# Relationship between magneto-capacitance-voltage characteristics and magnetoresistance of $Au/Cr_2O_3/Cr_2O_{3-x}/FeCr/CeO_2/Si$ metal-insulator-semiconductor capacitor

T. Yokota,<sup>a)</sup> S. Murata, S. Kito, and M. Gomi

Materials Science and Engineering, Nagoya Institute of Technology, Gokiso-cho, Showa-ku, Nagoya City, Aichi 466-8555, Japan

(Presented 11 November 2008; received 17 September 2008; accepted 21 October 2008; published online 4 February 2009)

We investigated the relationship between the magneto-capacitance-voltage characteristics and the magnetoresistance of a metal (Au)/insulator  $(Cr_2O_3/Cr_2O_{3-x}/FeCr/CeO_2)$ /semiconductor (Si) capacitor, in which the insulator consists of magnetic materials. By applying an electric field, electrons propagating through the FeCr/CeO<sub>2</sub> layer from Si were injected into the  $Cr_2O_{3-x}$  layer. When a magnetic field was applied, the resistance of this capacitor above the flat-band voltage was reduced, causing the hysteresis window to become large. This result indicates that this capacitor, which contains a magnetic gate insulator, has the potential to be used in multilevel memories by the application of an external magnetic field. © 2009 American Institute of Physics. [DOI: 10.1063/1.3059406]

## **I. INTRODUCTION**

Information-processing devices that use semiconductor random access memories generally operate using electric charge, without utilizing electric spin. In these devices, however, spin becomes important for dimensions smaller than about 100 nm. Consequently, a new information technology that utilizes spin, known as spintronics, has begun to be developed. In order to make efficient use of electric spin in semiconductor information-processing devices, we have proposed a metal-insulator-semiconductor (MIS) capacitor that contains a magnetic insulator and a floating gate.<sup>1</sup> The magnetic insulating layer consists of an antiferromagnetic thin film  $(Cr_2O_3)$ , a ferromagnetic film (FeCr), and a paramagnetic film (CeO<sub>2</sub>). Cr<sub>2</sub>O<sub>3</sub> is also a magnetoelectric (ME) material, which means that its magnetic and ferroelectric properties can be controlled by the application of an external electric and magnetic field, respectively.<sup>2,3</sup> Binek et al. reported the possibility of creating spintronic devices by turning on a magnetic interaction by using the ME effects of  $Cr_2O_3$ .<sup>4-6</sup> Thus, a Si-MIS capacitor that contains a  $Cr_2O_3$ film could be used as a spintronic device with semiconductor characteristics. Previously, we reported the carrier injection process and the memory effects of this capacitor.' However, the effect of the magnetic properties on this carrier injection process is unclear. Hence, in this present study, we investigate the relationship between the magneto-capacitancevoltage characteristics and the magnetoresistance (MR) of the Au/Cr<sub>2</sub>O<sub>3</sub>/Cr<sub>2</sub>O<sub>3-x</sub>/FeCr/CeO<sub>2</sub>/Si MIS capacitor.

# **II. EXPERIMENTAL PROCEDURE**

The sample was prepared using the three-gun radiofrequency magnetron sputtering method.  $Cr_2O_3$  and  $CeO_2$  sintered ceramics were used as the target. The base pressure before introducing the sputtering gas was  $3.0 \times 10^{-4}$  Pa and the gas pressure during deposition was  $8.0 \times 10^{-1}$  Pa. The first layer was a 5-nm-thick CeO<sub>2</sub> tunnel layer and it was deposited on a *n*-type Si (111) substrate cleaned using improved Radio Corporation America methods. A 0.25-nm-thick FeCr layer was then deposited using only Ar as the sputtering gas. In order to prevent this FeCr layer from oxidizing, a 1-nm-thick  $Cr_2O_{3-x}$  layer was deposited, also by using only Ar as the sputtering gas; this layer also functions as a floating layer. Finally, a 45-nm-thick Cr<sub>2</sub>O<sub>3</sub> layer was deposited. The magnetic properties of the sample were measured using a superconducting quantum interference device. Structural analysis of the films was performed by x-ray diffraction (XRD) utilizing Cu  $K\alpha$  radiation and reflected highenergy electron diffraction. The surface morphology was measured by atomic force microscopy (AFM). The leakage current was measured using a picoammeter (Keithley, 6487), while the capacitance was measured using an LCR meter (Agilent, E6480A). The electric properties at low temperature were measured using a cryostat system (Nagase, PS25SR). The top and bottom electrodes consisted of Au and were 0.7 mm in diameter. To avoid deviations in the measurements of the electric properties, they were performed using about 40 electrodes, which were constructed on the sample.

