Single-Crystal like MgB₂ thin films grown on *c*-cut sapphire substrates

Pham Van Duong, Mahipal Ranot, and Won Nam Kang*

BK21 Physic Division and Department of Physic, Sungkyunkwan University, Suwon 440-746, Republic of Korea

(Received 4 September 2014; revised or reviewed 29 September 2014; accepted 29 September 2014)

Abstract

Single-crystal like MgB_2 thin film was grown on (000*l*) Al_2O_3 substrate by using hybrid physical-chemical vapor deposition (HPCVD) system. Single crystal properties were studied by X-ray diffraction (XRD) and the full width at half maximum (FWHM) of the (0001) MgB_2 peak is 0.15° , which is very close to that has been reported for MgB_2 single-crystal. It indicates that the crystalline quality of thin film is good. Temperature dependence on resistivity was investigated by physical property measurement system (PPMS) in various applied fields from 0 to 9 T. The upper critical field (H_{c2}) and irreversibility field (H_{irr}) were determined from PPMS data, and the estimated values are comparable with that of MgB_2 single-crystals. The thin film shows a high critical temperature (T_c) of 40.4 K with a sharp superconducting transition width of 0.2 K, and a high residual resistivity ratio (RRR=21), it reflects that MgB_2 thin film has a pure phase structure.

Keywords: MgB2 thin film, Single-crystal, HPCVD.

1. INTRODUCTION

Since Nagamatsu et al., have discovered in 2001 that MgB_2 is a superconductor with high critical temperature (T_c) of 39K [1], a huge effort was expended in basic research to understand and enhance the properties of this material. Due to these efforts, many interesting results about MgB_2 were found.

MgB₂ is a conventional BCS superconductor with two superconducting gaps $(\Delta_{\sigma} \text{ and } \Delta_{\pi})$, in which the Cooper pairs are formed through electron-phonon coupling [2], [3]. It has a simple crystal structure, less anisotropy and larger coherence length than the high- T_c superconductors [4-5], and high critical current density (J_c) [6-7]. This implies that MgB2 is a very promising material for application in superconducting devices. However, the of investigations maiority were performed polycrystalline samples. Therefore, to understand fully the intrinsic properties of this material, investigations on single crystals and high quality thin films should be performed. The fabrication of high quality thin film is the first crucial step towards device applications. We used hybrid physical-chemical vapor deposition (HPCVD) technique because it meets the requirement of hightemperature and pressure required for the synthesis of single-crystal like MgB₂ thin film. In this work, we deposited single-crystal like MgB₂ thin films on c-cut sapphire substrates. The θ - 2θ and ϕ -scans were carried on the thin films by X-ray diffraction (XRD), and superconducting properties were studied by physical property measurement system (PPMS).

2. EXPERIMENT

The HPCVD technique is a combination of physical vapor deposition and chemical vapor deposition, and it is very effective in making high quality MgB₂ thin films. The working process of HPCVD system has been described in detail in some articles [7-9]. The high purity (99.999%) Mg pieces were placed into the groove of stainless-steel susceptor. A c-cut Al₂O₃ substrate of 10 mm x 10 mm in size was also placed at the center of the top surface of susceptor. The total pressure inside the quartz tube was kept stable at 210 Torr with 100 sccm flow rate of H₂ gas (6N purity). The substrate and Mg pieces were inductively heated to 650 °C. Then, we introduced B₂H₆ gas (5% concentration in H₂) with flow rate of 50 sccm and the deposition of MgB₂ started. The deposition was stopped when B₂H₆ flow was shut down. The susceptor was cooled down to room temperature in flowing H₂ gas. The phase purity and crystallinity of thin film were analyzed by x-ray diffraction (XRD). The PPMS system was used to investigate superconducting and transport properties of thin film.

3. RESULT AND DISCUSSION

Fig. 1 shows the temperature dependence of resistivity of the single-crystal like MgB_2 thin film. The T_c onset of the film is observed to be 40.4 K. The origin of high T_c in our film is most probably the epitaxial strain generated in the growth process [10]. The magnified view of a sharp superconducting transition from 40.4 K to 40.2 K is also presented in the inset of figure. The resistivity value of

^{*} Corresponding author: wnkang@skku.edu

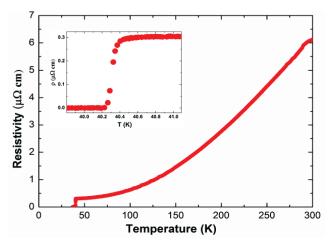


Fig. 1. Temperature dependence of resistivity for single-crystal like MgB_2 thin film grown on Al_2O_3 substrate. The inset shows the magnified view of superconducting transition in the temperature region from 39 to 41 K.

the film at room-temperature (300 K) is 6.2 $\mu\Omega$ cm, and it decreased to 0.3 $\mu\Omega$ cm at 42 K. The low residual resistivity suggests a very long electron mean free path [11]. The residual resistivity ratio (RRR) of MgB₂ thin film is as high as 21, it indicates that MgB₂ thin film has a high purity phase [12].

