Applied Physics Express **4** (2011) 043005
DOI: 10.1143/APEX.4.043005

Quantitative Evaluation of Voltage-Induced Magnetic Anisotropy Change by Magnetoresistance Measurement

Yoichi Shiota[1,2], Shinichi Murakami[1,2], Frédéric Bonell[1,2], Takayuki Nozaki[1,2], Teruya Shinjo[1], and Yoshishige Suzuki[1,2]

[1] *Graduate School of Engineering Science, Osaka University, Toyonaka, Osaka 560-8531, Japan*
[2] *CREST, Japan Science Technology Agency, Kawaguchi, Saitama 332-0012, Japan*

Received February 5, 2011; accepted March 7, 2011; published online March 25, 2011

We investigated the voltage-induced perpendicular magnetic anisotropy change in an epitaxial magnetic tunnel junction (MTJ) with an ultrathin FeCo layer. Tunneling magnetoresistance (TMR) curves were measured under various bias voltage applications for different FeCo thicknesses. Clear changes in the shape of TMR curves were observed depending on the voltage-controlled perpendicular magnetic anisotropy. By evaluating the relative angle of two ferromagnetic layers, we could estimate the anisotropy energy change quantitatively. The realization of voltage-induced anisotropy change in the MTJ structure makes it possible to control the magnetization dynamics, leading to a new area of electric-field-based spintronics devices.

One of the technological issues in the area of spintronics is to realize magnetization switching with low-power consumption. There are some successful approaches of "electric-current" based one, such as using a current-induced magnetic field or spin-transfer torque,[1,2)] however, these approaches still require a high current density. Therefore, magnetization manipulation using an "electric-field" is expected as a promising and ideal method for future magnetic writing technology. Voltage control of magnetic properties has already been demonstrated in several materials and stacking structures, including multilayered stacks with piezoelectric materials,[3,4)] ferromagnetic semiconductors,[5)] single-phase multiferroic materials[6,7)] or heterostructures consisting of artificial ferroelectric/ferromagnetic layers,[8–11)] transition of the magnetic state at metal surfaces,[12)] and anisotropy change in ultrathin ferromagnetic metal layers.[13–20)] The surface magnetoelectric effect in an ultrathin 3*d* ferromagnetic layer has recently received a lot of interests because it can easily be applied to the conventional MgO-based magnetic tunnel junctions.[21,22)]

We first succeeded to observe the voltage-induced surface magnetic anisotropy change in Au/ultrathin Fe(Co)/MgO/polyimide/indium tin oxide (ITO) junctions.[14,15)] However, because of the hysteretic behavior of the anisotropy change (see the supplementary information of ref. 14), it was difficult to evaluate the intrinsic anisotropy energy change quantitatively. To avoid this problem, a multilayer structure with clean interfaces is required. Furthermore, to realize the voltage-induced dynamic magnetization switching proposed in our previous paper,[14)] we need a sample with a radio frequency (RF) compatible structure. For this purpose, a magnetic tunnel junction (MTJ) structure is a good candidate. Very recently, we have succeeded in detecting the voltage effect in a fully epitaxial MTJ by using a spin-transfer torque induced ferromagnetic resonance.[16)] However, the FeCo thickness was limited in the thinner range of the perpendicularly magnetized film due to the difficulty in measurement technique. In this paper, we report a direct and simple approach to evaluate the voltage-induced magnetic anisotropy change in the epitaxial MTJs from tunneling magnetoresistance (TMR) measurement under various bias voltage applications.

An MTJ structure of MgO (10 nm)/Cr (10 nm)/Au (50 nm)/$Fe_{80}Co_{20}$ (t_{FeCo}: 0.55–0.75 nm)/MgO (1.7 nm)/Fe (10 nm)/Au cap (5 nm) was epitaxially grown on a MgO (001) substrate by molecular beam epitaxy. All the layers were deposited at room temperature (RT), and only the Au buffer layer was annealed at 200 °C for 10 min. after the RT deposition to improve the surface flatness. The film was fabricated into junctions of $0.8 \times 0.2\,\mu m^2$ in size by electron beam lithography, Ar-ion etching technique, and lift-off processes. The sign of the bias voltage is defined with respect to the top Fe electrode. TMR measurements were carried out using a conventional two-terminal technique under magnetic fields applied in-plane or perpendicular to the film plane directions depending on the designed FeCo thickness. All the measurements were performed at RT.

