Hall effect of epitaxial double-perovskite $\text{Sr}_2\text{FeMoO}_6$ thin films

W. Westerburg, F. Martin, and G. Jakob

Institute of Physics, University of Mainz, 55099 Mainz, Germany

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We prepared high epitaxial thin films of the compound $\text{Sr}_2\text{FeMoO}_6$ $(\text{SFMO})$ with narrow rocking curves by pulsed laser deposition. The diagonal and nondiagonal elements of the resistivity tensor were investigated at temperatures from 4 K up to room temperature in magnetic fields up to 8 T. An electronlike ordinary Hall effect and a holelike anomalous Hall contribution are observed. Both coefficients have reversed sign compared to the colossal magnetoresistive manganites. We found at 300 K an ordinary Hall coefficient of $-1.87 \times 10^{-10}$ m$^3$/As, corresponding to a nominal charge carrier density of four electrons per formula unit. At low temperature only a small negative magnetoresistance is observed which vanishes at higher temperatures. The temperature coefficient of the resistivity is negative over the whole temperature range. A Kondo like behavior is observed below 30 K while above 100 K variable range hopping like transport occurs.

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I. INTRODUCTION

Half-metallic ferromagnetic oxides as the manganites have reattributed much interest. Recently, a large negative magnetoresistance (MR) in a further oxide $\text{Sr}_2\text{FeMoO}_6$ (SFMO) was observed. This compound has an ordered double-perovskite structure and is, like the manganites, a ferrimagnetic (or ferromagnetic) oxide with a Curie-temperature of 410-450 K and a highly spin-polarized conduction band. For applications as a magnetic field sensor at room temperature a high spin-polarized conduction band is necessary to obtain a large magnetoresistance in low fields. Therefore spin-polarized magnetic compounds with Curie temperatures well above 300 K are interesting. We prepared epitaxial thin films of SFMO and investigated their structural and magnetic properties. Further, the longitudinal and transverse resistivity were measured as a function of temperature and magnetic field.

II. EXPERIMENT

Using pulsed laser deposition we prepared epitaxial thin films of SFMO from a stoichiometric target on (100) $\text{SrTiO}_3$ (STO) substrates. The substrate temperature during deposition was 700$^\circ$C in an oxygen partial pressure of $10^{-3}$ Pa. In x-ray diffraction only film reflections corresponding to a $(00\ell)$ orientation are visible. In Fig. 2 a detail of the $\theta/2\theta$-scan, showing the $(008)$ reflection peak of SFMO nearby the $(004)$ reflection peak of STO, is presented. A high degree of orientation of the $c$-axis is achieved. Rocking angle analysis of the SFMO (004) reflection shows an angular spread of 0.04$^\circ$, as can be seen in the inset of Fig. 2. The in-plane orientation was investigated by $\phi$-scans using the $\{224\}$ reflections. The film axes are parallel to the substrate axes with a 4-fold in-plane symmetry, demonstrated in Fig. 2. With this preparation conditions the $c$-axis is elongated to 7.998 Å compared to bulk material. This indicates either epitaxial strain between SFMO and the STO substrate or (and) a non stoichiometric oxygen content of the films.

With scanning electron microscopy and atomic force microscopy we found a smooth surface with a roughness of 10 nm and the average grain size to be 50 nm. By Rutherford backscattering on a reference sample on MgO substrate the metal atom stoichiometry was determined to be $\text{Sr}_2\text{Fe}_{1.06}\text{Mo}_{0.90}\text{O}_x$. This is within the experimental errors identical to the nominal composition of the target ($\text{Sr}_2\text{FeMoO}_{6+\delta}$). The film thickness was 250 nm, measured with a Mireau interferometer. The samples, not resistant against water, were patterned photolithographically and etched to a Hall bar structure. The longitudinal and transverse resistivity were measured in magnetic fields up to 8 T from liquid helium temperature to 300 K. A standard four point technique with DC current was used. The Hall coefficient was determined by slowly sweeping the magnetic field in positive and negative field direction with an asymmetric current injection to minimize the parasitic longitudinal voltage on the Hall contacts. Spontaneous magnetization was determined in small fields $(B = 100$ mT$)$ with a SQUID magnetometer.

