Thick polycrystalline MgB_2 film on Cu substrate by hybrid physical–chemical vapour deposition

INSTITUTE OF PHYSICS PUBLISHING SUPERCONDUCTOR SCIENCE AND TECHNOLOGY
Supercond. Sci. Technol. **19** (2006) 1196–1199 doi:10.1088/0953-2048/19/11/018

Thick polycrystalline MgB_2 film on Cu substrate by hybrid physical–chemical vapour deposition

Fen Li, Tao Guo, Kaicheng Zhang, Li-ping Chen, Chinping Chen and Qing-rong Feng[1]

Department of Physics and State Key Laboratory for Artificial Structure and Mesoscopic Physics, Peking University, Beijing 100871, People's Republic of China

E-mail: qrfeng@pku.edu.cn

Received 14 July 2006, in final form 26 September 2006
Published 18 October 2006
Online at stacks.iop.org/SUST/19/1196

Abstract
Thick MgB_2 films have been grown on Cu substrates by the technique of hybrid physical–chemical vapour deposition (HPCVD). The films are about 2–3 μm thick and are quite dense. The T_c (onset) is as high as 37–38 K and sharp, ∼0.7 K. X-ray diffraction indicates that the films show a highly textured polycrystalline character. The upper critical field, $H_{c2}(0)$ at $T = 0$ K, is extrapolated as 15.3 T. The controlled growth of MgB_2 film on Cu substrate provides an alternative route for the preparation of MgB_2 tape materials.

1. Introduction

Since the discovery of the 39 K superconductivity in the simple binary compound MgB_2 [1], extensive research has been carried out on the fabrication of this superconductor in various forms, such as polycrystalline bulk samples, single crystals, metal clad tapes, wires, thin epitaxial films and thick polycrystalline films [2–10]. Compared to low-T_c superconductors, i.e., Nb, $NbSn_3$, etc, MgB_2 has a much higher T_c and larger energy gaps [11, 12]. This means a potentially higher speed. Increasing the operation temperature from helium temperature for Nb and $NbSn_3$ to 20 K for MgB_2 is a 'big deal'. A MgB_2 technology with high speed and 20 K operation, using a much cheaper and much reliable cryocooler than the facility necessary for the helium temperature operation, will make superconducting electronics much more competitive.

Various techniques have been applied, including powder-in-tube (PIT) [13–16] and diffusion of Mg vapour into B fibres [17], in an effort to find out an effective and efficient process to produce superconducting elements such as wires and tapes. One of the routes may rely on the fabrication of thick film on various substrates as a pre-stage toward the mass production of these superconducting elements. For example, thick MgB_2 films deposited on sapphire [9, 18], stainless steel [8], silicon carbide [10], etc, by HPCVD have been reported with the film thickness ranging from 1 to over 20 μm. The next step is to limit the flux jumping and stabilize the transport current in MgB_2. One of the solutions is to coat the MgB_2 film on a Cu layer, and then incorporate it into multifilamentary (MF) strands [19]. Recently, however, much attention has been drawn to the study of Cu-sheathed MgB_2 wires or tapes [20–23] due to the fact that Cu is superior to Fe in terms of the following four advantages: (1) high thermal and electrical conductivity, (2) high ductility, (3) nonmagnetic nature, and (4) cost effectiveness. These four factors are crucial to the cryogenic stability, wire drawing workability, AC loss, and large scale production of practical superconductors, respectively. In spite of the efforts, several questions remain open and need to be answered. For example, how to improve the uniformity of the superconducting phase in MgB_2 wires or tapes, how to improve the density of the superconducting wires or tapes, etc. Recently, we studied the reaction between Cu and Mg in detail, and have synthesized MgB_2 thick films on Cu substrate by HPCVD. In this report, we illustrate the fabrication of MgB_2 films on Cu substrate and their properties.

2. Experimental details

Experimentally, the reaction chamber inside which the vapour deposition takes place is similar to that reported

[1] Author to whom any correspondence should be addressed.

