

## Recent progresses and future challenges in voltage-controlled magnetic anisotropy effect

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The development of electric-field manipulation of magnetism is strongly demanded for the reduction in operation power of future spintronic devices. The voltage-controlled magnetic anisotropy (VCMA) effect in an ultrathin ferromagnetic metal layer [1, 2] is a promising and practical approach due to its high applicability in an MgO-based magnetic tunnel junction (MTJ) with high-speed response [3,4]. The VCMA effect originates from voltage-induced charge accumulation/depletion and induction of electron redistribution at the interface between ultrathin ferromagnet and dielectric layers [5]. To show the feasibility of MRAM controlled by voltage, called voltage-torque MRAM [6], we need further improvement in VCMA coefficient. For example, for giga-bit class memory applications, VCMA coefficient of more than a few hundreds or even 1000 fJ/Vm is required [7]. However, high speed VCMA effect is limited to be 100 fJ/Vm at present [8].

In this talk, recent progresses in materials research for the enhancement in the VCMA effect, especially focusing on an epitaxial Fe/MgO MTJs, will be reviewed. Large VCMA coefficient of about 300 fJ/Vm has been achieved by interface engineering using a transition metal doping at the ultrathin Fe/MgO interface.

We'll also introduce the evaluation of write error rate (WER) of precessional magnetization switching induced by VCMA effect in perpendicularly magnetized MTJs [9]. By optimizing the thermal stability and VCMA coefficient in the voltage-controlled free layer, lowest WER of  $2 \times 10^{-5}$  has been demonstrated [10]. Future strategy to realize the practical low WER value will also be discussed.

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## Perpendicular magnetic anisotropy at Fe/MgAl<sub>2</sub>O<sub>4</sub> interfaces and its voltage effect

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Voltage-controlled magnetic anisotropy (VCMA) [1] in magnetic heterostructures is expected as a key technology for achieving low-power consumption spintronic devices such as voltage-torque magnetoresistive random access memories (MRAMs). However, increase of both the interface perpendicular magnetic anisotropy (PMA) energy ( $K_i$ ) and the VCMA coefficient ( $\beta$ ), i.e.,  $K_i > 2-3$  mJ/m<sup>2</sup> and  $\beta > 1000$  fJ/(Vm), is necessary for high density memory applications. In order to achieve such a giant VCMA effect, exploring the origin of the VCMA effect using “standard PMA heterostructures” without any interfacial defects can be indispensable. Recently, large PMA energies were reported in lattice-matched Fe/MgAl<sub>2</sub>O<sub>4</sub> [2] and Co<sub>2</sub>FeAl/MgAl<sub>2</sub>O<sub>4</sub> heterostructures [3]. Therefore, we focused in this study on ultrathin Fe/MgAl<sub>2</sub>O<sub>4</sub>(001) epitaxial interfaces to achieve high  $K_i$  and  $\beta$  using an electron-beam evaporation technique. Especially, we precisely investigated the Fe thickness dependence using Fe/MgAl<sub>2</sub>O<sub>4</sub>/CoFeB orthogonally magnetized MTJs. We report that only a monolayer thickness difference has a significant impact on the PMA energy and VCMA effect.

MTJ stacks of Cr buffer (30)/Fe ( $t_{Fe} = 0.70, 0.84, 1.0 = 5, 6, 7$  monolayers (MLs))/MgAl<sub>2</sub>O<sub>4</sub> (2)/Co<sub>20</sub>Fe<sub>60</sub>B<sub>20</sub> (5)/Ru (10) (unit in nm) were prepared on an MgO(001) substrate by electron-beam evaporation. The top 5-nm CoFeB is the reference layer with in-plane magnetization for evaluating the VCMA effect of the bottom Fe. The Cr, Fe, MgAl<sub>2</sub>O<sub>4</sub>, and CoFeB layers were post-annealed to improve their crystallinity and flatness. Magnetic properties were investigated using a vibrating sample magnetometer-superconducting quantum interference device. After microfabrication (10  $\mu$ m scale), magnetotransport properties of MTJs were characterized by a physical property measurement system at room temperature. The positive bias was defined with respect to CoFeB (electron tunneling from the lower to upper electrode).

