Extraordinary Hall effect in $Ba_{1-x}Sr_xRuO_3$ films

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(Received 18 August 2010; revised manuscript received 20 October 2010; published 22 November 2010)

We have investigated the extraordinary Hall effect of $Ba_{1-x}Sr_xRuO_3$ thin films with $x=1$ and 0.8. For $x=1$, the extraordinary Hall resistivity can be interpreted by the skew scattering mechanism in the paramagnetic region above about 160 K. In the ferromagnetic region above about 60 K, the extraordinary Hall coefficient R_s can be expressed by the combination of the skew and the side-jump scattering mechanisms as $R_s=a\rho+b\rho^2$ using the electrical resistivity ρ, as usually observed in ordinary ferromagnetic metals. Below 60 K, the field dependence of the Hall resistivity ρ_H at intermediate field region cannot be expressed as $\rho_H=R_0B+R_sM$, suggesting an additional term in the field dependence of the extraordinary Hall resistivity. R_s below 60 K derived from the high field region does not follow the relation $R_s=a\rho+b\rho^2$, suggesting that the coefficients a and b of the skew and the side-jump scattering depends on temperature below 60 K. On the other hand in the sample with $x=0.8$, no anomalies were observed below 60 K both in temperature and field dependence. According to the structural analysis of the samples, we infer that the modulation of the Fermi surface due to the structural change plays an important roll on the anomaly of the temperature dependence of the extraordinary Hall effect for $x=1$.

DOI: 10.1103/PhysRevB.82.174430 PACS number(s): 73.50.Jt, 75.47.Lx, 61.05.cp

I. INTRODUCTION

The Hall effect in magnetic metals consists of two components; the ordinary Hall effect and the extraordinary one.[1] The former contains information on the Fermi surface such as the carrier concentration and the k dependence of the conduction electron scattering. The latter provides information on the left-right asymmetry of the scattering. The Hall resistivity ρ_H under an external field B is expresses by the sum of the ordinary Hall component R_0B and the extraordinary one R_sM as

$$\rho_H = R_0B + R_sM, \tag{1}$$

where R_0 and R_s are the ordinary and the extraordinary Hall coefficients, respectively, and M the magnetization. In ordinary ferromagnetic metals, the origin of the left-right asymmetric scattering responsible for the extraordinary Hall effect has been classified into two terms; the skew scattering proportional to the resistivity ρ and the side-jump scattering proportional to ρ^2.[1–3] Both the asymmetric scatterings are based on the spin-orbit interaction between the conduction-electron spin and the orbital of the scattering center such as the magnetic impurities. The extraordinary Hall coefficient R_s in ferromagnetic state is described as

$$R_s = a\rho + b\rho^2, \tag{2}$$

where the coefficients a and b are the characteristics of the asymmetric scatterings.[1–3] Contrary to the conventional theory based on the left-right asymmetric scatterings, Karplus and Luttinger have proposed the intrinsic mechanism of the extraordinary Hall effect, in which the interband transition of the conduction electron combined with the spin-orbit interaction induces a current perpendicular to both the electric field and the magnetization.[4]

Recently, It has been reported that the extraordinary Hall effect shows an anomaly, which cannot be explained by the left-right asymmetric scatterings in several strongly correlated electron systems such as ferromagnetic oxides.[5–7] In these reports, the origin of the extraordinary Hall effect has been attributed to the intrinsic mechanism arising from its electronic structure[8–11] based on the theory by Karplus and Luttinger.[4] In real materials, however, both R_0 and R_s are affected by the modulation of the Fermi surface arising from the change in the structure and the magnetic properties.[12–14] The experimental works asserting the intrinsic mechanism of the extraordinary Hall effect have ignored the possibility.

In order to reveal the anomaly in the extraordinary Hall effect, it is important to decompose the Hall resistivity into the ordinary and extraordinary Hall components carefully and to compare both R_0 and R_s with other properties.[12–14] In addition, the investigation of the crystalline structure is necessary since the modulation of the Fermi surface resulting from the structural change induces the change in the characteristics of the left-right asymmetric scattering responsible for the extraordinary Hall effect. We have investigated the Hall resistivity, the resistivity and the magnetization measured on the same sample of $Ba_{1-x}Sr_xRuO_3$ epitaxial films with $x=0.8$ and 1.0. Furthermore, we have investigated the crystalline structure at low temperatures on the same films using a synchrotron x-ray diffraction, in order to clarify the structural change associated with the modulation of the Fermi surface responsible for the anomalous temperature dependence of the extraordinary Hall effect in $Ba_{1-x}Sr_xRuO_3$. The preliminary results of the Hall effect measurements have been reported in Ref. 15.

