Large Anisotropic Normal-State Magnetoresistance in Clean MgB₂ Thin Films

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We report a large normal-state magnetoresistance with temperature-dependent anisotropy in very clean epitaxial MgB₂ thin films (residual resistivity much smaller than 1 $\mu\Omega$ cm) grown by hybrid physicalchemical vapor deposition. The magnetoresistance shows a complex dependence on the orientation of the applied magnetic field, with a large magnetoresistance ($\Delta\rho/\rho_0 = 136\%$) observed for the field $H \perp ab$ plane. The angular dependence changes dramatically as the temperature is increased, and at high temperatures the magnetoresistance maximum changes to $H \parallel ab$. We attribute the large magnetoresistance and the evolution of its angular dependence with temperature to the multiple bands with different Fermi surface topology in MgB₂ and the relative scattering rates of the σ and π bands, which vary with temperature due to stronger electron-phonon coupling for the σ bands.

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The existence of multiple conduction bands in MgB_2 is the most remarkable characteristic of this 40-K superconductor [1,2]. The Fermi surface of MgB₂ consists of four disconnected sheets formed by the boron orbitals: two holelike σ bands, a holelike and an electronlike π bands [3]. The interband scattering between the σ and π bands is small [4]. The electron-phonon coupling with the boron bond-stretching E_{2g} mode is stronger for the σ bands than the π bands, giving rise to two superconducting energy gaps [1,2,5]. It also leads to a stronger contribution to the temperature dependence of resistivity from the σ bands than the π bands [4]. The multiband effect not only influences various properties of MgB₂ [6-8] but also presents opportunities to observe new physical phenomena that do not exist in single-band superconductors [9-11]. In this Letter, we report a large normal-state magnetoresistance with complex and temperature-dependent anisotropy in MgB₂ thin films grown by hybrid physical-chemical vapor deposition (HPCVD) [12]. The angular dependence of magnetoresistance has been studied for decades as a powerful probe into the Fermi surface of crystals [13-15], and its application to MgB₂ can provide valuable insight into the unique properties of multiband systems. However, the existing MgB2 single crystals are not clean enough (residual resistivity above 1 $\mu\Omega$ cm [16,17] and mean free path ≤ 1000 Å [17,18]) for observing large magnetoresistance. The HPCVD films used in this work are very clean (residual resistivity much less than 1 $\mu\Omega$ cm) and epitaxial (single crystal-like), allowing us to observe the temperature-dependent anisotropy which could not be observed in the previous magnetoresistance studies using polycrystalline MgB₂ samples [19–22].

When a magnetic field (in the z direction) is applied perpendicular (transverse) to an electric current flowing along a thin conductor strip (in the x direction), the Lorentz force deflects the electrons resulting in a Hall field in the y direction. For a single-band free-electron system, the Hall field exactly balances the Lorentz force, and the electron moves as if in zero field without being deflected; thus, there is no magnetoresistance. However, if there are two or more bands, in particular, if the carriers are of different types, the Hall field cannot exactly cancel the Lorentz force, and there will be magnetoresistance [13-15]. As MgB₂ has four Fermi surfaces with both electrons and holes [3], large transverse magnetoresistance can be expected in MgB₂. In the reciprocal space, electrons under a magnetic field orbit the Fermi surface in a plane perpendicular to the magnetic field with a cyclotron frequency $\omega_c \equiv eB/m^*$ [15]. Using an m^* of 0.3–1.2 m_e [18], a v_F of 4.4 \times 10⁵ m/s [7], and the residual resistivity of samples in this work, we estimate that $\omega_c \tau$, where τ is the relaxation time, is mostly ≥ 1 at low temperatures for the fields studied in this work. In other words, the electron orbits the Fermi surface one or more times before it is scattered. In this condition, the details of the Fermi surface are becoming more important in affecting the magnetoresistance than the scattering process. Because the magnitude of magnetoresistance scales with $(\omega_c \tau)^2$ [13], large magnetoresistance requires very clean samples with large τ and thus low resistivity.

