

# Cr<sub>2</sub>O<sub>3</sub> BASED MAGNETOELECTRIC FERRIMAGNET TOWARD MRAM APPLICATIONS

Tomohiro NOZAKI<sup>1</sup> and Masashi SAHASHI<sup>1,2</sup>

1) Department of Electronic Engineering, Tohoku University, Sendai 980-0845, Japan

2) ImPACT Program, Japan Science and Technology Agency, Tokyo 102-0076, Japan

## I. INTRODUCTION

Recently the voltage control of a magnetization direction has received considerable attentions as the technology to achieve an ultra-low power consumption magnetic random access memory (MRAM). One of the promising technologies is the voltage control of a magnetic anisotropy (VCMA) in ferromagnetic metals. Since the dynamic switching of magnetization was demonstrated by utilizing the VCMA effect<sup>1,2</sup> in MgO-based magnetic tunnel junctions (MTJ), it has been intensively investigated. It is called “voltage torque MRAM”. VCMA value of as large as  $\sim 300$  fJ/Vm was already reported<sup>3</sup>. Thus it is expected to realize voltage torque MRAM in the not-too-distant future.

The other candidate is that using magnetoelectric (ME) antiferromagnet Cr<sub>2</sub>O<sub>3</sub>. When we apply parallel (anti-parallel) electric ( $E$ ) and magnetic field ( $H$ ), the antiferromagnetic domain of Cr<sub>2</sub>O<sub>3</sub> align as  $\uparrow\downarrow\uparrow\downarrow$  ( $\downarrow\uparrow\downarrow\uparrow$ ). If we apply fixed positive  $H$ , by changing the direction of  $E$  from positive to negative, we can switch the direction of surface spin of Cr<sub>2</sub>O<sub>3</sub> from up ( $\uparrow$ ) to down ( $\downarrow$ ). In addition, the surface spin information can be transferred to neighbor ferromagnet via exchange coupling. Thus by combining both the ME effect and exchange coupling, we can achieve electric control of magnetization. The concept of so called “magnetoelectric random access memory (MERAM)” was proposed in 2006<sup>4</sup>. After that, its primitive operations were demonstrated for Cr<sub>2</sub>O<sub>3</sub> bulk<sup>5,6</sup> and thin film<sup>7,8</sup> systems. Recently the reduction of the large switching energy was proposed<sup>9</sup>. In additions, ferromagnet free purely antiferromagnetic random access memory was demonstrated<sup>10</sup>. These reports make the MERAM more realistic and more interesting. Although Cr<sub>2</sub>O<sub>3</sub> is an antiferromagnet, sometimes finite magnetization has been observed for Cr<sub>2</sub>O<sub>3</sub> films. We found relative large parasitic magnetic moment, which comparable to the ferrimagnet, for doped Cr<sub>2</sub>O<sub>3</sub>. In this study, we investigated the parasitic magnetic moments of Al- and Ir-doped Cr<sub>2</sub>O<sub>3</sub> film and discussed the usability.

## II. EXPERIMENTAL PROCEDURES

The non-dope, Al-doped, and Ir-doped Cr<sub>2</sub>O<sub>3</sub> films are fabricated by RF reactive sputtering method. The Al and Ir contents and lattice parameters of the films were confirmed by X-ray fluorescence (XRF) and X-ray diffraction measurements, respectively. Magnetic and magnetoelectric properties were measured by using a superconducting quantum interference device (SQUID) magnetometer or anomalous Hall effect (AHE). The detail of the magnetoelectric properties measurements are described in<sup>11</sup>. Uncompensated surface spin of Cr<sub>2</sub>O<sub>3</sub> film was measured by X-ray magnetic circular dichroism (XMCD) spectroscopy. The XMCD measurements were carried out at beam line BL25SU of the SPring-8 synchrotron radiation facility.

## III. RESULTS AND DISCUSSIONS

By Al- and Ir-doping, relative large volume magnetization were obtained. The Cr<sub>2</sub>O<sub>3</sub> volume magnetization increase with increasing both Al- and Ir-contents; by about 3.7% Al-(Ir-) dope, volume magnetization of as large as 59 (4.9) emu/cc were obtained, which correspond to 0.61 (0.05)  $\mu_B$ /Cr. Interestingly, the doped Cr<sub>2</sub>O<sub>3</sub> film still exhibit ME properties, and the parasitic magnetization is coupled with the ME order parameter of Cr<sub>2</sub>O<sub>3</sub>. Fig. 1 shows the ME coefficient  $\alpha$  of (a) Al-doped and (b) Ir-doped Cr<sub>2</sub>O<sub>3</sub> film against  $H$  at 170 K. With increasing applied  $H$ , the parasitic magnetization reverse and simultaneously the ME order parameter also reverse. Fig. 1 indicate that in Al-doped sample case, Cr<sub>2</sub>O<sub>3</sub> parasitic magnetization is coupled with F<sup>+</sup> state ( $\uparrow\downarrow\uparrow\downarrow$ ), while in Ir-doped sample case, Cr<sub>2</sub>O<sub>3</sub> parasitic magnetization is coupled with F<sup>-</sup> state ( $\downarrow\uparrow\downarrow\uparrow$ ). These fact were confirmed by combined study of