II. SAMPLE PREPARATION AND EXPERIMENTAL PROCEDURE

The polycrystalline $Ba_{1-x}Sr_xRuO_3$ for the targets of the films were synthesized at 1300 °C by a solid-state reaction of predried $BaCO_3$, $SrCO_3$, and RuO_2 with purity of 99.9%. The epitaxial films of $Ba_{1-x}Sr_xRuO_3$ were grown on $SrTiO_3(001)$ substrates by pulsed laser deposition using targets of polycrystalline $Ba_{1-x}Sr_xRuO_3$ samples with about 30 mm diameter. The substrate temperature was about 800 °C and the oxygen partial pressure was about 350 mTorr during the deposition. The thickness of the films is 500 nm and 20 nm for $x=1$ and 0.8, respectively. The orientation and the crystallinity of the films were characterized using Philips X'pert multi-purpose x-ray diffractometer (MPD) x-ray diffractometer at room temperature. The structure of the samples was tetragonally distorted. The lattice constants are $a=b=3.89$ Å and $c=3.94$ Å, and $a=b=3.89$ Å and $c=3.97$ Å for $x=1$ and 0.8, respectively, where c axis is perpendicular to the film plane.

The magnetization measurements were performed with a Quantum Design superconducting quantum interference device magnetometer and also with a Quantum Design physical property measurement system (PPMS) up to 9 T. The Hall and the resistivity measurements were performed by a conventional dc four-probe method under a magnetic field up to 9 T. For all the measurements, the magnetic field was applied perpendicular to the film plane.

The synchrotron x-ray diffraction measurements at low temperatures were performed on a diffractometer at BL46XU in SPring8 with the photon energy of 12.6 keV. The x-ray diffraction was measured under the magnetic field about 0.2 T perpendicular to the film plane using Neodymium permanent magnet.

III. EXPERIMENTAL RESULTS

A. Hall effect measurements

The temperature dependence of the Hall resistivity ρ_H, the resistivity ρ, and the magnetization M of $SrRuO_3$ are shown in Fig. 1. First, we show the analysis of the Hall effect in the paramagnetic region above $T_c(\simeq 160$ K). Above 160 K, $\rho_H(T)$ measured at $B=7$ T decreases with increasing temperature as $M(T)$. In the paramagnetic region, only the skew scattering contributes to the extraordinary Hall effect and ρ_H is described as $\rho_H(T)=R_0B+a\rho(T)M(T)$. This formula is transformed as $R_H=R_0+a\rho(T)\chi(T)$, where $R_H(=\rho_H/B)$ is the Hall coefficient and $\chi(=M/B)$ is the magnetic susceptibility.[1] In this case, the measured Hall coefficient $R_H(T)$ is expected to lie on a straight line as a function of measured $\rho(T)\chi(T)$, where the temperature is an implicit parameter. We plotted $R_H(T)$ against $\rho(T)\chi(T)$ and confirmed the linear relation between R_H and $\rho(T)\chi(T)$ above 160 K for $x=1$ as shown in Fig. 2. This finding ascertains that the skew scattering is dominant on the extraordinary Hall effect of $SrRuO_3$ in the paramagnetic region. From the plot, R_0 and a are obtained as $R_0=0.01(10^{-9}$ m^3/C) and $a=88(10^{-3}$ T$^{-1})$ for $x=1$. The analysis of ρ_H in the paramagnetic region was not performed for $Ba_{0.2}Sr_{0.8}RuO_3$ since the accuracy of the magnetization measurement in the paramagnetic region was too poor on the film with the thickness of only 20 nm.

