## Insight into new magnetic recording principle with magnetoelectric writing

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The recording areal density has continued increasing to nearly 1Tbpsi with Perpendicular Magnetic Recording (PMR). However, it seems that the growth rate of a recording areal density is fairly reduced for the trilemma problem due to particulate magnetic recording principle. Of course, such alternative technologies or methods as energy assisted recording (HAMR, MAMR etc.) have been worked on with energy towards realizing the next generation recording system where they are still based on a particulate magnetic recording principle. Considering a magnetic recording (i.e. head/media system), STT and SOT are not suitable for magnetic recording principle. So in this paper we propose a new magnetic recording principle with voltage effect on magnetization-switching, especially Magnetoelectric Effect of Cr2O3 sesquioxide, that is to say that magnetic bits are written on Cr2O3 antiferromagnetic media by E(electric field) • H(magnetic field) product.

Magnetoelectric (ME) effect has so far been paid attention to be applied to a nonvolatile memory (NVM). Cr2O3 is a typical sesquioxide with ME effect and its antiferromagnetic Neel temperature is TN =307 K, which is higher than RT. A robust isothermal electric control of exchange-bias at RT is actually reported for bulk Cr2O3 single crystal sample when both of electric field E = 0.02 [MV/cm] and magnetic field H = -1.54 [kOe] was applied [1]. But ME effect has not yet been clarified in Cr2O3 thin films because of its large leakage current and imperfect antiferromagnetic-ordering while ME effect like behavior up to 200K is reported to be observed in an ultrathin Cr2O3/Fe2O3 Nano-Oxide Layer (NOL) [2]. When considering the application of ME effect to magnetic recording technology with voltage-controlled magnetization switching, there are some problems except the above issue, which should be resolved. The first is to realize and design an effectually high exchange-bias filed between antiferromagnetic (AFM) Cr2O3 and ferromagnetic (FM) thin film multilayers in the higher temperature range than RT, which means higher Neel temperature (TN) and higher blocking temperature (TB), where the properly low coercive force of FM is also required. The second is to invest FM layer with a perpendicular anisotropy which is thought to be caused by both of the hybridization of FM 3d and O 2p orbitals and the magnetic coupling at the interface between FM and Cr2O3. The third is to confirm ME effect in the thin film Cr2O3 after getting Cr2O3 thin film which shows good electrical properties.

In this paper, electrical and magnetic performances of the thin film Cr2O3/Fe2O3 sesquioxide were investigated. We successfully fabricated the Cr2O3 and Fe2O3 thin films with small leakage current and good magnetic properties. We successfully confirmed ME effect of Cr2O3 thin films (100nm~500nm) and the switching of both exchange bias field and residual magnetization using Co/Cr2O3 exchange bias bilayer with low FM layer coercivity (~20 Oe) structure, shown in Fig.2 under both of ME filed cooling and isothermal process conditions for the first time in the world [3],[4]. In addition, we succeeded in enhancing TM of Fe2O3 higher than 400K by Ir-doping where perpendicular-spin-alignment of Fe2O3 was also confirmed in both of Mossbauer spectroscopy and weak ferromagnetic moment measurement with SQUID magnetometer. These results support our new magnetic recording principle concept with voltage controlled magnetization switching of AFM Cr2O3 thin film above room temperature at the first step. In addition, we also propose low EH product writing with positive exchange coupling in Chromia/Co bilayer system where an induced weak ferromagnetic moment is also observed of antiferromagnetic Chromia. The related experimental

where an induced weak ferromagnetic moment is also observed of antiferromagnetic Chromia. The related experimental results and phenomenologically analytical consideration will be discussed.

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# Characterization of magneto-electric switching energy in Cr<sub>2</sub>O<sub>3</sub> antiferromagnetic thin films

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**Introduction:** Towards applications in voltage-controlled HDD and MRAM, usually the ferromagnetic layer is considered as the "active" media. A possibility of using antiferromagnetic (AFM) media is hindered by the difficulty of controlling and accessing the antiferromagnetic state. Recently, there is a revival in utilizing the magnetoelectric (ME) antiferromagnet  $Cr_2O_3$ , where the electric control of perpendicular exchange-bias in a  $Cr_2O_3$ /Co multilayer system has been realized under the simultaneous application of electric and magnetic fields<sup>1-4)</sup>. However, The required energy of the product of electric and magnetic fields (EH) is 2 to 3 orders of magnitude higher in thin-film systems compared with bulk crystals<sup>1,5)</sup>. In this presentation, we will present the investigation of the origin of this increase, and a novel way to control it.

