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.

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 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 hololike, 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 ($\rho_{xx}$) is determined by the former and the Hall resistivity ($\rho_{xy}$) 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 [9].

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 the first 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$_2$O$_3$ (1 1 0 2) single crystals under a high-vacuum condition of $\sim 10^{-7}$ Torr by using the pulsed laser deposition and the post-annealing 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). The narrow sidearm width of 0.1 mm was patterned so that the
sidearms would have an insignificant effect on the equipotential. Using this 6-probe configuration, we were able to measure simultaneously the $\rho_{xx}$ and $\rho_{xy}$ at the same $T$; thus the $\cot \Theta_H$ was obtained very precisely. To achieve good ohmic contacts ($<1 \, \Omega$), we coated 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 $10^2 - 10^4 \, \text{A/cm}^2$. 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 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 $10^5 \, \text{A/cm}^2$ [2]. 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 $\rho_{xx}$ for a MgB$_2$ film at $H = 0$ and 5 T. The upper inset shows the 6-probe Hall-bar pattern. Pads 3 - 4 and 5 - 6 were used to measure $\rho_{xy}$ and $\rho_{xx}$, 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, $\rho_{xx}$ was $3.4 \, \mu\Omega \, \text{cm}$, giving a residual resistivity ratio $[\text{RRR} = \rho_{xx}(300 \, \text{K})/\rho_{xx}(40 \, \text{K})]$ of 3, which was smaller than the value observed for MgB$_2$ single crystals [38]. 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 2 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 rigorously. 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 $3 \times 10^{-11} \, \text{m}^2/\text{C}$, and the hole carrier density, calculated from $1/eR_H$, was $2 \times 10^{23} \, \text{holes/cm}^3$. The absolute value of the hole density was two orders of magnitude larger than that of YBa$_2$Cu$_3$O$_7$ [12], indicating that MgB$_2$ has a metallic superconductor. The average value of the $R_H$ was consistent with the 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 [1] 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 [13]: the thermoelectric power increased linearly with increasing $T$ up to around 150 and then showed a downward deviation from linearity. This behavior is believed to be due to the different $T$ dependences of the multi-band 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 $\cot \Theta_H$ 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 [2], which is based on charge-spin separation, charge transport is governed by two separate scattering times with different $T$ dependences. The longitudinal conductivity ($\sigma_{xx}$) is proportional to the transport scattering time ($\tau_{tr}$) whereas the Hall conductivity ($\sigma_{xy}$) is determined by $\tau_H \tau_{tr}$ where the Hall relaxation time ($\tau_H$) is proportional to $1/T^2$. The $\tau_H$ 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][13]. 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 $\cot \theta_H$ 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 $\rho_{xx}$ 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 $\rho_{xy}$ 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 [8] 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 ($\sim 50$ Å) and a relatively large mean free path (250–600 Å) have been reported for this compound [13, 24].

An interesting microscopic approach based on the time-dependent Ginzburg-Landau theory has been proposed in a number of papers [20, 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 [20, 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 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 $T$ dependences 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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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.

FIG. 3. $R_H$ vs Temperature of MgB$_2$ thin films at 5 T. Distinct temperature dependences 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 $\cot\theta_H$ at 5 T. A clear $T^2$ law was observed above 130 K.
Fig. 1. Kang et al.
Fig. 2. Kang et al.
$J = 10^4 \text{ A/cm}^2$

$H = 5 \text{ T}$

$R_H (10^{-12} \text{ m}^3/\text{C})$

$T^*$

$T = 40 \text{ K}$

$IH (T)I$

$V_{xy} (\mu\text{V})$

Fig. 3. Kang et al.
$H = 5 \, T$

$J = 10^4 \, \text{A/cm}^2$

**Fig. 4. Kang et al**
Fig. 5. Kang et al