Large Voltage-induced Magnetic Anisotropy Change in a Few Atomic Layers of Iron

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Large voltage-induced magnetic anisotropy change in a few atomic layers of iron

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In the field of spintronics, researchers have manipulated magnetization using spin-polarized currents[1-3]. Another option is to use a voltage-induced symmetry change in a ferromagnetic material to cause changes in magnetization or in magnetic anisotropy[4-14]. However, a significant improvement in efficiency is needed before this approach can be used in memory devices with ultralow power consumption. Here, we show that a relatively small electric field (less than 100 mV nm^{-1}) can cause a large change (~40%) in the magnetic anisotropy of a bcc Fe(001)/MgO(001) junction. The effect is tentatively attributed to the change in the relative occupation of 3*d* orbitals of Fe atoms adjacent to the MgO barrier. Simulations confirm that voltage-controlled magnetization switching in magnetic tunnel junctions is possible using the anisotropy change demonstrated here, which could be of use in the development of low-power logic devices and non-volatile memory cells.

To develop voltage-driven spintronic devices, several areas of investigation have been suggested, including voltage control of magnetic anisotropy[4,7], ferromagnetism in ferromagnetic semiconductors[8,9], magnetoelectric switching of exchange bias[10,11] and anisotropy[12], magnetoelectric interface effects[5,6], multiferroic properties[13] and magnetostriction in a hybrid system with piezoelectric materials[14]. Most work to date has been carried out only at low temperatures, using GaMnAs or perovskite systems, or needed piezoelectric distortions, which may limit the endurance of any devices. Weisheit and colleagues[7] observed up to 4.5% coercivity change in FePt(Pd) films with the application of voltage. However, they required the use of a liquid electrolyte to apply a high electric field at the surface. In this study, we have overcome these difficulties by using ultrathin Fe/MgO junctions. The system is built from all-solid-state and distortion-free materials that have controllable perpendicular surface anisotropy[15-20].

As shown in Fig. 1, the sample structure stack layers comprised a MgO substrate/MgO(10 nm)/Cr(10 nm)/Au(50 nm)/Fe(2–4 ML)/MgO(10 nm)/polyimide(1,500 nm)/ITO(100 nm) (ML, monatomic layer; see Methods). Because the influence of the electric field on the perpendicular anisotropy is effective only at the interface, the ferromagnetic layer had to be composed of only a few monatomic layers. In addition, to control the perpendicular anisotropy, the film had to have a moderate crystalline and surface anisotropy in the absence of the bias voltage. In this regard, we used an ultrathin Fe layer for the anisotropy change. Because the Fe grew almost in a layer-by-layer mode onto the Au(001) surface at room temperature, we were able to precisely control the layer thickness[21]. The ultrathin epitaxial Fe layer deposited on the Au(001) buffer layer exhibited a transition in magnetic anisotropy from in-plane to perpendicular, depending on the film thickness[20]. Such features are ideal for the observation of the anisotropy change in response to the electric field, because we could systematically control the strength of the perpendicular anisotropy. The insulating layers comprised MgO and polyimide. MgO was used because it can be epitaxially grown onto an Fe(001) surface and exhibits a high breakdown voltage as a barrier material in magnetic tunnel junctions[22]. The polyimide layer was used to ensure a pinhole-free barrier over an extended area.

Figure 2 shows representative magnetic hysteresis loops in a 0.48-nm-thick Fe layer under the application of a bias voltage, obtained from Kerr ellipticity, η_K, measurements. Under two different bias voltages, $U = +200$ and -200 V, a significant change in perpendicular anisotropy was observed. The perpendicular magnetic anisotropy energy per unit volume of the film, E_{perp}, was calculated from the $\eta_K - H$ curve, assuming a linear relation between η_K and magnetization:

$$E_{perp} = -\mu_0 \frac{M_s}{\eta_s} \int_0^{\eta_s} H \mathrm{d}\eta_K, \tag{1}$$

where μ_0, M_S, H, η_K and η_s are permeability of free space, saturation magnetization, external magnetic field, Kerr ellipticity and saturation Kerr ellipticity, respectively. If the film possesses uniaxial crystalline anisotropy K_u, and surface anisotropy K_s, the E_{perp} is expressed as

