Transport and magnetic properties on MgB₂ thin films

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Magnetotransport data on MgB $_2$ thin films, fabricated on Al $_2$ O $_3$ substrates using electron – bean deposition and Mg diffusion method are reported for applied magnetic field up to 9T. The upper critical field anisotropy, lower critical field and irreversibility field versus temperature were determined. The Hall coefficient is slightly temperature dependent and positive in normal state. Using the extracted data, the electronic mean free path, coherence length ξ_0 , anisotropic coefficient γ and penetration depth λ were calculated.

(Received December 2, 2005; accepted May 18, 2006)

Keywords: MgB2 thin films, Magnetotransport properties, Hall effect, Magnetization

1. Introduction

In the last 8 years there has been a renew interest in the intermetalic superconductors which incorporate light elements such as boron, due to the discovery of the new class of borocarbides RE-TM₂B₂C where RE=Y, Lu, Er, Dy and other rare earth and TM= Ni or Pd [1,2]. The main characteristics of these compounds are the very high critical temperature $T_{\rm c}$ among intermetalics ($T_{\rm c}\sim23$ K), the anisotropic layered structure and a strong interplay between magnetism and superconductivity [3].

 MgB_2 known since early 1950's, but only recently discovered to be superconductor [4] at a high critical temperature about 39 K has a simple hexagonal structure. This discovery of superconductivity in MgB_2 , have sparked renewed interest in the connection between superconducting properties and electronic structural features of non-copper oxide materials. Subsequent, experimental studies showed that this material not only has a much higher T_c than ordinary metallic superconductors, but also has significantly different superconductivity properties [4-13].

MgB₂ superconductivity announcement was the catalyst for the discovery of several superconductors, TaB₂ with $T_c = 9.5$ K, BeB_{2.75} with $T_c = 0.7$ K, graphite sulphur composites with T_c=35 K. The C-S composites are similar materials with MgB_2 , electronically crystallographically [14]. The critical temperature of T_c=39 K is close to or above the theoretical value predicted from BCS theory [15], but the isotope effect is reduced [11] from the BCS values of 1/2. The specific heat measurements [12], photoemission [16] and tunneling spectra [9], shown low energy excitations suggesting a secondary energy gap. Two-gap superconductivity in MgB2 was first predicted theoretically. The two gaps in MgB₂ are associated with different parts of Fermi surface, which is composed of four separates sheets [17].

The manifestation of MgB_2 electronic structure in macroscopic quantities such as the superconducting critical fields and the anisotropic parameter

$$\gamma = \frac{H_{c2}^{ab}}{H_{c2}^{c}} = \frac{\xi_{ab}}{\xi_{c}}$$
 has attracted much attention. Here

 H_{c2} and ξ are upper critical fields and Ginsburg-Landau coherence length, respectively. Reported values of the anisotropy coefficients are ranging between 1 - 6 depending on the measurements technique and on sample type, i.e. single crystals, oriented films, aligned crystallites or powders [18].

In this article magnetoresistivity, Hall effect and magnetic properties have been investigated on MgB₂ thin films fabricated on Al₂O₃ (1001) substrates using electronbeam deposition and Mg diffusion method.

2. Experimental

Thin films of MgB₂ were fabricated on Al_2O_3 (1001) substrates, using electron-beam deposition and Mg diffusion method. An amorphous B film was first deposited at the room temperature in a vacuum of 10^{-6} Torr. The B films with excess Mg were sealed in a Ta tube, in 3 kPa Argon atmosphere.

The sealed Ta tube was sealed in quartz tube under vacuum argon and heated into furnace to $600\,^{\circ}\text{C}$ in 5 minutes. The temperature was then increased to $900\,^{\circ}\text{C}$ and held approximately 20-30 minutes at this temperature. The reaction ampoule was then removed from furnace and quenched to room temperature. X-ray diffraction analyses confirmed the samples to be MgB₂ single phase.

Measurements of temperature and field dependent electrical resistivity and magnetization were performed in Quantum Design MPMS and PPMS systems. Resistivity and Hall constant measurements were performed using a standard DC five probe geometry technique in the

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temperature range 4-300 K, using silver epoxy to make contacts.