**Experimental procedure:** Two samples were used, sample A was a commercial bulk  $Cr_2O_3$  substrate, sample B was a sputter-deposited thin-film sample of the following structure: c-Al<sub>2</sub>O<sub>3</sub> sub./Pt 25 nm/Cr<sub>2</sub>O<sub>3</sub> 500 nm/Pt 25 nm. The average domain state of  $Cr_2O_3$  was measured by the linear magnetoelectric susceptibility ( $\alpha$ ) in a SQUID magnetometer. The application of a small electric field (E) results in an induced magnetization (M =  $\alpha$ E), and the sign of  $\alpha$  indicates the AFM domain state of  $Cr_2O_3$ .

**Results and discussions:** Figure 1 shows the comparison between the temperature-dependence of  $\alpha$  in samples A and B. Except for a small decrease in Neel temperature, the magnitude of  $\alpha$  is same and the ME property is intrinsically similar in thin films as to bulk crystals. The increase of EH switching product in thin films is mostly due to the increase of exchange-bias energy density as the thickness becomes reduced orders of magnitude.

Additionally, we surprisingly found a weak magnetization from  $Cr_2O_3$  films, which was coupled to AFM order parameter. This weak magnetization required an additional EH switching energy for single-layer  $Cr_2O_3$ . However, the sign of switching EH energy required to overcome the weak magnetization is opposite to the EH energy required to overcome the exchange-coupling energy with the Co layer. We could design a balance between both, and we could decrease the switching energy 2 orders of magnitude compared to other reports on thin-film  $Cr_2O_3$ .



Fig. 1 Temperature-dependence of  $\alpha$  in samples A and B

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## Magnetic field dependence of threshold electric field for switching exchange bias polarity

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Electric field control of magnetization in magnetoelectric (ME) insulators plays an important role in spintronic applications owing to various advantages such as the high processing speed and the low power consumption. Antiferromagnetic (AFM)  $\alpha$ -Cr<sub>2</sub>O<sub>3</sub> is a typical ME material which showed a fascinating exchange bias as coupled with a ferromagnetic layer [1, 2]. The isothermal ME switching of the perpendicular exchange bias in an all-thin-film system was reversibly achieved with the change in polarity of exchange bias from negative-to-positive (N-to-P) and positive-to-negative (P-to-N) by tuning the applied electric field while maintaining the magnetic field [1]. At a temperature, the threshold electric field ( $E_{th}$ ) at which the polarity of exchange bias is reversed depends on the applied magnetic field [1]. However the study on magnetic field dependence of  $E_{th}$  for switching exchange bias polarity, which is indispensable for future spintronic devices, is still insufficient.

In this study, we investigated the isothermal ME switching of perpendicular exchange bias in  $Cr_2O_3$ . Pt/Co/spacer/Cr<sub>2</sub>O<sub>3</sub>/Pt stacked films were prepared on an  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> substrate using DC magnetron sputtering system. The isothermal switching of exchange bias was investigated by the anomalous Hall effect (AHE) measurement using a Hall-bar device with a 2-µm-width and a 40-µm-length. At 275 K, the exchange bias field was reversibly switched by reversing the electric field under a fixed magnetic field. Fig. 1 shows the hysteretic electric field dependence of the exchange bias field (left) and remanence ratio (right) under -60 kOe at 275 K. The rectangular hysteresis is in agreement with the isothermal switching of AFM domain state in Cr<sub>2</sub>O<sub>3</sub>. Fig. 2 shows the magnetic field dependence of  $E_{th}$  for switching exchange bias polarity, in which  $E_{th}$  was evaluated from the cross point of remanence ratio curve with the horizontal axis for both N-to-P and P-to-N processes. The switching condition, simply expressed by  $EH_{th}$  = constant, followed the coherent model [1, 2] in which the ME effect leads to the energy gain for switching exchange bias polarity. The asymmetry of  $EH_{th}$  between N-to-P and P-to-N was attributed to the uniaxial nature of magnetic anisotropy of AFM layer and the unidirectional nature of exchange coupling of the FM layer. The detail of the dependence of  $E_{th}$  on magnetic field (magnetic field and direction) will be discussed in the presentation.