The MgB₂ films used in this work are grown by HPCVD [12,23] on both (0001) 4H-SiC and (0001) sapphire substrates. The films grow epitaxially with the *c* axis normal to the substrate surface. The films are around 2000–4000 Å in thickness, and their T_c values are around 40 K. The magnetoresistance was measured at Penn State (up to 9 T), Institute of Physics, CAS (up to 12 T), and NHMFL, Tallahassee (up to 18 T) either by fixing the temperature and sweeping the field or by fixing the field and sweeping



FIG. 1. Resistivity versus temperature curves for a MgB₂ film under different magnetic fields. (a) $H \perp ab$. (b) $H \parallel ab$. In both figures, the magnetic fields are, from the bottom curves to the top curves, 0, 3, 5, 7, and 9 T, respectively.

the temperature. MgB₂ films made with HPCVD are very clean with residual resistivity as low as 0.26 $\mu\Omega$ cm [24], which makes it possible to observe large magnetoresistance. In addition, the single crystal-like structure in the epitaxial films allows magnetoresistance measurement with the field along different crystallographic orientations.

Figure 1 shows resistivity versus temperature curves for a MgB₂ film on a SiC substrate at different magnetic fields applied (a) perpendicular $(H \perp ab)$ and (b) parallel $(H \parallel$ *ab*) to the film plane. A large magnetoresistance $\Delta \rho / \rho_0$, where ρ_0 is the zero-field resistivity, was observed in the normal state when a perpendicular magnetic field $H \perp ab$ was applied. The magnitude of the magnetoresistance became smaller with increasing temperature as the zero-field resistivity became higher. There was also magnetoresistance when $H \parallel ab$, but it was considerably smaller than for $H \perp ab$. In Fig. 2, the magnetic field dependence of the resistivity (left scale) and magnetoresistance $\Delta \rho / \rho_0$ (right scale) of the film is plotted for T = 45 K. Again, one sees a large anisotropic normal-state magnetoresistance. At $\mu_0 H = 18$ T, $\Delta \rho / \rho_0$ is 136% for $H \perp ab$ and 42% for $H \parallel ab$. The low resistivity (0.34 $\mu\Omega$ cm) of the MgB₂ film is critical for the large magnetoresistance. For samples with residual resistivity $\geq 1-2 \ \mu\Omega$ cm, the maximum magnetoresistance is only a few percent.

The anisotropy of the magnetoresistance was further measured by applying the magnetic field at a different angle θ with respect to the film surface, i.e., the *ab* plane of the MgB₂ film. Figure 3 shows the angular dependence of the resistivity at different magnetic fields and T = 45 K. The measurement temperature is sufficiently far from the superconducting transition; therefore, the result is not influenced by the superconducting fluctuations. A complex structure is found in the angular dependence at high magnetic fields, where the Fermi surface effects are more significant. Besides a minimum at $H \parallel ab \ (\theta = 0^\circ)$ and a



FIG. 2. Resistivity of a MgB₂ film as a function of the applied magnetic field measured at T = 45 K. The results are shown for $H \perp ab$ and $H \parallel ab$. A magnetoresistance as large as 135% is obtained for $H \perp ab$.

maximum at $H \perp ab$ ($\theta = 90^{\circ}$), there is an additional dip near 80° and a shoulder near 40°. At low magnetic fields, the angular dependence is simpler, with a dip at 90° and the broad shoulder shifting towards the $H \perp ab$ direction.

The angular dependence changes dramatically when the temperature increases. In Fig. 4, the magnetic field dependence (left panels) and angular dependence (right panels) of the magnetoresistance for a MgB₂ film are shown for 60 K [(a) and (b)], 100 K [(c) and (d)], and 120 K [(e) and (f)]. At low temperatures, the magnetoresistance is larger for $H \perp ab$ than for $H \parallel ab$ and the angular dependence is rather complex. A completely opposite behavior is observed at high temperatures, where magnetoresistance is larger for $H \parallel ab$ than for $H \perp ab$. The resistivity follows



FIG. 3. Resistivity of a MgB₂ film as a function of the angle between the applied magnetic field and the film surface measured at T = 45 K. The results are shown for different applied magnetic fields.



FIG. 4. Magnetic field dependence [(a), (c), and (e)] and angular dependence [(b), (d), and (f)] of magnetoresistance of a MgB₂ film. The results are for T = 60 K [(a) and (b)], 100 K [(c) and (d)], and 120 K [(e) and (f)]. A change of angular dependence is seen as the temperature is increased.

a simple angular dependence. The crossover occurs at around 100 K for this sample, when minima are seen for both $H \parallel ab$ and $H \perp ab$ directions.

