Hall effect in *c*-axis-oriented MgB_2 thin films

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We have measured the longitudinal resistivity and the Hall resistivity in the ab plane of highly c-axis-oriented MgB_2 thin films. In the normal state, the Hall coefficient (R_H) behaves as $R_H \sim T$ with increasing temperature (T) up to 130 K and then deviates from that linear T dependence at higher temperatures. The T^2 dependence of the cotangent of the Hall angle is only observed above 130 K. The mixed-state Hall effect reveals no sign anomaly over a wide range of current densities from 10^2 to 10^4 A/cm^2 and for magnetic fields up to 5 T.

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I. INTRODUCTION

The recent discovery of superconductivity in MgB_2, with a transition temperature (T_c) of about 39 K,[1] has attracted extensive scientific interest in the fields of basic research and applications. Already, several physical properties, such as the Hall effect, the thermoelectric power, the magnetization, and the magnetoresistance, have been investigated using polycrystalline samples.[2–6] However, many experimental results still remain controversial because of the relatively high anisotropic nature of this compound. Based on measurements of the upper critical field for different crystal planes of MgB_2 single crystals[7,8] and of highly c-axis-oriented thin films,[9] they have confirmed the anisotropic nature of the MgB_2 superconductor. These results strongly suggest that the physical properties of MgB_2 should be investigated by either using single crystals or high-quality thin films having preferred orientations. For the Hall measurements, since sizable single crystals are not available and the Hall signal is very small due to its metallic character, the thin-film form is the best candidate for achieving accurate experimental results.

In our earlier work on polycrystalline samples,[2] we confirmed that the majority charge carriers were holelike, which was consistent with theoretical estimates;[10] subsequently, similar results were also reported for polycrystalline MgB_2 thin films.[11] To the best of our knowledge, the in-plane Hall effect for MgB_2 has not been previously studied; thus, measurement of the ab-plane Hall effect for c-axis-oriented MgB_2 thin films should provide significant input for future investigations of its electronic transport properties and vortex dynamics.

For high-T_c cuprate superconductors (HTS), a universal T^2 dependence of the cotangent of the Hall angle ($\cot\Theta_H$) has been extensively discussed. Anderson[12] has proposed that this behavior can be explained if two different scattering rates, a transport scattering time and a Hall (transverse) scattering time, are considered, where the longitudinal resistivity (ρ_{xx}) is determined by the former and the Hall resistivity (ρ_{xy}) is determined by both. Most experimental results for HTS have supported this theory,[13–15] and it is generally accepted that, in the normal state, a $\cot\Theta_H \sim T^2$ law is universal over a wide temperature range. Similar behavior has also been reported for polycrystalline MgB_2 superconductors.[2,11]

Another interesting feature concerning the mixed-state Hall effect as a probe of superconductivity is the anomalous sign change near T_c as a function of the T and the magnetic field, and its origin has remained an unsolved subject for over 30 years. The sign anomaly has been observed in some conventional superconductors,[16] as well as in most HTS.[16–18] However, in clean superconductors, such as pure Nb, V, and $2H$-$NbSe_2$, no sign anomaly has been found.[16] Our Hall data for MgB_2 are more similar to the behavior seen in Nb, V, and $2H$-$NbSe_2$, suggesting that MgB_2 might be a clean-limit superconductor.[19]

In this paper, we report a measurement of the in-plane Hall effect of MgB_2. The measurement was carried out using highly c-axis-oriented thin films, and we found that the sign of the R_H was positive like those of HTS. Also, the R_H appeared to follow a linear behavior for the T region from 30 to 130 K, which is different from the behaviors of polycrystalline MgB_2 and of HTS. The Hall effect in the mixed state showed no sign anomaly over a wide range of current densities from 10^2 to 10^4 A/cm^2 and for magnetic field up to 5 T, which is in contrast to the observations in most HTS and polycrystalline MgB_2 thin films.

II. EXPERIMENT

The MgB_2 thin films were grown on Al_2O_3 ($1\,\bar{1}\,0\,2$) single crystals under a high-vacuum condition of $\sim 10^{-7}$ Torr by using the pulsed laser deposition and the postannealing techniques reported in an earlier paper.[20] Typical samples were 10 mm in length, 10 mm in width, and 0.4 μm in thickness. The film thickness was measured using scanning electron microscopy. Standard photolithographic techniques were used to produce thin-film Hall-bar patterns, which consisted of a rectangular strip (1 mm in width and 3 mm in length) of MgB_2 with three pairs of sidearms (the upper inset of Fig. 2 below). The narrow sidearm width of 0.1 mm was patterned so that the sidearms would have an insignificant effect on the equipotential. Using this six-probe configuration, we were able to measure simultaneously the ρ_{xx} and ρ_{xy} at the same T; thus the $\cot\Theta_H$ was obtained very precisely. To achieve good ohmic contacts (<1 Ω), we

