Unusual Hall Effect in Superconducting MgB$_2$ Films: Analogy to High-$T_c$ Cuprates

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We have investigated the temperature and magnetic field dependence of the Hall coefficient of two well-characterized superconducting MgB$_2$ films ($T_{c,0}=38.0$ K) in both the normal and superconducting states. Our results show that the normal-state Hall coefficient $R_H$ is positive and increases with decreasing temperature, independent of the applied magnetic field. Below $T_c(H)$, $R_H$ decreases rapidly with temperature and changes sign before it reaches zero. The position and magnitude at which $R_H$ shows a minimum depends on the applied field. Quantitative analysis of our data indicates that the Hall response of MgB$_2$ behaves very similarly to that of high-$T_c$ cuprates: $R_H \propto T$ and $\cot \theta_H \propto T^2$ in the normal state, and a sign reversal of $R_H$ in the mixed state. This suggests that the B-B layers in MgB$_2$, like the Cu-O planes in high-$T_c$ cuprates, play an important role in the electrical transport properties.

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The recent discovery of unexpectedly high temperature superconductivity in MgB$_2$ ($T_c = 39$ K) [12] has stimulated a great deal of interest in its mechanism. One of the central issues is whether MgB$_2$ is related to other well-known superconductors or represents a new class of superconductor. Although superconductivity was found in other borides with the same crystal structure as MgB$_2$ [2, 3], the $T_c$ of these other materials does not exceed 0.6 K. What makes the $T_c$ of MgB$_2$ almost two orders of magnitude higher? One school of thought proposes a phonon-mediated BCS pairing mechanism. Evidence for this view is provided by isotope effect experiments [12], an isotropic energy gap [13, 14], NMR studies [15, 16], and specific heat measurements [17, 18].

Evidence for unconventional superconductivity is found in the non-BCS-like temperature dependence of both penetration depth $\lambda(T)$ and microwave surface resistance $R_{\perp}$ [19]. Theoretically, Hirsch argues that the B-B planes in MgB$_2$ should play a role similar to the Cu-O planes in high-$T_c$ cuprates, and predicts hole superconductivity in MgB$_2$ [20]. A recent Hall study on bulk MgB$_2$ shows a positive normal-state Hall coefficient [21], confirming hole conduction in this material.

The anomalous Hall response of high-$T_c$ cuprates has been considered as one of their most remarkable and puzzling properties [22]. In this Letter, we report for the first time a surprisingly similar Hall response observed in superconducting MgB$_2$ films. The experiments show that $R_H$ is positive in the normal state and has a strong temperature dependence that can be described using the model Anderson proposed for high-$T_c$ cuprates [23]. Further analysis yields a $T^2$-dependence of the Hall angle from $T_c$ to at least 300 K. We also find that $R_H$ changes sign as the sample is cooled below $T_c$; the temperature and magnetic field dependence of $R_H$ in the mixed state resembles that observed in hole-doped high-$T_c$ cuprates. We discuss the possible origin of unusual Hall behaviors based on theoretical models for high-$T_c$ cuprates.

The MgB$_2$ films were prepared using a precursor post-processing approach and extensively characterized as described in Ref. [24]. For the films used in the Hall measurements, the zero-field resistivity indicates an onset $T_c^{onset}=38.6$ K with a transition width of $\Delta T=0.4$ K (see Fig. 2b), indicating the good quality of our samples. For the Hall measurements, we cut the samples into rectangular shape with dimensions of 4 mm $\times$ 2 mm. The Hall and longitudinal resistivities were measured using a standard six-point method. Stable low-resistance contacts were achieved by heating the sample with fresh contacts (Epotek H-20E silver epoxy) at 120 $^\circ$C for 4 hours. The experiments were performed in a Quantum Design Physical Property Measurement System using a horizontal rotator and magnetic fields up to 8 Tesla. In order to exclude the longitudinal contribution due to misalignment of Hall-voltage contacts, the Hall resistivity was derived from the antisymmetric part of the transverse resistivity under magnetic field reversal at a given temperature, $i.e., \rho_{H}=\rho_H(+H)-\rho_H(-H)/2$. Finally, the Hall coefficient $R_H$ is evaluated from $R_H=\rho_H/H$. In the normal state, checks were made at several temperatures to ensure that the Hall coefficient was linear in applied current and field.

