Voltage control of interfacial antiferromagnetic spins based on magnetoelectric effect

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Efficient manipulation of antiferromagnetic (AFM) state is desirable for advanced spintronic devices with fast operation and robustness against the magnetic field perturbation. Using the magnetoelectric Cr_2O_3 epitaxial film, we demonstrate the giant voltage modulation of the antiferromagnetic spin reversal. We obtained the significant modulation efficiency of the switching field, $\Delta\mu_0 H_{SW}/\Delta V$ ($\Delta\mu_0 H_{SW}/\Delta E$), −500 mT/V (−4.80 T·nm/V) in maximum. We also found that the stable spin direction was determined depending on the sign of the electric and magnetic field product, yielding the checkerboard-like assignment of the spin state in the *E*-*H* plane. Our findings not only advance the voltage-based insulating AFM spintronics but also provide the basis of the device function such as a deterministic logic operation by means of the field combination.

***Index Terms*—Antiferromagnetic materials, Hall effect, Magnetoelectric (ME) effect**

I. Introduction

Voltage control of magnetism has attracted much attention as the energy efficient spin manipulation principle as well as a benefit from CMOS compatibility. However, since the voltage (the electric field *E*) is not directly coupled with the magnetic moment, the elaborate method is required to couple them. So far, two distinct physical phenomena underlying this coupling have been attempted: Voltage-controlled magnetic anisotropy (VCMA) effect [1] and magnetoelectric (ME) effect [2]. The VCMA effect is defined as a change in the magnetic anisotropy energy density by *E*, and it typically occurs at a metallic magnetic layer/nonmagnetic insulator interface. Since the VCMA effect reduces the energy barrier and does not change the symmetry of the energy landscape of magnetic anisotropy, the finally obtained magnetization state is not deterministic [3].

The ME effect is an alternative principle which couples the magnetic moment and the voltage (or *E*). The ME effect is defined as the induction of the net magnetization *M* by *E* or the induction of the dielectric polarization *P* by the magnetic field *H*, expressed as

$$M_i = \alpha_{ij} E_j \,,\; P_i = \alpha'_{ij} H_j \qquad (1)$$

where α_{ij} and α'_{ij} are ME coefficient (*i, j = x, y, z*). The non-zero α_{ij}, i.e., the emergence of the finite ME effect is involved by the crystal structure with the broken spatial inversion symmetry. The *E*-induced magnetization via eq. (1) can obtain the Zeeman energy gain as in the ordinal ferromagnetic (FM) materials. Therefore, the combination of *E* and *H* in the ME materials switches the spin state in a deterministic way.

Another significance to use the ME effect is that some ME materials exhibit the antiferromagnetic (AFM) order. Although no net magnetization in the AFM materials hinders controlling its spin state by conventional techniques, the ME-based mechanism would be a solution. Using Cr_2O_3, prototypical ME-AFM material, we have been investigating the ME-based spin manipulation [4,5]. In this digest, we summarize our progress on the ME-based control of AFM spin state.

II. Detection of Interfacial AFM spins

As described above, the ME feature involves the specific crystal structure with the broken special inversion symmetry. At the surface/interface, the structural inversion symmetry is inherently broken. The symmetry-arguments [6] predict the emergence of the sizable spin density at the surface in conjunction with the ME effect, called the surface magnetization or the boundary magnetization coupled with the bulk AFM order parameter (the Néel vector). By utilizing the interface-sensitive principle, we can detect the surface magnetization, consequently the orientation of the Néel vector. We employed spin-dependent transport at the nonmagnetic heavy metal/magnetic insulator interface such as $Pt(111)/Cr_2O_3(0001)$. Since the electric conductivity is relied on the Pt layer, we can access the interfacial AFM (Cr) spin state separated from the bulk site.

Figure 1 shows the R_{xy} as a function of magnetic field for $Pt(111)/Cr_2O_3(0001)/Pt(111)$ film with the 8-nm thick Cr_2O_3. [4] According to the magnetocrystalline anisotropy, the magnetic easy axis should be perpendicular to the film due to the magnetocrystalline anisotropy; The field direction was normal to the film plane, i.e. along the *c*-axis of Cr_2O_3. The R_{xy}-*H* curve shows the rectangular hysteresis below 250 K, resembling the magnetization curve along the easy axis. The emergence of hysteresis is due to the enhanced contribution of the surface magnetization by reducing the Cr_2O_3 thickness. The hysteresis in the R_{xy}-*H* curve becomes absent for the Cr_2O_3 film thicker than 40 nm wherein the bulk AFM nature is dominant and consequently, the energy gain by the Zeeman effect is insufficient to switch the surface magnetization. We confirmed that the interfacial Cr spin reversal occurred using the soft X-ray magnetic circular dichroism. Notably, no appreciable signal was detected by the conventional magnetization measurements, which supports that Cr_2O_3 maintains the antiferromagnetic nature in whole film for all studied films.

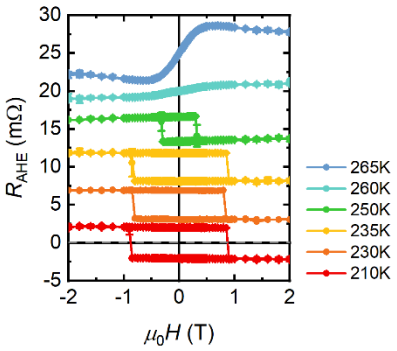

Fig.1 Series of R_{xy}-H curves measured at various temperatures [4].