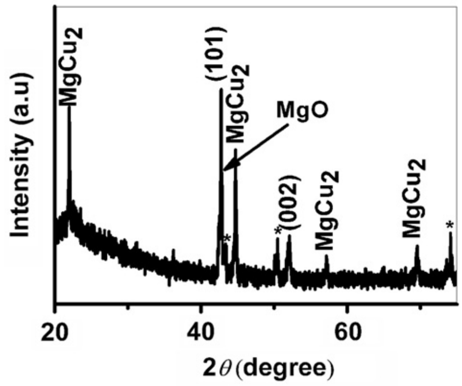

Figure 1. X-ray diffraction pattern of MgB_2 film grown on the Cu substrate.

previously [24]. A gas mixture of 75% B_2H_6 in H_2 along with Mg ingots was used as the active sources. The Mg ingots were placed around the substrate located on an iron sample holder, which can be heated up inductively by an rf generator to vapourize the Mg. The iron sample holder is supported by a columnar graphite with a larger heat capacity. So the temperature of the sample holder can be controlled more easily. Additional pure H_2 gas also flowed in the reaction chamber, serving to reduce the oxygen content and suppress any further oxidation of the sample during the deposition process. Also, the H_2 would cut down the decomposition rate of B_2H_6. The flow rate of the B_2H_6 mixture gas was about 5 sccm at a pressure of 1 kPa. A background gas mixture, H_2 + Ar (90%), ∼100 sccm at 19 kPa, was also flowing through the reaction chamber. In this case, the Mg has a lower melting point, ∼around 590 °C, than the reaction temperature of Mg with Cu, ∼630 °C. With this condition, the deposition temperature was regulated at 600 °C for about 10 min. The resulting deposition rate of a typical film was estimated to be of the order of 4 nm s^{-1}. This high growth rate can be ascribed to the higher concentration of B_2H_6, ∼75%.

The x-ray diffraction (XRD) analysis was performed using a Philip's X'pert-MRD diffractometer. The scanning electron microscope (SEM) observations were carried out with an FEI QUANTA 200 FEG SEM. The temperature-dependent resistance (ρ–T) was performed by a standard four-probe measurement using a Quantum Design PPMS-9 system, and the M–T measurement was performed on a Quantum Design MPMS-7.

3. Results and discussion

Figure 1 shows the highly textured MgB_2 crystal structure. Besides the diffraction peaks of MgB_2, MgO, and the Cu substrate marked by '*', there are some clear $MgCu_2$ peaks showing up in the spectrum, which come from the reaction between Mg and Cu in the deposition process. The (101) peak of the MgB_2 film is higher than that of the Cu substrate. This can be explained by two reasons. First, the thickness of the MgB_2 film is large and the crystallites are mostly (101) oriented. Second, the presence of the MgO peak also enhances the intensity of the MgB_2(101) peak.

The general feature of the film surface is shown in figures 2(a) and (b), in which (b) is a magnification showing a part of (a). The surface morphology reveals that the film is composed of many irregular MgB_2 crystallites. Their sizes are not uniform. The biggest crystallite is over 1 μm. Though the film surface is not smooth, it is dense. Figure 2(c) shows the cross-sectional image of the film, as marked by the two arrows. The average thickness is about 2.5 μm. A close view of the interface between the MgB_2 film and the substrate, marked by '*', has revealed that there exists a very thin interface layer in a lighter colour consisting of nanocrystallites, possibly $MgCu_2$. The thickness is difficult to estimate in the photograph. The presence of a $MgCu_2$ interface layer is

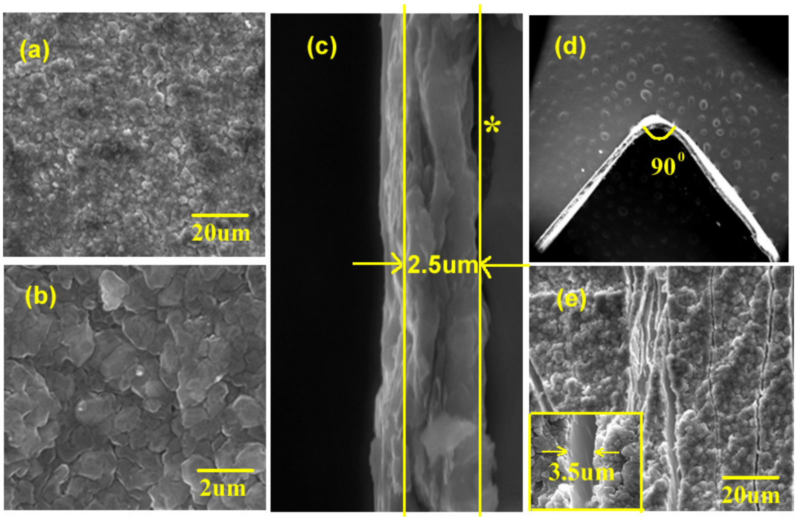

Figure 2. SEM images of the fabricated thick MgB_2 film. (a) Characteristic surface morphology of the MgB_2 thick film. (b) Magnified image of the surface morphology. (c) Cross-sectional image of the grown film; the mark '*' indicates the side of the film that is nearer to the substrate. (d) The sample bent to 90° with the film surface facing outwards at a curving radius of about 0.63 mm. (e) The surface morphology after the bending. The inset shows the magnified view for the largest crack, ∼3.5 μm.