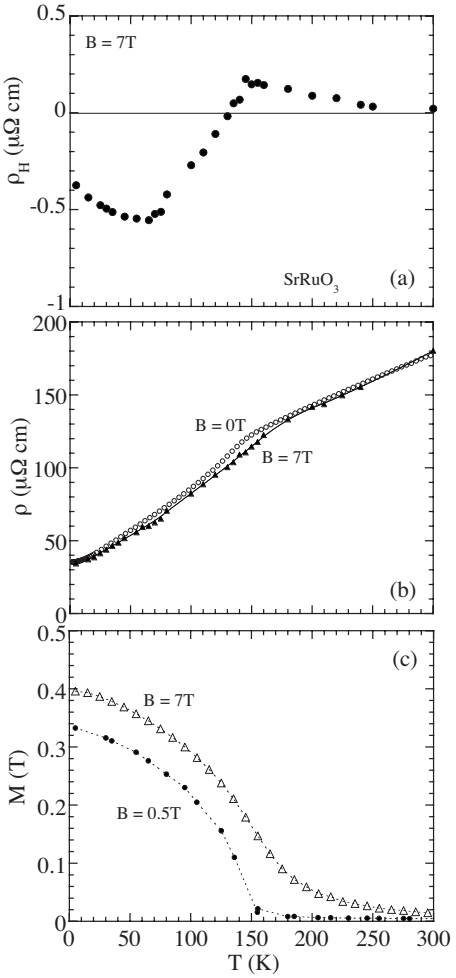

FIG. 1. Temperature dependence of the Hall resistivity (ρ_H), the resistivity (ρ), and the magnetization (M) of $SrRuO_3$ epitaxial film.

Next, we show the analysis of ρ_H in the ferromagnetic region below $T_c(\simeq 160$ K). The field dependence of ρ_H for $x=1$ measured at $T=5$, 100, and 160 K are shown in Fig. 3. In order to obtain R_0 and R_s, we performed the fitting of the field dependence of ρ_H using $\rho_H(B)=R_0B+R_sM(B)$, where R_0 and R_s are the fitting parameters and $M(B)$ is the measured magnetization. Above 60 K, the measured $\rho_H(B)$ was

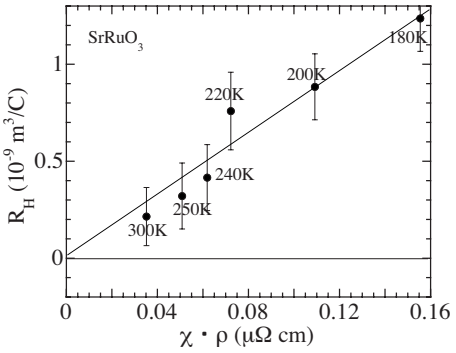

FIG. 2. The Hall coefficient ($R_H=\rho_H/B$) versus $\chi\cdot\rho$ for $SrRuO_3$ above 160 K.

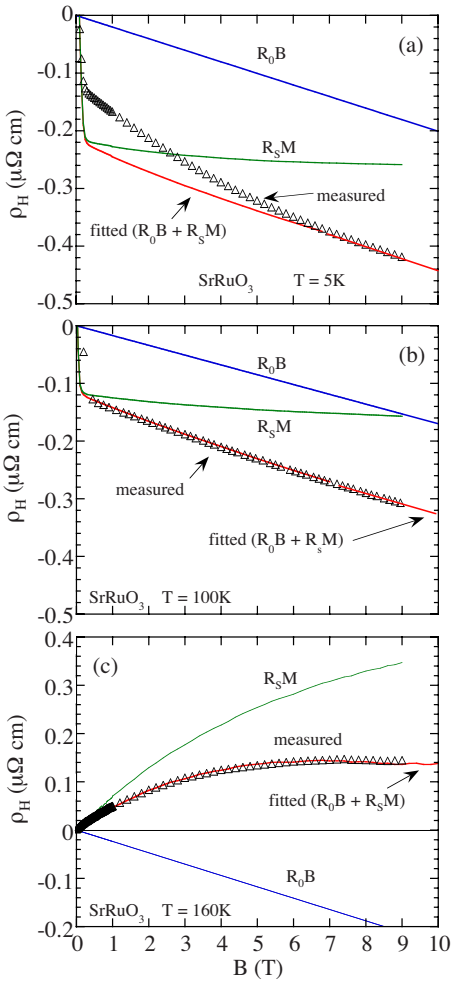

FIG. 3. (Color online) Field dependence of the Hall resistivity (ρ_H) of $SrRuO_3$. The ordinary component R_0B, the extraordinary one R_sM and the fitted $\rho_H(B)=R_0B+R_sM$ with the fitting parameters R_0 and R_s are also shown.

