## Challenges toward voltage-torque MRAM

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A magnetic tunnel junction (MTJ) consisting of a thin insulating layer (a tunnel barrier) sandwiched between two ferromagnetic electrodes exhibits the tunnel magnetoresistance (TMR) effect due to spin-dependent electron tunneling. Since the discovery of room-temperature TMR,<sup>1,2)</sup> MTJs with an amorphous aluminum oxide (Al–O) tunnel barrier, which exhibit magnetoresistance (MR) ratios of several tens percent, have been studied extensively. In 2004, MR ratios of about 200% were obtained for fully epitaxial MTJs with single-crystal MgO(001) tunnel barrier<sup>3)</sup> and textured MTJs with (001)-oriented MgO tunnel barrier<sup>4)</sup>. MTJs with a CoFeB/MgO/CoFeB structure were also developed for practical application.<sup>5)</sup> In the CoFeB/MgO/CoFeB MTJ, a highly textured MgO(001) barrier layer is grown on an amorphous CoFeB bottom electrode layer. By post-annealing the MTJs, the amorphous CoFeB layers are crystallized in bcc(001) structure due to the solid-phase epitaxial growth from the MgO interfaces<sup>6)</sup>. Then, the (001)-textured CoFeB/MgO/CoFeB MTJ exhibit giant MR ratios as well as other practical properties such as low resistance-area (RA) product <sup>7,8)</sup> and/or interfacial perpendicular magnetic anisotropy (PMA).<sup>9)</sup> Because of the high manufacturability and practical magneto-transport properties, the CoFeB/MgO/CoFeB MTJs are widely used as the read heads of hard disk drives (HDDs), memory cell of non-volatile memory (STT-MRAM) especially with perpendicular magnetization, spin-torque oscillator (STO), and physical random number generator (Spin Dice).<sup>10,11</sup>

Although the textured CoFeB/MgO/CoFeB MTJs have been very successful, the properties are not sufficient for future device applications. Novel voltage-driven MRAM or voltage-torque MRAM based on voltage-induced dynamic switching<sup>11</sup> requires not only very high MR ratio (>>300%) but also very large voltage-control of magnetic anisotropy (VCMA) effect and PMA at the same time.<sup>12</sup> For satisfying these requirements, we need to develop novel MTJs with new materials for barrier and magnetic layers by using epitaxial growth on Si substrate as well as the wafer bonding and three-dimensional integration technologies to integrate the epitaxial MTJs in practical LSI. This paper summarizes challenges toward the voltage-torque MRAM.

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## Large voltage-controlled magnetic anisotropy change in epitaxial Cr/ultrathin Fe/MgO/Fe magnetic tunnel junctions

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Technological development in electric-field control of magnetic properties is strongly demanded to realize novel spintronic devices with ultralow operating power. Voltage-controlled magnetic anisotropy (VCMA) effect in an ultrathin ferromagnetic metal layer<sup>1), 2)</sup> is the most promising approach, because it can be applied in MgO based magnetic tunnel junction (MTJ). We have demonstrated fast speed response of VCMA effect through the voltage-induced ferromagnetic resonance<sup>3)</sup> and pulse-voltage induced dynamic magnetization switching<sup>4)</sup> so far. One of the outstanding technical issues in the VCMA effect is the demonstration of scalability. For example, for the development of G-bit class memory applications, high VCMA coefficient of more than 1000 fJ/Vm is required with sufficiently high thermal stability. However, the VCMA effect with high speed response is limited to be about 100 fJ/Vm at present.<sup>5)</sup>

In this study, we investigated the VCMA effect in an ultrathin Fe layer sandwiched between epitaxial Cr(001) buffer and MgO(001) barrier layers.<sup>6)</sup> High interface anisotropy energy,  $K_{i,0}$  of about 2 mJ/m<sup>2</sup> was recently demonstrated in Cr/ultrathin Fe/MgO structure,<sup>7)</sup> probably due to the atomically flat interfaces and suppression of surface segregation from the buffer material. We applied this structure in the voltage-driven MTJ and performed systematic investigations on perpendicular magnetic anisotropy (PMA) and VCMA effect through the tunnel magnetoresistance (TMR) properties. Fully epitaxial MTJ of MgO seed (3 nm)/Cr buffer (30 nm)/ultrathin Fe ( $t_{Fe}$ )/MgO ( $t_{MgO}$ )/Fe (10 nm)/Ta/Ru were deposited on MgO (001) substrates by molecular beam epitaxy. Here, the ultrathin Fe layer is the voltage-controlled free layer with perpendicular magnetic easy axis and top thick Fe layer is the reference layer with in-plane magnetic easy axis. The PMA energy,  $K_{PMA}$  and VCMA properties were evaluated from the normalized TMR curves measured under in-plane magnetic fields with various bias voltage applications. Saturation magnetization value was obtained by SQUID measurement.