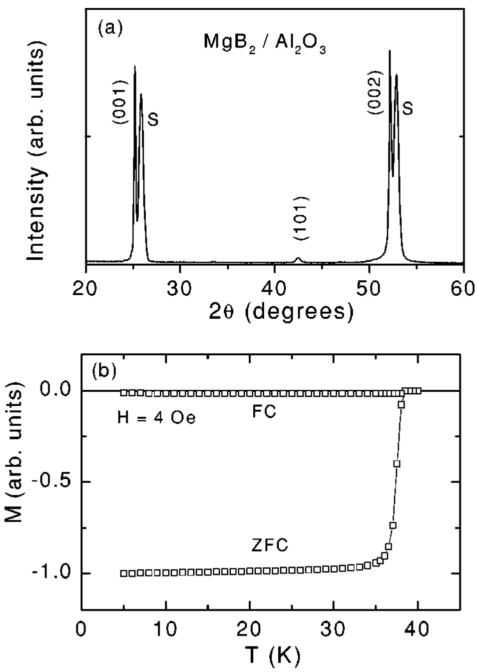

FIG. 1. (a) X-ray-diffraction patterns of MgB_2 thin films. A highly *c*-axis-oriented crystal structure normal to the substrate surfaces was observed. *S* denotes the substrate peaks. (b) Magnetization at $H=4$ Oe in the ZFC and FC states.

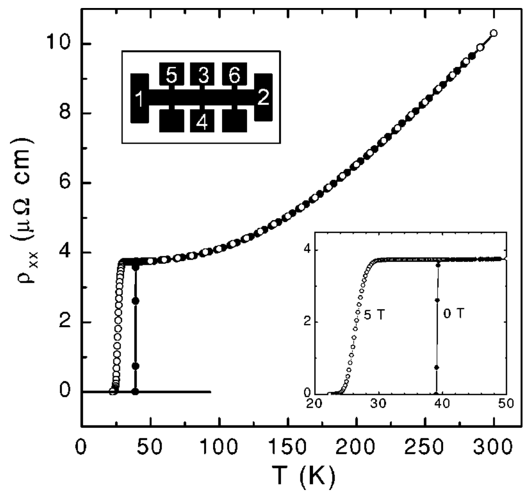

FIG. 2. Temperature dependence of the resistivity of MgB_2 thin films for $H=0$ and 5 T. The lower inset shows a magnified view near the T_c. A sharp transition, with a narrow transition width of $\sim$0.15 K, was observed. The upper inset is a schematic diagram of the Hall-bar pattern.

coated an Au film on the contact pads after cleaning the sample surface by using Ar-ion milling. After installing a low-noise preamplifier prior to the nanovoltmeter, we achieved a voltage resolution of below 1 nV. The magnetic field was applied perpendicular to the sample surface by using a superconducting magnet system, and the applied current densities were 10^2-10^4 A/cm^2. The Hall voltage was found to be linear in both the current and the magnetic field.

III. RESULTS AND DISCUSSION

The structural analysis was carried out using x-ray diffractometry, and the results are shown in Fig. 1(a). The MgB_2 thin film showed a highly *c*-axis-oriented crystal structure, and the sample purity exceeded 99% and had only a minor {101} oriented phase. Figure 1(b) shows the low-field magnetization at $H=4$ Oe for both the zero-field-cooled (ZFC) and field-cooled (FC) states of an MgB_2 film. A very sharp diamagnetic transition is observed. Even at a high *T* of 37 K and under self-field conditions, the critical current density determined by direct current vs voltage measurements was observed to be on the order of 10^5 A/cm^2.[21] These results indicate that the MgB_2 films used in the present study were homogeneous and of very high quality.

Figure 2 shows the *T* dependence of ρ_{xx} for a MgB_2 film at $H=0$ and 5 T. The upper inset shows the six-probe Hall-bar pattern. Pads 3 and 4 and 5 and 6 were used to measure ρ_{xy} and ρ_{xx}, respectively, while the current was applied between 1 and 2. The lower inset shows a magnified view near the superconducting transition. The onset T_c was 39.2 K and had a narrow transition width of $\sim$0.15 K, as judged from the 10% to 90% superconducting transition. At 40 K, ρ_{xx} was 3.4 $\mu\Omega$ cm, giving a residual resistivity ratio (RRR) [RRR $=\rho_{xx}(300\ \mathrm{K})/\rho_{xx}(40\ \mathrm{K})$] of 3, which was smaller than the value observed for MgB_2 single crystals.[7,8] A very small (less than 0.5%) magnetoresistance was observed in the normal state at 5 T.