3. Results and discussions

Fig. 1 presents temperature dependence of the electrical resistivity data for MgB_2 films taken at a variety of applied magnetic fields, from zero up to 9 T. It is clearly seen the effect of the magnetic field: a suppression of the superconducting phase to lower temperatures for increasing applied magnetic field, and there is a clear large magnetoresistivity in the normal state. On the other hand what can be clearly seen is that there is a substantial loss of the scattering associated with cooling the samples from 300 K down to 39 K.

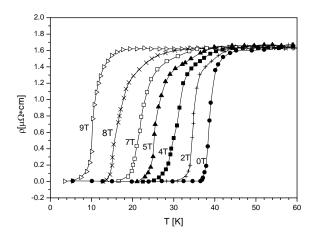


Fig. 1. Temperature dependent resistivity of the MgB₂ thin film in magnetic field up to 9 T.

The residual resistivity ratio $\rho(300 \text{ K})/\rho(40 \text{ K})$ is close to 18. The substantial magnetorestivity observed in the normal state is a consequence of the very low normal state resistivity. Using these and taking $v_F = 4.7 \times 10^7 \, cm/s$ we can calculate electronic mean free path 1=530 Å. When this length scale is compared with the superconducting coherence length ξ_0 , it becomes clear that MgB₂ is deep at the limit of superconductivity, $1 >> \xi_0$. A low resistivity is also very important for high current application. The upper critical field H_{c2}(T) data extracted from the magnetotransport data are plotted in Fig. 2.

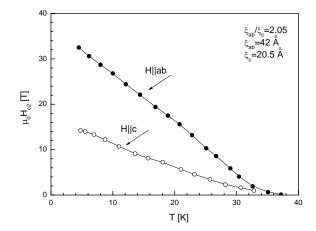


Fig. 2. Temperature dependence of H_{c2} for MgB_2 thin film with H||ab| and H||c.

 $H_{c2}(T)$ with $H\|ab$ rises to slightly above 37 T and $H_{c2}(T)$ with $H\|c$ rises to slightly above 18 T about two times higher.

Anisotropy is very important both for basic understanding of this material and practical applications, strongly affecting the pinning and critical currents. The anisotropy degree of MgB₂ is still unresolved, reports giving values ranging between 1.1 and 9.

The anisotropy ratio $\gamma = H \frac{||ab|}{c^2} / H \frac{||c|}{c^2}$ is reported to be between 1.1 and 1.7 for textured bulk and partial oriented crystallites; 1.2 -2.0 for c-axis oriented films, 1.7-2.7 in single crystals and 5-9 for powders [19-23].

From our data we found $\gamma = H_{c2}^{\parallel ab} / H_{c2}^{\parallel c} = 2.05$, in agreement with the values reported by others authors.

In order to deduce the values of the anisotropic coherence lengths from the upper critical fields we used the anisotropic Ginsburg-Landau theory equations: for the magnetic field applied along c-axis $H \begin{vmatrix} c \\ c \end{vmatrix}^2 = \frac{\Phi_0}{2\pi\xi_{ab}}^2$, and for the magnetic field applied in the ab-plane $H \begin{vmatrix} ab \\ c \end{vmatrix}^2 = \frac{\Phi_0}{2\pi\xi_{ab}\xi_c}$, where Φ_0 is the flux quantum, ξ_{ab} , ξ_c are the coherence lengths along ab-plane and c-axis.

Using these relations, we can extracted and estimate $\xi_{ab} = 42\,$ Å and $\xi_{c} = 20.5\,$ Å.

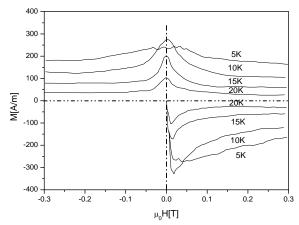


Fig. 3. Magnetization hysteresis loops for a MgB₂ thin film and different temperature between 5 and 20 K using a SQUID magnetometer.