High interface anisotropy energy,  $K_{i,0}$  of 2.1 mJ/m<sup>2</sup> was confirmed in our sample. Figure 1 shows an example of applied electric field dependene of surface anisotropy energy,  $K_{PMA}t_{Fe}$  for the MTJ with  $t_{Fe}$ = 0.45 nm and  $t_{MgO}$  = 2.8 nm. We observed large VCMA coefficient of about 400 fJ/Vm under the negative electric field application, while non-linear behavior appeared under the positive direction. In the presentation, we'll discuss the possible origin of the enhanced VCMA effect and non-linearity including the evaluation results of structural analysis at the Cr/ultrathin Fe/MgO interfaces.

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Figure 1 Example of VCMA effect observed in epitaxial Cr/ultrathin Fe/MgO/Fe MTJ with  $t_{\rm Fe}$ =0.45 nm and  $t_{\rm MgO}$  = 2.8 nm. Perpendicular magnetic anisotropy,  $K_{\rm PMA}$  was evaluated from normalized TMR curves and saturation magnetization value measured by SQUID.

## Write error rate of voltage-driven dynamic magnetization switching

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Voltage-control of magnetic anisotropy [1,2] is a promising technique for ultimate spintronic devices with ultra-low power consumption. To apply the voltage-induced magnetic anisotropy change to the writing process, the dynamic magnetization switching triggered by the sub-ns pulse voltage has been demonstrated. [3,4] One of the important issues for the practical application is the evaluation and improvement of the write error rate (WER). However precise control of the magnetization dynamics is not easy because the proper pulse duration is about 1ns or shorter. In this study, we investigated the WER of voltage-induced dynamic magnetization switching in perpendicularly magnetized magnetic tunnel junctions (p-MTJs). [5]

A film for p-MTJ, consisting of buffer layer /  $[Co (0.24 \text{ nm})/Pt (0.16 \text{ nm})]^7$  / Co (0.24 nm) / Ru (0.46 nm) /  $[Co (0.24 \text{ nm})/Pt (0.16 \text{ nm})]^5$  / CoB (0.4 nm) / W (0.15 nm) / Co<sub>12</sub>Fe<sub>68</sub>B<sub>20</sub> (1.0 nm) / MgO barrier / FeB (1.8 nm) / W (2.0 nm) / cap layer, was prepared by using ultra-high vacuum sputtering machine (Canon-Anelva C-7100). The film was annealed at 350°C for 1 hour and micro-fabricated into a 120-nm-diameter p-MTJ. The magnetoresistance ratio and resistance-area product are 101% and 370  $\Omega \cdot \mu m^2$ , respectively. We investigated the WER from the 10<sup>5</sup> repeated events at various conditions of pulse duration and pulse amplitude and external magnetic field.

First, we observed the bidirectional switching and oscillatory behavior of switching probability. These results clearly indicate that the observed switching originates from the voltage-induced magnetic anisotropy change. Figures 1 (a) – (c) show the WER as a function of pulse duration under different conditions of the in-plane magnetic field strength. The minimum of WER, (WER)<sub>min</sub>, was obtained at the half period of the magnetization precession, which becomes shorter as increasing the in-plane magnetic field. Increase of switching time results in low (WER)<sub>min</sub> because the effect of thermal agitation becomes negligible. However further increase of an in-plane magnetic field increases the (WER)<sub>min</sub> of  $4 \times 10^{-3}$  was obtained as shown in Fig. 1 (b). The comparison between the results of the experiment and simulation based on a macro-spin model shows a possibility of ultralow WER (< 10<sup>-15</sup>).

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Figure 1 Write error rate (WER) as a function of pulse duration under in-plane magnetic fields of (a) 14 mT, (b) 20 mT, and (c) 38 mT. Blue and red curves represent the WER from P to AP state and AP to P state, respectively.

