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Unusual Hall effect in superconducting MgB_2 films

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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 ($T_{c0}=38.0$ 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 (to 8 T). We find that $R_H^{-1} \propto T(40–300\ \mathrm{K})$ and $\cot\theta_H \propto T^2(100–300\ \mathrm{K})$. As the sample is cooled 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.

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The recent discovery[1] of unexpectedly high temperature superconductivity in MgB_2—a material with no *d* electrons—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,[4] an isotropic energy gap,[5,6] NMR studies,[7,8] and specific heat measurements.[9,10] Evidence that the superconductivity may be unconventional can be found in the non-BCS-like temperature dependence of both penetration depth[11,12] and microwave surface resistance.[13]

In the study of high-T_c cuprates, the T-linear behavior of the inverse Hall coefficient is one of their most remarkable and puzzling properties.[14] In a one band model with an isotropic scattering rate τ^{-1}, the Hall coefficient R_H is predicted to be T independent because it is only the anisotropy of τ, and not its magnitude, that contributes to R_H.[15] In a multiband metal containing both electrons and holes, a strongly T-dependent R_H is much less surprising. If, for example, the mobility of each band has a different temperature dependence, or if thermal expansion produces a redistribution of the carriers among the bands, then a strong temperature dependence of R_H can be easily rationalized. Nevertheless, although not prohibited by theory, $R_H^{-1} \propto T$ is rare in metals.[16]

In this communication, we report an inverse Hall coefficient $R_H^{-1} \propto T$ in superconducting MgB_2 films. The experiments show that R_H is positive in the normal state. Further analysis yields a T^2 dependence of the Hall angle from 100 to 300 K. Although this behavior may be due to the complex Fermi surface of MgB_2, the data bear a qualitative resemblance to the Hall response of high-T_c cuprates and may reflect similar underlying physics if, for example, two quasi-two-dimensional (2D) bands (see below) are the major contributors to the Hall coefficient of MgB_2. 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, although similar behaviors have been observed in a few conventional BCS superconductors as well.

The MgB_2 films were prepared using a precursor post-processing approach and extensively characterized as described in Ref. 17. For the films used in the Hall measurements, the zero-field resistivity indicates an onset $T_c^{\mathrm{onset}} = 38.6$ K with a transition width of $\Delta T = 0.4$ K [see Fig. 2(b)], indicating the good quality of our samples. For the Hall measurements, we cut the samples into a rectangular shape with dimensions of $4\times 2\ \mathrm{mm}^2$. 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 °C for 4 h. The experiments were performed in a Quantum Design Physical Property Measurement System using a horizontal rotator and magnetic fields up to 8 T. 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., $\rho_H = [\rho_H(+H)\text{-}\rho_H(-H)]/2$. 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 μm thick MgB_2 film. The applied field was 8 T. To emphasize the variation of R_H in a superconducting state, we plot the data on a semilogarithmic scale. Note that R_H reveals strong T dependence in both the normal and superconducting states. Above $T_c(8\ \mathrm{T}) \sim 25$ K, R_H is positive and increases with decreasing temperature. Similar to previous observations on bulk MgB_2,[18] the magnitude of the normal-state R_H is essentially two orders of magnitude smaller than that of high-T_c cuprates. 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 a minimum $R_H^{\min}$ at $T_{\min}$, it increases again until zero is approached. The general behavior is qualitatively the same as that observed in hole-doped high-T_c cuprates.

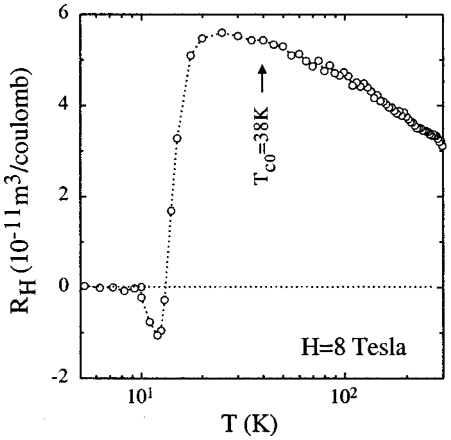

FIG. 1. Temperature dependence of $R_H(H=8\text{T})$ of a MgB_2 film plotted on a logarithmic temperature scale. Note the sign change of R_H in the mixed state.

