}{\rho_M/\rho_m + \rho_m/\rho_M}. \]  

(6)

Thus, the bias voltage and temperature dependence of the resistances can be easily deduced from Eq.(1) by reciprocal transformation between the conductance and resistance.

\[ \frac{R^\gamma(V,T)}{R^\gamma(0,0)} = \frac{1}{1 + QSC^\gamma[ev/E_m + (2k_BT/E_m)ln(k_BT/E_c^\gamma)]}, \quad \text{for } eV < E_m \text{ and } k_BT > E_c^\gamma, \]  

(7)

\[ \frac{R^\gamma(V,T)}{R^\gamma(0,0)} = \frac{1}{1 + QSC^\gamma[(2 - E_m/eV) + (2k_BT/E_m)ln(k_BT/E_c^\gamma)]}, \quad \text{for } eV > E_m \text{ and } k_BT > E_c^\gamma, \]  

(8)

\[ \frac{R^{AP}(0,0)}{R^P(0,0)} = \frac{\rho_M^2 + \rho_m^2}{2\rho_M\rho_m} = \frac{1}{\xi}. \]  

(9)

When \( eV << E_m \) and \( k_BT > E_c^\gamma \), the bias voltage and temperature dependence of TMR ratio can be deduced as follows:

\[ TMR(V,T) = \frac{R^{AP}(V,T) - R^P(V,T)}{R^P(V,T)} = \left( \xi \right) \left[ \frac{1}{\xi} \left[ 1 + QS\xi[ev/E_m + (2k_BT/E_m)ln(k_BT/E_c^P)] \right] \right] - 1, \]  

(10)

\[ TMR(0,0) = \frac{R^{AP}(0,0) - R^P(0,0)}{R^P(0,0)} = \frac{1}{\xi} - 1 = 2P^2/(1 - P^2). \]  

(11)

In which, \( TMR(0,0) \) is the TMR ratio of the MTJs at 0 K and zero bias. It can be seen from Eqs.(4), (5) and (6) that the \( TMR(0,0) \) can be only determined by the density of
states (DOS) of the itinerant electrons (i.e. spin-polarization $P$) of the two FM electrodes, although the magnetoresistances for the P and AP configurations at 0 K and zero bias are closely related with the direct and spin-dependent matrix-elements (i.e. $|T^d|$ and $|T^J|$).\textsuperscript{15,19} Therefore, the Julli`ere’s formula is only valid at 0 K and zero bias,\textsuperscript{1} and in such extreme cases the inelastic magnon excitation is quenched and the inelastic phonon excitation is absent.\textsuperscript{20}

When $eV > E_m$ and $k_B T > E_c^\gamma$, the similar explicit function expression for the bias voltage and temperature dependence of TMR ratio also can be deduced as follows:

$$TMR(V,T) = \frac{R^{AP}(V,T) - R^P(V,T)}{R^P(V,T)} = \frac{1}{\xi} \left[ \frac{1}{1 + QS\xi[(2 - E_m/eV) + (2k_BT/E_m)\ln(k_BT/E_c^P)]} \right] - 1. \quad (12)$$

Hence, the main intrinsic magnetoelectric properties of one MTJ can be self-consistently evaluated by using these explicit function formulations with an unique set of intrinsic parameters. First parameter is the Curie temperature $T_C$ of the ferromagnetic electrode, which can determine the maximum energy of the magnon excitation by $E_M = 3k_BT_C/(2S + 1)$. Second parameter is the density of state (DOS) for itinerant majority and minority electrons $\xi(\rho_M/\rho_m)$ of ferromagnetic electrodes at the interfaces between FM/I/FM layers, which is determined by the value of effective spin-polarization of two FM electrodes. These two parameters are the eigen properties of ferromagnetic electrode material. The other parameters are the spin-dependent matrix-element ratio (i.e. $|T^d|^2/|T^J|^2$), which is determined by resolving the barrier Hamiltonian,\textsuperscript{15} and the anisotropic-wavelength-cutoff energy $E_c^{AP}$ and $E_c^P$ of spin-wave spectrum in the MTJ. They are the structure parameters of the MTJ.