The *T* dependence of the R_H at 5 T is shown in Fig. 3. The offset voltage due to the misaligned Hall electrodes was eliminated by reversing the field from -5 T to 5 T (inset of Fig. 3), and the Hall voltage was taken as the average value, $V_{xy}=(V_{+H}-V_{-H})/2$, for all data points. The offset voltage at $H=0$ T was very small compared to V_{xy} at 5 T, indicating excellent alignment of the Hall electrodes. The value of V_{xy} was about two orders-of-magnitude larger than that of polycrystalline bulk samples.[2] Due to our high-resolution measurements, we were able to interpret our Hall data rigor-

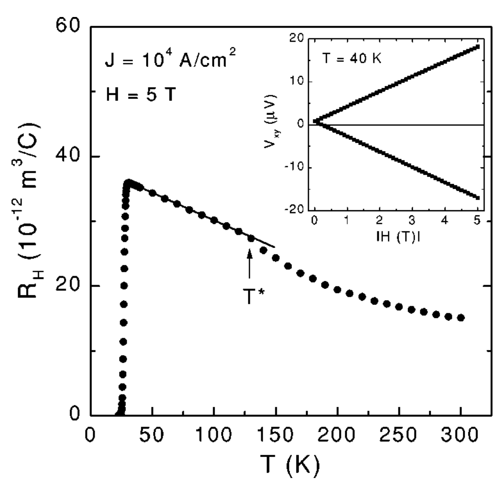

FIG. 3. R_H vs temperature of MgB_2 thin films at 5 T. Distinct temperature dependencies of the R_H are evident below and above 130 K. The data were measured by reversing the magnetic field from -5 T to 5 T at a fixed temperature, as shown in the inset.

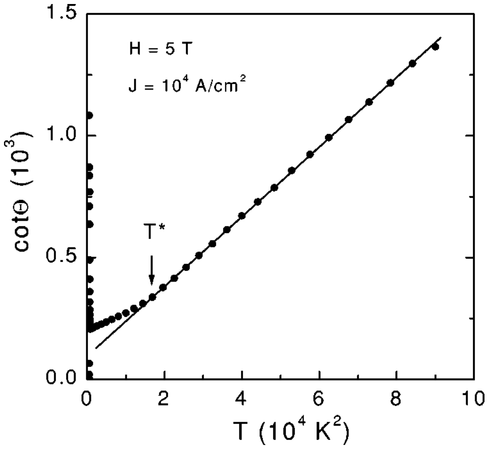

FIG. 4. Temperature dependence of cot θ_H at 5 T. A clear T^2 law was observed above 130 K.

ously. The value of R_H was positive over the entire T range, which is consistent with the result of band calculations.[10] Although the charge-carrier density cannot be determined simply within the context of the Boltzmann theory because of the anisotropic band structure and the complex Fermi surface of MgB_2, such a calculation would be meaningful for comparison with other superconductors. At 100 K, the R_H was 3×10^{-11} m^3/C, and the hole carrier density, calculated from $1/eR_H$, was $\sim 2\times10^{23}$ holes/cm^3. The absolute value of the hole density was two orders-of-magnitude larger than that of $YBa_2Cu_3O_7$,[22] indicating that MgB_2 has a metallic superconductor. The average value of the R_H was consistent with theoretical estimates.[10]

As the T was increased from the T_c, the R_H decreased linearly up to 130 K (T^*) and then deviated from that linear behavior at higher T, suggesting that the electronic transport mechanism changes at around 130 K. The T^* was observed to be independent of magnetic fields up to 5 T. This feature is somewhat different from previous results for polycrystalline bulk[2] and thin-film samples[11] for which the R_H exhibited the same T dependence over the entire T range from the T_c to 300 K. These results suggest that MgB_2 might have different transport mechanisms in the in-plane and the out-of-plane directions. A similar distinct T dependence around 150 K was also observed in the thermoelectric power measurements;[3,4] the thermoelectric power increased linearly with increasing T up to around 150 K and then showed a downward deviation from linearity. This behavior is believed to be due to the different T dependencies of the multiband contributions to the transport properties; at low T (below 150 K), charge transport is governed mainly by hole carriers whereas at higher T, the contribution of electron carriers must be considered.[3]

In Fig. 4, we show the T dependence of cot θ_H at 5 T. A good linear fit to AT^2+B is observed for the T range from 130 to 300 K; clear deviation from a T^2 dependence is seen below 130 K. According to the Anderson theory,[12] which is based on charge-spin separation, charge transport is governed by two separate scattering times with different T dependencies. The longitudinal conductivity (σ_{xx}) is propor-