We first discuss the normal-state Hall response. In a single-band model, as mentioned above, R_H is T independent if the anisotropy of τ^{-1} is independent of T. In a multiband model, R_H is a weighted sum of the contributions from each band; if the scattering rate of each band has a different T dependence, the weighted sum changes with temperature and can produce a T dependent R_H. Recent band structure calculations predict that the Fermi surface of MgB_2 consists of four sheets: three are holelike and one is electronlike.[19] Two of the holelike bands, derived from $p_{x,y}$ orbitals, have 2D character and contribute over 30% of the states at the Fermi level.[19] At present, it is unclear whether the T-dependent R_H of MgB_2 is caused by multiband effects or is a reflection of an unusual transport mechanism. In high-T_c cuprates, though a one-band model is thought to be appropriate, R_H is considered to be controlled by both a transport scattering rate τ^{-1} and a transverse scattering rate τ_H^{-1}.[14] Within this picture, $R_H(T)$ is expected to vary as[14]

$$R_H^{-1}=aT+b, \tag{1}$$

where a and b are constants. Can Eq. (1) also be applied to MgB_2? In Fig. 2(a), we plot the temperature dependence of the inverse Hall coefficient R_H^{-1} at $H=8$ T. Interestingly, R_H^{-1} varies approximately linearly with T at all temperatures between $T_c(H)$ and 300 K. By fitting the data with Eq. (1), we obtain $a=5.06\times10^7$ C/m^3 K and $b=1.66\times10^{10}$ C/m^3. The fitting result is illustrated in Fig. 2(a) as the solid line.

The above fitting procedure demonstrates the validity of Eq. (1) for MgB_2. The fact that $R_H^{-1}\propto T$ is rare in metals[16] supports the notion that the Hall response of MgB_2 may be anomalous. However, we recall that Eq. (1) holds if $\tau^{-1}\propto T$ and $\tau_H^{-1}\propto T^2$.[14] In this framework a linear T dependence of the longitudinal resistivity ρ is expected. Figure 2(b) shows the temperature dependence of ρ at $H=0$ and 8 T. Though it decreases with decreasing T, the normal-state ρ deviates from linearity below $\sim$100 K. Measurements on bulk samples[18] and wires[4] are qualitatively similar. However, we cannot exclude the possibility that the measured ρ may not represent the in-plane resistivity ρ_{ab}. If the T dependence of

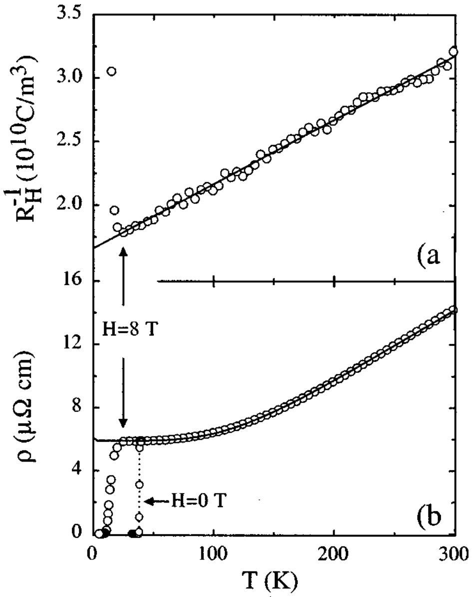

FIG. 2. Temperature dependence of (a) R_H^{-1} and (b) longitudinal resistivity ρ. The solid lines are fits to experimental data using Eqs. (1) and (2), respectively.

c-axis resistivity ρ_c is distinctly different from ρ_{ab}, the measured ρ for polycrystals can deviate significantly from ρ_{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-Grüneisen (BG) expression.[20] The BG formula for the resistivity can be written as

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

where ρ_0 is the residual resistivity, A is a T-independent constant, and Θ_D is the Debye temperature. Using $\Theta_D=746$ K,[9] 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. 2(b), Eq. (2) (solid line) describes our data fairly well, indicating the importance of electron-phonon interaction in MgB_2. In this circumstance, it is surprising that the linear T dependence of R_H^{-1} persists down to $T_c(H)$ even in the region that $\tau^{-1}\propto T$ no longer holds.