In Fig. 1, we show the temperature dependence (5-300 K) of $R_H$ obtained on a 0.6$\mu$m thick MgB$_2$ film. The applied field was 8 T. To emphasize the variation of $R_H$ in superconducting state, we plot the data on a semi-logarithmic scale. Note that $R_H$ reveals strong temperature dependence in both the normal and superconducting states. Above $T_c(8\text{T}) \approx 25$ K, $R_H$ is positive and increases with decreasing temperature. Similar to previous observations on bulk MgB$_2$ [25], the magnitude of the normal-state $R_H$ is essentially two orders of magnitude smaller than that of high-$T_c$ cuprates, reflecting a much higher carrier concentration in MgB$_2$. More striking is
the behavior of $R_H$ in the mixed state. Below $T_{c}(H)$, $R_H$ decreases rapidly and changes sign from positive to negative. After reaching minimum $R_H^\text{min}$ at $T_{\text{min}}$, it increases again until zero is approached. The general behavior is qualitatively the same as that observed in high-$T_c$ cuprates.

We first discuss the normal-state Hall response. The temperature dependence of $R_H$ is anomalous when analyzed in terms of conventional transport theory. In a single-band Drude model, $R_H$ is $T$-independent if the anisotropy of the transport scattering rate $\tau^{-1}(k)$ is independent of $T$, where $k$ is the wave vector [18]. At present, it is unclear whether the anisotropy in $\tau^{-1}(k)$ varies with temperature for MgB$_2$. Recent band structure calculations predict that the Fermi surface of MgB$_2$ consists of four sheets: three are hole-like and one is electron-like [19, 20]. With such a complex Fermi surface, it is not surprising that $R_H$ is $T$-dependent. High-$T_c$ cuprate superconductors are the examples. In the latter system, $R_H$ is considered to be controlled by both transport scattering rate $\tau^{-1}$ and transverse scattering rate $\tau_H^{-1}$ [16]. Within this picture, $R_H(T)$ is expected to vary as

$$1/R_H = aT + b,$$  \hspace{1cm} (1)

where $a$ and $b$ are constants. Can Eq. 1 also be applied to MgB$_2$? In Fig. 2a, we plot the temperature dependence of the inverse Hall coefficient $1/R_H$ at $H = 8$ T. Interestingly, $1/R_H$ varies approximately linearly with $T$ at all temperatures between $T_c(H)$ and 300 K. By fitting the data with Eq. 1, we obtain $a = 5.06 \times 10^7$ C$^2$/m$^3$.K and $b = 1.66 \times 10^{10}$ C/m$^3$. The fitting result is illustrated in Fig. 2a as the solid line.

The above fitting procedure demonstrates the validity of Eq. 1 for MgB$_2$. However, we recall that Eq. 1 holds if $\tau^{-1} \propto T$ and $\tau_H^{-1} \propto T^2$ [16]. In this framework a linear $T$-dependence of the longitudinal resistivity $\rho$ is expected. Fig. 2b shows the temperature dependence of $\rho$ at $H = 0$ and 8 T. Though it decreases with decreasing $T$, the normal-state $\rho$ deviates from linearity below $\sim 200$ K and flattens as $T_c$ is approached from high temperature. Measurements on bulk samples [15] and wires [3] are qualitatively similar. However, we cannot exclude the possibility that the measured $\rho$ may not represent the in-plane resistivity $\rho_{ab}$. If the temperature dependence of $c$-axis resistivity $\rho_c$ is distinctly different from $\rho_{ab}$, the measured $\rho$ for polycrystals can deviate significantly from $\rho_{ab}$. While experimental investigation of the resistivity anisotropy is lacking, theoretical calculations predict a nearly isotropic resistivity, which can be described by a standard Bloch-Gr"uneisen (BG) expression [21]. The BG formula for the resistivity can be written as

$$\frac{\rho - \rho_0}{A} = (4\pi)^2 \left(\frac{2T}{\Theta_D}\right)^5 \int_0^{\Theta_D/2T} dx \frac{x^5}{\sinh^2(x)},$$  \hspace{1cm} (2)

where $\rho_0$ is the residual resistivity, $A$ is a $T$-independent constant and $\Theta_D$ is the Debye temperature. Using $\Theta_D = 746$ K [8], we model our resistivity data between 40 and 300 K using Eq. 2, yielding $\rho_0 = 5.9$ $\mu\Omega$ cm and $A = 0.37$ $\mu\Omega$ cm. As shown in Fig. 2b, Eq. 2 (solid line) describes our data fairly well, indicating the importance of electron-phonon interaction in MgB$_2$. In this circumstance, it is of surprise that the linear $T$-dependence of $1/R_H$ persists down to $T_c(H)$ even in the region that $\tau^{-1} \propto T$ no longer holds.