III. RESULTS AND DISCUSSION

The zero field resistivity is presented in Fig. 3. The room temperature resistivity value of 3 m$\Omega$mcm is comparable with other reported results but down to 4 K the resistivity increases almost an order of magnitude. Depending on preparation the sign of the temperature coefficient and the sign of the MR changes. We observed in our semiconducting films a negative MR of -3.5 % at a temperature of 4 K and a magnetic field of 8 T, similar to the result of Asano et al. At higher
temperatures the MR vanishes. While in the plot of Fig. 3 the resistivity shows a smooth temperature variation with a negative temperature coefficient, closer inspection indicates a change in conduction mechanism. Below 30 K the resistivity increases strictly logarithmic with falling temperature. This behavior is known from Kondo systems, where it results from carrier scattering at uncorrelated magnetic impurities. Above 100 K the temperature dependence of the resistivity is best described by $\rho \propto \exp((T_0/T)^{0.25})$ with $T_0 = 4900$ K, as can be seen in the inset of Fig. 3. The origin of the opposite behavior in transport properties between different epitaxial films remains up to now unclear. One cause may be disorder-effects in the B-cation arrangement.

For the double-perovskites random, rock salt and layered systems, where it results from carrier scattering at uncorrelated magnetic impurities. Above 100 K the temperature dependence of the resistivity is best described by $\rho \propto \exp((T_0/T)^{0.25})$ with $T_0 = 4900$ K, as can be seen in the inset of Fig. 3. The origin of the opposite behavior in transport properties between different epitaxial films remains up to now unclear. One cause may be disorder-effects in the B-cation arrangement.

A segregation into clusters of compositions SrMoO$_3$ ($a = 3.975$ Å) and SrFeO$_3$ ($a = 3.869$ Å) should be visible in x-ray diffraction as severe peak broadening for small clusters or as peak splitting for large clusters. Both effects are not observed. The peak splitting visible in Fig. 4 for substrate and film peak is due to the Cu $K_\alpha$ doublet only. For the double-perovskites random, rock salt and layered B',B'' sublattice types are known.

In neutron powder diffraction of SFMO Rietveld refinement showed perfect B',B''-rock salt structure. Due to extinction conditions the x-ray diffraction in Bragg-Brentano geometry cannot resolve the sublattice structure for (00$\ell$) oriented films. However, the saturation magnetization of our films is with one $\mu_B$ per formula unit (f.u.) much smaller than the moment of 3 $\mu_B$/f.u. observed by Kobayashi et al. in a bulk sample. This discrepancy is a hint to cation site disorder. Other possible influences as non stoichiometric oxygen content and substrate induced strain will be the subject of future investigations.