Though the in-plane resistivity of high-T_c materials can also be described by Eq. (2),[21] their Hall response is unusual in a one-band picture because $\tau_H\neq\tau$. For MgB_2, the similar T dependence of R_H^{-1} to high-T_c cuprates may be accidental owing to multiband effects. Due to the lack of detailed information about each individual band, any model we construct would contain such a large number of adjustable parameters that no conclusions could be drawn from such an analysis. On the other hand, if the quasi-2D bands derived from $p_{x,y}$ orbitals dominate the Hall response of MgB_2, it is plausible that the qualitatively similar behavior of R_H in high-T_c cuprates and MgB_2 is a manifestation of similar underlying physics. In this case, the Hall angle $\cot\theta_H=\omega_c^{-1}\tau_H^{-1}$ (ω_c is

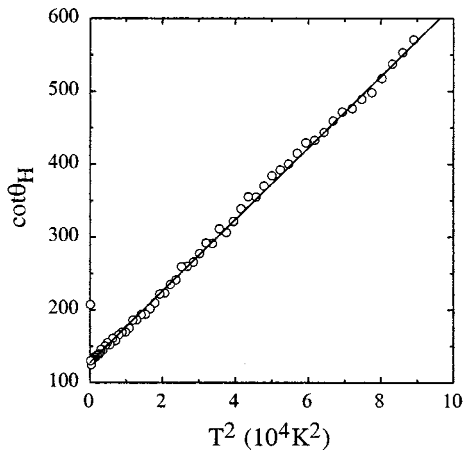

FIG. 3. Hall angle cot θ_H vs temperature between $T_c(H)$ and 300 K at $H=8$ T. The solid line is fit to experimental data above 100 K using Eq. (3).

the cyclotron frequency) is expected to exhibit a quadratic temperature dependence in the normal state,[14] i.e.,

$$\cot\theta_H = \alpha T^2 + \beta, \quad T > T_c, \tag{3}$$

where α and β are constants. For MgB_2, $\cot\theta_H = \rho/R_H H$ 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 temperature range between 100 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 τ_H^{-1} indicates that $\tau_H \neq \tau$, suggesting an unconventional Hall response in MgB_2.

We now consider the Hall effect in the superconducting state. Shown in Fig. 4(a) is the temperature dependence of R_H at $H=2$, 4, 6, and 8 T, respectively. At each applied field,

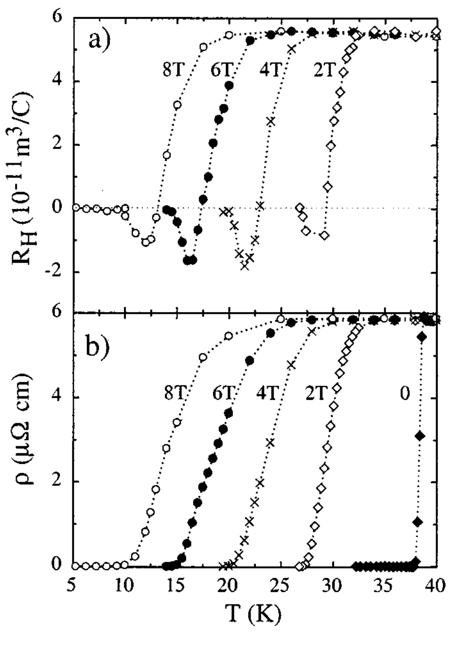

FIG. 4. Temperature dependence of (a) R_H and (b) longitudinal resistivity ρ at $H=2$, 4, 6, and 8 T.

