Four-State Magnetoresistance in Epitaxial CoFe-Based Magnetic Tunnel Junctions

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A four-state magnetic random access memory (MRAM) was developed using an epitaxial $Co_{50}Fe_{50}$–MgO–$Co_{50}Fe_{50}$ magnetic tunnel junction (MTJ) with a tunnel magnetoresistance (TMR) ratio of 145% at room temperature (RT). Four remanent magnetization states in the single-crystalline $Co_{50}Fe_{50}$ electrode, due to the cubic anisotropy with easy axes of the $\langle 110 \rangle$ directions, result in four possible angular-dependent TMRs, each separated by more than 20% at RT. Analysis of the asteroid curve for $Co_{50}Fe_{50}$ indicated that the magnetic field along 22.5° from the $\langle 110 \rangle$ directions made it possible to change the magnetization direction of the selected cell without disturbing those of the half-selected cells.

***Index Terms*—CoFe, magnetic tunnel junction (MTJ), magnetoresistance (MR), multistate magnetic random access memory (MRAM).**

I. INTRODUCTION

VERY large scale magnetic random access memory (MRAM) is indispensable for future electronics and information network. As the lateral size of the ferromagnet becomes smaller, the switching-field increases due to the demagnetizing effect [1], and this increases the WRITE signal current. Multilevel signal storing is a promising approach to increasing the storage density without downsizing the ferromagnet, and several multistate MRAMs have been reported [2]–[5]. These devices, however, are not adequate for practical use in terms of their magnetoresistance (MR) ratio or operating temperature. Zheng, *et al.* reported a four-state giant magnetoresistance (GMR) device that uses a NiFe–CoFe–Cu system with an MR ratio of approximately 3.0% [4]. Tanaka, *et al.* reported a four-state magnetic tunnel junction (MTJ) that uses a NiMnSb with a tunnel magnetoresistance (TMR) ratio of approximately 4.5% at 77 K [5]. We recently proposed a four-valued MRAM consisting of two MTJs and a resonant tunnel diode (RTD) stacked upon each other and showed that a high effective MR ratio was possible due to the negative differential resistance characteristics of the RTD [6], [7].

Very high TMR ratios of more than 100% were recently achieved at room temperature (RT) in MTJs with an epitaxial or highly-oriented MgO tunnel barrier [8]–[12]. This enables a practical multistate MRAM. In our current study, we developed a four-state MRAM using a fully epitaxial $Co_{50}Fe_{50}$–MgO–$Co_{50}Fe_{50}$ MTJ with a TMR ratio of 145% at RT. Furthermore, we propose a novel writing method using a combination of orthogonal magnetic fields, which enables us to change the magnetization direction of the selected cell without disturbing those of the half-selected cells.

II. ANGULAR-DEPENDENT MR

The TMR of the MTJ depends on the angle θ between the magnetization vector of the free layer and that of the pinned layer. Based on Julliere's model, the TMR value R can be written as

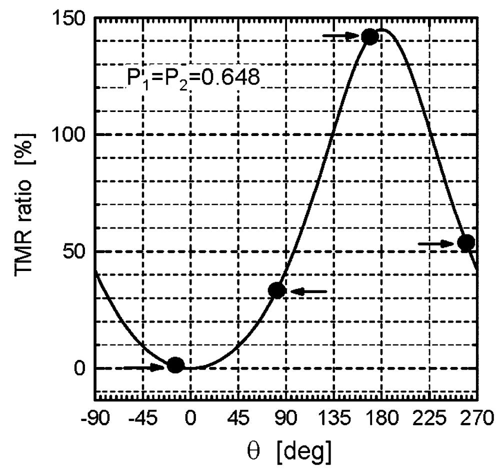

Fig. 1. Calculated TMR ratio as a function of θ, the angle between the magnetization vectors for the free and the pinned layers. The solid circles indicate four possible θ values $-\phi_0, \phi_0+90^\circ, \phi_0+180^\circ$, and $\phi_0+270^\circ-$ for ϕ_0 set to a slightly smaller value than 0°, where ϕ_0 is the angle of the magnetization vector of the pinned layer.