Tomohiro NOZAKI

E-mail: nozaki@ecei.tohoku.ac.jp

tel: +81-22-795-7067

magnetization measurements, measurements of  $\text{Cr}_2\text{O}_3$  surface spin by XMCD, and ME coefficient measurements. These results suggest the parasitic magnetic moment can be controlled by magnetic and electric fields through the ME effect. That is “magnetoelectric ferrimagnet”. XRD results indicate both  $a$  and  $c$  value expansion for Ir-doped sample, and  $a$  and  $c$  value compression for Al-doped case. These different kind of lattice strain may related to the difference in  $\text{Cr}_2\text{O}_3$  volume magnetization direction, since similar magnetization direction change was also observed for  $\text{Cr}_2\text{O}_3$  films with various buffer layers<sup>10</sup>. In this study, we clarified the co-existence of ME and ferrimagnetic properties in doped  $\text{Cr}_2\text{O}_3$  films. Such an electrically controllable magnetization have a great potential for developing new electric field controlled memory concepts, in additions to the utilization of reduction of ME switching energy<sup>9</sup>.

#### ACKNOWLEDGEMENTS

This work was partly funded by ImPACT Program of Council for Science, Technology and Innovation (Cabinet Office, Japan Government) and JSPS KAKENHI Grant Number 16H05975.

#### REFERENCES

- 1) Y. Shiota et al., “Induction of coherent magnetization switching in a few atomic layers of FeCo using voltage pulses”, *Nat. Mater.*, 11, 39 (2012).
- 2) S. Kanai et al., “Electric field-induced magnetization reversal in a perpendicular-anisotropy CoFeB-MgO magnetic tunnel junction”, *Appl. Phys. Lett.*, 101, 122403 (2012).
- 3) T. Nozaki et al., “Large Voltage-Induced Change in the Perpendicular Magnetic Anisotropy of an MgO-Based Tunnel Junction with an Ultrathin Fe Layer” *Phys. Rev. Appl.*, 5, 044006 (2016).
- 4) X. Chen et al., “Magnetoelectric exchange bias systems in spintronics”, *Appl. Phys. Lett.*, 89, 202508 (2006).
- 5) P. Borisov et al., “Magnetoelectric Switching of Exchange Bias”, *Phys. Rev. Lett.*, 94, 117203 (2005).
- 6) X. He et al., “Robust isothermal electric control of exchange bias at room temperature”, *Nat. Mater.*, 9, 579 (2010).
- 7) T. Ashida et al., “Observation of magnetoelectric effect in  $\text{Cr}_2\text{O}_3/\text{Pt}/\text{Co}$  thin film system”, *Appl. Phys. Lett.*, 104, 152409 (2014).
- 8) T. Ashida et al., “Isothermal electric switching of magnetization in  $\text{Cr}_2\text{O}_3/\text{Co}$  thin film system”, *Appl. Phys. Lett.*, 106, 132407 (2015).
- 9) M. Al-Mahdawi et al., “Low-energy magnetoelectric control of domain states in exchange-coupled heterostructures”, *Phys. Rev. B*, accepted.
- 10) T. Kosub et al., “Purely antiferromagnetic magnetoelectric random access memory”, *Nat. Commun.*, 8, 13985 (2017).
- 11) M. Al-Mahdawi et al., “Apparent critical behavior of sputter-deposited magnetoelectric antiferromagnetic  $\text{Cr}_2\text{O}_3$  films near Neel temperature”, *J. Phys. D: Appl. Phys.*, 50, 155004 (2017).

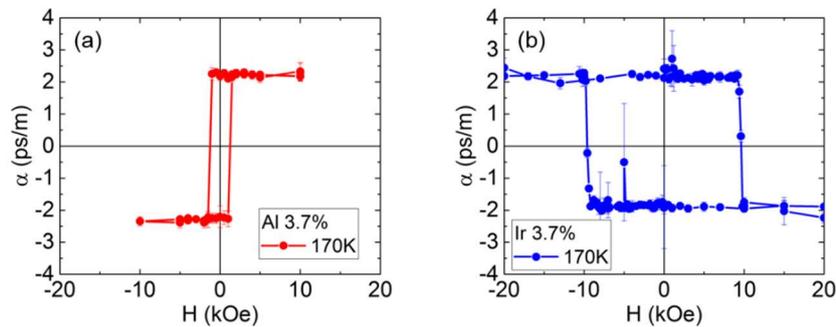


Fig. 1  $H$  dependence of ME coefficient  $\alpha$  of (a) Al-doped and (b) Ir doped samples measured at 170K.