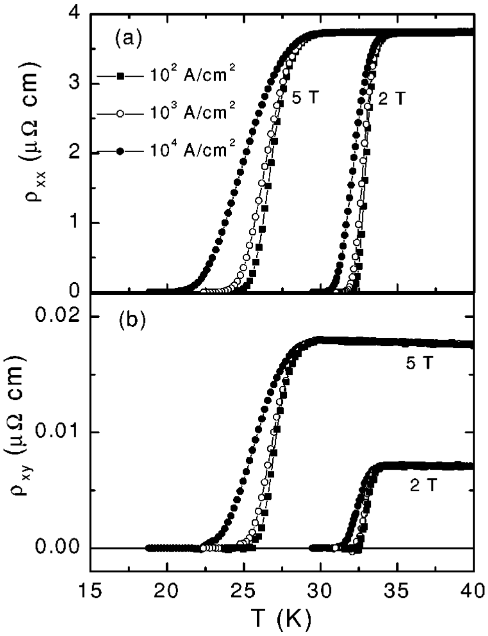

FIG. 5. Mixed-state (a) ρ_{xx} and (b) ρ_{xy} measured at applied current densities of 10^2, 10^3, and 10^4 A/cm^2 and for $H=2$ and 5 T. No sign change was observed, which is different from the case of HTS.

tional to the transport scattering time (τ_{tr}) whereas the Hall conductivity (σ_{xy}) is determined by $\tau_H\tau_{tr}$ where the Hall relaxation time (τ_H) is proportional to $1/T^2$. The τ_H is mainly governed by spinon-spinon interactions; thus, its T dependence is not affected by impurities. As a result, the cot $\theta_H(=\sigma_{xx}/\sigma_{xy})$ should follow a T^2 law. Such a universal temperature dependence has been observed in most HTS,[13] and the T^2 law has been confirmed not to depend on impurities.[14,15] Above 130 K, our experimental data are also in good agreement with a T^2 law as observed in most HTS. However, our data cannot be interpreted within the Anderson theory because MgB_2 does not have active spins. We also observe a distinct T^2 dependence of cot θ_H at around 130 K.

Finally, we address the transport properties in the superconducting state. In Fig. 5(a), we show the T dependence of the ρ_{xx} for magnetic fields of 2 and 5 T and at current densities of 10^2 to 10^4 A/cm^2. A broad superconducting transition is observed, which implies the existence of a relatively wide vortex-liquid phase in this compound. This result is quite similar to those for HTS. In a separate paper,[23] we reported that this vortex phase could be interpreted well by using two distinct regions: a thermal fluctuation region at high T near T_c and a vortex-glass region at low T. Moreover, we found a very narrow thermally activated flux-flow region, which was different from the case of HTS.

The corresponding ρ_{xy} data are plotted in Fig. 5(b). No sign reversal was detected in the Hall data measured for magnetic fields from 1 to 5 T and over two orders of magnitude of the current density. A puzzling sign reversal has been observed in the mixed-state Hall effect for most HTS (Ref. 18) and even for polycrystalline MgB_2 films.[11] In conventional superconductors, this sign change occurs mostly in moderately clean superconductors, but is not seen in either clean-limit superconductors, such as V, Nb, and $NbSe_2$, or dirty-limit superconductors, such as superconducting alloys.[16] Therefore, the absence of the Hall sign anomaly suggests that MgB_2 should be categorized as a clean-limit superconductor. Indeed, a short superconducting coherence length (~ 50 Å) and a relatively large mean free path (250–600 Å) have been reported for this compound.[19,24]

An interesting microscopic approach based on the time-dependent Ginzburg-Landau theory has been proposed in a number of papers.[25–27] According to this model, the mixed-state Hall conductivity in type-II superconductors is determined by the quasiparticle contribution and the hydrodynamic contribution of the vortex cores. Since the hydrodynamic contribution is determined by the energy derivative of the density of states,[26,27] if that term is negative and dominates over the quasiparticle contribution, a sign anomaly can appear. This theory is consistent with experimental data for HTS.[28] For the mixed-state Hall effect in the MgB_2 compound, since no sign anomaly was detected, we may suggest that the hydrodynamic contribution is very small or negligible in this superconductor.

IV. SUMMARY

Using high-quality *c*-axis-oriented MgB_2 thin films, we studied the in-plane Hall effect as a function of the magnetic field over a wide range of current densities. The normal-state R_H increased linearly with increasing T up to 130 K and then showed a downward deviation from its linear dependence at higher T, which is probably due to the distinct T dependencies of the electronic states of the MgB_2 compound. Our Hall data were also in good agreement with a $\cot \theta_H \sim T^2$ law above 130 K. The mixed-state Hall effect revealed no sign anomaly for magnetic fields from 1 to 5 T over two orders of magnitude of the current density.

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