$$R = R_0\left(1+\frac{P_1P_2}{1+P_1P_2\cos\theta}(1-\cos\theta)\right) \tag{1}$$

where P_1 and P_2 are the respective spin polarizations of the free and the pinned layers, and R_0 is the TMR value when both magnetization vectors are parallel. Note that we used Julliere's model as only a first approximation since it does not take into account the spin filtering effect of the MgO barrier in the single-crystalline CoFe–MgO–CoFe MTJs. Since an epitaxial $Co_{50}Fe_{50}$ electrode has four in-plane remanent magnetization ($\boldsymbol{M}_\mathbf{R}$) directions $-[110], [1-10], [-110]$, and $[-1-10]-$, due to the cubic magnetocrystalline anisotropy, θ takes four possible values, i.e., $\phi_0, \phi_0+90^\circ, \phi_0+180^\circ$, and ϕ_0+270°, in the absence of an applied magnetic field, where ϕ_0 is the angle of the magnetization vector of the pinned layer with respect to the $Co_{50}Fe_{50}$ [110] direction. Fig. 1 shows the calculated TMR values as a function of θ. We used $P_1 = P_2 = 0.648$ for the

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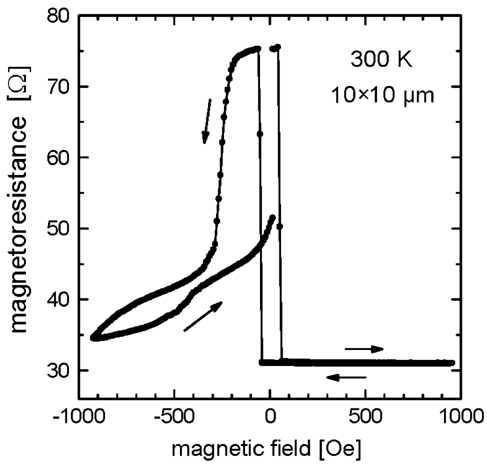

Fig. 2. Major loop of the TMR curve of the fabricated device. The magnetic field was applied along the pinned direction.

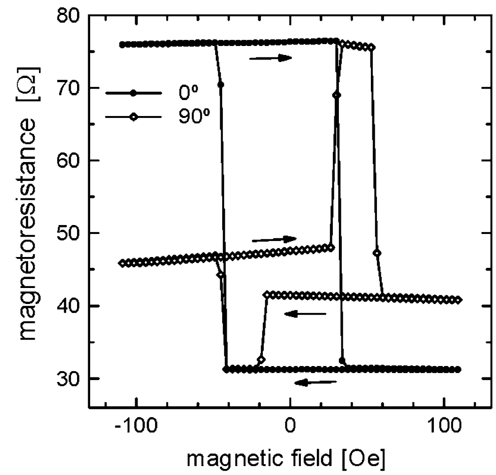

Fig. 3. Minor loop of the TMR curves of the fabricated device. The magnetic field was applied along 0° and 90° with respect to the $Co_{50}Fe_{50}$ [110] direction.

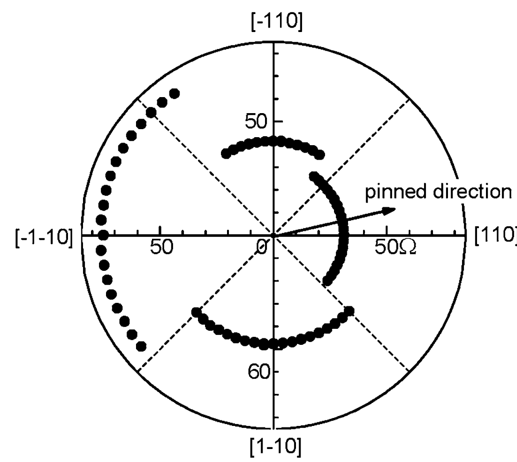

Fig. 4. Polar plot of MR in the absence of a magnetic field. Four distinct MR values corresponding to the magnetization directions for the free layer were obtained.

spin polarizations, as determined from the TMR value obtained experimentally in the fully epitaxial $Co_{50}Fe_{50}$–MgO–$Co_{50}Fe_{50}$ MTJ used in this study. The solid circles in Fig. 1 indicate four possible θ values, $\phi_0, \phi_0 + 90°, \phi_0 + 180°$, and $\phi_0 + 270°$, for ϕ_0 set to be slightly smaller than 0°. These four possible values provide four-state TMR values. If ϕ_0 is set to 0°, three different TMR values are obtained, since two of the four states among $\phi_0 + \alpha(\alpha = 0°, 90°, 180°,$ and $270°)$ provide an equal TMR value.