In ferromagnetic materials the transverse resistivity is given by

$$\rho_{xy} = R_H B + R_A \mu_0 M \quad (1)$$

with the magnetization $M$ and the ordinary and anomalous Hall coefficients $R_H$ and $R_A$, respectively. The Hall voltage $U_{hall}$ was measured at several constant temperatures between 4 K and 300 K in magnetic fields up to 8 T. Fig. 4 shows the results for the Hall resistivity and the Hall voltage. The error in the data is smaller than the symbol size. The low magnetoresistivity of both the sample and the Pt-thermometer allows at high temperatures the elimination of the parasitic longitudinal part of the Hall voltage and a quantitative analysis of $R_H$. Due to the increasing resistance the measurement current was reduced from 1 mA to 100 $\mu$A for the data taken at $T = 4$ K, leading to a worse signal to noise ratio. In low fields, a steep increase of $U_{hall}$ with increasing field is seen. This part, where the magnetization of the sample changes, is dominated by the anomalous Hall contribution. In the case of SFMO $R_A$ is holelike, in contrast to the manganites. At 1 T a maximum occurs and at higher fields the data show a linear negative slope. In this high field regime the magnetization of the sample is constant and therefore, according to Eq. (1), the ordinary Hall effect becomes visible. This behavior, positive $R_A$ and negative $R_H$, was also observed in iron and ferromagnetic iron alloys. The linear negative slope $d\rho_{xy}/dB$ indicates an electronlike charge-carrier concentration. The Hall coefficient at 300 K is $-1.87 \times 10^{-10}$ m$^3$/A, corresponding to a charge carrier density in a one-band model of 4.1 electrons/f.u.. The value of $R_H$ increases with decreasing temperature to $-1.15 \times 10^{-10}$ m$^3$/A at 80 K. If one assumes that there exists a residual magnetization increase in the high-field regime its anomalous contribution is holelike. Therefore it will lead to an underestimation of $R_H$, but not to a sign change.

The anomalous Hall effect in ferromagnetic materials has two possible origins, an asymmetry of scattering (skew scattering) or a sideward displacement of the center of weight of an electron wave packet during the scattering process (side-jump), both due to spin-orbit interaction. The anomalous Hall effect is closely related to the longitudinal resistivity $\rho_{xx}$ by

$$R_A \mu_0 M = \gamma \rho_{xx}^n \quad (2)$$

but with a different exponent $n = 1$ and $n = 2$ for skew scattering and side jump, respectively. The anomalous Hall coefficient can be extracted from the data by extrapolation the linear high-field data to $B = 0$. The obtained value is then, according to Eq. (2), $R_A \mu_0 M_{Sat} := \rho_{xx}^*$. The resistivites $\rho_{xx}$ versus $\rho_{xy}^*$ in a double logarithmic plot show indeed in the case of SFMO a linear slope with $n = 0.75$, indicating skew scattering. Due to the opposite sign of the temperature coefficient of the resistivity between SFMO and the manganites in the ferromagnetic regime, the anomalous Hall coefficient increases for SFMO with decreasing temperature. In the manganites the anomalous Hall effect vanishes for very low temperatures, because of increasing magnetic order, as expected by theory. This is a further hint that in our SFMO thin films a full magnetic order is not obtained.

**IV. CONCLUSION**

In summary we prepared high epitaxial thin films of the compound Sr$_2$FeMoO$_6$ with narrow rocking curves by pulsed laser deposition. We performed detailed transport measurements of the diagonal and nondiagonal elements of the resistivity tensor from 4 K up to room temperature in magnetic fields up to 8 T. An electronlike ordinary Hall effect and a holelike anomalous Hall contribution were observed. These signs are reversed compared to the colossal magnetoresistive manganites. The value of the nominal charge carrier density at 300 K is four electrons.
per formula unit. A full magnetic order was not observed in our samples.

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FIG. 1. θ/2θ-scan of the SFMO thin film showing the (008) reflection peak (Cu Kα doublet). The three peaks at higher angles are due to the substrate. In the inset a rocking curve (2θ = 45.32°) with an extremely narrow linewidth (Δω = 0.04°), indicating a high degree of epitaxy, is shown.

FIG. 2. φ-scan of the SFMO(224) reflections of the thin film grown on a SrTiO3 substrate. The 4-fold in-plane symmetry, the film axes are parallel to the substrate axes, is visible.
FIG. 3. Temperature dependence of the resistivity $\rho_{xx}$ in zero field. Below 30 K the resistivity increases logarithmic with falling temperature. The variable range hopping model in the inset describes the data above 100 K. The lines are in both cases a guide to the eye.

FIG. 4. Hall voltages $U_{\text{hall}}$ (left axis) and Hall resistivities $\rho_{xy}$ (right axis) as functions of magnetic field for several constant temperatures.