Figure 1 shows the typical in-plane magnetization curves for the MTJ stacks with different Fe thicknesses. It was found that the 5- and 6-ML Fe layers had perpendicular magnetization. Areal PMA energy density  $E_{pma}$  (PMA energy density  $K_{eff}$  [unit in J/m<sup>3</sup>]  $\times t_{Fe}$ ) for the 5-ML (6-ML) Fe sample was determined to be 0.85 mJ/m<sup>2</sup> (0.77 mJ/m<sup>2</sup>). We investigated the bias voltage dependence of  $E_{pma}$  for the 5- and 6-ML Fe samples using normalized tunnel magnetoresistance ratios as functions of both bias voltage and in-plane magnetic field. As clearly seen in Fig. 2,  $E_{pma}$  values for both the samples show complicated bias voltage dependence. Importantly, the  $E_{pma}$  curve shape significantly depends on the Fe thickness; a local minimum appears near +0.2 V for the 5-ML Fe sample, whereas a peak appears at the zero-bias voltage for the 6-ML one. We found that the complicated VCMA effect was associated with the formation of quantum well states [4] for the  $\Delta_1$  states in the ultrathin Fe layers between Cr and MgAl<sub>2</sub>O<sub>4</sub>. This work was partly supported by the ImPACT Program of Council for Science, Technology and Innovation, Japan, and JSPS KAKENHI Grant No. 16H06332.

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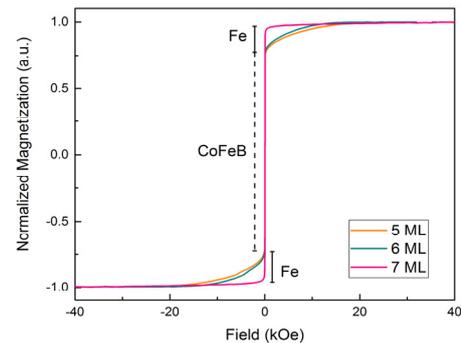


Fig. 1. Magnetizations as a function of in-plane magnetic fields for ultrathin-Fe/MgAl<sub>2</sub>O<sub>4</sub>/CoFeB MTJs with 5-7 ML thick Fe.

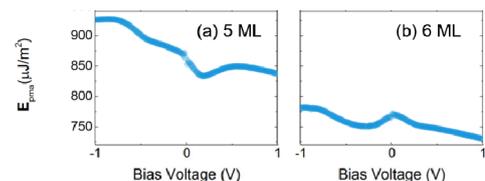


Fig. 2. Bias voltage dependences of  $E_{pma} = K_{eff} \times t_{Fe}$  for (a) 5-ML and (b) 6-ML Fe sample.

# The effect of Os or Ir layer insertion into MgO/Fe interface on the electric-field modulation of magnetic anisotropy

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The voltage-torque magnetoresistive random access memory is the ultra-low energy consumption non-volatile memory based on voltage-controlled magnetic anisotropy (VCMA). The VCMA coefficient was reported to be 30-40 fJ/Vm for the MgO/Fe/Au and MgO/CoFeB/Ta films [1, 2]. Recently, large VCMA of 290 fJ/Vm was demonstrated for the MgO/Fe/Cr film [3]. However, VCMA effect larger than 1000 fJ/Vm is required for realizing the voltage-induced magnetization switching in magnetic tunnel junctions below 30nm. The purpose of this work is to design the magnetic film exhibiting larger VCMA. We theoretically investigated the effect of *5d* transition-metal layer insertion into the MgO/Fe interface on the electric-field modulation of magnetic anisotropy.

We have carried out first-principles electronic-structure calculations employing the projector augmented-wave with plane wave basis set by using the Vienna ab initio simulation package [4]. We estimated magnetic anisotropy energy (MAE) and its electric-field modulation for MgO/Os(Ir)/bcc-Fe/Cu(001) films. The MAE was estimated by using the magnetic force theorem.

Figures 1(a) and (b) show the electric-field modulation of MAE for the Os/Fe and Ir/Fe films, respectively, with and without MgO capping layer. The VCMA coefficient is estimated to be -173, 298 fJ/Vm for the MgO/Os/Fe and MgO/Ir/Fe film, respectively, and these values are one order of magnitude larger than that for the MgO/Fe interface. These VCMA coefficients are comparable with that of Os- and Ir-monolayer on the Fe surface. However, perpendicular MAE is drastically decreased in both Os/Fe and Ir/Fe film by MgO capping. In the case of Ir/Fe film, opposite sign of VCMA is obtained for the film with and without MgO. These results indicate that the bonding between *5d* transition-metal and oxygen plays an important role for the MAE and its electric-field modulation. At the MgO/Os and MgO/Ir interfaces, the density of states (DOS) projected on the majority-spin *5d*( $3z^2-r^2$ ) orbital, which contributes to the in-plane MAE, is increased near the Fermi level by the hybridization between *5d*( $3z^2-r^2$ ) and O-*2p*(*z*) orbitals. This is the origin of the reduction of perpendicular MAE by the MgO capping. In particular, MgO/Ir/Fe film shows the huge in-plane MAE, since the DOS of *5d*( $3z^2-r^2$ ) orbital is located just at the Fermi level. In the presentation, we also discuss the origin of the sign change of VCMA coefficients for the Ir/Fe and MgO/Ir/Fe films.