高次の磁気異方性を有する自由層の磁化反転特性

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Magnetization switching property in a free layer having higher-order magnetic anisotropy R. Matsumoto<sup>\*</sup>, H. Arai<sup>\*\*,\*</sup>, S. Yuasa<sup>\*</sup>, and H. Imamura<sup>\*</sup> (\*AIST, \*\*JST-PRESTO)

#### 1 はじめに

CPU の SRAM や DRAM を置き換えるための STT-MRAM を開発するためには、その熱耐性 ( $\Delta_0$ ) を 60 以上に保持しつつスピントランスファー・トルク磁化反転の閾値電流密度 ( $J_{sw}$ ) を 1 MA/cm<sup>2</sup> 以下に低減させることが求められる. 最近我々は、等しい熱耐性  $\Delta_0$ (=60) で比較した場合、コーン磁化の自由層 (c-FL) は従来の垂直磁化の自由層より閾値電流密度は 22% 小さく磁化反転時間は 56% 短くなることなど、c-FL の優位性を理論的に明らかにしてきた <sup>1,2)</sup>. コーン磁化とは、低次の磁気異方性 (その定数を  $K_{u1,eff}$  と呼び、反磁場エネルギーを含めたものとする) と高次の磁気異方性 (その定数を  $K_{u2}$  と呼ぶ) の競合で発現する磁化状態で、その磁化は面内方向と面直方向の間の方向を向く. コーン磁化にならずとも  $K_{u2}$  は  $J_{sw}$  低減に有利であると考えられる. 本研究では、 $K_{u2}$  を有する自由層を利用した STT-MRAM 素子の  $\Delta_0$  と  $J_{sw}$  を解析的に計算し、その効果を調べた.

本研究で考慮する STT-MRAM 素子を図 1(a) に図示した.参照 層は垂直磁化で,自由層は垂直磁化かコーン磁化である.極角 ( $\theta$ ) は z 軸から測った角度である.正の電流のとき電子 (電気素量を e と する) は自由層から参照層へ流れる.

自由層のエネルギー密度 ( $\epsilon$ ) は次のように書き表される:  $\epsilon = K_{u1,eff} \sin^2 \theta + K_{u2} \sin^4 \theta$ . 図 1(b) に磁化状態の  $K_{u1,eff}$ ,  $K_{u2}$  依存性を示している.  $K_{u1,eff} < 0$  かつ  $K_{u2} > -(1/2)K_{u1,eff}$ のときにコーン磁化が安定状態となる.  $K_{u1,eff} > 0$ のときに垂直磁化が安定状態か準安定状態となる.

 $\Delta_0$ の解析式は  $\epsilon$  から得られる.図 1(c)の①の領域す なわち [ $K_{u1,eff}$  < 0 かつ  $K_{u2}$  >  $-(1/2)K_{u1,eff}$ ]のとき  $\Delta_0$  =  $\left(K_{u1,eff} + K_{u2} + \frac{K_{u1,eff}^2}{4K_{u2}}\right)V/(k_BT)$ ,②の領域すなわち [ $K_{u1,eff}$  > 0 か つ  $K_{u2} \ge -(1/2)K_{u1,eff}$ ]のとき  $\Delta_0$  = ( $K_{u1,eff} + K_{u2}$ ) $V/(k_BT)$ ,③ の領域すなわち [ $K_{u1,eff}$  > 0 かつ  $K_{u2} \le -(1/2)K_{u1,eff}$ ]のとき  $\Delta_0$  =  $\left[-K_{u1,eff}^2/(4K_{u2})\right]V/(k_BT)$ である<sup>3)</sup>.解析式から計算した  $\Delta_0$ の  $K_{u1,eff}$ ,  $K_{u2}$ 依存性を図 1(d) に示す. $K_{u1,eff}$  と  $K_{u2}$  は大きいほど  $\Delta_0$  は大きい.

 $J_{sw}$ の解析式はランダウ-リフシッツ-ギルバート方程式から得られる. 図 1(e) の ① の領域すなわち [ $K_{u1,eff} > 0$ かつ  $K_{u2} \ge (1/4)K_{u1,eff}$ ] または [ $K_{u1,eff} < 0$ かつ  $K_{u2} > -(1/2)K_{u1,eff}$ ] のとき  $J_{sw} = \frac{8}{3\sqrt{6}} \frac{\alpha d|e|}{\hbar P} \sqrt{\frac{(K_{u1,eff} + 2K_{u2})^3}{K_{u2}}}$ であり、 $K_{u1,eff}$  と  $K_{u2}$  は大きいほど  $J_{sw}$ も大きい. 一方で ② の領域すなわち [ $K_{u1,eff} > 0$ かつ  $K_{u2} \le (1/4)K_{u1,eff}$ ] のとき  $J_{sw} = 4 \frac{\alpha d|e|}{\hbar P} K_{u1,eff}$ であり、 $J_{sw}$  は  $K_{u1,eff}$  のみに 比例する. 解析式から計算した  $J_{sw}$  の  $K_{u1,eff}$ 、 $K_{u2}$  依存性を図 1(f) に 示す. 図 1(e), (f) から [ $K_{u1,eff} > 0$ かつ 0 <  $K_{u2} \le (1/4)K_{u1,eff}$ ] のと きは、 $K_{u2}$ は $\Delta_0$ の上昇に寄与するものの  $J_{sw}$ を上昇させないこと がわかる.  $\Delta_0$ を保持させつつ  $J_{sw}$  を低減させる観点からはこの領 域が最も有利であると考えられる.