$$E_{perp} d = \left(-\frac{1}{2}\mu_0 M_s^2 + K_u\right) d + K_{S,MgO/Fe} + K_{S,Fe/Au} + \Delta K_S(V) \tag{2}$$

where d is the film thickness. $\Delta K_S(V)$ is a surface anisotropy, induced by application of a voltage. When a positive voltage $U = +200$ V was applied, then decreased to $U = -200$ V, perpendicular anisotropy was induced and the magnetic anisotropy energy was changed from -31.3 to -13.7 kJ m^{-3}. The ratio of the magnetic anisotropy energy change, defined as $\Delta E_{perp}/(2E_{perp,ave}) = (E_{perp,200V} - E_{perp,-200V})/(E_{perp,200V} + E_{perp,-200V})$, was 39%. If we count this change as a change in the surface anisotropy energy, that is, $\Delta K_s(V)$ in equation (2), it corresponds to 8.4 μJ m^{-2}.

To precisely measure the thickness dependence of the effect, we used a modulation technique (see Methods). The inset in Fig. 2 shows the external field dependence of the $\mathrm{d}\eta_K/\mathrm{d}V$ signal that was obtained for the same sample. The $\mathrm{d}\eta_K/\mathrm{d}V$ signal displays a maximum value, $(\mathrm{d}\eta_K/\mathrm{d}V)_{max}$, where the η_K hysteresis curves, obtained for positive (B) and negative (A) bias voltages, show a

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The following is a comment on the published paper shown on the preceding page.

Large Voltage-induced Magnetic Anisotropy Change in a Few Atomic Layers of Iron

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Introduction

From ancient times in the history of mankind, magnetic field produced by a permanent magnet or current has been used to magnetize ferromagnetic materials. Recently, especially in the field of spintronics, researchers have started to manipulate magnetization with spin polarized currents.[1-3] In both cases, however, the current consumes extremely large energy compared to the stabilization energy of single bit information. Since the voltage, as it is proven in the C-MOS technology, doesn't consume energy in principle, a voltage control of the magnetization will be an ideal information writing method. In order to develop voltage-driven spintronic devices, several approaches have been suggested, such as voltage control of magnetostriction in a hybrid system with piezoelectric materials,[4] ferromagnetism in ferromagnetic semiconductors,[5] multiferroic properties[6] and magnetoelectric interface effects.[7,8] Most work has been carried out only at low temperatures, using the GaMnAs or perovskite systems, or needed piezoelectric distortions, which may limit the endurance of the device. As for the magnetoelectric interface effect, Weisheit *et al.*[7] observed up to 4.5% coercivity change in FePt(Pd) films with voltage application. However, they required a liquid electrolyte to apply a high electric field at the surface and it is not suitable for practical applications. In this study, we demonstrate the large voltage-induced magnetic anisotropy change in few atomic bcc-Fe(001) layer / MgO(001) junctions. The effect is tentatively attributed to the change in the relative occupation of 3d-orbitals of Fe atoms adjacent to the MgO barrier.

Experimental procedure

Multilayers of MgO(10 nm) / Cr(10 nm) / Au(50 nm) / Fe(2-4 ML) / MgO(10 nm) were grown on single crystal MgO(001) substrates by molecular beam epitaxy. After depositing the top MgO layer, the sample was removed from the deposition chamber and the surface was coated with a polyimide layer (1500 nm in thickness) by using a spin-coater. An ITO (Indium tin oxide) electrode of 1mm in diameter was fabricated using a metal mask. Since the influence of the electric field on the perpendicular anisotropy is effective only at the interface, the ferromagnetic layer needed to be composed of only several monatomic layers. In addition, to control the perpendicular anisotropy, the film required a moderate crystalline and surface anisotropy in absence of the bias voltage. For this purpose, we employed an ultrathin Fe layer on Au for the anisotropy change, because we could systematically control the strength of perpendicular anisotropy by the Fe layer thickness. The bias voltage was applied between the top ITO and the bottom Au electrodes. The polyimide layer was used to assure a pinhole-free barrier over an extended area. The bias direction was defined with respect to the top ITO electrode (see Fig. 1). To investigate the magnetic hysteresis curve of the Fe layer, Kerr ellipticity, η_K was measured in a polar configuration under the application of voltage.

