Available online at www.sciencedirect.com
ScienceDirect
Physica C 468 (2008) 426–431
PHYSICA C
www.elsevier.com/locate/physc

Angular dependent transport properties of MgB_2 films with columnar grains

Dong Ho Kim [a,*], Hye-Young Kim [a], Tae-Jong Hwang [a], Won-Kyung Seong [b], Won Nam Kang [b]

[a] Department of Physics, Yeungnam University, Gyeongsan 712-749, Republic of Korea
[b] BK21 Physics Division and Department of Physics, Sungkyunkwan University, Suwon 440-746, Republic of Korea

Received 8 November 2007; received in revised form 27 December 2007; accepted 9 January 2008
Available online 16 January 2008

Abstract

We studied the angular dependence of the transport properties of MgB_2 films with columnar grains grown by hybrid physical chemical vapor deposition method, one sample with unreacted boron in the volume and the other sample with no traceable impurity phase. The angular dependence of resistivity and critical current density in applied magnetic fields for both samples showed a flux pinning effect by the grain boundaries between columnar grains. The temperature dependence of the upper critical fields was analyzed by using the dirty-limit two-gap model. We found that the unreacted boron in the body of the film had negative effect on flux pinning and intraband electron diffusivities.

PACS: 74.70.Ad; 74.25.Fy

Keywords: MgB_2 film; Transport properties

1. Introduction

Magnesium diboride (MgB_2) has been one of the most extensively studied superconductors recently. For basic properties, MgB_2 has been known to have two superconducting gaps associated with two distinct Fermi surfaces [1,2]. The gap energy for the σ band from the σ orbitals of B is 7.2 meV and that for the π band from the π orbitals of B is 2.3 meV. The quasiclassical Usadel equations for MgB_2 have been solved for the dirty-limit with accounting the intraband and interband scattering by nonmagnetic impurities. The normal-state resistivities and the upper critical fields (H_{c2}) were found to be essentially dependent on the intraband electron diffusivities [3]. The temperature dependences of H_{c2} of MgB_2 single crystals [4] and films [5,6] have been analyzed based on the dirty-limit two-band model and the resulting intraband electron diffusivities have been reported. In the above samples, the in-plane electron diffusivity of the π band has been found to be always higher than that of the σ band.

MgB_2 films have a great potential for many applications in the areas of high performance devices and sensors. Intensive research work has been performed to fabricate MgB_2 films with high critical current density (J_c) [7–12]. MgB_2 films have been grown by using hybrid physical chemical vapor deposition (HPCVD) [7], pulsed laser deposition (PLD) [8], reactive evaporation [9], electron-beam evaporation [10], or molecular beam epitaxy (MBE) [11,12]. The substrate is another important factor in fabricating high-quality MgB_2 films. It has been known that (001) Al_2O_3 substrate is suitable for the epitaxial growth of hexagonal MgB_2 films [13]. For MgB_2 films epitaxially grown on the basal plane of a (001) sapphire single crystal substrate, a very high transport J_c has been observed [14]. In addition to that, a distinct growth structure with columnar grains

* Corresponding author. Tel.: +82 53 810 2340; fax: +82 53 810 4616.
E-mail address: dhkim@ynu.ac.kr (D.H. Kim).

0921-4534/$ - see front matter © 2008 Elsevier B.V. All rights reserved.
doi:10.1016/j.physc.2008.01.003

has often been observed [10,12,15]. The angular dependence of J_c revealed that the highest J_c occurred when the applied magnetic field (H) was applied parallel to the grain boundaries of the columnar-shape grains, and these results were interpreted as a direct proof of flux pinning by the grain boundaries [10,12].

For MgB_2 films grown by HPCVD in this work, we easily obtained a growth rate as high as 4 nm/s [16]. Such a fast growth often results in slightly off-stoichiometric MgB_2 films, which are either Mg-rich or B-rich. In this work we studied the superconducting properties of two MgB_2 films epitaxially grown on (001) Al_2O_3 by HPCVD, one with unreacted B in the bulk and the other with no traceable impurity phase. We have measured the angular dependence of resistivity and J_c in the applied fields and determined H_{c2}. The J_c of both films showed a specific feature related to the columnar growth along the c-axis, which was also confirmed by the cross-sectional images from transmission electron microscope (TEM) [15]. By applying the dirty-limit two-gap model [3] to the temperature dependence of H_{c2}, we determined intraband diffusivity for both the σ and π bands for both films. We discuss the influence of the unreacted B on the transport properties mainly in terms of the intraband electron diffusivities.