(This figure is in colour only in the electronic version)

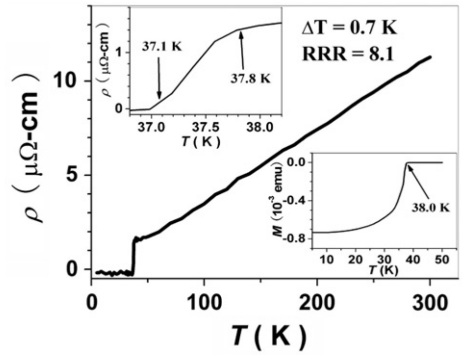

Figure 3. ρ–T measurement of the MgB_2 thick film. The upper left inset shows a magnified view near the transition region. The lower right inset is for the M–T measurement.

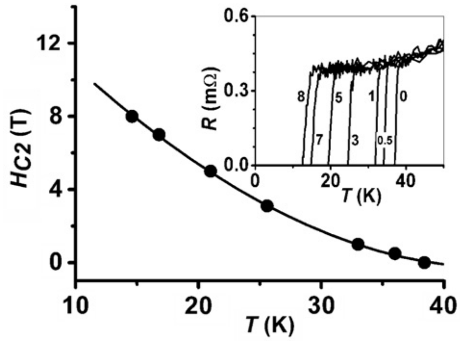

Figure 4. Upper critical field H_{C2} as a function of temperature. The inset is for the temperature-dependent resistance measurements under applied fields up to 8 T. The number by the side of each curve indicates the applied field in units of tesla.

consistent with the result reported by Yao *et al* [25]. It may be a crucial factor for the MgB_2 film strongly attaching to the Cu substrate. To examine the tenacity and the bonding strength of the film to the substrate, we bent the substrate as much as 90° with the film surface facing outward at a curving radius of about 0.63 mm; see figure 2(d). The corresponding surface morphology attributed to the bending is shown in figure 2(e). Although cracks appear in the film (the largest one is ~3.5 μm, shown in the inset of figure 2(e)), it still adheres to the Cu substrate firmly, showing much better mechanical quality of the film than that reported by Wang *et al*: 'the film appeared not to be firmly attached to the Cu substrate, a plate-like layer of the film could be easily removed from the substrate by a blade' [26].

The ρ–T curve is plotted in figure 3. The transition temperatures are determined as $T_C(\text{onset}) = 37.8$ K, higher than that obtained by Wang *et al* [26] and comparable with the Cu sheathed MgB_2 tapes reported by Wang [20]. Shown in the upper left inset of figure 3, the corresponding transition width is as narrow as 0.7 K. The lower right inset is for the M–T measurement performed on a Quantum Design MPMS-7. The superconducting transition is also sharp. This demonstrates that the superconducting phase is uniform in our film.

The upper critical field $H_{C2}(T)$ shown in figure 4 is obtained from the temperature-dependent resistance measurements under applied fields up to 8 T shown in the inset of figure 4. Then, $H_{C2}(0) = 15.3$ T is determined by fitting the data points of $H_{C2}(T)$ with the polynomial $H_{C2}(T) = H_{C2}(0) + A_1T + A_2T^2 + A_3T^3 + A_4T^4$ and extrapolating to $T = 0$ K. Figure 4 shows that $H_{C2}(T)$ is curved upwards near T_C. This agrees with the result of Bud'ko *et al* [27]. $H_{C2}(0)$ for the present sample is not very high in comparison with the value of 28 T obtained for an MgB_2 film electroplated to a stainless steel substrate [28]. This is possibly due to the presence of small impurities in our film.

4. Conclusion

We have deposited MgB_2 films on Cu substrates by HPCVD. The average thickness of the film is about 2.5 μm, and the films are quite dense. The transition temperature is T_C (onset), ~37.8 K, with a sharp transition width as narrow as 0.7 K. XRD analysis indicated that the films have a highly (101) and (002) textured structure. The upper critical field $H_{C2}(0)$ at $T = 0$ K was extrapolated as 15.3 T. The bending test indicated that the MgB_2 film adhered on the substrate tightly as the substrate was bent to 90° with a curving radius of about 0.63 mm. We believe that the deposition of thick MgB_2 films on Cu substrates is a potential technique leading to the mass production of MgB_2 superconducting wires, tapes, etc.