Figure 1 shows the TMR curves of the MTJs with $t_{FeCo} = 0.68$, 0.63, and 0.60 nm measured under the magnetic field (a)–(c) in-plane (hard axis), $H_{in\text{-}plane}$, or (d)–(f) perpendicular to the film plane, H_{perp}, directions. The nominal MgO barrier thickness is 1.70 nm and the tunneling resistance at the parallel magnetization state is about 740 Ω for all the samples, and the TMR ratios are about 15.2% ($t_{FeCo} = 0.68$ nm), 11.1% ($t_{FeCo} = 0.63$ nm), and 9.6% ($t_{FeCo} = 0.60$ nm) from Figs. 1(a)–1(c), respectively. $t_{FeCo} = 0.63$ nm is around the critical thickness of the perpendicular anisotropy for this sample, i.e., the magnetic easy axis is in-plane for $t_{FeCo} > 0.63$ nm, and perpendicular to the film plane for $t_{FeCo} < 0.63$ nm. For the thickness range of $t_{FeCo} > 0.63$ nm, because of the magnetostatic coupling with the top thick Fe layer, the remanent magnetization configuration is anti-parallel. On the other hand, the relative angle is 90° at the remanent state for the thickness range of $t_{FeCo} < 0.63$ nm. In both cases, the tunneling resistance takes a maximum value at zero field. As for the in-plane magnetized case ($t_{FeCo} = 0.68$ nm), a clear change in the saturation property was observed at around $H_{perp} = 1200$ Oe in Fig. 1(d). Since the magnetization direction of the thick Fe layer cannot be tilted so much by this small perpendicular magnetic field, this saturation property should reflect the perpendicular magnetic anisotropy of the ultrathin FeCo layer. The linear reduction in the resistance observed in the higher field range of more than 2 kOe comes from the tilting magnetization of the top thick Fe layer. As for the perpendicular magnetized case ($t_{FeCo} = 0.60$ nm), only the linear reduction in the resistance was observed with increasing magnetic field, as shown in Fig. 1(f), which means that only the magnetization of the top thick Fe was tilted in the perpendicular direction.

Figures 2(a) and 2(b) show the bias voltage dependence of the TMR curves. Since the tunneling resistance and MR ratio strongly depend on the bias voltage, the vertical axis is

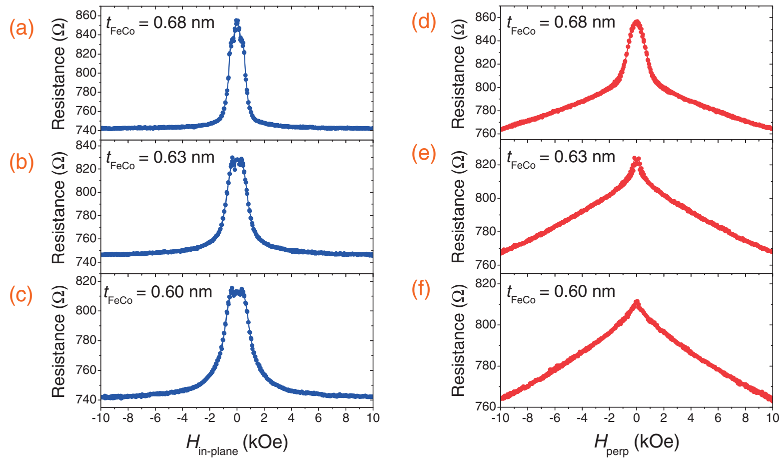

Fig. 1. TMR curves of the MTJs with $t_{FeCo} = 0.68$, 0.63, and 0.60 nm measured under the magnetic field (a)–(c) in-plane (hard axis) directions or (d)–(f) perpendicular to the film plane.

