# Anomalous Hall effect with giant hysteresis loop in La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub>|SrRuO<sub>3</sub> superlattices

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We report anomalous Hall effects exhibiting a hysteresis loop as large as about 10 T in a ferromagnetic superlattice comprising  $La_{0.67}Sr_{0.33}MnO_3$  and  $SrRuO_3$  layers. The superlattices grown by pulsed laser deposition exhibit a strong antiferromagnetic interlayer coupling below 110 K, where both  $La_{0.67}Sr_{0.33}MnO_3$  and  $SrRuO_3$  layers show anomalous Hall effects. With increasing magnetic-field strength, the anomalous Hall resistivity in the superlattices changes its sign depending on the magnetization directions of the  $La_{0.67}Sr_{0.33}MnO_3$  and  $SrRuO_3$  layers. As a consequence of competition among the antiferromagnetic interlayer coupling, the Zeeman effect, and magnetic anisotropies, the width of the hysteresis loop in the anomalous Hall resistivity in the superlattices becomes larger than 8 T at 10 K, clearly greater than those observed in  $La_{0.67}Sr_{0.33}MnO_3$  and  $SrRuO_3$  single layer films.

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#### I. INTRODUCTION

Anomalous Hall effects are one of the fundamental and typical spin-dependent transport phenomena in ferromagnets [1]. While normal Hall effects are caused by Lorentz forces, the origin of anomalous Hall effects is the spin-orbit interaction, which connects spin polarizations with orbital motions of conduction electrons. A full understanding of the physical mechanism of anomalous Hall effects has long been challenging, and many studies have been conducted for various ferromagnetic conductors [1].

Hall effects are characterized by the Hall resistivity,  $\rho_{yx} = (V_H/I)t$ , where  $V_H$  is the Hall voltage generating along the y axis, I is the electric current applied along the x axis, and t is the sample thickness [see Fig. 1(a)]. The Hall resistivity in ferromagnetic conductors consists of normal and anomalous Hall resistivities. Namely,  $\rho_{yx}$  is described by the relation

$$\rho_{yx} = R_0 \mu_0 H + R_s M, \tag{1}$$

with *H* and *M* being the magnetic field and the magnetization along the direction perpendicular to the sample (*z* axis), respectively.  $R_0$  is the normal Hall coefficient, while  $R_s$  is the anomalous Hall coefficient. Since the magnitude of the anomalous Hall resistivity ( $R_s M$ ) is usually greater than that of the normal Hall one ( $R_0 \mu_0 H$ ) in a low-*H* range,  $\rho_{yx}$  in ferromagnets is usually proportional to the magnetization.

In the present paper, we have investigated the anomalous Hall effect in a ferromagnetic superlattice comprising La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> and SrRuO<sub>3</sub> layers. La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> and SrRuO<sub>3</sub> are ferromagnetic metals, but their magnetic properties are different; bulk La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> is a soft ferromagnet with a high  $T_C \approx 350$  K and a moment of  $\sim 3.5 \mu_B/Mn$ , while bulk SrRuO<sub>3</sub> is a hard ferromagnet with a low  $T_C \approx 150$  K and  $\sim 1.5 \mu_B/Ru$ . Although the anomalous Hall effects in La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> and SrRuO<sub>3</sub> have been investigated well so far [2–12], the anomalous Hall effect in hybrid structures made of the two ferromagnetic metals is an intriguing issue.

Importantly, magnetizations of the  $La_{0.67}Sr_{0.33}MnO_3$  and  $SrRuO_3$  layers are known to be coupled antiferromag-

netically to each other [13-20], which should affect the anomalous Hall effect in La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub>|SrRuO<sub>3</sub> multilayer systems. The interfacial antiferromagnetic coupling between La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> and SrRuO<sub>3</sub> layers [13-21] has been attributed to the hybridization of 2p states of O atoms with 3d states of Mn atoms and 4d states of Ru atoms [13,14]. The antiferromagnetic interlayer coupling associates the magnetization process of La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> layers with that of SrRuO<sub>3</sub> layers, and thus will affect the anomalous Hall effect in La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub>|SrRuO<sub>3</sub> superlattices. In fact, here we observe a very large hysteresis loop of  $\sim 10$  T in the Hall resistivity in La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub>|SrRuO<sub>3</sub> superlattice samples at 10 K, which is produced by magnetic competition among the antiferromagnetic interlayer coupling, the Zeeman effect driven by an external magnetic field, and the magnetic anisotropies of SrRuO<sub>3</sub> layers developing at low temperatures.