well reproduced by the fitting as shown in Figs. 3(b) and 3(c). Below 60 K, however, $\rho_H(B)$ cannot be fitted for the whole field region measured [see Fig. 3(a)]. We determined R_0 and R_s from $\rho_H(B)$ above 7 T, where $\rho_H(B)$ is linear to the field B. As a result of the procedure, a difference was observed at intermediate field region between the measured and the fitted $\rho_H(B)$ in the sample with $x=1$. The difference between the measured and the fitted $\rho_H(B)$ in the sample with $x=1$ is shown in Fig. 4. In the sample with $x=0.8$, $\rho_H(B)$ was well reproduced by the fitting even below 60 K (not shown). Temperature dependence of R_0 and R_s for $SrRuO_3$ and $Ba_{0.2}Sr_{0.8}RuO_3$ is shown in Fig. 5, which were obtained by the fitting of $\rho_H(B)$ at each temperature. The temperature dependence of R_0 is small and the typical value of R_0 is about $-0.2(10^{-9}\ \mathrm{m^3/C})$ for $x=1$. The magnitude of R_0 in the ferromagnetic region is one order larger than that in the paramagnetic region ($0.01[10^{-9}\ \mathrm{m^3/C}]$) shown in Fig. 2. For $x=0.8$, the temperature dependence of R_0 and R_s is similar to that for $x=1$ but the values of R_0 and R_s are several times smaller than those for $x=1$, which might be due to the inaccuracy of the size factor of the film responsible for the very thin and nonuniform thickness.

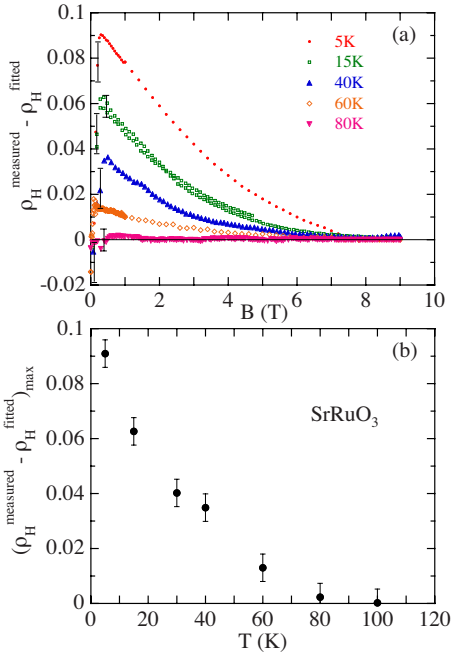

FIG. 4. (Color online) (a) Field dependence of the difference between the measured and the fitted Hall resistivity ($\rho_H^{\mathrm{measured}}$-$\rho_H^{\mathrm{fitted}}$) and (b) temperature dependence of the maximum value of $\rho_H^{\mathrm{measured}}$-$\rho_H^{\mathrm{fitted}}$ of $SrRuO_3$.

In ordinary ferromagnetic metals where R_s is expressed as $R_s=a\rho+b\rho^2$,[1] R_s/ρ against ρ lies on a straight line. Figures 6(a) and 6(b) show R_s/ρ versus ρ plots of $Ba_{1-x}Sr_xRuO_3$ with $x=1$ and $x=0.8$, respectively. In the plots, the temperature is

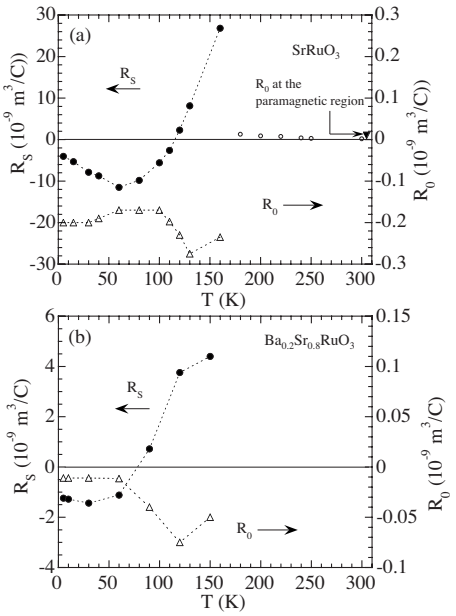

FIG. 5. Temperature dependence of the ordinary and the extraordinary Hall coefficients (R_0 and R_s, respectively) of (a) $SrRuO_3$ and (b) $Ba_{0.2}Sr_{0.8}RuO_3$ in the ferromagnetic region. The Hall coefficients R_0 and R_s are obtained by the fitting of $\rho_H(B)$ at each temperature (see the text). In Fig. 5(a), R_0 and R_s in the paramagnetic region derived from Fig. 2 are also shown by full triangle and open circles, respectively.