III. Experiment

MTJ layer structures consisting of (from the top) Ru(5 nm)–IrMn(10)–$Co_{90}Fe_{10}$(2)–Ru(0.8)–$Co_{50}Fe_{50}$ (3)–MgO(1.7)–$Co_{50}Fe_{50}$(50)–MgO(10) were grown on a MgO(001) substrate. Each layer was successively deposited in an ultrahigh vacuum chamber (with a base pressure of about 8×10^{-8} Pa) through the combined use of magnetron sputtering and electron beam evaporation. All layers except for the MgO layers were deposited at RT using magnetron sputtering. The MgO buffer layer and MgO tunnel barrier were deposited by electron beam evaporation at 400 °C and RT, respectively.

The MTJs with 10 × 10-μm size were fabricated using photolithography and Ar ion milling. After fabrication, the MTJs were annealed at 300 °C for 60 min under a magnetic field of 5 kOe in a vacuum of 5×10^{-2} Pa. To obtain four TMR values, the magnetization direction of the upper $Co_{50}Fe_{50}$ layer was pinned through exchange biasing along a direction approximately 10° off from that of the $Co_{50}Fe_{50}$ [110]. A synthetic antiferromagnetic (SAF) trilayer structure consisting of $Co_{90}Fe_{10}$ (2)–Ru(0.8)–$Co_{50}Fe_{50}$ (3) was used to lower the net saturation magnetization in the pinned structure with IrMn, enabling a high exchange bias field H_{ex}. The TMR characteristics of the fabricated devices were measured through a direct current (dc) four-probe method at RT.

IV. Results and Discussions

Fig. 2 shows a major loop of the TMR curve for the fabricated device. The magnetic field $\boldsymbol{H}$ was applied from −950 to 950 Oe along the pinned direction. The maximum TMR ratio, defined by $(R_{\mathrm{AP}} - R_{\mathrm{P}})/R_{\mathrm{P}}$, was approximately 145% at RT, where R_{AP} and R_{P} were the TMR values with θ of 180° and 0°, respectively. Since the switching fields for the pinned layer were approximately 0 and −300 Oe, a relatively large H_{ex} of approximately 150 Oe was obtained because of the SAF structure.

Fig. 3 shows minor loops of the TMR curves for the same device with two different directions. The H ranging from −120 to 120 Oe was applied along [110] and [−110] directions. Note that the magnetization vector of the $Co_{50}Fe_{50}$ pinned layer was always fixed to the same direction at $|\boldsymbol{H}| = 0$ (10° off from [110]) due to a large H_{ex}. The TMR at $|\boldsymbol{H}| = 0$ showed four distinct values, i.e., 31, 41, 49, and 76 Ω, each separated by more than 20%. These values correspond to the states where the $\boldsymbol{M}_{\mathrm{R}}$ lies for the [110, [−110], [1−10], and [−1−10] directions, respectively. We will refer to these four states as "0," "1," "2," and "3."

Fig. 4 shows a polar plot of the TMR at $|\boldsymbol{H}| = 0$. The angle of the polar plot corresponds to the direction of $\boldsymbol{H}$, which was applied before measurement with various directions from 0° to 360° in steps of 5° in the (001) plane to set the direction of $\boldsymbol{M}_{\mathrm{R}}$. Since the pinned direction lay 10° off from the [110] direction, the TMR for $\boldsymbol{M}_{\mathrm{R}}$ with the [−110] direction (the "1" state) was

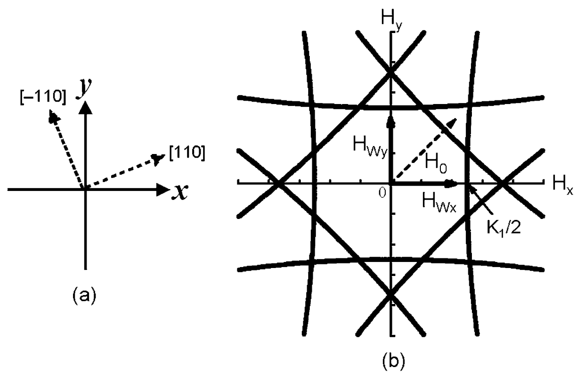

Fig. 5. (a) Definition of the x and y directions and (b) calculated asteroid curve for $Co_{50}Fe_{50}$ with cubic magnetocrystalline anisotropy.