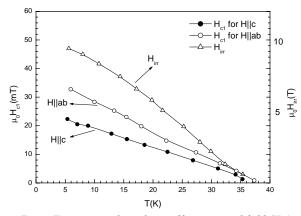


Fig. 4. Temperature dependence of lower critical field (H_{cl}) and irreversibility field (H_{irr}) .

In Fig. 3 are plotted the magnetization hysteresis loops for a MgB_2 thin films at different temperatures between 5 and 20 K using a SQUID magnetometer (MPMS). The curves present the fluctuations in magnetization at T=10 and 5 K, while only regular behavior is seen at the higher temperatures. These fluctuations indicate that numerous flux jumps are taking place. The amplitude of the fluctuations is found to be the largest near 10 K. This unexpected behavior is typical for such films, when space-resolved magnetic measurements are performed.

In Fig. 4 are plotted the anisotropic lower critical fields for MgB₂ thin films. Most of the values for all samples are situated between 28 mT and 38 mT. The London penetration depth deduced from the lower critical field data range between 900 Å and 1500 Å for $H_{c1}^{\parallel c}(0)$ and $H_{c1}^{\parallel ab}(0)$.

The irreversibility fields at zero Kelvin for MgB₂ thin films samples are about 10 T, as can be seen in Fig. 4. The critical current densities at 4.2 K and 1T magnetic field, calculated from our data, are of order of 10⁶-10⁷ A/cm², in

agreement with data have been reported by others [21, 24, 25].

Fig. 5 shows the temperature dependence of Hall coefficient in MgB_2 thin films. The fact that the normal state Hall coefficient R_H is positive, the charge carriers in magnesium diboride are holes with a density at 300 K of about $2.5 \times 10^{26} \, holes/m^3$, for an magnetic field applied parallel with c-axis oriented film.

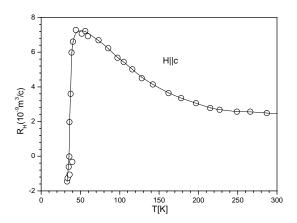


Fig. 5. Hall coefficient versus temperature for MgB₂ thin film.

4. Conclusions

In this article we present detailed the transport properties of MgB_2 thin films in magnetic field up to 9T with the critical temperature of 39 K.

The MgB_2 thin films were fabricated on Al_2O_3 substrates using electron-beam deposition and Mg diffusion method.

In the superconducting state we find that MgB₂ epitaxial thin films has a relatively linear H_{c2}(T) curves which leads to H_{c2}(0)=37T for H||ab and H_{c2}(0)=18T for H||c with an anisotropic coefficient $\gamma = 2.05$ and coherence lengths $\xi_{ab} = 42 \text{Å}$ and $\xi_c = 20.5 \text{Å}$. The penetration depth deduced from the lower critical field data range between 900 Å and 1500 Å for $H_{c1}^{\parallel c}(0) = 28 \, \text{mT}$ and $H_{c1}^{\parallel ab}(0) = 38 \, \text{mT}$. The critical current densities at 4.2 K and H=1T range between 10^6 - 10^7 Å/cm². The Hall

4.2 K and H=1T range between 10^6 - 10^7 A/cm². The Hall coefficient is positive and holes density at 300 K is about 2.5×10^{26} holes/m³ for magnetic field applied parallel with the c-axis of oriented films. The superconductivity in MgB₂ is not predicted a priori and for understanding the mechanism of superconductivity of these new materials more experimental and theoretical research are necessary.

References

[1] R. J. Cava, H. Takagi, H. W. Zandbergen, J. J. Krajewski, W. F. Peck, T. Siegrist, B. Batlogg, R. B.