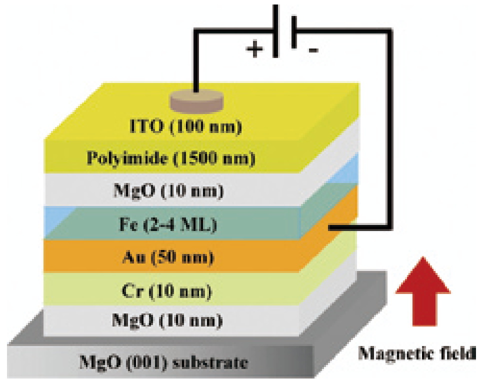

Fig.1 Schematic illustration of the sample. Bias voltage was applied between the top ITO and bottom Au layers.

Voltage effect on the magnetic hysteresis curve

Figure 2(a) shows representative magnetic hysteresis loops in a 0.48nm thick Fe layer under the bias voltage application. Under the two different bias voltages, U = +200 V and -200 V, a significant change in perpendicular anisotropy was observed. The perpendicular magnetic anisotropy energy per unit volume of the film, E_{perp}, was calculated from η_K -H curve, assuming a linear relation between η_K and magnetization.

$$E_{perp} = -\mu_0 \frac{M_s}{\eta_s} \int_0^{\eta_s} H d\eta_K \,, \qquad (1)$$

where μ_0, M_S, H, η_K and η_s are permeability of free space, saturation magnetization, external magnetic field, Kerr ellipticity and saturation Kerr ellipticity, respectively. If the film possesses uniaxial crystalline anisotropy, K_u, and surface anisotropy, K_s, the E_{perp} is expressed as follows.

$$E_{perp} d = \left(-\frac{1}{2}\mu_0 M_s^{\,2} + K_u \right) d + K_{S,MgO/Fe} + K_{S,Fe/Au} + \Delta K_S(V), \qquad (2)$$

where d is the film thickness. $\Delta K_S(V)$ is a surface anisotropy, induced by a voltage application. When we applied a positive voltage U =+200 V and decreased it to U =-200 V, perpendicular anisotropy was induced and the magnetic anisotropy energy was changed from -31.3 kJ/m^3 to -13.7 kJ/m^3. If we count this change as a change in the surface anisotropy energy, i.e. $\Delta K_s(V)$ in eq. (2), it corresponds to 8.4 μJ/m^2.

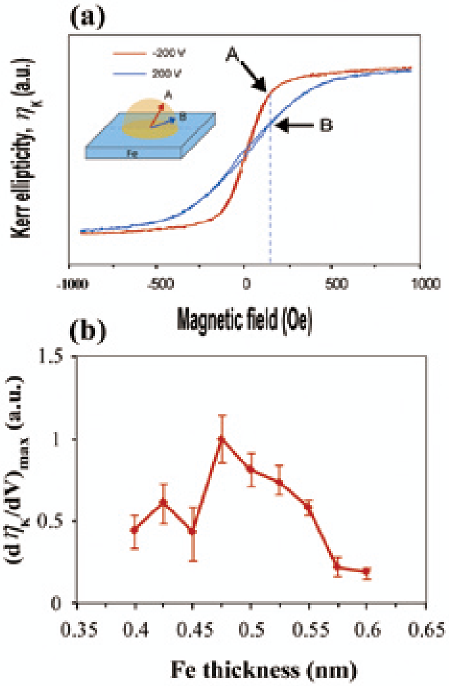

Fig.2 (a) Representaitve Magneto-optical Kerr ellipticity,η_K curves measured under the bias voltage application of $U = \pm$ 200 V. The ultrathin Fe layer thickness is 0.48 nm. (b) Fe layer thickness dependencies of voltage modulation response of η_K. The lock-in modulation technique was used for the precise measurement of the voltage response.

Figure 2(b) shows $(d\eta_K/dV)_{max}$ as a function of film thickness (see the original paper for the detailed information of the measurement technique). The effect was largest for an Fe film with a thickness of 0.48nm, and was smaller for both in thinner and thicker Fe films. Since the influence of the electric field is effective only at the metal / insulator interface, it is natural to observe a smaller effect for the thicker Fe films. The reduction of the amplitude in the thinner region may be caused by a deterioration of the film quality.