#### **II. METHODS**

La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub>|SrRuO<sub>3</sub> superlattice samples were grown by pulsed laser deposition using a KrF excimer laser on SrTiO<sub>3</sub> (001) substrates at 800 °C in 0.3 Torr O<sub>2</sub> [22]. After deposition, samples were annealed at 800 °C in the 400 Torr O<sub>2</sub> atmosphere for 1 h and then cooled to room temperature. The fabricated superlattices consist of ten periods of a La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> layer and a SrRuO<sub>3</sub> layer. The superlattice structure was confirmed by transmission electron microscopy (TEM), as shown in Figs. 1(b) and 1(c). From the TEM measurements, the thicknesses of the La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> and SrRuO<sub>3</sub> layers are estimated to be 2.4 and 3.5 nm, respectively [Fig. 1(c)]. The total thickness of the superlattice sample is about 57 nm. The longitudinal resistivity, Hall resistivity, and magnetization for the superlattice samples were measured in a Physical Property Measurement System (Quantum Design, Inc.), where an external magnetic field was applied perpendicular to the superlattices (i.e., parallel to the z axis). For reference, the magnetization of a SrTiO<sub>3</sub> substrate was measured in the same experimental condition; the substrate



FIG. 1. (Color online) (a) A schematic illustration of the Hall effect measurement for a  $La_{0.67}Sr_{0.33}MnO_3|SrRuO_3$  superlattice sample. LSMO and SRO denote  $La_{0.67}Sr_{0.33}MnO_3$  and  $SrRuO_3$  layers, respectively. (b) An overview cross-sectional TEM image for a superlattice sample. A magnified view is shown in (c).

magnetization was subtracted from the magnetization data for superlattice samples.

## **III. RESULTS AND DISCUSSION**

Figure 2(a) shows the temperature (*T*) dependence of resistivity  $\rho_{xx}$  for a representative superlattice sample. In the entire *T* region below 300 K,  $\rho_{xx}$  shows a metallic *T* dependence  $(d\rho_{xx}/dT > 0)$ .  $\rho_{xx}$  at the lowest *T* is 0.4 m $\Omega$  cm, which is almost the same as that of similar superlattices [23]. The residual resistance ratio is larger than 2.

Magnetization *M* measured under  $\mu_0 H = 1$  T in a direction perpendicular to a superlattice sample is shown as a function of *T* in Fig. 2(b). Here, the diamagnetic contribution of the SrTiO<sub>3</sub> substrate was subtracted from the raw data. When *T* decreases, *M* increases rapidly below ~300 K, which is assigned to the ferromagnetic transition of La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub>



FIG. 2. (Color online) (a) Temperature (*T*) dependence of resistivity ( $\rho_{xx}$ ) for a La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub>|SrRuO<sub>3</sub> superlattice sample. (b) Temperature (*T*) dependence of magnetization (*M*) for a La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub>|SrRuO<sub>3</sub> superlattice sample. The magnetization was measured in a magnetic field of 1 T applied perpendicular to the film plane.



FIG. 3. (Color online) (a) Magnetic-field (*H*) dependence of Hall resistivity ( $\rho_{yx}$ ) for a La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub>|SrRuO<sub>3</sub> superlattice sample at some temperatures below 300 K. (b) Magnetic-field (*H*) dependence of magnetization (*M*) at 75 K. The dotted lines are just guides for the eyes. Here, the diamagnetic contribution of the SrTiO<sub>3</sub> substrate was subtracted from the raw data.

layers. *M* monotonically increases with decreasing *T* below 300 K, but below 110 K, where  $SrRuO_3$  layers exhibit a ferromagnetic transition, a clear decrease in *M* is observed. This is evidence that the magnetization for the  $SrRuO_3$  layers antiferromagnetically couples to that for the  $La_{0.67}Sr_{0.33}MnO_3$  layers.