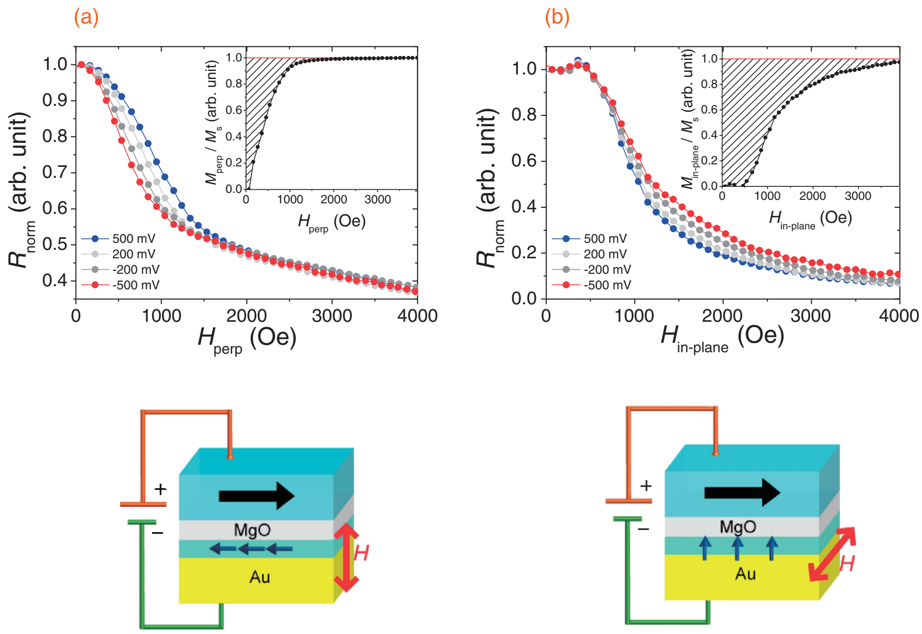

Fig. 2. Bias voltage dependence of the normalized TMR curves of (a) $t_{FeCo} = 0.68$ nm, and (b) $t_{FeCo} = 0.60$ nm. Only the positive magnetic field region is shown here. The perpendicular anisotropy energy, E_{perp}, was estimated from the shaded area shown in the inset. The bottom figures represent the magnetization configuration of two ferromagnetic layers at zero magnetic field, and the direction of the external magnetic field.

normalized by the maximum ($H_{ext} = 0$ Oe) and minimum ($H_{ext} = 18$ kOe) values in Fig. 2(a). The blue (red) curves were measured under a bias voltage of +500 mV (−500 mV). Clear changes in the saturation property were observed in the MTJs with $t_{FeCo} = 0.68$ nm. In a similar way, changes in the saturation property were also observed in the MTJs with $t_{FeCo} = 0.60$ nm as shown in Fig. 2(b). Here the magnetic field was applied in the in-plane direction. The constant resistance state in the small magnetic field range originates from the dipole coupling.

The influence of the current, i.e., current-induced magnetic field or spin-transfer torque, is negligibly small in the investigated structure due to its large junction size and high RA value (maximum current and current density are 0.7 mA and 4.3×10^9 A/m^2, respectively). We can conclude that the observed shift in the saturation field is caused by the change in the perpendicular magnetic anisotropy field. In all cases, when the positive (negative) bias voltage is applied, the perpendicular magnetic anisotropy is suppressed (induced). This sign is consistent with our previous works.[14–16]

We estimate the magnetization of ultrathin FeCo layer of perpendicular component, M_{perp}, and in-plane component, $M_{in\text{-}plane}$, from the relative angle of two ferromagnetic layers obtained in TMR curves by the following equations,

$$\frac{M_{perp}}{M_s} = \cos\left\{\cos^{-1}\left[\frac{2R_{AP}R_P}{R(R_{AP}-R_P)} - \frac{R_{AP}+R_P}{R_{AP}-R_P}\right] + \cos^{-1}\left[\frac{H_{perp}}{H_{s,Fe}}\right]\right\}$$

under perpendicular field,

$$\frac{M_{in\text{-}plane}}{M_s} = \left[\frac{2R_{AP}R_P}{R(R_{AP}-R_P)} - \frac{R_{AP}+R_P}{R_{AP}-R_P}\right]$$

under in-plane field,

where M_s, R_P (R_{AP}), and $H_{s,Fe}$ are saturation magnetization of ultrathin FeCo layer, resistance in parallel (anti-parallel)