*Fig. 1*: Hysteretic electric field dependence of the exchange bias field (left) and remanence ratio (right) under -60 kOe at 275 K.



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*Fig.* 2: Magnetic field dependence of threshold electric field  $E_{\rm th}$  for switching exchange bias polarity measured at 275 K.

## Al 置換による電気磁気材料 Cr<sub>2</sub>O<sub>3</sub> 薄膜の磁気異方性向上

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## Enhanced magnetic anisotropy of magnetoelectric Cr<sub>2</sub>O<sub>3</sub> film by Al-doping T. Nozaki, Y. Shiokawa, S. P. Pati, S. Ye, M. Al-Mahdawi, and M. Sahashi (Tohoku University)

## <u>序論</u>

電気磁気材料である Cr<sub>2</sub>O<sub>3</sub> は電圧による磁化反転を可能とする電気磁気効果デバイス、電圧制御 HDD や MRAM の候補として注目を集めている。近年、Cr<sub>2</sub>O<sub>3</sub>/Co 薄膜を用いた系で垂直交換バイアスの電界制御が実 現されたことから<sup>1,2</sup>、実用化がより現実味を帯びてきている。しかし Cr<sub>2</sub>O<sub>3</sub>/Co 交換結合膜では、Cr<sub>2</sub>O<sub>3</sub>の磁 気異方性が低い(K<sub>AF</sub> ~ 2 × 10<sup>5</sup> erg/cc)ため、特に薄い Cr<sub>2</sub>O<sub>3</sub> 薄膜を用いた場合に、交換バイアスのブロッキング 温度がネール温度よりも大幅に下がってしまうことが問題となっている。我々は Cr<sub>2</sub>O<sub>3</sub> と同じコランダム構 造を持つ a-Fe<sub>2</sub>O<sub>3</sub> 薄膜への Ir 少量置換による垂直磁気異方性の向上に成功しており<sup>3,4</sup>、本研究では元素置換 による Cr<sub>2</sub>O<sub>3</sub> 薄膜の磁気異方性の向上を目指した。

## <u>実験方法</u>

膜構成は Al<sub>2</sub>O<sub>3</sub> 基板/Pt 25/Cr<sub>2</sub>O<sub>3</sub> または Al-doped Cr<sub>2</sub>O<sub>3</sub> t<sub>Cr2O3</sub>/Co 1/Pt 5 (nm)である。(Al 置換)Cr<sub>2</sub>O<sub>3</sub> 薄膜は Cr または Al-Cr 合金ターゲットを用い、反応性スパッタ法で作製した。薄膜の Al 組成は XRF により確認した。 Al 5atm%の Al-Cr 合金ターゲットを用いて作製した薄膜の Al 含有量は 3.7atm%程度であった。磁気特性の評価には SQUID 磁力計を用いた。

### <u>実験結果</u>

図1に無置換試料およびAI置換試料の交換バイアスの温度依存性を示す。無置換試料ではCr<sub>2</sub>O<sub>3</sub>薄膜の膜厚が250nmであっても、その小さな磁気異方性と大きな交換バイアスのため、ブロッキング温度は130K程度とネール温度~300Kよりもかなり小さい値となった。それに対して、AI置換試料では、膜厚が同じ程度であるにも関わらず280K程度の大きなブロッキング温度が得られた。さらに、交換バイアスの大きさ自体も無置換試料よりも高い値が得られ、最大で4500Oeを超える巨大な交換バイアスが得られた。Meiklejohn -Beanのモデルによるとブロッキング温度は磁気異方性が大きいほど、また、交換バイアスの大きさが小さいほど大きくなることから、AI置換試料では大幅な磁気異方性の増大が起こっていると考えられる。このような磁気異方性の増大は、電気磁気効果デバイスの熱安定性を確保するためにも不可欠である。



図1 無置換試料および Al 置換試料の 交換バイアスの温度依存性.