Figure 3(a) shows the magnetic-field (*H*) dependence of the Hall resistivity ( $\rho_{yx}$ ) for a superlattice sample at various temperatures below 300 K. With decreasing *T* below 300 K, the magnitude of  $\rho_{yx}$  increases.  $\rho_{yx}$  shows a kink structure around ±1 T at 200 and 150 K, corresponding to the magnetization saturation field of the La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> layers. In this *T* range from 300 to 150 K, the SrRuO<sub>3</sub> layers are paramagnetic, and only the La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> layers show an anomalous Hall effect. As shown in Fig. 3(a), the sign of the anomalous Hall resistivity is negative in a positive magnetic field, which is consistent with that for La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> single layer films [23].

As T decreases below 110 K, the SrRuO<sub>3</sub> layers also show ferromagnetism. At 100 and 75 K, where both La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> and SrRuO<sub>3</sub> layers are ferromagnetic, a nonmonotonic H dependence of  $\rho_{yx}$  is observed in Fig. 3(a);  $\rho_{yx}$  shows the anomalous Hall effect of a negative sign in a low-H regime, while it becomes positive in a high-H range. This sign change around  $\pm 3$  T is caused by the anomalous Hall effect in the SrRuO<sub>3</sub> layers. The positive sign of anomalous Hall resistivity for the SrRuO<sub>3</sub> layers in positive magnetic fields is consistent with the anomalous Hall effect in very thin SrRuO<sub>3</sub> films grown on SrTiO<sub>3</sub> (001) substrates [11,12]. Hence, the complex H dependence of  $\rho_{yx}$  at 100 and 75 K is caused by two anomalous Hall contributions from the La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> and the SrRuO<sub>3</sub> layers, where the anomalous coefficients have different signs between the La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> and the SrRuO<sub>3</sub> layers.

The *H* dependence of  $\rho_{yx}$  observed at 100 and 75 K [Fig. 3(a)] can be explained by a sum of anomalous Hall contributions from La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> and SrRuO<sub>3</sub> layers,

$$\rho_{yx} = \alpha \left( R_0^{\text{LSMO}} \mu_0 H + R_s^{\text{LSMO}} M^{\text{LSMO}} \right) + \beta \left( R_0^{\text{SRO}} \mu_0 H + R_s^{\text{SRO}} M^{\text{SRO}} \right),$$
(2)

where  $R_0^{\text{LSMO}}$  ( $R_0^{\text{SRO}}$ ),  $R_s^{\text{LSMO}}$  ( $R_s^{\text{SRO}}$ ), and  $M^{\text{LSMO}}$  ( $M^{\text{SRO}}$ ) are normal and anomalous Hall coefficients and magnetization for La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> (SrRuO<sub>3</sub>) layers, respectively. Because of shunting of the electric current inside the superlattices, prefactors  $\alpha$  and  $\beta$  are required. In an equivalent circuit model [24],  $\alpha$  and  $\beta$  are expressed using resistivities and thicknesses of the La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> and SrRuO<sub>3</sub> layers:  $\alpha = \rho_{\text{SRO}}^2 t_{\text{LSMO}} t / (\rho_{\text{SRO}} t_{\text{LSMO}} + \rho_{\text{LSMO}} t_{\text{SRO}})^2$ and  $\beta = \rho_{\text{LSMO}}^2 t_{\text{SRO}} t / (\rho_{\text{SRO}} t_{\text{LSMO}} + \rho_{\text{LSMO}} t_{\text{SRO}})^2$ . Since resistivities in low temperature region below 100 K and thicknesses have similar magnitudes between the La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> and SrRuO<sub>3</sub> layers, respectively,  $\alpha \sim \beta$  in the present case.  $R_s^{\rm LSMO}$  and  $R_s^{\rm SRO}$  are constants of negative and positive signs, respectively;  $\rho_{yx}$  depends strongly on the magnetization directions of the La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> and SrRuO<sub>3</sub> layers.