Since $R_H$ depends on two scattering rates for high-$T_c$ cuprates, more attention has been paid to a simpler model. As shown in Fig. 2b, Eq. 2 (solid line) describes our data fairly well, indicating the importance of electron-phonon interaction in MgB$_2$. In this circumstance, it is of surprise that the linear $T$-dependence of $1/R_H$ persists down to $T_c(H)$ even in the region that $\tau^{-1} \propto T$ no longer holds.

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turate dependence in the normal state, i.e.,
\[
cot \theta_H = \alpha T^2 + \beta, \quad T > T_c,
\]
where \(\alpha\) and \(\beta\) are constants. For MgB\(_2\), \(\cot \theta_H = R_H/H/\rho\) is shown in Fig. 3 plotted as \(\cot \theta_H\) vs. \(T^2\). It should be noted that the data fall on an approximately straight line in the whole temperature range between \(T_c(8T)\) and 300 K. The solid line in Fig. 3 shows a fit to Eq. 3 with \(\alpha = 4.9e-3\) K\(^{-2}\) and \(\beta = 128\). The \(T^2\)-dependence of \(\cot \theta_H\) and thus of \(\tau_H^{-1}\) suggests that \(\tau_H \neq \tau\), leading to the unconventional Hall response in MgB\(_2\).

We now consider the Hall effect in the superconducting state. Shown in Fig. 4a is the temperature dependence of \(R_H\) at \(H=2, 4, 6\) and 8 T, respectively. At each applied field, the \(R_H\) vs. \(T\) curve exhibits the same feature. Surprisingly, the sign reversal of \(R_H\) appears not only in high fields but also persists in low fields. An increase of \(H\) pushes \(R_H(T)\) curve to lower temperatures, i.e., \(T_{\min}\) decreases with increasing \(H\). However, the magnitude of \(R_H^{\min}\) seems to depend on \(H\) non-monotonically. It first increases and then decreases with increasing \(H\). Although similar features have been observed in all high-\(T_c\) cuprates and few BCS superconductors, it is interesting that sign change of \(R_H\) also occurs in MgB\(_2\). To assure that the sign change of \(R_H\) is not due to inhomogeneous superconductivity, we simultaneously measured the longitudinal resisitivty. As shown in Fig. 4b, \(\rho\) does not only reveal a sharp transition in zero field but also decreases smoothly with \(T\) in applied fields without showing any step or kink throughout the entire transition regime. As reported previously \([24]\), an increase of applied magnetic field results in a shift of the resistive transition to lower temperatures with appreciable broadening.

Fig. 4 clearly indicates that the sign change of \(R_H\) occurs before \(\rho\) reaches zero. This strongly suggests that the Hall anomaly is a consequence of vortex dynamics. For conventional superconductors, a phenomenological flux-flow model was developed by Nozieres and Vinen \([23]\) that takes into account the hydrodynamic magnus force. In this model, the mixed-state Hall resistivity \((\rho_H = E_y/j_x)\) is given by:
\[
\rho_H = (\epsilon \tau/m) \rho_n H, \quad H < H_{c2}.
\]
Here \(m\) is the effective mass of the normal electrons, \(\rho_n\) is the normal-state longitudinal resistivity, and \(H_{c2}\) is the upper critical field. As presented in Fig. 5, \(\rho_H\) varies perfectly linearly with \(H\) at \(T = T_c(H = 0) = 38\) K. Below 38 K, it deparls markedly from this behavior at fields smaller than \(H_{c2}\). In this regime, \(\rho_H\) increases rapidly with \(H\) after reaching a negative peak \(\rho_H^{\min}\) at \(H_{\min}\) to merge with the normal-state behavior. It is interesting to note that the value of \(\rho_H^{\min}\) remains more or less the same below 34 K, though \(H_{\min}\) increases with decreasing temperature. It seems that a lower limit for \(\rho_H^{\min}\) sets in at a temperature slightly below 38 K. To the best of our knowledge, such a feature has not been seen in any other superconductors. Nevertheless, our data in Fig. 5 demonstrates that Eq. 4 fails to describe the unusual H-dependence of \(\rho_H\) of MgB\(_2\).