### <u>謝辞</u>

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## Effect of heavy metal doping on the Morin transition of

## epitaxial a-Fe<sub>2</sub>O<sub>3</sub> (0001) thin films

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Hematite (a-Fe<sub>2</sub>O<sub>3</sub>), which is an antiferromagnetic material with high Néel temperature ( $T_{\rm N} = 950$  K), is attracting great interest, because this material can be applied for assisting layer of perpendicular magnetic devices, e.g., electric-field-writing-hard-disk drives. In pure a-Fe<sub>2</sub>O<sub>3</sub>, the Morin transition, which is the antiferromagnetic to weak ferromagnetic transition, occurs at 253 K, so that c-axis oriented a-Fe<sub>2</sub>O<sub>3</sub> thin films have the in-plane spin configuration and show weak ferromagnetism at room temperature. Recently, Shimomura *et al.* reported that the Morin transition temperature ( $T_{\rm M}$ ) can be enhanced above 400 K in c-axis oriented a-Fe<sub>2</sub>O<sub>3</sub> thin films by doping 1% Ir <sup>1</sup>). The Morin transition can be explained by the competition between magnetic dipolar anisotropy  $K_{\rm MD}$  and single ion anisotropy  $K_{\rm FS}$ . The enhancement of  $T_{\rm M}$  is caused by the increase of  $K_{\rm FS}$ . In this study, we report the heavy-metal-doping effect for Morin transition of c-axis oriented a-Fe<sub>2</sub>O<sub>3</sub> thin films.

Heavy metal (Ru, Ir and W) doped a-Fe<sub>2</sub>O<sub>3</sub> films of about 80 nm in thickness were deposited on Al<sub>2</sub>O<sub>3</sub>(0001) substrates by pulsed laser deposition method at various substrate temperatures in background oxygen pressure of 10 Pa. X-ray diffraction (XRD) measurement and conversion electron Mössbauer spectroscopy were performed for these samples at room temperature.

XRD measurement clarified that the growth direction of the heavy-metal-doped a-Fe<sub>2</sub>O<sub>3</sub> films on Al<sub>2</sub>O<sub>3</sub>(0001) substrates was along c-axis. Figure 1 shows the Mössbauer spectra of 5% Ru doped a-Fe<sub>2</sub>O<sub>3</sub> films deposited at the substrate temperature of 300°C, 400°C and 500°C. The angle *q* between the average spin direction and the vertical axis of film plane can be estimated by the intensity ratio of the six peaks of the Mössbauer spectra. When the spin direction is perpendicular to the film plane (*q* = 0°), the peaks indicated by the arrows disappear. The results show the peaks indicated by red arrows decrease with increasing growth temperature. The Mössbauer spectra indicated that the spin

direction of 5% Ru doped a-Fe<sub>2</sub>O<sub>3</sub> films deposited at the substrate temperature of 300°C is nearly in-plane ( $q = 76^{\circ}$ ) and that the  $T_{\rm M}$  is below room temperature, while the spin direction of the 5% Ru doped a-Fe<sub>2</sub>O<sub>3</sub> films deposited at the substrate temperature of 500°C is almost perpendicular ( $q = 16^{\circ}$ ) and the  $T_{\rm M}$  is higher than room temperature. The spin direction angle of 5% Ru doped a-Fe<sub>2</sub>O<sub>3</sub> films deposited at the substrate temperature of 300°C is 31°, indicating that the  $T_{\rm M}$  of this sample is near room temperature. These results imply that the lattice location of Ru ions in a-Fe<sub>2</sub>O<sub>3</sub> films varies with changing deposition temperature and that the difference of spin-orbit interaction of Ru ions affects the enhancement of  $K_{\rm FS}$ .

In this presentation, we will also demonstrate the effect of other heavy metal (Ir and W) doping on Morin transition of a-Fe<sub>2</sub>O<sub>3</sub> films.

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Fig. 1 Mössbauer spectra of 5% Ru doped a-Fe<sub>2</sub>O<sub>3</sub> films deposited at the substrate temperature of 300°C, 400°C, and 500°C