To examine the magnetization processes of the La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> and SrRuO<sub>3</sub> layers under an external magnetic field applied perpendicular to the superlattice, we present in Fig. 3(b) the isothermal magnetization curve for a superlattice at 75 K. At high magnetic fields above  $\pm 4$  T, since the Zeeman energy overwhelms the energy of the antiferromagnetic interlayer coupling, the magnetizations of the La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> and the SrRuO<sub>3</sub> layers ferromagnetically align along the magnetic-field direction  $(M^{\text{LSMO}}||M^{\text{SRO}})$ . In a low-H range below  $\sim 2$  T, however, the strength of the antiferromagnetic coupling at the interface exceeds that of the Zeeman effect, and thus the magnetization of SrRuO<sub>3</sub> layers flips to the antiferromagnetic direction ( $M^{\text{SRO}} \rightarrow -M^{\text{SRO}}$ ), as illustrated in Fig. 3(b). This magnetization flip in the SrRuO<sub>3</sub> layers at  $\sim$ 3 T induces the sign reversal of the anomalous Hall contribution from the SrRuO<sub>3</sub> layers ( $R_s^{\text{SRO}}M^{\text{SRO}} \rightarrow -R_s^{\text{SRO}}M^{\text{SRO}}$ ). Since the magnitude of the anomalous Hall resistivity for the  $SrRuO_3$  layers is greater than that for the  $La_{0.67}Sr_{0.33}MnO_3$ layers ( $|R_s^{\text{SRO}}M^{\text{SRO}}| > |R_s^{\text{LSMO}}M^{\text{LSMO}}|$ ), the sign reversal of  $\rho_{yx}$  is observed around  $\pm 3$  T at 100 and 75 K, as shown in Fig. 3(a).

As T decreases from 75 to 50 K, the hysteresis loops observed in the Hall resistivity  $(\rho_{vx})$  and magnetization (M) curves become wider, as shown in Figs. 4(a) and 4(b). At 50 K, the magnetization process is similar to that at 75 K [compare Figs. 3(b) and 4(b)], and a sign change in  $\rho_{yx}$  around  $\pm 3$  T is also observed [Fig. 4(a)]. With a further decrease in T from 50 K, however, the overall H dependence of  $\rho_{yx}$ becomes quite different from that above 50 K. As shown in Fig. 4(c), the hysteresis loops in  $\rho_{yx}$  merge at 10 K, and one large hysteresis loop appears. The magnitude of the hysteresis loop is as large as 10 T, which is much larger than that observed in La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> and SrRuO<sub>3</sub> single-component systems [2-12]. Even if the strong demagnetization field for La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> layers, ~1 T obtained at 150 K and 200 K in Fig. 3(a), is taken into account, the magnitude of the hysteresis loop at 10 K is estimated to still be over 8 T.

At 10 K, as shown in Fig. 4(d), an inverted hysteresis is observed in the isothermal magnetization curve [25], which is not observed at high temperatures above 50 K [see Figs. 3(b) and 4(b)]. The inverted hysteresis observed at 10 K [Fig. 4(d)] means that the magnetization of the  $La_{0.67}Sr_{0.33}MnO_3$  layers points in the opposite direction to the external magnetic field



FIG. 4. (Color online) Magnetic-field (*H*) dependence of (a),(c) Hall resistivity ( $\rho_{yx}$ ) and (b),(d) magnetization (*M*) for a La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub>|SrRuO<sub>3</sub> superlattice sample at 50 K [(a) and (b)] and 10 K [(c) and (d)]. In (b) and (d), the diamagnetic contribution of the SrTiO<sub>3</sub> substrate was subtracted from the raw data. As for the magnetization data at 10 K shown in (d), see also Ref. [25].