We now discuss the possible origin of the large magnetoresistance and the temperature-dependent anisotropy in the MgB₂ films. For a single-band free-electron system, the solution of the Boltzmann equation in a magnetic field, satisfying the condition $j_y = 0$, predicts a zero magnetoresistance. However, in a multiband solid, assuming that each band has an average σ_i , ω_{ci} , and τ_i , the total conductivity is [14,15]

$$\sigma = \frac{\left(\sum_{i} \frac{\sigma_{i}}{1+\omega_{ci}^{2}\tau_{i}^{2}}\right)^{2} + \left(\sum_{i} \frac{\sigma_{i}\omega_{ci}\tau_{i}}{1+\omega_{ci}^{2}\tau_{i}^{2}}\right)^{2}}{\sum_{i} \frac{\sigma_{i}}{1+\omega_{ci}^{2}\tau_{i}^{2}}}.$$
 (1)

In order to illustrate the essential physical picture, we simplify Eq. (1) for the low-field limit $\omega_c \tau \ll 1$, to yield the magnetoresistance $\Delta \rho / \rho_0$ as

$$\frac{\Delta\rho}{\rho_0} \simeq \frac{1}{2} \frac{\sum_i \sum_{j \neq i} \sigma_i \sigma_j (\omega_{ci} \tau_i - \omega_{cj} \tau_j)^2}{\left(\sum_i \sigma_i\right)^2}.$$
 (2)

For an electronlike band $\omega_{ci} < 0$, and for a holelike band $\omega_{ci} > 0$. If *i* and *j* bands are of the same types of carriers, $\omega_{ci}\tau_i$ and $\omega_{cj}\tau_j$ in the $(\omega_{ci}\tau_i - \omega_{cj}\tau_j)^2$ term reduce each other; they add up when the carriers are electrons for one band and holes for the other. The weighting factor $\sigma_i\sigma_j$ suggests that a cleaner band with larger conductivity will have a larger contribution. In MgB₂, there are three hole-like bands $(\sigma_1, \sigma_2, \text{ and } \pi_2)$ and one electronlike band (π_1) with different effective masses [3,18], leading to large $(\omega_{ci}\tau_i - \omega_{cj}\tau_j)^2$ terms and large magnetoresistance.

The magnetoresistance anisotropy can be explained by the highly anisotropic Fermi surface of MgB₂. When the applied field direction changes, the orbiting plane of the electron intercepts the Fermi surface at different angles. As a result, the effective masses and conductivities for each bands have to be considered as tensors and the Fermi velocities as vectors. However, the principle described above should still be valid: The interplay between the σ_i , ω_{ci} , and τ_i of the four bands for the specific field direction determines the magnetoresistance for that direction. A full account of the angular dependence with fine structures should be accessible by a numerical solution to the Boltzmann equation in field, taking into account the Fermi surface topology and charge scattering of the four bands in MgB₂. Qualitatively, when $H \perp ab$, ω_{ci} is larger for the σ bands, in which the carriers orbit the small circumference of the tubular Fermi surface, than for the π band, in which the carriers orbit the large circle of the honeycomb around the Γ point. For $H \parallel ab$, the σ -band orbits are open ($\omega_{ci} = 0$) and σ_i is small for the 2dimensional σ bands, while the π carriers orbit the small circumference of the tube forming the honeycomb Fermi surface (larger ω_{ci}). The diminishing contributions from the σ bands result in the smaller magnetoresistance for $H \parallel$ ab at low temperatures. Recently, Pallecchi et al. have obtained solutions for $H \perp ab$ and $H \parallel ab$ by considering only the diagonal elements of the effective mass tensors [25], which are in qualitative agreement with the result in Fig. 2.

Assuming that the Fermi surface does not vary much with temperature, the change in the angular dependence with temperature can be explained by the relative change of scattering in the σ and π bands. As the temperature increases, σ_i is reduced more rapidly for the σ bands than for the π bands due to the stronger electron-phonon interaction, and the contributions of the π bands to magnetoresistance become more important. Therefore, the angular dependence of the magnetoresistance is dominated by the anisotropy of the σ -band Fermi surface at low temperatures and by the π bands at high temperatures. In that case, the larger π band ω_{ci} for $H \parallel ab$ than for $H \perp ab$ gives rise to a larger magnetoresistance at high temperatures



FIG. 5. Kohler plot for a MgB₂ film: (a) $H \perp ab$ and (b) $H \parallel ab$. The insets are results at low magnetic fields. The Kohler rule is not obeyed.

when $H \parallel ab$. Such a profound change of angular dependence of magnetoresistance with temperature is a vivid demonstration of new physical phenomena that cannot be observed in single-band systems.