## **III. RESULTS AND DISCUSSION**

Figure 1(a) shows an XRD pattern, which reveals that the film consists of polycrystalline  $Cr_2O_3$  and highly oriented  $CeO_2$ . Figure 1(b) shows an AFM image of the surface morphology and the surface profile along the dotted line in this image; they reveal that the sample surface is smooth. The sample has a root-mean-square roughness of approximately 3.5 nm. Figure 2 shows the capacitance-voltage (*C-V*) char-

105, 07D905-1

<sup>&</sup>lt;sup>a)</sup>Author to whom correspondence should be addressed. Electronic mail: yokota.takeshi@nitech.ac.jp.



FIG. 1. (a) XRD pattern and (b) AFM image showing the surface morphology of the  $Cr_2O_3/Cr_2O_{3-x}/FeCr/CeO_2/Si$  capacitor and the surface profile along the dotted line in the AFM image.

acteristics measured at 250 K. During the C-V measurements, the sample was irradiated with a laser beam having a wavelength of 532 nm to activate the carriers. This measurement was also performed under a magnetic field of 0.1 T. The C-V curves exhibit hysteresis with a clockwise trace, indicating that electrons have been injected into the  $Cr_2O_{3-r}$ layer. When a magnetic field of 0.1 T is applied, only the increasing capacitance trace from -1 to 1 V, which corresponds to the carrier injection process, is shifted by about 0.01 V. Figure 3 shows the retention properties of this capacitor. The capacitance, both with and without the application of a magnetic field, did not change within the measurement period of 3 h. This implies that the change in capacitance produced by the application of a magnetic field is an intrinsic property of this capacitor. In order to investigate the effect of a magnetic field, the MR was measured at various magnetic fields. The measurement direction of the MR was the same as that for the C-V measurement, which means that a current indicates carrier injection into the  $Cr_2O_{3-x}$  floating layer. It is also important to take the measurement voltages into account, because carrier injection occurs above the flat-band voltage. According to the depletion layer approximation, the experimental flat-band voltage of this capacitor is about  $0.1 \text{ V}^{.8}$  Figure 4(a) shows the MR measured for applied voltages of 0.15, 0.3, and 1.0 V. For comparison, the hysteresis window widths (HWWs) of the



FIG. 2. *C-V* characteristics measured at 250 K. The solid and dashed lines indicate the sample with/without the application of a magnetic field of 0.1 T, respectively.



FIG. 3. Retention properties of the sample. Carriers were injected by applying a voltage of 3 V with/without a magnetic field. The capacitance was then measured at the flat-band voltage.

C-V curves as a function of the magnetic field are plotted in the same graph. The magnetization curve of the sample measured at 250 K is also shown in Fig. 4(b). The resistance increases up to about 7 mT as a result of spin scattering due to the magnetic moments aligning with the magnetic field. This magnetic field corresponds to the coercive field of FeCr indicated in the magnetization curve. Above 8 mT, the resistance decreases with an increase in the magnetic field. The resistances are almost saturated above 20 mT. With the exception of the MR measured at 1 V, this behavior is similar to other measured voltages. Since the charge injection process is almost complete above 1 V, hardly any current flows in the capacitor after carrier injection into the floating gate has finished. Therefore, the resistance due to the magnetic field did not change. On the other hand, the MR behaviors for 0.15 and 0.3 V correspond to the magnetization process of FeCr. At the same applied magnetic fields,  $\Delta$ HWW also increased. These results indicate that the charge injection process of this capacitor can be controlled by the magnetization of the FeCr layer and by the amount of the injected carriers, which correspond to the HWW, which in turn can be controlled by the applied external magnetic field. Finally, the influence of the magnetic state of this capacitor was investigated using the exchange bias effect. As mentioned above, our capacitor contains an antiferromagnetic layer and a ferromagnetic layer. In this interface, we can expect a magnetic interaction, the so-called exchange interaction.<sup>9</sup> Therefore, the sample was subjected to magnetic field cooling (FC) of



FIG. 4. (a) Relationship between the MR measured at 0.15, 0.3, and 1.0 V and  $\Delta$ HWW by the application of a magnetic field. (b) Magnetization curves of the sample measured at 250 K.