The X-ray diffraction (XRD) θ -2 θ scan of MgB₂ film grown on c-cut sapphire substrate is shown in Fig. 2. There are only (000l) diffraction peaks from MgB₂ except the substrate peak, this suggesting the phase purity and high c-axis orientation in our film. The full width at half maximum (FWHM) obtained from the rocking curve of (0001) peak is 0.15° (inset of figure 2), which is very close to that has been reported for MgB₂ single-crystal [13], indicating that the crystalline quality of the film is good.

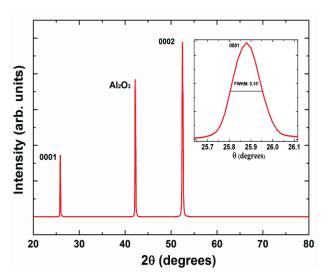
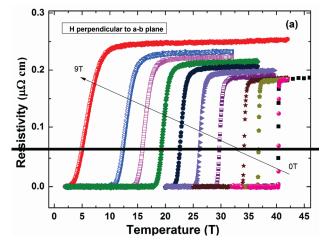


Fig. 2. The X-ray diffraction of single-crystal like MgB_2 thin film grown on (0001) Al_2O_3 substrate. The inset shows the full width at half maximum (FWHM) obtained from the (0001) peak of MgB_2 .



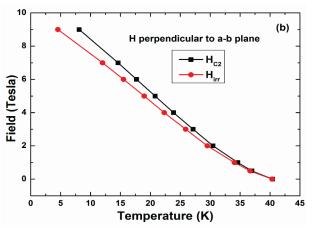


Fig. 3. (a) Resistivity transitions measured at various fields from 0 to 9 T when H was applied perpendicular to a-b plane of MgB_2 thin film. (b) Temperature dependence of H_{c2} and H_{irr} .

The upper critical field (H_{c2}) and irreversibility field (H_{irr}) were determined from resistivity measurements. Resistive transitions measured under applied field varying from 0 to 9 T are presented in Fig. 3a. The fields were applied perpendicular to the a-b plane of single-crystal like MgB_2 thin film. The H_{c2} and H_{irr} were determined from the criteria of 90 and 10% of its normal state resistance. Fig. 3b shows the temperature dependence of H_{c2} and H_{irr} .

For a two band superconductor, like MgB₂, the formula $H_{c2}(0) = 0.69T$ (dH_{c2}/dT)_{T=Tc}, which is derived from single-band Ginzburg-Landau theory could not be apply to calculate $H_{c2}(0)$. However, $H_{c2}(0)$ value for MgB₂ can be calculated by applying a theory developed for the two band superconductors [14]. Here, just for comparison, $H_{c2}(0)$ is estimated simply by extrapolation to zero K. We obtained $H_{c2}^{\perp ab} = 11$ T, which is slightly higher than that reported for MgB₂ single-crystal [5] but much lower than that reported for clean MgB₂ thin film grown on 6H-SiC substrate [15]. This result probably reflectes that the quality and the properties of our thin film is like that of MgB₂ single crystals.

4. CONCLUSION

In conclusion, we have grown c-axis oriented MgB₂ thin films with single-crystal quality on (0001) Al₂O₃ substrates. The crystalline structure and transport properties of the film were studied. The thin film shows a high critical temperature ($T_c = 40.4 \text{ K}$) and a sharp superconducting transition (0.2 K). The highly c-axis oriented peaks and high-purity phase without any impurity or secondary peaks were observed in the θ -2 θ scan of XRD. The H_{c2} value of thin film was found comparable with the value reported for MgB2 single crystal. These results would provide a new horizon in fabricating high-quality MgB2 thin film on inexpensive substrate sapphire for superconducting applications.

ACKNOWLEDGMENT

This work was supported by Mid-career Researcher Program through NRF grant funded by the Ministry of Education, Science & Technology (MEST) (No. 2010-0029136).