As a result, these parameters mentioned above are only determined by the ferromagnetic electrode materials and the MTJ barrier structure. Each parameter of them is intrinsic and unique in the identical case for one MTJ. This means that the main magnetoelectric properties of an MTJ can be self-consistently evaluated and explained using such concise explicit function formulations and an unique set of intrinsic parameters.\textsuperscript{19}
III. EXPERIMENTAL METHOD

For fabricating the MTJs with the contact-shadow-mask method, the deposition processes were done at a base pressure of about $3 \times 10^{-6}$ Pa with breaking vacuum twice. The bottom electrode of Ta(5 nm)/Cu(25 nm)/Ni$_{79}$Fe$_{21}$(5 nm)/Ir$_{22}$Mn$_{78}$(10 nm)/Co$_{75}$Fe$_{25}$(4 nm) was deposited through the first contact shadow mask to form two sets of long, narrow strips onto a Si(100)/SiO$_2$ substrate with the size of $25 \times 25 \text{mm}^2$ by rf magnetron sputtering. Each set consists of 7 strips (the active width of junction section is 100 $\mu$m). Then, the samples were taken out from the vacuum main chamber to change the second contact shadow mask, thereafter put samples into the vacuum Al-deposition and oxidation chamber for depositing the square Al layer with the thickness of 1.0 nm and length of 800 $\mu$m. The Al-O insulating layer, i.e. Al(1.0 nm)-oxide, was formed by inductively coupled plasma (ICP) oxidation with an oxidation time of 60 s in a mixture of oxygen and argon at a pressure of 1.0 Pa. Finally, the samples were taken out from the Al-deposition and oxidation chamber to change the third contact shadow mask, once again put samples into the vacuum main chamber for depositing the top electrode of Co$_{75}$Fe$_{25}$(4 nm)/Ni$_{79}$Fe$_{21}$(20 nm)/Ta(5 nm) to form a cross strip with the tunnel section of $100 \mu m \times 100 \mu m$. Each sample consists of 14 junctions.

To fabricate the MTJs with the lithography method, all the deposition processes were done at a base pressure of about $3 \times 10^{-6}$ Pa without breaking vacuum in any process. 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 by using rf magnetron sputtering on Si(100)/SiO$_2$ substrate. The Al-O insulating layer is formed by ICP oxidation with an oxidation time of 40 s in a mixture of oxygen and argon at a pressure of 1.0 Pa. The optical lithography combined with Ar ion-beam etching was used to pattern the junction area. The active area of the junctions was $8 \times 8 \mu m^2$. The etching depth was down into a few nanometer below the insulating layer. Then, the junctions were coated with 250 nm SiO$_2$ by rf magnetron sputtering. The contact holes were opened using lithographic technique combined with Ar ion-beam etching and CF$_4$ reactive
etching. Finally, a top conducting layer of 300 nm Cu was deposited and patterned using metal mask to make a connection with the top magnetic electrode. Each sample consists of 9 sets and each set contains 9 junctions. 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. Both the TMR curves with a dc bias voltage of 1 mV and the normalized TMR ratio including junction resistance $R$ versus dc bias voltage curves at RT for the tunnel junction were measured using the dc four-probe method in a magnetic field up to 1000 Oe.

IV. RESULTS AND DISCUSSION

As an example, figure 1 shows the TMR curves measured at RT for the MTJ, fabricated with the contact-shadow-mask method, at the as-deposited state. The junction area $S$ is $100 \times 100 \, \mu m^2$. The experimental curve was measured by a dc four-probe method with a dc bias of 1.0 mV. A high TMR ratio of 41% and low resistance-area product $R_S$ of $1.9 \times 10^4$ $\Omega \mu m^2$ at the as-deposited state were observed at RT.

Figure 2 shows the TMR curves versus the low magnetic field measured at RT for the same MTJ as that shown in Fig.1. 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 -60 to 60 Oe, is 6.2 Oe. The TMR ratio jumps from 0.5% to 33.5% with one step when the magnetic field increases from -4.6 to 6.2 Oe, which shows that the magnetic field sensitivity of the junction reached at 20.6%/Oe. Such MTJs can be used to fabricating the magnetic field sensors after further optimizing the size and resistance.

Figure 3 shows the TMR curves measured at RT for the MTJ, fabricated with the lithography method, after annealing at 300°C for an hour. The junction area $S$ is $8 \times 8 \, \mu m^2$. The TMR ratio increased more than two times from 22% at the as-deposited state to
50% after annealing at RT. It is attributed to the improvements in the junction properties of the interface between FM/I/FM layers and barrier homogenization and as the effect of the reduction in the defect density of the Al-oxide barrier upon annealing. The resistance-area product $R_S$ increases from 3022 Ω$\mu$m$^2$ at the as-deposited state to 4364 Ω$\mu$m$^2$ after annealing.

Figure 4 shows the TMR curves versus the low magnetic field measured at RT for the same MTJ as that shown in Fig.3. The coercivity of the free layer is 25.7 Oe. The TMR ratio jumps from 10.0% to 47.8% with one step when the magnetic field increases from 21.8 to 25.7 Oe, which shows that the magnetic field sensitivity of the junction reached at 9.7%/Oe. Such MTJs can be used to fabricate the cell of MRAM after further decreasing the coercivity.