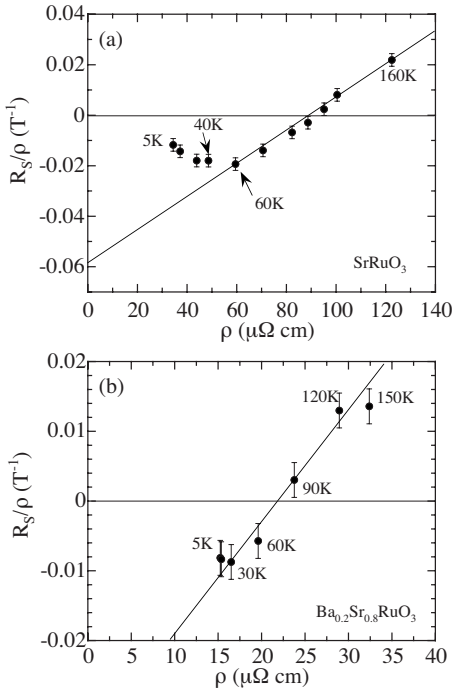

FIG. 6. R_s/ρ versus ρ plots for (a) $SrRuO_3$ and (b) $Ba_{0.2}Sr_{0.8}RuO_3$.

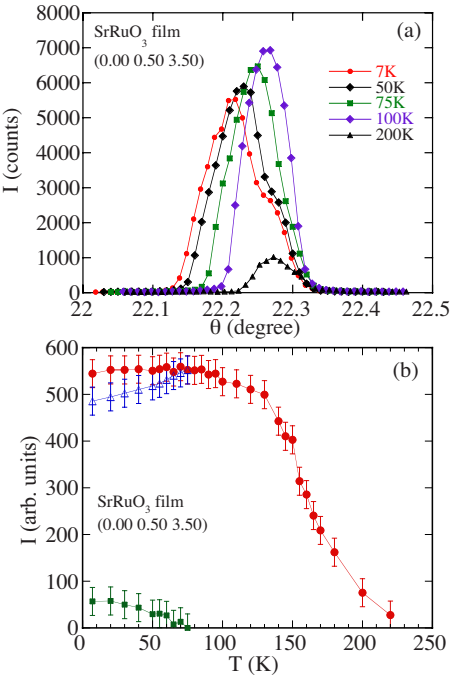

FIG. 7. (Color online) (a) The profiles of the superlattice reflection at $\boldsymbol{q}=(0,0.5,3.5)$ of $SrRuO_3$. (b) Temperature dependence of the area intensity of the superlattice reflection. Note that two reflections were observed below about 60K. In Fig. 7(b), the intensities of each reflections are also shown (open triangles and full squares for the profile at the lower and at the higher angles, respectively) along with the total intensity (full circles).

an implicit parameter and ρ is the resistivity measured at 9 T. The linear relation was observed in R_s/ρ versus ρ plots for the sample with $x=1$ above 60 K and for the sample with $x=0.8$ in the whole temperature region. The linear relation between R_s/ρ and ρ implies that the extraordinary Hall effect can be explained by the skew and the side-jump scatterings with constant a and b, as ordinary ferromagnetic metals. It holds above 60 K for the sample with $x=1$ and in the whole temperature region for the sample with $x=0.8$. The obtained coefficients from the plots are $a=-61(10^{-3}\ \mathrm{T}^{-1})$ and $b=6.7(10^{-4}\ \mu\Omega^{-1}\ \mathrm{cm}^{-1}\ \mathrm{T}^{-1})$ for $x=1$, and $a=-10(10^{-3}\ \mathrm{T}^{-1})$ and $b=45(10^{-4}\ \mu\Omega^{-1}\ \mathrm{cm}^{-1}\ \mathrm{T}^{-1})$ for $x=0.8$, which are comparable with those of the ordinary ferromagnetic metals such as Fe and Co.[2,16] The sign of a for $x=1$ is opposite to that in the paramagnetic region. Below 60 K, R_s/ρ for $x=1$ does not follow the linear relation against ρ. This fact suggests that the coefficients a and b of the skew and the side-jump scatterings are temperature dependent in $SrRuO_3$ at low temperatures. For $x=0.8$, the coefficients a and b are independent of temperature in the whole temperature region (Fig. 6).

