Letter

Nonlinear electric field effect on perpendicular magnetic anisotropy in Fe/MgO interfaces

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Received 7 July 2017, revised 20 August 2017
Accepted for publication 22 August 2017
Published 14 September 2017

Abstract
The electric field effect on magnetic anisotropy was studied in an ultrathin Fe(001) monocrystalline layer sandwiched between Cr buffer and MgO tunnel barrier layers, mainly through post-annealing temperature and measurement temperature dependences. A large coefficient of the electric field effect of more than 200 fJ (Vm)−1 was observed in the negative range of electric field, as well as an areal energy density of perpendicular magnetic anisotropy (PMA) of around 600 µJ m−2. More interestingly, nonlinear behavior, giving rise to a local minimum around +100 mV nm−1, was observed in the electric field dependence of magnetic anisotropy, being independent of the post-annealing and measurement temperatures. The insensitivity to both the interface conditions and the temperature of the system suggests that the nonlinear behavior is attributed to an intrinsic origin such as an inherent electronic structure in the Fe/MgO interface. The present study can contribute to the progress in theoretical studies, such as ab initio calculations, on the mechanism of the electric field effect on PMA.

Keywords: magnetoresistance, ultrathin-Fe, magnetic tunnel junction, perpendicular magnetic anisotropy, voltage controlled magnetic anisotropy, electric field effect

(Some figures may appear in colour only in the online journal)

1. Introduction
The electric field effect on magnetic anisotropy, which is often called ‘voltage-control of magnetic anisotropy (VCMA)’, in ferromagnetic metal layers attracts much interest in recent years [1–24]. This technology can help to realize low-power magnetization switching in devices, which is a key to next generation magnetic random access memories (MRAMs). In fact, by using the VCMA, direct manipulation of magnetization by voltage [25–27] and assistance to spin transfer torque switching [28, 29] have been demonstrated.

Cr-buffered Fe/MgO heterostructures are of particular importance in perpendicular magnetic anisotropy (PMA) and relevant studies, and Nozaki et al have recently achieved a remarkable progress in the VCMA study using magnetic tunnel junctions (MTJs) with the Cr/Fe/MgO structure [23]. First, a very large interface PMA was predicted for the Cr/Fe/MgO structure by ab initio calculations [30, 31], followed by an experimental demonstration of PMA of ~1.4 MJ m−3 (~1000 µJ m−2 for areal energy density) in previous studies [32, 33]. Then, Nozaki et al successfully reproduced such a large PMA in their MTJs including the Cr/Fe/MgO structure, so that its
VCMA was examined. The VCMA coefficient obtained for the PMA reached 290 J/(Vm)$^{-1}$ [23].

The mechanism of the large VCMA in Cr/Fe/MgO has not been well understood, although ab initio studies have described the effect of charge accumulation and depletion at the interface that affects the spin-dependent screening and electron’s occupancy of 3d orbitals [32, 34]. Furthermore, an open question is the nonlinear behavior observed unexpectedly in the electric field dependence of PMA energy density [23]. Since it appears to be a unique feature for the VCMA coefficient more than 200 J/(Vm)$^{-1}$, the nonlinear behavior could be a key to develop further large VCMA. At the same time, one may wonder if the nonlinearity can be associated with possible sample-to-sample variation, since no theoretical explanation has been provided. As Cr diffusion into the Fe layer (also into the Fe/MgO interface region) and possible interface contamination with carbon and/or oxygen are somehow discussed in [23], it may be difficult to obtain the well-defined Fe/MgO interface repeatedly and systematically.

In this work, we examined the VCMA in Cr/Fe/MgO under different conditions, i.e. temperature for annealing the structure and temperature of the VCMA measurement, to confirm the presence of the nonlinear VCMA behavior occurring characteristically in the Cr/Fe/MgO with a large VCMA coefficient. The nonlinear VCMA that was obtained together with a large areal PMA energy of ~600 µJ m$^{-2}$ and a large VCMA coefficient of more than 200 J/(Vm)$^{-1}$ gave rise to a local minimum at around 100 mV nm$^{-1}$ in the VCMA curve, being independent of both the post-annealing temperature (i.e. interface quality) and the measurement temperatures. The results insensitive to the interface quality and the measurement conditions suggest that the nonlinear VCMA has an intrinsic origin such as a basic feature of the interface electronic structure.