In spite of many different approaches, the mechanism responsible for the Hall sign reversal remains controversial. For cuprate superconductors, models have been proposed \([24, 25, 26, 27, 28]\) suggesting that the sign change is related to flux pinning, backflow of thermally excited quasiparticles, layered structure, a vortex-glass transition, or imbalance of the electron density between the center and the far outside the region of the vortices. In the absence of a more detailed analysis than is available for these scenarios, their usefulness in explaining our results re-
mains uncertain. However, it should be pointed out that the scaling behavior between $\rho_H$ and $\rho$ does not seem to hold for MgB$_2$, suggesting that the sign reversal is not a consequence of a vortex-glass transition [27]. Given the fact that the sign change persists at temperatures well below $T_c$, the backflow scenario should also be ruled out [25]. In addition, we find that the Hall conductivity $\sigma_H$ cannot be described by $\sigma_H = -C/H + DH$, where C and D are positive constants. Therefore, models based on time-dependent Ginzburg-Landau theory [29, 30, 31] may fail to quantitatively explain our data in mixed state. Clearly, to find out whether the Hall anomaly arises from the layered nature of MgB$_2$, further experiments on single crystals would be desirable.

In summary, we have measured the Hall effect on well-characterized films of MgB$_2$ in both the normal and superconducting states. The Hall quantities exhibit many features that are strikingly similar to those found in high-$T_c$ cuprates. Although these similarities may be accidental, it is more probable that these similarities are clues to understanding the high transition temperatures found in both MgB$_2$ and the cuprates.

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[1] J. Nagamatsu, N. Nakagawa, T. Muranaka, Y. Zenitani, and J. Akimitsu, Nature 410, 63 (2001).
[2] A.S. Cooper et al., Proc. National Acad. Sci. USA 67, 313 (1970).
[3] L. Leyarovska and E. Leyvarovski, J. Less Common Metals 67, 249 (1979).
[4] S.L. Bud’ko et al., Phys. Rev. Lett. 86, 1877 (2001).
[5] G. Karapetrov et al., cond-mat/0102312 (2001).
[6] A. Sharoni, I. Felner, and O. Milo, cond-mat/0102325 (2001).
[7] H. Kotegawa et al., cond-mat/0102334 (2001).
[8] H. Tou et al., cond-mat/0103484 (2001).
[9] C. Walti, E. Felder, C. Degen, G. Wigger, R. Monnier, B. Delley, and H. Ott, cond-mat/0102522 (2001).
[10] B. G. R.K. Kremer and K. Ahn, cond-mat/0102432 (2001).
[11] S.L. Li and H.H. Wen and Z.W. Zhao et al., cond-mat/0103032 (2001).
[12] C. Panagopoulos, B. Rainford, T. Xiang, C. Scott, M. Kambara, and I. Inoue, cond-mat/0103060 (2001).
[13] A.A. Zhukov, K. Yates, G.K. Perkins et al., cond-mat/0103587 (2001).
[14] J. Hirsch, cond-mat/0102115 (2001).
[15] W.N. Kang, C.U. Jung, K.H.P. Kim et al., cond-mat/0102479 (2001).
[16] P. Anderson, Theory of Superconductivity in High-$T_c$ Cuprates (Princeton University Press, Princeton, 1997).
[17] M. Paranthaman et al., Appl. Phys. Lett. (in press) (2001).
[18] C. Kurd, The Hall effect and its applications (Plenum, New York, 1980).
[19] J. Kortus et al., cond-mat/0101446 (2001).
[20] S.V. Shulga et al., cond-mat/0103154 (2001).
[21] Y. Kong, O. Dolgov, O. Jepsen, and O. Anderson, cond-mat/0102499 (2001).
[22] Y. Takano et al., (unpublished) (2001).
[23] P. Nozieres and W. Vinen, Philos. Mag. 14, 667 (1966).
[24] Z.D. Wang et al., Phys. Rev. Lett. 72, 3875 (1994).
[25] R. Ferrell, Phys. Rev. Lett. 68, 2524 (1992).
[26] J.M. Harris et al., Phys. Rev. Lett. 73, 610 (1994).
[27] J. Luo et al., Phys. Rev. Lett. 68, 690 (1992).
[28] Z.D. Wang et al., Phys. Rev. Lett. 72, 3875 (1994).
[29] M.V. Feigel’man et al., JETP Lett. 62, 834 (1995).
[30] A. Dorsey, Phys. Rev. B 46, 8376 (1994).
[31] V. Geshkenbein and A. Larkin, Phys. Rev. Lett. 73, 609 (1994).
[32] N.B. Kopnin et al., J. of Low Temp. Phys. 90, 1 (1993).