#### 2 結果および考察



**Fig. 1** (a) STT-MRAM 素子の模式図. (b): 磁化状態, (c), (d):  $\Delta_0$ , (e), (f):  $J_{sw}$  の  $K_{ul,eff}$ ,  $K_{u2}$  依存性. (c) と (e) は解析 式の区分を表す.

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- 3) 本文中の数式における記号の意味は次の通りである:  $V \ge d$  は自由層の体積と厚さ,  $k_{\rm B}$  はボルツマン定数, T は絶対温度,  $\alpha$  はギルバート・ダンピング定数,  $\hbar$  はディラック定数, P はスピン分極率.

## Deep etching microfabrication of perpendicularly magnetized MTJ

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Low damage microfabrication is one of the most importance issue to fabricate higher density magnetic memory devices. Etching process of the pillar part of magnetic tunnel junction (MTJ) is thought to be the main origin of the processing damage. Argon ion beam etching has been used widely to fabricate the pillar part of MTJs because its etching rate is not much sensitive to film materials. Reduction of the beam voltage of Ar ion beam etching is a straight way to decrease the processing damage. Here, we report the deep etching microfabrication using low voltage Ar ion beam etching, and some of the deep etched MTJs show enhancement of coercive field (H<sub>c</sub>) and keep thermal activated energy ( $\Delta$ ).

We introduced new fabrication machine which is combining one etching chamber and two deposition chambers. This machine makes possible to etch the pillar of MTJs and then to transfer the deposition chamber without breaking the vacuum. The beam voltage and current of Ar ion beam is set to 150V and 45 mA, respectively. Low resistance perpendicularly-magnetized MTJs [1] were used to estimate the process damage. We prepared a film of perpendicularly-magnetized MTJ, which is consisting of buffer layer / [Co (0.24 nm)/Pt (0.16 nm)]<sup>9</sup> / Co (0.24 nm) / Ru (0.52 nm) / [Co (0.24 nm)/Pt (0.16 nm)]<sup>4</sup> / W (0.1 nm) / CoB (0.4 nm) / W (0.1 nm) / FeB (1.1 nm) / MgO barrier / FeB (~2 nm) / MgO cap / cap layer by ultra-high vacuum sputtering machine (Canon-Anelva C-7100). The top of the buffer layer is about 50 nm-thick Ta layer. The film was annealed at 330°C for 1 hour, and then microfabricated into circular MTJs with etching masks of 85, 75, 65 nm diameters. The resistance-area (RA) product of the film was 2.0  $\Omega$ ·µm<sup>2</sup>.

Two etching processes are tested; the first is standard etching where the MTJ film was etched down to just top of the buffer layer; and the second is deep etching where the film was over etched into the middle of the buffer layer. The etching depth was monitored by secondary ion mass spectrometer, but we need to care that the etching depth near the pillar tends to be smaller than that of the plane part. The typical etching time for the standard etching is 30 min. and that for the deep etching is about 50 min. After that, the pillar was covered by  $SiO_2$  layer without breaking the vacuum, and then lift-offed the etching mask and made the top electrode.

For both cases, the magnetoresistance (MR) ratios of the MTJs were 110~120% and well coincident. Diameters of the MTJs were estimated from the resistance of parallel state and the RA value. Reduction of the diameter was about 15nm for standard etching and that was about 25 nm for deep etching. We found the deep etched MTJs tend to have larger coercive field (H<sub>c</sub>) that standard etched one and those MTJs have relative large thermal activation energy ( $\Delta$ ) where  $\Delta$  was evaluated from the current dependence to the switching probability [2].

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Figure 1 Relationships between (a) estimated diameter of MTJ and  $H_c$ , (b) estimated diameter and  $\Delta$ , (c)  $H_c$  and  $\Delta$ .