2. Experimental procedures

Figure 1 shows a schematic design of the MTJ used to examine the VCMA of Cr/Fe/MgO in this study. A fully epitaxial stack of MgO (5 nm)/Cr (30 nm)/Fe (0.7 nm)/MgO (2.2 nm)/Fe (2 nm)/Ru (15 nm) was deposited on a MgO (001) substrate. The bottom 0.7 nm Fe layer was characterized by using a vibrating sample magnetometer to determine the saturation magnetization ($M_s$). The typical resistance-area product (RA) of the MTJs was the order of 10$^4$ Ωcm$^2$. The largest current density applied during the measurement was estimated to be approximately 3 × 10$^4$ A cm$^{-2}$ (from the cross-sectional area of ~40 µm$^2$), in which almost no spin-transfer-torque was exerted. Separately, the magnetization curve of the ultrathin Fe layer was characterized by using a vibrating sample magnetometer to determine the saturation magnetization ($M_s$).

3. Results and discussion

Figures 1(b) and (c) show the RHEED patterns of the ultrathin Fe (0.7 nm) and the Cr buffer layers with the incident electron beam parallel to the [100] azimuth of the MgO (001) substrate. Formation of c(2 × 2) surface structure is observed for both the Cr and Fe surfaces, as the additional streaks pointed by red arrows. Such a surface structure may improve the surface flatness and the magnitude of PMA of the ultrathin Fe layer [31]. In addition, it is noted that the absence of the c(2 × 2) structure for Fe was reported in [23], in contrast to the present study.

Figure 2(a) shows a full TMR curve of a prepared MTJ annealed at 400 °C in the in-plane magnetic field ($H_{ex}$). Due to the small shape magnetic anisotropy energy of the ultrathin Fe layer and the sufficiently large interface PMA induced at
the Fe/MgO interface, the easy axis of the ultrathin Fe layer is aligned perpendicular to the film plane; meanwhile, the top Fe layer has an easy axis parallel to the film plane. This orthogonal-easy-axis design enables us to electrically detect the rotation of the magnetization in the ultrathin Fe layer by applying an $H_{\text{ex}}$ [11]. As described in figure 2(a), the two Fe layers take the orthogonal magnetization configuration at $H_{\text{ex}} = 0$, which brings about a high tunnel resistance state of the MTJ. By applying an $H_{\text{ex}}$, the magnetization of the top Fe layer (reference layer) immediately turns parallel to the $H_{\text{ex}}$ due to its in-plane magnetic anisotropy and small in-plane coercivity (< a few Oe), i.e. the magnetization of the top layer is almost always parallel to the $H_{\text{ex}}$ direction. On the other hand, the magnetization of the bottom Fe layer (free layer) gradually tilts and finally becomes parallel to the $H_{\text{ex}}$ when the $H_{\text{ex}}$ reaches the anisotropy field of the bottom Fe layer ($H_k$). Therefore, the TMR curve reflects the magnetization process of the free Fe layer, i.e. the tunnel resistance takes the maximum at $H_{\text{ex}} = 0$ (orthogonal magnetization configuration) and gradually decreases down to the minimum with the increase of $H_{\text{ex}}$ (parallel magnetization configuration). The TMR ratio in figure 2(a) corresponds to a half of the whole TMR change between parallel and antiparallel magnetization configurations.

Figure 2(b) shows the normalized TMR curves in the negative $H_{\text{ex}}$ region for MTJs annealed at different temperatures. The TMR ratio is normalized by using the maximum (at $H_{\text{ex}} = 0$) and minimum (at $H_{\text{ex}} > H_k$) values, respectively. In the TMR curves, one can clearly see that the saturation behavior changes with the annealing temperature. This means that $H_k$ strongly depends on the annealing temperature, indicating that the annealing process governs the Fe/MgO interface conditions that determine the PMA characteristics.

From the normalized TMR curves, we can evaluate the effective PMA energy density ($K_{\text{eff}}$), including the contribution of the shape magnetic anisotropy, as follows: the tunnel resistance is given by the relative angle between the magnetizations of the free and reference magnetic layers. In the sample design, the maximum resistance occurs at the $90^\circ$ configurations ($R_0$ at $H_{\text{ex}} = 0$), while the minimum resistance does in the parallel configuration ($R_p$ at $H_{\text{ex}} = H_k$). The resistance at a given relative angle $\theta$ is expressed as [23]:

\[
R(\theta) = \frac{R_0 R_p}{R_p + (R_0 - R_p) \cdot \cos \theta}.
\]

Since the magnetization direction of the top Fe layer is considered to be parallel to $H_{\text{ex}}$ (i.e. in-plane direction), the ratio of the in-plane component of the magnetization $M_{\text{in-plane}}$ to its saturation magnetization $M_s$ in the bottom ultrathin Fe layer can be determined as:

\[
\frac{M_{\text{in-plane}}}{M_s} = \cos \theta = \frac{R_0 - R(\theta)}{R(\theta)} \frac{R_0}{R_0 - R_p}.
\]