For $x=1$, the magnitude of R_0 and the sign of the skew scattering changes below and above T_c, which suggests that some modulation of the Fermi surface occurs through the ferromagnetic transition. For $x=1$, the coefficients a and b of the skew and the side-jump scattering varies below 60 K. This observation also suggests the modulation of the Fermi surface since both the sign and the magnitude of the left-right asymmetric scattering are largely affected by the Fermi surface of the materials. Therefore, the present Hall effect measurements suggest that a two-stage modulation of the Fermi surface occurs at low temperatures for $x=1$. On the other hand, the coefficients a and b are temperature independent down to the lowest temperature investigated for $x=0.8$, which suggests that the modulation of the Fermi surface below 60 K vanishes.

B. Synchrotron x-ray diffraction measurements

In order to clarify the possible structural change responsible for the modulation of the Fermi surface in the sample with $x=1$, we performed the x-ray diffraction experiments. As shown in Fig. 6(a), we found a superlattice reflection at $\boldsymbol{q}=(0,0.5,3.5)$ of the tetragonal unit cell at lower temperatures, which implies the lowering of the lattice symmetry accompanied with the ferromagnetic transition. With decreasing temperature through T_c [see Fig. 6(b)], the superlattice reflection intensity drastically increases. Note that the superlattice reflection was observed above T_c, which is possibly due to the applied magnetic field about 0.2 T. Below around 100 K, the reflection shifts to lower angle with decreasing temperature. The linewidth increases with decreasing temperature and the superlattice reflection shows a shoulder below about 60 K. The development of the double peak becomes apparent with further decreasing temperature as shown in Fig. 6(a), which suggests the development of another superlattice reflection due to the further change of the lattice. Also in the sample with $x=0.8$, a superlattice reflection at $\boldsymbol{q}=(0,0.5,3.5)$ was observed below around T_c as shown in Fig. 8. In contrast to $x=1$, however, the superlattice reflection of the sample with $x=0.8$ does not show the remarkable change down to the lowest temperature investigated (see Fig. 8). These observations suggest the change in

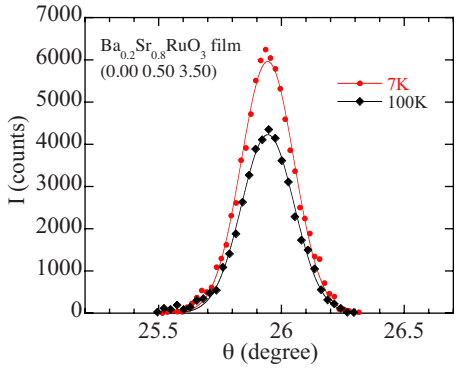

FIG. 8. (Color online) The superlattice reflection at $\boldsymbol{q}=(0,0.5,3.5)$ of $Ba_{0.2}Sr_{0.8}RuO_3$.

the lattice below 60 K vanishes in $x=0.8$ though the change through T_c is basically the same as that in $x=1$.

IV. DISCUSSION

The present experiment suggests the two-stage modulation of the lattice structure for $x=1$; one around T_c and another around 60 K, which possibly induces the two-stage modulation of the Fermi surface. The superlattice reflection at $\boldsymbol{q}=(0,0.5,3.5)$ suggests that the periods of the lattice below T_c in the ferromagnetic region becomes longer than that in the paramagnetic region for b and c axes. When the periods of the lattice becomes longer, the shape of the Fermi surface changes, in general, due to the superzone gap formation. The modulation of the shape of the Fermi surface possibly change the ordinary Hall coefficient R_0, which is correlated with the cross section of the Fermi surface. This consideration is consistent with the fact that the magnitude of R_0 changes remarkably between the ferromagnetic and the paramagnetic regions in the present experiment. However, the change of the resistivity ρ around T_c is small compared with the remarkable change of R_0. In general, the transport coefficients such as ρ and R_0 reflect the magnitude and the anisotropy of the Fermi surface. In $SrRuO_3$, the change in the cross section of the Fermi surface in ab plane which contributes to the change of R_0 in the present experiment may large while the change of the area of the whole Fermi surface responsible for the resistivity change may be comparatively small around T_c. In order to discuss the magnitude of ρ and R_0, the detailed structural analysis and the band calculation are necessary. The development of another superlattice reflection below 60 K suggests another change in the lattice structure, leading to a further modulation of the Fermi surface. Since the left-right asymmetric scattering of the conduction electrons depends on the Fermi surface,[1–3] the change in the coefficients of the left-right asymmetric scatterings a and b is possibly induced by the Fermi surface modulation below 60 K. In this situation, R_0 is also expected to change, however, the temperature dependence of R_0 below 60 K is small in the present experiment. This observation implies the change in the area of the Fermi surface by the formation of the superstructure below 60 K is small.