 $(M^{\text{LSMO}} < 0 \text{ and } M^{\text{SRO}} > 0 \text{ in } H > 0)$  in a low-H range at 10 K. At high T's, the magnetization of the SrRuO<sub>3</sub> layers tends to flip in a low-H range ( $M^{\text{LSMO}} > 0$  and  $M^{\text{SRO}} < 0$ in H > 0), because the net positive magnetization ( $M^{\text{LSMO}}$  +  $M^{\text{SRO}} > 0$ ) is favorable for the energy gain of the Zeeman effect. By contrast, owing to an additional energy gain related to the perpendicular magnetic anisotropy of SrRuO<sub>3</sub> layers which develops at low T's [26], the inverted hysteresis loop is stabilized at 10 K [20]. The antiferromagnetically coupled magnetization of the  $La_{0.67}Sr_{0.33}MnO_3$  layers ( $M^{LSMO} < 0$ in H > 0) induces a positive anomalous Hall resistivity in a positive H; this sign is the same as that of anomalous Hall resistivity for the SrRuO<sub>3</sub> layers. Hence, multiple hysteresis loops merge and one large hysteresis loop appears in the Hall resistivity of the superlattice sample at 10 K, as shown in Fig. 4(c). Because of the strong antiferromagnetic coupling in our superlattice samples, the magnitude of the hysteresis loop becomes larger than 8 T; such a large hysteresis has not been achieved in other perovskite-oxide superlattices [27].

#### **IV. CONCLUSION**

In conclusion, we measured the anomalous Hall effect for  $La_{0.67}Sr_{0.33}MnO_3|SrRuO_3$  superlattices. The anomalous Hall effect in  $La_{0.67}Sr_{0.33}MnO_3$  layers appears below 300 K, while that in SrRuO\_3 layers appears below 100 K. The anomalous Hall effect in the superlattices is well explained by a sum of anomalous Hall resistivities of  $La_{0.67}Sr_{0.33}MnO_3$  and SrRuO\_3 layers, regardless of the antiferromagnetic coupling between the  $La_{0.67}Sr_{0.33}MnO_3$  and SrRuO\_3 layers below 110 K. Since the signs of the anomalous Hall coefficients are different between the  $La_{0.67}Sr_{0.33}MnO_3$  and SrRuO\_3 layers, regardless of the anomalous Hall coefficients are different between the  $La_{0.67}Sr_{0.33}MnO_3$  and SrRuO\_3 layers, regardless of the anomalous Hall coefficients are different between the  $La_{0.67}Sr_{0.33}MnO_3$  and SrRuO\_3 layers, regardless of the anomalous Hall coefficients are different between the  $La_{0.67}Sr_{0.33}MnO_3$  and SrRuO\_3 layers, regardless of the anomalous Hall coefficients are different between the  $La_{0.67}Sr_{0.33}MnO_3$  and SrRuO\_3 layers, regardless of the anomalous Hall coefficients are different between the  $La_{0.67}Sr_{0.33}MnO_3$  and SrRuO\_3 layers, regardless of the anomalous Hall coefficients are different between the  $La_{0.67}Sr_{0.33}MnO_3$  and SrRuO\_3 layers, regardless of the anomalous Hall coefficients are different between the  $La_{0.67}Sr_{0.33}MnO_3$  and SrRuO\_3 layers, regardless of the anomalous Hall coefficients are different between the  $La_{0.67}Sr_{0.33}MnO_3$  and SrRuO\_3 layers, regardless of the anomalous Sr\_0.33MnO\_3 and Sr\_0.33MnO\_3 and Sr\_0.33MnO\_3 and Sr\_0.33MnO\_3 and Sr\_0.33MnO\_3 anomalous Sr\_0.33MnO\_3 anomalous Sr\_0.33MnO\_3 anomalous

and since the magnetization directions of the La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> and SrRuO<sub>3</sub> layers are opposite to each other in a low-*H* range due to anitiferromagnetic interlayer coupling, the Hall resistivity in the La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub>|SrRuO<sub>3</sub> superlattices shows a nonmonotonic magnetic-field dependence. Especially at 10 K, multiple hysteresis loops merge to form one hysteresis loop that is wider than 8 T in the Hall resistivity. In many cases, hysteresis in anomalous Hall resistivity results from magnetic anisotropies in materials; for ferromagnetic materials with a large anisotropy, the anomalous Hall resistivity often shows a large hysteresis loop. In the present La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub>|SrRuO<sub>3</sub> superlattices, by contrast, the large hysteresis loop is produced by magnetic competition among the antiferromagnetic interlayer coupling, the Zeeman effect, and magnetic anisotropies.

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