Downloaded 25 Aug 2010 to 133.68.192.95. Redistribution subject to AIP license or copyright; see http://jap.aip.org/about/rights\_and\_permissions



FIG. 5. (a) Magnetization curves of the samples with (solid line)/without (broken line) FC. (b) Relationship between the MR measured at 0.15 V and  $\Delta$ HWW by the application of a magnetic field after FC.

0.8 T from 350 K. Figure 5(a) shows magnetization curves of the samples obtained with and without FC. The magnetization curves were shifted slightly, approximately 1.5 mT, by the FC treatment. This implies that the ferromagnetic FeCr layer was pinned by the antiferromagnetic Cr<sub>2</sub>O<sub>3</sub> layer. Figure 5(b) shows the MR measured at 0.15 V. The  $\Delta$ HWW in the of C-V curve as a function of the external magnetic field after FC is also plotted on the same graph. Both the MR and  $\Delta$ HWW do not change much compared with the sample that was not subjected to FC by the application of a magnetic field. In particular, the values of  $\Delta$ HWW are almost the same as those when a magnetic field is applied (see Fig. 4). These results support the charge injection mechanism under an applied magnetic field. After FC, the magnetic moment of FeCr was fixed parallel to the magnetic field applied during FC. Consequently, the MR effect became small. In addition, the amount of electrons was unaffected by the application of a magnetic field. Eventually, no changes in the HWW in the C-V curve under an applied magnetic field were observed.

#### **IV. CONCLUSIONS**

We investigated the relationship between the MR and C-V characteristics under an applied magnetic field. On applying a magnetic field, the HWW in the C-V curve increased. A negative magnetoresistance was observed in the same magnetic field range. The increase in the HWW is most likely due to the increase in the injection carrier caused by the reduction in the resistance that occurs when a magnetic field is applied.

### ACKNOWLEDGMENTS

This research was supported in part by a Grant-in-Aid for Young Scientists (B) (Grant No. 20760197) from the Japan Society for the Promotion of Science

- <sup>1</sup>T. Yokota, T. Kuribayashi, M. Gomi, T. Shundo, and Y. Sakakibara, Adv. Mater. Res. **11-12**, 133 (2006).
- <sup>2</sup>T. Lottermoser, T. Lonkai, U. Amann, D. Hohlwein, J. Ihringer, and M. Fiebig, Nature (London) 430, 541 (2004).
- <sup>3</sup>E. Kita, A. Tasaki, and K. Siratori, Jpn. J. Appl. Phys. 18, 1361 (1979).

<sup>4</sup>Ch. Binek, P. Borisov, X. Chen, A. Hochstrat, S. Sahoo, and W. Kleemann, Eur. Phys. J. B 45, 197 (2005).

<sup>5</sup>P. Borisov, A. Hochstrat, X. Chen, W. Kleemann, and Ch. Binek, Phys. Rev. Lett. **94**, 117203 (2005).

<sup>6</sup>Ch. Binek, A. Hochstrat, X. Chen, P. Borisov, W. Kleemann, and B. Doudin. J. Appl. Phys. **97**, 10C514 (2005).

<sup>7</sup>T. Yokota, T. Kuribayashi, and M. Gomi, J. Ceram. Soc. Jpn. **116**, 1204 (2008).

<sup>8</sup>S. M. Sze, *Semiconductor Physics*, 2nd ed., (Wiley InterScience, 1981 pp. 496–500.

<sup>9</sup>W. H. Meiklejohn and C. P. Bean, Phys. Rev. **102**, 1413 (1956); Phys. Rev. **105**, 904 (1957).