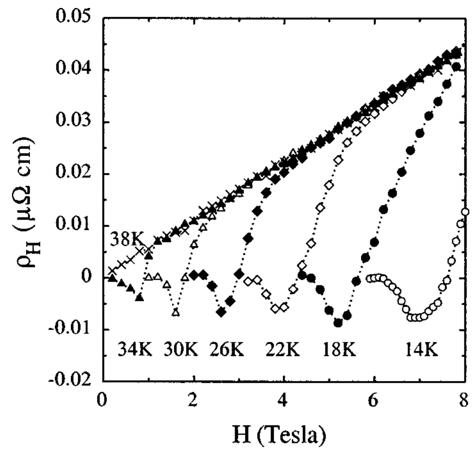

FIG. 5. Magnetic field dependence of the Hall resistivity ρ_H of MgB_2 at $T=14$, 18, 22, 26, 30, 34, and 38 K.

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 the $R_H(T)$ curve to lower temperatures, i.e., $T_{\min}$ decreases with increasing H. However, the magnitude of $R_H^{\min}$ seems to depend on H nonmonotonically. 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 the 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 resistivity. As shown in Fig. 4(b), ρ 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,[22] an increase of applied magnetic field results in a shift of the resistive transition to lower temperatures with appreciable broadening.

Figure 4 clearly indicates that the sign change of R_H occurs before ρ 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

$$\rho_H = (e\tau/m)\rho_n H, \quad H < H_{c2}. \tag{4}$$

Here m is the effective mass of the normal electrons, ρ_n is the normal-state longitudinal resistivity, and H_{c2} is the upper critical field. As presented in Fig. 5, ρ_H varies perfectly linearly with H at $T=T_c(H=0)=38$ K. Below 38 K, it departs markedly from this behavior at fields smaller than H_{c2}. In this regime, ρ_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 ρ_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[24–26] 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 region of the vortices. In the absence of a more detailed analysis than is available for these scenarios, their usefulness in explaining our results remains uncertain. However, it should be pointed out that the scaling behavior between ρ_H and ρ does not seem to hold for MgB_2, suggesting that the sign reversal is not a consequence of a vortex-glass transition.[25] Given the fact that the sign change persists at temperatures well below T_{c0}, the backflow scenario should also be ruled out.[26] In addition, we find that the Hall conductivity σ_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[27] may fail to quantitatively explain our data in mixed state. Clearly, to find out whether the Hall anomaly arises from the layered nature or electronic band structure 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 unusual electrical transport properties of both MgB_2 and the cuprates.

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[1] J. Nagamatsu, N. Nakagawa, T. Muranaka, Y. Zenitani, and J. Akimitsu, Nature (London) **410**, 63 (2001).

[2] A.S. Cooper, E. Corenzwit, L.D. Longinotti, B.T. Matthias, and W.H. Zachariasen, Proc. Natl. Acad. Sci. U.S.A. **67**, 313 (1970).

[3] L. Leyarovska and E. Leyvarovski, J. Less-Common Met. **67**, 249 (1979).

[4] S.L. Bud'ko, G. Lapertot, C. Petrovic, C.E. Cunningham, N. Anderson, and P.C. Canfield, Phys. Rev. Lett. **86**, 1877 (2001).

[5] G. Karapetrov, M. Iavarone, W.K. Kwok, G.W. Crabtree, and D.G. Hinks, Phys. Rev. Lett. **86**, 4374 (2001).

[6] A. Sharoni, I. Felner, and O. Millo, Phys. Rev. B **63**, 220508 (2001).

[7] H. Kotegawa, K. Ishida, Y. Kitaoka, T. Muranaka, and J. Akimitsu, Phys. Rev. Lett. **87**, 127001 (2001).

[8] H. Tou, H. Ikejiri, Y. Maniwa, T. Ito, T. Takenobu, K. Prassides, and Y. Iwasa, cond-mat/0103484 (unpublished).

[9] Ch. Walti, E. Felder, C. Degen, G. Wigger, R. Monnier, B. Delley, and H. R. Ott, Phys. Rev. B **64**, 172515 (2001).