Acknowledgments

This work is supported by NSFC under contract No. 50572001. We appreciated the technical supports of Li Chen, and Yong-zhong Wang.

References

[1] Nagamatsu J, Nakagawa N, Muranaka T, Zenitani Y and Akimitsu J 2001 *Nature* **410** 63–4
[2] Buzea C and Yamashita T 2001 *Supercond. Sci. Technol.* **14** R115
[3] Larbalestier D C *et al* 2001 *Nature* **410** L53
[4] Canfield P C, Bud'ko S L and Finnemore D K 2003 *Physica* C **385** 1
[5] Flükiger R, Suo H L, Musolino N, Beneduce C, Toulemonde P and Lezza P 2003 *Physica* C **385** 286
[6] Kang W N, Choi E M, Kim H J and Lee S I 2003 *Physica* C **385** 24
[7] Lee S 2003 *Physica* C **385** 31
[8] Chen C P *et al* 2005 *Chin. Sci. Bull.* **50** 719
[9] Zhang K C, Ding L L, Zhuang C G, Chen L P, Chen C P and Feng Q R 2006 *Phys. Status Solidi* a **203** 2463
[10] Li F, Guo T, Zhang K C, Chen L P, Chen C P and Feng Q R 2006 *Supercond. Sci. Technol.* **19** 1073
[11] Tsuda S, Yokoya T, Kiss T, Takano Y, Togano K, Kito H, Ihara H and Shin S 2001 *Phys. Rev. Lett.* **87** 177006
[12] Schmidt H, Zasadzinski J F, Gray K E and Hinks D G 2002 *Phys. Rev. Lett.* **88** 127002
[13] Feng Y *et al* 2001 *Supercond. Sci. Technol.* **15** 12
[14] Fu B Q *et al* 2002 *J. Appl. Phys.* **92** 7341–4
[15] Eyidi D, Eibl O, Wenzel T, Nickel K G, Schlachter S I and Goldacker W 2003 *Supercond. Sci. Technol.* **16** 778–88
[16] Serquis A *et al* 2003 *Appl. Phy. Lett.* **82** 2847
[17] Canfield P C, Finnemore D K, Bud'ko S L, Ostenson J E, Lapertot G, Cunningham C E and Petrovic C 2001 *Phys. Rev. Lett.* **86** 2423
[18] Chen C P, Wang X F, Lu Y, Jia Z, Guo J P, Wang X N, Zhu M, Xu X Y, Xu J and Feng Q R 2004 *Physica* C **416** 90
[19] Sumption M D, Bhatia M, Wu X, Rindfleisch M, Tomsic M and Collings E W 2005 *Supercond. Sci. Technol.* **18** 750

Sumption M D, Bhatia M, Rindfleisch M, Tomsic M and Collings E W 2006 *Supercond. Sci. Technol.* **19** 155
[20] Wang X L, Yao Q W, Horvat J, Qin M J and Dou S X 2004 *Supercond. Sci. Technol.* **17** L21
[21] Soltanian S, Wang X L, Horvat J, Li A H, Liu H K and Dou S X 2002 *Physica* C **382** 187
[22] Fang H, Gijavanekar P, Zhou X Y, Liang G, Putman P T and Salama K 2005 *IEEE Trans. Appl. Supercond.* **15** 3215
[23] Liang G, Fang H, Katz D, Tang Z and Salama K 2006 *Physica* C **442** 113
[24] Zeng X H *et al* 2002 *Nat. Mater.* **1** 35–8
[25] Yao Q W, Wang X L, Soltanian S, Li A H, Horvat J and Dou S X 2004 *Ceram. Int.* **30** 1603
[26] Wang S F, Zhou Y L, Zhu Y B, Zhang Q, Liu Z, Chen Z H, Lu H B, Dai S Y and Yang G Z 2003 *Physica* C **390** 6
[27] Bud'ko S L, Lapertot G, Petrovic C, Cunningham C E, Anderson N and Canfield P C 2001 *Phys. Rev. Lett.* **86** 1877
[28] Hideki A, Kenji N, Motoharu I and Hideak Ki 2004 *Appl. Phys. Lett.* **85** 6197