2. Experiment

The principles and setup of the HPCVD system are basically similar to that reported by Zeng et al. [7]. The HPCVD system used in the present work has been described in detail elsewhere [17]. In brief, the system consists of a vertical quartz reactor with a uniquely designed inductive heating susceptor to compensate for the low sticking coefficient of Mg. Before the film deposition, the quartz tube was first purged with high purity (99.9999%) Ar and H_2 gas. The susceptor, along with the Al_2O_3 substrate and Mg pieces, was heated inductively to 520–600 °C in 150 Torr of the hydrogen flow. Then the B_2H_6 gas (5%) in H_2 was introduced at a flow rate of 50 sccm into the reactor to start the deposition of the MgB_2 films. In our present work, the total gas flow rate was kept at a 150 sccm and the react total pressure was 150 Torr. Finally, the MgB_2 sample was cooled in the H_2 carrier gas to room-temperature. As grown MgB_2 thick films were investigated by scanning electron microscope (SEM), X-ray diffraction (XRD), and cross-sectional TEM of the representative samples. Films were patterned by using conventional photolithography and Ar-ion milling to form a bridge of $20 \times 600\ \mu m^2$ for resistivity and J_c measurements. The thicknesses of the films were measured by using an alpha-step.

3. Results and discussion

We studied two MgB_2 films with thicknesses of 1.2 μm (M12) and 0.86 μm (M09) with the latter showing an unreacted B phase. In the XRD patterns as shown in Fig. 1, the

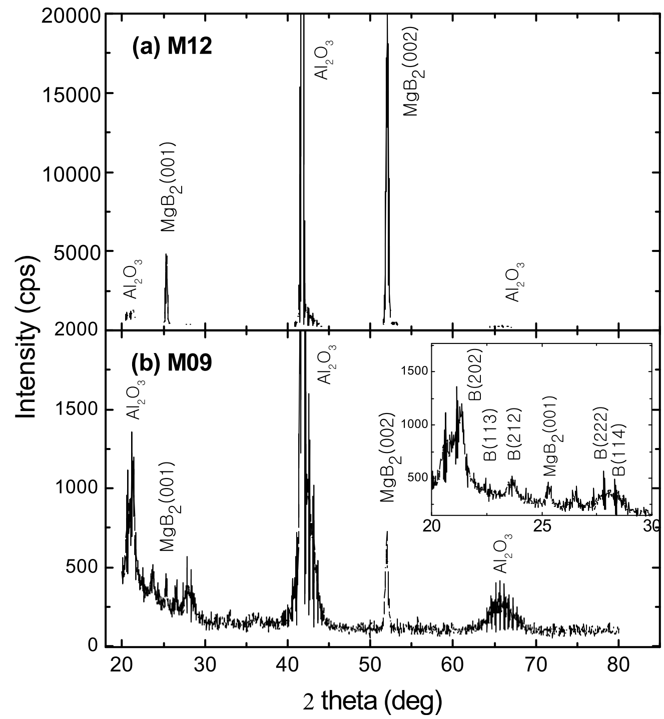

Fig. 1. X-ray diffraction patterns for two MgB_2 films, (a) 1.2 μm thick and (b) 0.9 μm thick, both grown on (001) Al_2O_3 substrates, show the c-axis oriented growth perpendicular to the substrate. In film M09, extra B peaks were observed as shown in the inset of (b).

(001) and (002) peaks of MgB_2 were detected indicating that all films were c-axis oriented perpendicular to the substrates. The c-axis lattice constant measured by using the (002) peak was found to be 0.3510 nm, slightly smaller than that of the bulk value of 0.3524 nm. The compressed c-axis lattice has been usually observed in MgB_2 films grown on sapphire substrate [18,15]. Cross-sectional TEM images of the part of M09 showed columnar grains with their long axis along the c-axis [17], similar to the earlier works in films with columnar grains [10,12]. A top SEM view of the sister film showed that the grain diameter varied approximately from 0.1 to 1.0 μm [16]. No impurity phase was detected in the XRD patterns of the M12 sample, however, for M09, small but clear B peaks were identified as shown in the inset of Fig. 1b. Those B peaks did not disappear after thinning a part of film by about 0.1 μm by using Ar-ion milling, which suggested that unreacted B was distributed throughout the volume.

Resistive transitions of the two MgB_2 films in an ambient field from room-temperature to a superconducting state are shown in Fig. 2. The details of the transition near the onset are shown in the inset. The room-temperature resistivity (ρ_{300}) and residual resistivity (ρ_0), measured at 41 K, of the two samples were quite different from each other. For M09, $\rho_{300} = 35\ \mu\Omega$ cm and $\rho_0 = 11\ \mu\Omega$ cm, and for M12, $\rho_{300} = 12\ \mu\Omega$ cm and $\rho_0 = 0.34\ \mu\Omega$ cm. The residual resistance ratio (RRR) of M09 was 3.2, much smaller than 35 of M12. Residual resistivity has been found to be linearly dependent on 1/thickness [19,18] in MgB_2 films