III. Voltage-control of AFM spin reversal field

Advancing the results shown in Section II., we measured the R_{xy}-H curves upon applying the gate voltage V_G. Figure 2(a) shows the R_{xy}-H curves with $V_G = 0$ and ±0.3 V for the film with 8-nm thick Cr_2O_3 [5]. The switching field H_{SW} is modulated by V_G; the $\mu_0 H_{SW}$ value was changed from 680 mT at $V_G = -0.3$ V to 550 mT at $V_G = +0.3$ V. As plotted in Fig. 2(b), the switching field monotonically decreases with increasing V_G. The linear fitting gives the modulation efficiency $\Delta\mu_0 H_{SW}/\Delta V_G$ ($\Delta\mu_0 H_{SW}/\Delta E$) as −180 mT/V (−1.4 T·nm/V). The temperature dependence of the modulation efficiency revealed that the highest efficiency reached −500 mT/V (−4.8 T·nm/V) which is about 50 times larger than the FM counterpart based on the VCMA mechanism. Based on this feature, the V_G-induced spin reversal under the constant magnetic field was also possible. In this switching protocol, the leakage current (the current density) at the switching condition about 1.2 nA (1.5 A/m^2) was achieved, about 9 orders lower than the current based mechanism such as the spin-orbit torque.

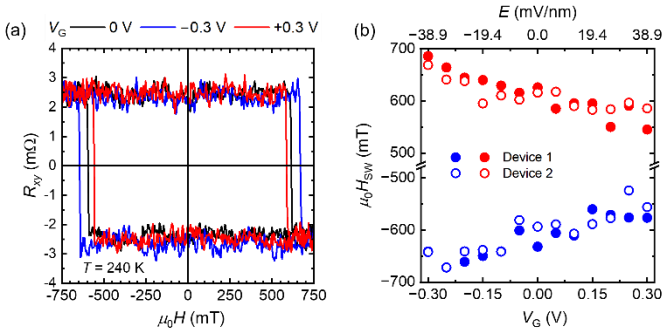

Fig. 2 (a) R_{xy}-H curve under V_G and (b) V_G dependence of switching field [5].

At $V_G = 0$, the thick Cr_2O_3 film did not show hysteresis. Since the Zeeman energy gain is proportional to EH, the thick Cr_2O_3 film would show similar hysteresis under the finite V_G. Figure 3(a) shows the R_{xy}-H curves at $V_G = +4.0$ V. Despite the absence of hysteresis at $V_G = 0$ V (black curve), the rectangular hysteresis emerged by the V_G application. Interestingly, when the negative V_G was applied (blue curve), the polarity of hysteresis is reversed. This polarity change is relevant to the difference in magnetic moments that couples with the magnetic field and that are responsible for the AHE. As long as the driving force of the spin reversal is Zeeman energy, the magnetic field responsiveness relies on the net magnetization. Thus, net magnetization defined by eq. (1) becomes parallel to the magnetic field in the saturated state. In contrast, AHE is relevant to surface magnetization coupled with the AFM order parameter and consequently, α. Therefore, the surface magnetization can be opposite to the magnetic field direction depending on E and H. In Fig. 3(c), the sign of R_{xy} was mapped in the E-H plane. Symbols represent the switching field at each E. The sign of R_{xy} in the saturated region is assigned in each four quadrants in a checkerboard manner. Each region is separated by two sets of hyperbolas with the positive/negative α. The inner region is the bi-stable regime corresponding to the inside of hysteresis shown in Figs. 1 and 2. In the functional viewpoints, when the map is intercepted along the horizontal axis, the E-sweep with the constant H would also switch the spin state, which would make this feature more fascinating, and it will be tackled in near future.

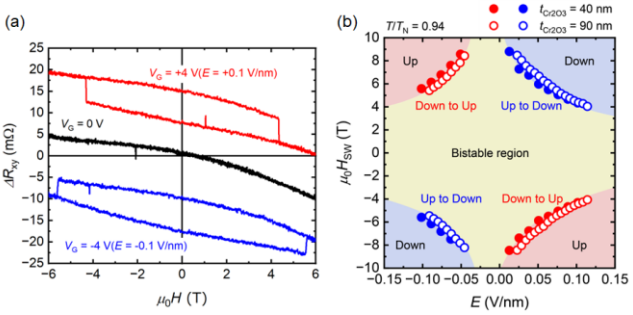

Fig. 3 (a) R_{xy}-H curve under $V_G = 0, \pm 4$ V, and (b) Map of spin state in E-H plane [5].

IV. Conclusions

We demonstrated voltage-based AFM spin manipulation by taking advantage of the surface magnetization relevant to the ME effect. The ME driven mechanism yields the quite high modulation efficiency exceeding hundred mT/V (a few T·nm/V). Besides, utilizing the sign change of ME coefficient, the novel functionality, e.g., the four quadrants assignment of spin state in E-H plane was demonstrated. The demonstration shown here will open a avenue on the voltage-driven AFM spintronics.

References

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