- van Dover, R. J. Felder, K. Mizuhashi, J. O. Lee, H. Eisaki, S. Uchida, Nature **367**, 252 (1994).
- [2] R. Nagarojan, C. Mazumdar, Z. Hossian, S. K. Dhar, K. V. Gopalakrishnan, L. C. Gupta, B. D. Padalia, R. Vijayraghavan, Phys. Rev. Lett. 72, 274 (1994).
- [3] H. Eisaki, H. Takaji, R. J. Cava, B. Batlogg, J. J. Krajewski, W. F. Jr. Phys. Rev. B. 50, 647 (1994).
- [4] J. Nagamatsu, N. Nakagawa, T. Muranaka, Y. Zenitani, J. Akimitsu, Nature 410, 63 (2001).
- [5] H. J. Choi, M. L. Cohen, S. G. Louie, Physica C 385, 66 (2003).
- [6] P. C. Canfield, S. L. Bud'ko, D. K. Finnemore, Physica C 385, 1 (2003).
- [7] S. L. Bud'ko, G. Lapertot, C. Petrovic, C. E. Cunningham, N. Anderson, P. C. Canfield, Phys. Rev. Lett. 86, 1877 (2001).
- [8] B. A. Glowaki, M. Majoros, M. Vickers, J. E. Evetts, Y. Shi, I. McDougall, Supercond. Sci. Technol. 14, 193 (2001).
- [9] M. R. Eskildsen, M. Kugler, G. Levy, S. Tanaka, J. Jun, S. M. Kazakov, J. Karpinski, O. Fischer, Physica C 385, 169 (2003).
- [10] T. Masui, S. Tajima, Physica C 385, 91 (2003).
- [11] D. G. Hinks, H. Claus, J. D. Jorgensen, Nature 411, 457 (2001).
- [12] Y. Wang, T. Plackowski, A. Jund, Physica C 385, 205 (2003).
- [13] S. Tsuda, T. Yokoya, T. Kiss, Y. Takano, K. Togano, H. Kito, H. Ihara, S. Shin, Phys. Rev. Lett. 87, 177006 (2001).
- [14] C. Buzea, T. Yamashita, Supercond. Sci. Techology, 14(11), 115 (2001).
- [15] W. L. McMillan, Phys. Rev. B 167, 331 (1998).

- [16] S. Tsuda, T. Kiss, Y. Takano, K. Togano, H. Kito, H. Ihara, S. Shin, Phys. Rev. Lett. 87, 177006 (2001).
- [17] A. Y. Liu, I. I. Mazin, J. Kortus, Phys. Rev. Lett. 87, 087005, (2001).
- [18] U. Welp, A. Rydh, G. Karapetrov, W. K. Kwok, G. W. Crabtree, C. Marcenat, L. M. Paulius, Lyard T. Klein, J. Marcus, S. Blanchard, P. Samuely, P. Szabo, A. G. M. Jansen, K. H. P. Kim, C. U. Jung, M. S. Lee, B. Kang, S. I. Lee, Physica C 385, 154 (2003).
- [19] O. F. de Lima, R. A. Ribeiro, M. A. Avila, C. A. Cardoso, A. A. Coelho, Phys. Rev. Lett 86, 5974 (2001).
- [20] C. U. Jung, M. S. Kang Park, W. N. Kang, M. S. Kim, S. Y. Lee, S. I. Lee, Physica C 353, 162 (2001).
- [21] S. Patnaik, L. D. Cooley, A. Gurevich, A. A. Polyanski, J. Jiang, X. Y. Cai, A. A. Squitieri, M. T. Naus, M. K. Lee, J. H. Choi, L. Belenky, S. D. Bu, J. Letteri, X. Song, D. G. Schlon, S. E. Babcock, C. B. Eom, E. E. Hellstrom, D. C. Larbalestier, Supercond. Sci. Technol, 14315 (2001).
- [22] S. L. Bud'ko, C. Petrovic, G. Lapertot, C. E. Cunningham, P. C. Canfield, M. H. Jung, A. H. Lacerda, Phys. Rev. B 63, 220503 (2001).
- [23] S. L. Bud'ko, V. G. Kogan, P. C. Canfield, Preprint, cod-mat/ 0106577.
- [24] Y. Bugoslavsky, L. F. Cohen, G. K. Perkins, M. Polichetti, T. J. Tate, R. Gwillian, A. D. Caplin, Nature, 411561, (2001).
- [25] M. Dhalle, P. Toulemonde, D. Beneduce, N. Musolino, M. Decroux, R. Flukiger, Physica C 363, 3, 155 (2001).

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