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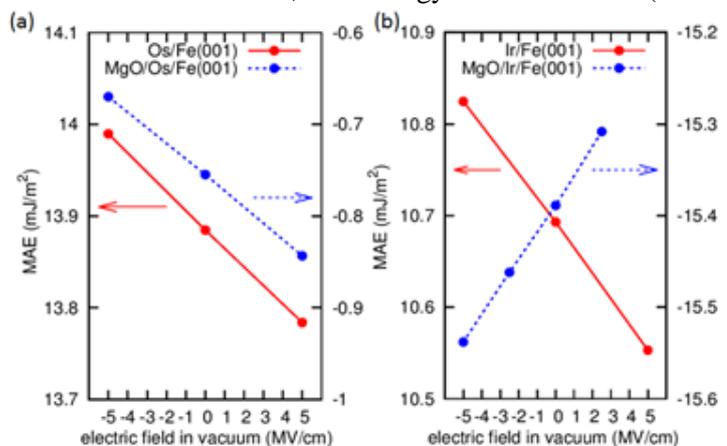


Fig. 1: Magnetic anisotropy energy (MAE) as a function of electric field in vacuum for the Os/Fe (a) and Ir/Fe (b) films with and without MgO capping layer.

# Electric field control of magnetic anisotropy in bilayer contacts with Rashba-type spin-orbit interaction

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Uniaxial magnetic anisotropy (MA) plays an important role in spintronic applications in which ferromagnetic (FM) thin films and heterojunctions are utilized. The MA in such magnetic materials originates from spin-orbit interaction (SOI) expressed by  $L$ - $S$  coupling and the low dimensionality of the lattice structure. As a result, out-of-plane MA often occurs at surfaces and interfaces of these magnetic heterojunctions. The magnetization direction in ferromagnets is usually controlled by an external magnetic field. Recently, control of magnetic ordering by using spin-transfer torque, the magnetostrictive effect, ferroelectricity, the piezoelectric effect, and electric field has attracted much attention in the field of spintronics.

Quite recently, another type of SOI, the Rashba-type SOI (R-SOI) was proposed to be a source of MA. Theoretical analysis of MA was performed for a two-dimensional layer by using exchange-split free electron and single-orbital TB models with R-SOI.<sup>1-3)</sup> The study using TB model<sup>3)</sup> predicted that the layer shows in-plane MA for both low and high electron densities, while it shows out-of-plane MA otherwise. The occurrence of MA by the R-SOI may be attributed to a characteristic change in the Rashba-split energy state under an exchange field produced by the FM layer itself or by magnetic ions/atoms in an FM material attached to a non-magnetic (NM) layer. It is interesting to note that the R-SOI may be controlled by an external electric field because of its intrinsic nature.

In this work,<sup>4)</sup> we theoretically study the uniaxial MA of a bilayer made of NM and FM layers putting an emphasis of relative role of the R-SOI on NM layer and  $L$ - $S$  coupling, that is, atomic-SOI (A-SOI) on the FM layer. We construct a simple model for the bilayer based on the first-principle calculation of the Rashba-split bands of the Au(111) surface. In this model, the electronic structure of NM layer is given by a single-orbital TB model, while that of FM layer is presented in the full  $3d$ -orbital TB model, in addition to the orbital mixing between NM and FM layers. After numerical calculation, we have shown that the R-SOI of the NM layer produces MA via  $p$ - $d$  mixing between the NM and FM layers. The MA energy caused by the R-SOI is less than 1 meV, while that caused by the A-SOI is a few meV per unit cell. Both interactions show an oscillatory dependence of the uniaxial MA energy on the electron number. Because the "phases" of these oscillations are different, the uniaxial MA originating from the R-SOI alone could be the same order of magnitude as that produced by A-SOI alone under certain conditions. The result indicates that an external electric field with reasonable magnitude may change the MA from being out-of-plane to in-plane, and vice versa.