The $K_{\text{eff}}$, $t_{\text{Fe}}$ as a function of applied electric field $E$ at RT for MTJs annealed at different temperatures. The dash lines indicate results of the linear fitting. The dotted line indicates the position of the local minimum.

\[
K_{\text{eff}} = \frac{M_{\text{in-plane}}}{M_s} = \cos \theta = \frac{R_0 - R(\theta)}{R(\theta)} \frac{R_0}{R_0 - R_p}.
\]
above the local minima. The linear fitting results were plotted for each MTJ (dashed line). The slope below the local minimum positions were in the range of $-133$ to $-266$ fJ (Vm)$^{-1}$. The values of the slopes are the so-called VCMA coefficient, and those observed in the present Fe/MgO interfaces are consistent with that reported in [23]. In the range of $E$ above the local minima, the VCMA coefficients are much smaller than those below the local minima. Furthermore, it is a new finding in the present study that the local minimum position around $+100$ mV nm$^{-1}$ is independent of the annealing temperature. This suggests that the nonlinear behavior is insensitive to the interface conditions, while the PMA energies and VCMA coefficients are very likely to depend on the interface conditions.

The appearance of the local minimum always observed at around $+100$ mV nm$^{-1}$ despite the variation in the annealing temperature would be specific to the VCMA characteristics of Cr/Fe (0.7 nm)/MgO structures. To explore it further, the VCMA characteristics for the MTJ annealed at 400 °C was evaluated at low measurement temperatures. Figure 4 shows the $K_{\text{eff}}$ vs. $E$ as a function of $E$ at 10, 100, 200 and 300 K. It is clearly seen that the $K_{\text{eff}}$ increases with decreasing the measurement temperature. Interestingly, the local minimum positions ($\sim +100$ mV nm$^{-1}$) are independent of the temperatures, while the nonlinearity becomes significant at lower temperatures. In addition, there might be a few fine structures, as implied by the faint peak at around $-50$ mV nm$^{-1}$ in the $E$ dependence of $K_{\text{eff}}$ at low temperatures.

As shown in figures 3 and 4, the nonlinear behavior, particularly the local minimum at around $+100$ mV nm$^{-1}$, has been found to be independent of both the annealing and measurement temperatures. The former temperature is likely to influence the interface conditions, and indeed the PMA and the VCMA coefficient vary with the annealing temperature. The latter temperature may be related with possible extrinsic effects such as an epitaxial strain induced in the MTJ and some kinds of artifacts. Thus, the observed insensitivity of the minimum position suggests that the nonlinear behavior is attributed to an intrinsic origin such as a basic feature of the electronic structure at the Fe/MgO interface. In fact, interface resonant states (IRSs) are formed in the minority spin band of the Fe/MgO system, and the IRSs may affect the transport properties in the Fe/MgO-based MTJs, as proposed by Belashchenko et al [35]. The effect of IRSs on the VCMA at the Fe/MgO interface was also studied by means of $\text{ab initio}$ calculations [36, 37]. Thus, the observed phenomena are expected to contribute to the progress in such theoretical studies, particularly in $\text{ab initio}$ calculations [30, 34, 36–40], on the mechanism of VCMA.

4. Conclusions

The nonlinear VCMA characteristics in orthogonally-magnetized MTJs with a Cr/Fe(0.7 nm)/MgO structure were studied by evaluating post-annealing and measurement temperature dependences. A large VCMA coefficient of more than 200 fJ (Vm)$^{-1}$ and a large areal PMA energy density of around 600 μJ m$^{-2}$ at RT were obtained at the 0.7 nm Fe/MgO interfaces. More interestingly, regardless of the post-annealing and measurement temperatures, a clear local minimum around +100 mV nm$^{-1}$ was observed in the electric field dependence of magnetic anisotropy. The present results imply that the origin of the local minimum is attributed to an inherent electronic structure in the Cr/Fe/MgO.

Acknowledgments

This study was partly supported by the ImPACT program of the Council for Science, Technology and Innovation (Cabinet Office, Government of Japan) and JSPS KAKENHI Grant Number 16H06332. A part of this work was performed under the Inter-University Cooperative Research Program of Institute for Materials Research, Tohoku University and the JSPS-ESPSRC-DFG Core-to-Core Program. QX acknowledges National Institute for Materials Science for the provision of a NIMS Junior Research Assistantship.

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