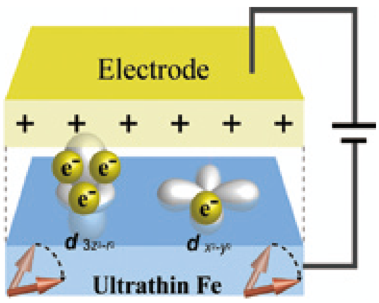

Fig.3 Schematic illustration of the effect of electric field on the electron filling of the 3*d* orbitals in the ultrathin Fe layer. Negative voltage application may increase number of electrons in the m_z = 0 states, leading to the decrease of the perpendicular magnetic anisotropy.

Origin of the voltage-induced anisotropy change

A possible origin of the observed effect is the influence of an electric field on electron filling of the Fe layer, which should affect on magnetic anisotropy. K. Kyuno *et al*. pointed out that the surface magnetic anisotropies in 3*d* ferromagnetic metal / noble metal interfaces were very sensitive to the electron filling of 3*d* orbitals.[9] In our case, from the capacitance of the junction, we could estimate that the change of electron filling about 2×10^{-3} electrons per Fe surface atom is induced by the application of 200 V. From the density of states, this corresponds to about 1 meV increase in chemical potential. This value seems very small, however, it can produce a non negligible change in the surface anisotropy energy. From our experiment, we could change anisotropy energy by 4 μeV per surface Fe atom. This magnitude of change can be reproduced from Kyuno's calculation taking 1 meV change in the chemical potential. Kyuno also pointed out that the effect mainly originates from the large DOS of a d_{xy} and $d_{x^2-y^2}$ character ($|m_z|$=2) at the Fermi energy in the Fe / Au (001) system in which Au has a large spin-orbit coupling. In our case, since the Fe has two interfaces, with Au(001) and MgO(001), the situation is not completely the same, but a similar mechanism may occur in our system. For example, negative voltage application can cause increase of the band filling in the $d_{3z^2-r^2}$ (m_z = 0) states, therefore, the electron occupancy in the d_{xy} and $d_{x^2-y^2}$ states could be changed relatively, leading to a modulation of the magnetic anisotropy (see Fig. 3).

In our experiment, we needed to apply the large voltage, because of the thick polyimide layer. The estimated voltage drop across the MgO layer, however, is about 45 mV/nm if we can neglect a charge accumulation in the barrier (see the Supplementary information of the original paper). As we know that more than 2 V can be applied to a 2 nm thick MgO barrier, a much larger effect can be expected for conventional tunnel magnetoresistance junctions with a MgO barrier.[10] We also proposed a novel magnetization switching method using the voltage-induced anisotropy change observed in this study. Due to limitations of space, please refer to the original paper for more information.

Our results have great significance from the viewpoint that we can control magnetic anisotropy by the application of the electric field in few atomic layers Fe to MgO junctions which can be combined with high quality Fe / MgO / Fe MTJs. This approach provides ideal technique for voltage-controlled magnetization switching and leads to great innovations in ultra-low-power spintronic devices.

Conclusions

We fabricated well-defined Au / few atomic Fe layer / MgO / Polyimide / ITO junctions and measured magnetic hysteresis curve under the voltage application. Clear and large magnetic anisotropy change was observed by the relatively small electric field of 45 mV/nm. One of the most important advantages of our proposed structure is that the basic structure can be applied to a magnetic tunnel junction, which is one of the most important spintronic devices. We hope our findings open up a novel approach of the low power writing technique for spintronics.

References

[1] J. C. Slonczewski, *J. Magn. Magn. Mater.* **159**, L1-L7 (1996).
[2] L. Berger, *Phys. Rev. B* **54**, 9353-9358 (1996).
[3] E. B. Myers *et al., Science* **285**, 867-870, (1999).
[4] V. Novosad *et al., J. Appl. Phys.* **87**, 6400 - 6402 (2000).
[5] H. Ohno *et al.,* Nature **408** (2000) 944.
[6] W. Eerenstein *et al., Nature* **442**, 759-765 (2006).
[7] M. Weisheit *et al., Science* **315**, 349-351 (2007).
[8] C. -G. Duan *et al.,* Phys. Rev. Lett. **101** (2008) 137201.
[9] K. Kyuno *et al., J. Phys. Soc. Jpn.* **65**, 1334-1339 (1996).
[10] S. Yuasa *et al., Nature Mater.* **3**, 868-871 (2004).