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電界による  $g$  因子の変調と磁気異方性の相関京大化研<sup>A</sup>, 東大工<sup>B</sup>水野隼翔<sup>A</sup>, 〇森山貴広<sup>A</sup>, 河口真志<sup>A</sup>, 田中健勝<sup>A</sup>, 小山知弘<sup>B</sup>, 千葉大地<sup>B</sup>, 小野輝男<sup>A</sup>Correlation between  $g$ -factor and magnetic anisotropy under the bias electric field<sup>A</sup>ICR, Kyoto University, <sup>B</sup>The University of TokyoH. Mizuno<sup>A</sup>, T. Moriyama<sup>A</sup>, M. Kawaguchi<sup>A</sup>, K. Tanaka<sup>A</sup>, T. Koyama<sup>B</sup>, D. Chiba<sup>B</sup>, and T. Ono<sup>A</sup>

## はじめに

磁性金属超薄膜に電界を印加し、垂直磁気異方性(PMA)を制御する試みが近年注目を集めている[1]。理論的には、界面での局在電子の軌道磁気モーメントの異方性と、スピン軌道相互作用が、磁性多層膜におけるPMAの起源であると予想されている(Bruno's model)[2]。軌道磁気モーメントの異方性は、強磁性共鳴(FMR)測定から得られる $g$ 因子の異方性を通してその評価が可能である[3]。今回、我々はPt/Co超薄膜にゲート電圧 $V_g$ を印加した状態でFMR測定を行い、電界による $g$ 因子の異方性の変調と、PMAとの相関について調査した。

## 実験方法

Ta(5)/Pt(3)/Co(0.8)/MgO(4) (単位: nm)層をGaAs基板にスパッタ成膜後、細線状に加工し、 $V_g$ 印加用の「絶縁層(HfO<sub>2</sub>)/ゲート電極(Au)」を作製した(図1(a))。高周波電流 $I_{rf}$ を細線に注入しながら外部磁場 $H_{ex}$ を掃引し、ホモダイン検出によるFMR測定を行った。試料に対する外部磁場 $H_{ex}$ の掃引方向( $\theta_H = 30, 90^\circ$ )を変えてFMR測定することで、 $g$ 因子の異方性を見積もった。

## 実験結果

図1(b)に示したように、共鳴条件のずれから、 $V_g$ 印加により磁気特性が変調されていることがわかる。これらの共鳴条件から見積もった $g$ 因子の異方性( $\Delta g = g^\perp - g^\parallel$ )及び、垂直異方性磁場( $H_{kl}$ )の電界による変化を図2に示す。ここで、 $g^\perp$ および $g^\parallel$ はそれぞれ面直方向および面内方向の $g$ 因子である。 $\Delta g$ 、 $H_{kl}$ 共に正の電界に対して増加しており、両者に正の相関があることが分かる。本講演ではBruno's model[2]を用いてこれらの相関の詳細について議論する。

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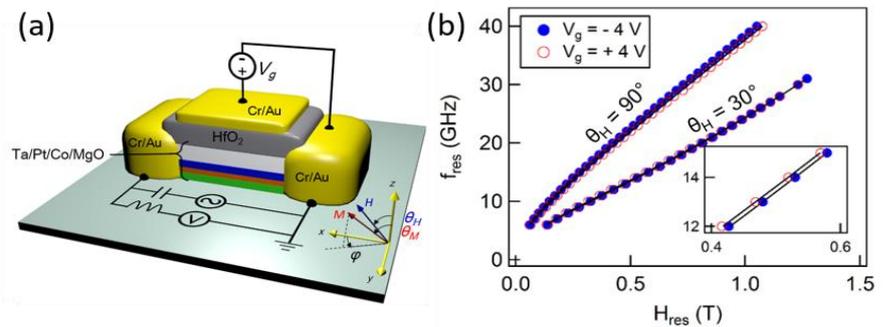
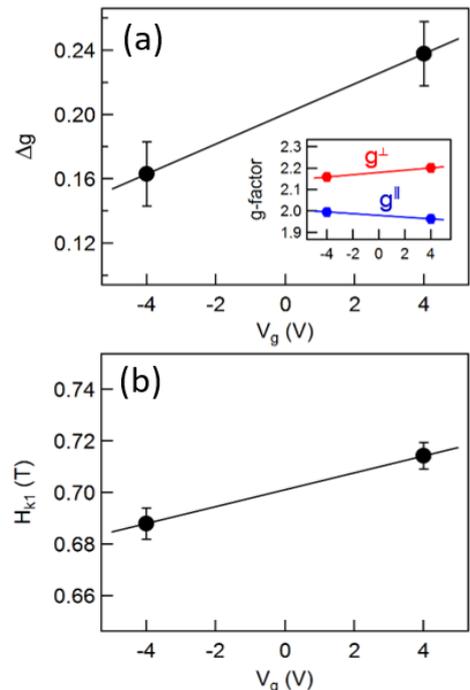


図1 (a)デバイスの模式図 (b)共鳴磁場と周波数の関係

図2 (a)  $g$  因子の異方性 ( $\Delta g$ ) および (b) 垂直異方性磁場 ( $H_{kl}$ ) のゲート電圧依存性