REFERENCES

- J. Nagamatsu, N. Nakagawa, T. Muranaka, Y. Zenitani, and J. Akimitsu, "Superconductivity at 39 K in magnesium diboride," Nature, 410, pp. 63-64, 2001.
- [2] J. Kortus, I. I. Mazin, K. D. Belashchenko, V. P. Antropov and L. L. Boyer, "Superconductivity of metallic Boron in MgB₂," *Phys. Rev. Lett.*, 86, pp. 4656, 2001.
- [3] S. Souma, Y. Machida, T. Sato, T. Takahashi, H. Matsui, S. C. Wang, H. Ding, A. Kaminski, J. C. Campuzano, S. Sasaki and K. Kadowaki, "The origin of multiple superconducting gaps in MgB₂," *Nature*, 423, pp. 65-67, 2003.
- [4] D. C. Larbalestier, L. D. Cooley, M. O. Rikel, A. A. Polyanskii, J. Jiang, S. Patnaik, X. Y. Cai, D. M. Feldmann, A. Gurevich, A. A. Squitieri, M. T. Naus, C. B. Eom, E. E. Hellstrom, R. J. Cava, K. A. Regan, N. Rogado, M. A. Hayward, T. He, J. S. Slusky, P. Khalifah, K. Inumaru and M. Haas, "Strongly linked current flow in polycrystalline forms of the superconductor MgB₂," *Nature*, 410, pp. 186-189, 2001.

- [5] M. Xu, H. Kitazawa, Y. Takano, J. Ye, K. Nishida, H. Abe, A. Matsushita, N. Tsujii and G. Kido, "Anisotropy of superconductivity from MgB₂ single crystals," *Appl. Phys. Lett.*, 79, pp. 2779, 2001.
- [6] S. H. Moon, J. H. Yun, H. N. Lee, J. I. Kye, H. G. Kim, W. Chung and B. Oh, "High critical current densities in superconducting MgB₂ thin films," *Appl. Phys. Lett.*, 79, pp. 2429, 2001
- [7] M. Ranot and W. N. Kang, "MgB₂ coated superconducting tapes with high critical current densities fabricated by hybrid physicalchemical vapor deposition," *Curr. Appl. Phys.*, 12, pp. 353-363, 2012.
- [8] W. K. Seong, J. Y. Huh, W. N. Kang, J. W. Kim, Y. S. Kwon, N. K. Yang and J. G. Park, "Growth of Epitaxial MgB₂ Thick films with Columnar Structures by Using HPCVD," *Chem. Vapor Deposition* 13, pp. 680-683, 2007.
- [9] W. K. Seong, W. N. Kang, S. J. Oh, J. K. Jung, C. J. Kim and J. Joo, "Superconducting property of single-crystal like MgB₂ thin film," *Physica C*, Vol. 470, pp. 1465–1467, 2010.
- [10] A. V. Pogrebnyakov, J. M. Redwing, S. Raghavan, V. Vaithyanathan, D. G. Schlom, S. Y. Xu, Qi Li, D. A. Tenne, A. Soukiassian, X. X. Xi, M. D. Johannes, D. Kasinathan, W. E. Pickett, J. S. Wu and J. C. H. Spence, "Enhancement of the Superconducting transition temperature of MgB₂ by a Strain-Induced Bond-Stretching mode softening," *Phys. Rev. Lett.*, 93, pp. 147006-1, 2004.
- [11] C. G. Zhuang, S. Meng, C. Y. Zhang, Q. R. Feng, Z. Z. Gan, H. Yang, Y. Jia, H. H. Wen and X. X. Xi, "Ultrahigh current-carrying capability in clean MgB₂ films," *J. Appl. Phys.*, 104, pp. 013924, 2008.
- [12] S. R. Chauhan and S. Chaudhary, "On the Residual Resistivity Ratio in MgB₂ Superconductors," *IEEE. Trans. Appl. Supercond.*, Vol. 20, No. 1, 2010.
- [13] T. Masui, S. Lee and S. Tajima, "Effect of the growing process on the electronic properties of MgB₂ single crystals," *Physica C*, Vol. 392–396, Part 1, pp. 281–285, 2003.
- [14] A. Gurevich, "Enhancement of the upper critical field by nonmagnetic impurities in dirty tow-gap superconductors," *Phys. Rev. B* 67, pp. 184515, 2003.
- [15] C. G. Zhuang, T. Tan, Y. H. Wang, S. S. Bai, X. B. Ma, H. Yang, G. H. Zhang, Y. S. He, H. Wen, X. X. Xi, Q. R. Feng and Z. H. Gan, "Clean MgB₂ thin films on different types of single-crystal substrate fabricated by hybrid physical-chemical vapor deposition," *Supercond. Sci. Technol.*, Vol. 22, pp. 025002, 2009.