Figure 5 shows the tunnel current $I$ as functions of the dc bias voltage at RT for the same MTJ as that shown in Fig.3. The barrier heights $\phi_1$ and $\phi_2$ and barrier width $d$ were obtained by fitting the current $I$ versus dc bias voltage $V$ curves to Simmons' equation with an asymmetric potential barrier in the insulating layer between the top and bottom magnetic electrodes$^{25,26}$. The effective barrier heights $\phi_1$ and $\phi_2$, barrier width $d$, and resistance-area product $R_S$ of the annealed MTJ were 2.33 and 2.53 eV, 0.78 nm, and 4364 Ω$\mu$m$^2$, respectively, at RT. The effective barrier width $d$ is near to the deposited Al thickness of 0.80 nm, which suggests 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 is reduced and sharpening of the FM/I interfaces is achieved upon annealing.

Figure 6 demonstrates the dc bias voltage dependence of $TMR(V=1.0\text{ mV})$ ratio from 0 to ±1000 mV for the same MTJ as that shown in Fig.3. 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. It is because the decrease of the resistance is in inverse proportion to $1/\xi$ for AP and to $\xi$ for P alignment while $1/\xi$ is always larger than $\xi$ as that shown in Eqs. (6), (7), and (8).
V. CONCLUSIONS

Magnetic tunnel junctions (MTJs) with the high magnetoresistance and low resistance were fabricated by using the contact-shadow-mask method and lithography method. The high magnetoresistance ratio between 40% and 50% and the low resistance-area product $R_S$ between $10^3$ and $10^4 \ \Omega \mu m^2$ were obtained at RT. The coercivity of the free layer was between 6.2 and 25.7 Oe and magnetic field sensitivity reached between 9.7%/Oe and 20.6%/Oe. Such MTJs can be used to fabricate the cell of MRAM and the magnetic field sensors after further optimizing.

Concise explicit function expression for the bias voltage or/and temperature dependences of TMR ratio and resistances were shown with an unique set of intrinsic parameters. These intrinsic parameters can be pre-determined using the experimental measurement or the first-principle calculation method for an MTJ. Therefore, the main magnetoelectric properties of an MTJ are solely dependent on the ferromagnetic electrode material and structure parameters.

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FIGURES

FIG. 1. TMR curves measured at RT for the MTJ, fabricated with the contact-shadow-mask method, at the as-deposited state.

FIG. 2. TMR curves versus the low magnetic field measured at RT for the same MTJ as that shown in Fig.1.

FIG. 3. TMR curves measured at RT for the MTJ, fabricated with the lithography method, after annealing at 300°C for an hour.

FIG. 4. TMR curves versus the low magnetic field measured at RT for the same MTJ as that shown in Fig.3.

FIG. 5. Tunnel current $I$ as functions of the dc bias voltage at RT for the same MTJ as that shown in Fig.3.

FIG. 6. dc bias voltage dependence of $TMR(V=1.0\text{ mV})$ ratio ranges from 0 to ±1000 mV at RT for the same MTJ as that shown in Fig.3.
$TMR = 41.5\%$
$R_p = 1.87\ \Omega$
$S = 100 \times 100\ \mu m^2$
$V_{DC} = 1\ mV$
As deposited state
Room temperature

Figure 1
Figure 2

- $TMR = 41.5\%$
- $R_p = 1.87\ \Omega$
- $S = 100 \times 100\ \mu m^2$
- $V_{DC} = 1\ mV$
- As deposited state
- Room temperature
After annealed at 300 °C
$R_S = 4364 \, \Omega \mu m^2$
$TMR = 50 \%$

Before annealing
$R_S = 3022 \, \Omega \mu m^2$
$TMR = 22 \%$

$S = 8 \times 8 \, \mu m^2$
$V_{DC} = 1 \, mV$
$T = 300 \, K$
$t = 40 \, s$

Figure 3
After annealed at $300 \, ^\circ\text{C}$
- $R_S = 4364 \, \Omega \mu\text{m}^2$
- $TMR = 50 \%$

Before annealing
- $R_S = 3022 \, \Omega \mu\text{m}^2$
- $TMR = 22 \%$

$S = 8 \times 8 \, \mu\text{m}^2$
$V_{DC} = 1 \, \text{mV}$
$T = 300 \, \text{K}$
$t = 40 \, \text{s}$

Figure 4
After annealed at 300 °C:
\[ R_S = 4364 \, \Omega \mu \text{m}^2 \]
\[ TMR = 50 \% \]

\[ S = 8 \times 8 \, \mu \text{m}^2 \]
\[ T = 300 \, \text{K} \]
\[ t = 40 \, \text{s} \]
Figure 6