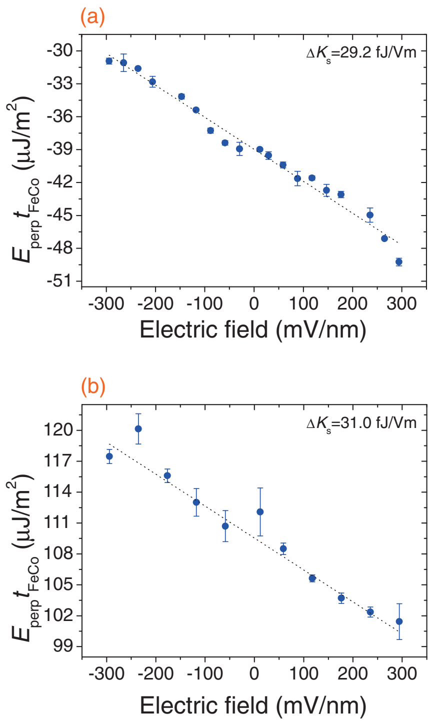

Fig. 3. Applied electric-field dependence of the surface magnetic anisotropy energy, $E_{perp}t_{FeCo}$, calculated from the TMR curves. (a) $t_{FeCo} = 0.68$ nm and (b) $t_{FeCo} = 0.60$ nm. The lines are a guided for the eye.

configuration, and saturation field of top thick Fe layer to the perpendicular direction, respectively. M_s has strong thickness dependence in these ultrathin regions, so here we used the value estimated in the previous experiments using the magneto-optical Kerr effect,[15] for example, the $\mu_0 M_s$ of the 0.68-nm-thick FeCo layer is 1.52 T in our sample. Then we calculate the perpendicular anisotropy energy, E_{perp}, from the shaded areas in the insets of Figs. 2(a) and 2(b).[23] In this calculation, since we can exclude the contribution of cubic anisotropy, only the uniaxial anisotropy change caused by the voltage application can be estimated. Figure 3 presents the electric field dependence of $E_{perp} \times t_{FeCo}$ for (a) $t_{FeCo} = 0.68$ nm and (b) $t_{FeCo} = 0.60$ nm. Linear changes of the anisotropy energy were observed in both cases, and the slope of the surface anisotropy energy change, ΔK_s(V), was evaluated to be 29.2 fJ V^{-1} m^{-1} for $t_{FeCo} = 0.68$ nm and 31.0 fJ V^{-1} m^{-1} for $t_{FeCo} = 0.60$ nm. Comparing the magnetic anisotropy energy at the zero bias voltage, extrapolated from the linear fitting, it increases with decreasing FeCo thickness and becomes positive at $t_{FeCo} = 0.60$ nm, indicating that the magnetic easy axis transits from the in-plane direction to the perpendicular direction. This tendency is almost consistent with our previous results.[15,16]

Here, let us compare the observed magnetic anisotropy energy change with theoretical works. There are several reports on the voltage effect on magnetic anisotropy. Comparing ΔK_s(V) with the results of the free-standing Fe case, the expected value is on the same order as our experimental results. On the other hand, a larger effect has been obtained in the multilayer structure, e.g., 196 fJ V^{-1} m^{-1} in Pt/Fe/Pt/MgO[24] and 100 fJ V^{-1} m^{-1} in Cu/Fe/MgO,[18] since the surface magnetic anisotropy and voltage effect on it should strongly depend on the material of the buffer and dielectric materials. Also, the condition of the interfaces, for example, the oxidation at the ultrathin Fe(Co) layer surface, may have a large influence.[20] Although further quantitative comparison requires first-principle calculation in the realistic structure, we have sufficient possibility to enhance the voltage effect.

In summary, we investigated the voltage effect on perpendicular magnetic anisotropy in an ultrathin FeCo layer using magnetoresistance measurement. In the bias voltage dependence of the TMR curves, a clear change in the perpendicular magnetic anisotropy was observed as a change in the saturation property depending on the bias voltage application. We found that the anisotropy energy changes linearly as a function of the applied electric-field. The slope of the anisotropy energy change was evaluated to be about 29.2 and 31.0 fJ V^{-1} m^{-1} for the FeCo thicknesses of 0.68 and 0.60 nm, respectively. The successful control of magnetic anisotropy in the MTJ structure opens the possibility for voltage-induced manipulation of magnetization switching and its application to novel electric-field based spintronics devices.

Acknowledgment Part of this research was conducted under the financial support of the Global Center of Excellence (G-COE) program of the Ministry of Education, Culture, Sports, Science and Technology, Japan (MEXT).

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