In contrast to the observation in $x=1$, the change in the lattice periodicity was observed only around T_c in $x=0.8$, which suggests that the modulation of the Fermi surface below 60 K vanishes in $x=0.8$. These observations corresponds to that no anomaly of the extraordinary Hall effect was observed below 60 K in $x=0.8$. In $Ba_{0.2}Sr_{0.8}RuO_3$, the chemical pressure from $SrTiO_3$ substrate is expected to be larger than that in $SrRuO_3$ since the lattice volume of $Ba_{0.2}Sr_{0.8}RuO_3$ is larger than that of $SrRuO_3$ due to the doping of Ba^{2+} with a larger ion radius than that of Sr^{2+}.[17] The stress from the substrate might suppress the modulation of the lattice structure responsible for the anomaly of the extraordinary Hall effect.

Finally we discuss the anomaly in the field dependence. For $x=1$ below 60 K, a deviation was observed at intermediate field region between the measured and the fitted $\rho_H(B)$ as shown in Fig. 4(a). The deviation becomes evident with decreasing temperature [see Fig. 4(b)]. Note that a slight deviation was still observed in the low field region at 60 K. The deviation suggests that the extraordinary Hall resistivity cannot be scaled by the magnetization $M(B)$ and has an additional term as $\rho_H(B)=R_0B+R_sM(B)+c(B)$. The additional term $c(B)$ is large at lower field region and decreases with increasing field. $c(B)$ almost vanishes at higher fields where $M(B)$ saturates. The field dependence can be understood if a spin chirality resulting from a noncollinear magnetic structure originates the additional term of the extraordinary Hall resistivity.[18–20] The additional term of the extraordinary Hall resistivity suggested in the present experiment implies the extraordinary Hall effect by an unconventional mechanism such as a spin chirality, though the evidence of the chiral spin structure was not observed in $SrRuO_3$. In order to elucidate the origin of the additional term of the extraordinary Hall resistivity in $SrRuO_3$, specific investigations of the magnetic structure under a magnetic field are indispensable.

V. CONCLUSION

We have investigated the extraordinary Hall effect of $Ba_{1-x}Sr_xRuO_3$ thin films with $x=1$ and 0.8. For $x=1$, the extraordinary Hall resistivity can be interpreted by the skew scattering mechanism in the paramagnetic region above about 160 K. In the ferromagnetic region above 60 K, the extraordinary Hall coefficient R_s can be expressed by the combination of the skew and the side-jump scattering mechanisms as $R_s=a\rho+b\rho^2$, which means that the mechanism of the extraordinary Hall effect in the system is basically the same as that of the ordinary ferromagnetic metals. Below 60 K, the field dependence of ρ_H shows an anomalous deviation from the relation $\rho_H=R_0B+R_sM$ at intermediate field region, suggesting an additional term in the field dependence of the extraordinary Hall resistivity. Below 60 K, the temperature dependence of R_s shows a deviation from the relation $R_s=a\rho+b\rho^2$. The observation suggests that the coefficients a and b of the skew and the side-jump scattering depends on temperature below 60 K. On the other hand in the sample with $x=0.8$, no anomalies were observed below 60 K both in the temperature and the field dependence. According to the structural analysis of the samples, we infer that the modulation of the Fermi surface due to the structural change plays an important roll on the anomaly of the tem-

perature dependence of the extraordinary Hall effect for $x=1$.

ACKNOWLEDGMENTS

This work was performed at BL46XU in SPring-8 with the approval of the Japan Synchrotron Radiation Research Institute (JASRI) (Proposals No. 2006A1326 and No. 2007A1381). This work was supported by Grant-in-Aid for Young Scientists (B) from the Ministry of Education, Science, Sports and Culture of Japan (Research Project No. 19740203).

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