TABLE I
WRITE DATA VERSUS SEQUENCE OF WRITE FILED

write data	sequence of write field
"0"	$H_1 \to H_3 \to H_2 \to H_0$
"1"	$H_3 \to H_2 \to H_0 \to H_1$
"2"	$H_0 \to H_1 \to H_3 \to H_2$
"3"	$H_2 \to H_0 \to H_1 \to H_3$

20% smaller than that for the $\boldsymbol{M}_{\mathbf{R}}$ with the $[1-110]$ direction (the "2" state). The $\boldsymbol{M}_{\mathbf{R}}$ direction can be controlled through the direction of the applied magnetic field.

Next, let us consider a WRITE operation. In MTJ arrays, each level must be written without disturbing other unselected cells. A combination of the orthogonal fields, $\boldsymbol{H}_{\boldsymbol{W}x}$ and $\boldsymbol{H}_{\boldsymbol{W}y}$, is used for selective writing. We defined the x and y directions as $-22.5°$ and $67.5°$ from the $[110]$ direction, as shown in Fig. 5(a). Fig. 5(b) shows the calculated asteroid curve of the free layer with cubic magnetocrystalline anisotropy. Let us consider the case when $\boldsymbol{H}_{\boldsymbol{W}x}$ and $\boldsymbol{H}_{\boldsymbol{W}y}$ with strength between $K_1/2\sqrt{2}$ and $K_1/2$ are applied, where K_1 is a cubic anisotropy constant divided by the saturation magnetization. The combination of $\boldsymbol{H}_{\boldsymbol{W}x}$ and $\boldsymbol{H}_{\boldsymbol{W}y}$ is then applied to the selected cell, while either $\boldsymbol{H}_{\boldsymbol{W}x}$ or $\boldsymbol{H}_{\boldsymbol{W}y}$ is applied to the half-selected cell. Application of either $\boldsymbol{H}_{\boldsymbol{W}x}$ or $\boldsymbol{H}_{\boldsymbol{W}y}$ does not switch any magnetization direction, while the $\boldsymbol{H}_{\boldsymbol{W}x}$ +$\boldsymbol{H}_{\boldsymbol{W}y}$ field, whose strength is between $K_1/2$ and $\sqrt{2}K_1/2$, can switch the magnetization vector with the $[1-10]$ direction (the "2" state) to the $[110]$ direction (the "0" state). Likewise, the $-\boldsymbol{H}_{\boldsymbol{W}x} + \boldsymbol{H}_{\boldsymbol{W}y}$ field can switch only "0" to "1," and so on. Based on this scheme, we propose a novel writing method as follows. Let us define the WRITE fields $\boldsymbol{H}_{\boldsymbol{W}x} + \boldsymbol{H}_{\boldsymbol{W}y}, -\boldsymbol{H}_{\boldsymbol{W}x} + \boldsymbol{H}_{\boldsymbol{W}y}, \boldsymbol{H}_{\boldsymbol{W}x} - \boldsymbol{H}_{\boldsymbol{W}y}$, and $-\boldsymbol{H}_{\boldsymbol{W}x} - \boldsymbol{H}_{\boldsymbol{W}y}$ as $\boldsymbol{H}_0, \boldsymbol{H}_1, \boldsymbol{H}_2$, and $\boldsymbol{H}_3$. To write the "0" state from any other states without disturbing other half-selected cells, the WRITE fields of $\boldsymbol{H}_1, \boldsymbol{H}_3, \boldsymbol{H}_2$, and $\boldsymbol{H}_0$ are applied in order. Table I summarizes the sequence of the applied field in a WRITE operation.

V. CONCLUSION

We have developed a four-state MRAM using an epitaxial CoFe-based MTJ with a TMR ratio of 145% at RT. The four remanent magnetization states in the single-crystalline $Co_{50}Fe_{50}$ electrode, due to the cubic anisotropy with easy axes of the $\langle 110 \rangle$ directions, result in four possible angular-dependent TMRs, each separated by more than 20% at RT. Furthermore, we have proposed a novel writing method, which enables the magnetization direction of the selected cell to be changed without disturbing those of the half-selected cells by applying the magnetic field along directions 22.5° from the $\langle 110 \rangle$ directions.

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