Another display of the multiband effect is the breakdown of the Kohler's rule. According to the Kohler's rule, if only one relaxation time τ exists in a solid, then $\Delta \rho / \rho_0$ should be a function of H/ρ_0 , and the results from different temperatures should collapse to a single curve in a Kohler plot, $\Delta \rho / \rho_0$ versus H / ρ_0 [15]. Figure 5 is the Kohler plot of a MgB₂ film for (a) $H \perp ab$ and (b) $H \parallel ab$. Clearly, the Kohler's rule is not obeyed: The results from different temperatures do not overlap with each other for both field orientations. This is the consequence of multiple bands with different relaxation times, which depend differently on temperature due to different electron-phonon coupling [4]. The result is contrary to the previous reports that the Kohler's rule is obeyed in MgB₂ [19–21], which may be explained by the defect, impurity scattering, and polycrystalline nature of those samples.

In conclusion, large anisotropic normal-state magnetoresistance is observed in clean epitaxial MgB_2 thin films. The large magnetoresistance is explained by the existence of the multiple bands of different types of carriers, and the complex angular dependence is attributed to the anisotropy of the Fermi surface of the different bands. The remarkable change in the angular dependence of magnetoresistance with temperature and the breakdown of the Kohler's rule are excellent examples of phenomena made possible by the multiband effect. The rich field, temperature, and angular dependencies of the magnetoresistance provide significant new insights into the interplay of the σ and π band scattering and electron-phonon coupling in MgB₂.

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- [1] H.J. Choi *et al.*, Nature (London) **418**, 758 (2002).
- [2] P.C. Canfield and G. Crabtree, Phys. Today 56, No. 3, 34 (2003).
- [3] J. Kortus et al., Phys. Rev. Lett. 86, 4656 (2001).
- [4] I.I. Mazin et al., Phys. Rev. Lett. 89, 107002 (2002).
- [5] A. Y. Liu, I. I. Mazin, and J. Kortus, Phys. Rev. Lett. 87, 087005 (2001).
- [6] A. Gurevich, Phys. Rev. B 67, 184515 (2003).
- [7] A. Brinkman et al., Phys. Rev. B 65, 180517(R) (2002).
- [8] T. Dahm and D. J. Scalapino, Appl. Phys. Lett. **85**, 4436 (2004).
- [9] D. F. Agterberg, E. Demler, and B. Janko, Phys. Rev. B 66, 214507 (2002).
- [10] A. Gurevich and V. Vinokur, Phys. Rev. Lett. 90, 047004 (2003).
- [11] E. Babaev, Phys. Rev. Lett. 89, 067001 (2002).
- [12] X.H. Zeng et al., Nat. Mater. 1, 35 (2002).
- [13] C. Kittel, *Quantum Theory of Solids* (Wiley, New York, 1993).
- [14] J. Callaway, *Quantum Theory of the Solid State* (Academic, New York, 1976).
- [15] J.M. Ziman, *Electrons and Phononss*, Classics Series (Oxford University Press, New York, 2001).
- [16] T. Masui et al., Physica (Amsterdam) 378C, 216 (2002).
- [17] U. Welp et al., Physica (Amsterdam) **385C**, 154 (2003).
- [18] A. Carrington et al., Phys. Rev. Lett. 91, 037003 (2003).
- [19] X.H. Chen et al., Phys. Rev. B 65, 024502 (2002).
- [20] D.K. Finnemore et al., Phys. Rev. Lett. 86, 2420 (2001).
- [21] S.L. Bud'ko et al., Phys. Rev. B 63, 220503(R) (2001).
- [22] I. Pallecchi et al., Phys. Rev. B 71, 104519 (2005).
- [23] X.X.Xi et al., IEEE Trans. Appl. Supercond. 13, 3233 (2003).
- [24] A. V. Pogrebnyakov *et al.*, Appl. Phys. Lett. **82**, 4319 (2003).
- [25] I. Pallecchi et al., Phys. Rev. B 72, 184512 (2005).