[10] R.K. Kremer, B.J. Gibson, and K. Ahn, cond-mat/0102432 (unpublished).

[11] S.L. Li, H.H. Wen, Z.W. Zhao, Y.M. Ni, Z.A. Ren, G.C. Che, H.P. Yang, Z.Y. Liu, and Z.X. Zhao, Phys. Rev. B **64**, 094522 (2001).

[12] C. Panagopoulos, B.D. Rainford, T. Xiang, C.A. Scott, M. Kambara, and I.H. Inoue, Phys. Rev. B **64**, 094514 (2001).

[13] A.A. Zhukov, L.F. Cohen, K. Yates, G.K. Perkins, Y. Bugoslavsky, M. Polichetti, A. Berenov, J.L.M. Driscoll, A.D. Caplin, L. Hao, and J. Gallop, Supercond. Sci. Technol. **14**, L13 (2001).

[14] P.W. Anderson, *The Theory of Superconductivity in High-T_c Cuprates* (Princeton University Press, New Jersey, 1997).

[15] C.M. Hurd, *The Hall Effect in Metals and Alloys* (Plenum Press, New York, 1972).

[16] W.W. Schulz, P.B. Allen, and N. Trivedi, Phys. Rev. B **45**, 10 886 (1992).

[17] M. Paranthaman, C. Cantoni, H.Y. Zhai, H.M. Christen, T. Aytug, S. Sathyamurthy, E.D. Specht, J.R. Thompson, D.H. Lowndes, H.R. Kerchner, and D.K. Christen, Appl. Phys. Lett. **78**, 3669 (2001).

[18] W.N. Kang, C.U. Jung, K.H.P. Kim, M.S. Park, S.Y. Lee, H.J. Kim, E.M. Choi, K.H. Kim, M.S. Kim, and S.I. Lee, cond-mat/0102313 (unpublished).

[19] J. Kortus, I.I. Mazin, K.D. Belashchenko, V.P. Antropov, and L.L. Boyer, Phys. Rev. Lett. **86**, 4656 (2001); S.V. Shulga, S.-L. Drechsler, H. Eschrig, H. Rosner, and W.E. Pickett, cond-mat/0103154 (unpublished).

[20] Y. Kong, O.V. Dolgov, O. Jepsen, and O.K. Andersen, Phys. Rev. B **64**, 020501 (2001).

[21] P.B. Allen, Nature (London) **412**, 494 (2001).

[22] Y. Takano, H. Takeya, H. Fujii, H. Kumakura, T. Hatano, K. Togano, H. Kito, and H. Ihara, Appl. Phys. Lett. **78**, 2914 (2001).

[23] P. Nozieres and W.F. Vinen, Philos. Mag. **14**, 667 (1966).

[24] Z.D. Wang, J. Dong, and C.S. Ting, Phys. Rev. Lett. **72**, 3875 (1994); J.M. Harris, N.P. Ong, and Y.F. Yan, *ibid.* **73**, 610 (1994); M.V. Feigel'man, V.B. Geshkenbein, A.I. Larkin, and V.M. Vinokur, Pis'ma Zh. Éksp. Teor. Fiz. **62**, 811 (1995) [JETP Lett. **62**, 834 (1995)]; Anne van Otterlo, M. Feigel'man, V. Geshkenbein, and G. Blatter, Phys. Rev. Lett. **75**, 3736 (1995).

[25] J. Luo, T.P. Orlando, J.M. Graybeal, X.D. Wu, and R. Muenchausen, Phys. Rev. Lett. **68**, 690 (1992).

[26] R.A. Ferrell, Phys. Rev. Lett. **68**, 2524 (1992).

[27] A.T. Dorsey, Phys. Rev. B **46**, 8376 (1994); V.B. Geshkenbein and A.I. Larkin, Phys. Rev. Lett. **73**, 609 (1994); N.B. Kopnin, B.I. Ivlev, and V.A. Kalatsky, J. Low Temp. Phys. **90**, 1 (1993).