Polycrystalline MgB_2 Films on Flexible YSZ Substrates Grown by Hybrid Physical-Chemical Vapor Deposition

A. V. Pogrebnyakov, E. Maertz, R. H. T. Wilke, Qi Li, A. Soukiassian, D. G. Schlom, J. M. Redwing, A. Findikoglu, and X. X. Xi

***Abstract*—We report properties of polycrystalline MgB_2 films deposited on thin flexible yttium-stabilized zirconia (YSZ) substrates by hybrid physical-chemical vapor deposition. The MgB_2 films show a transition temperature of 38.9 K with a narrow superconducting transition (0.1 K). The self-field critical current density in the films reaches 10^7 A/cm^2 at low temperatures. These properties do not change after repeated bending over a radius of 10 mm. Low microwave surface resistance comparable to those obtained in epitaxial MgB_2 films on single-crystalline sapphire substrate was observed. The result shows promise of polycrystalline MgB_2 films for applications in superconducting digital circuits as well as in coated conductor wires.**

***Index Terms*—Flexible YSZ substrates, hybrid physical-chemical vapor deposition, magnesium diboride, polycrystalline films.**

I. Introduction

MAGNESIUM diboride, with a relatively high critical temperature $T_c \sim 40$ K [1], is a promising superconducting material for high magnetic field and electronics applications [2], [3]. For electronics applications, besides the potential of MgB_2 Josephson junctions working over 20 K, which substantially reduces the cryogenic requirements compared to the Nb-based superconducting electronics [4], MgB_2 can also be used for flexible superconducting interconnect in high speed superconducting digital circuits. This requires depositing high quality MgB_2 thin films on flexible substrates with low loss at high frequencies (in the order of gigahertz). Previously, we have reported polycrystalline MgB_2 thin films on SiC fiber grown by hybrid physical-chemical vapor deposition (HPCVD) [5]. The pure MgB_2 fiber shows a high T_c of 39 K, while the carbon-alloyed fibers show high upper critical fields and high irreversibility fields [5]. In this paper, we present result on polycrystalline MgB_2 films on flexible yttium-stabilized zirconia (YSZ) substrates deposited by HPCVD. The MgB_2 films have T_c of 39 K, self-field critical current density J_c over 10^7 A/cm^2 at low temperatures, and low microwave surface resistance. The superconducting properties of these films do not degrade after repeated bending over a radius of 10 mm. The result is not only important for flexible superconducting interconnect, it is also significant for coated conductor wires where high J_c and ductility are essential.

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A. V. Pogrebnyakov and X. X. Xi are with the Department of Physics and Department of Materials Science and Engineering, The Pennsylvania State University, University Park, PA 16802 USA (e-mail: avp11@psu.edu).

E. Maertz, R. H. T. Wilke, and Q. Li are with the Department of Physics, The Pennsylvania State University, University Park, PA 16802 USA.

A. Soukiassian, D. G. Schlom, and J. M. Redwing are with the Department of Materials Science and Engineering, The Pennsylvania State University, University Park, PA 16802 USA.

A. Findikoglu is with the Los Alamos National Laboratory, Los Alamos, NM 87545 USA.

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II. Experimental Details

The MgB_2 films were grown by HPCVD on $1\ \mathrm{cm} \times 1\ \mathrm{cm} \times 0.1\ \mathrm{mm}$ polycrystalline substrates of flexible zirconia stabilized by 3 mole % of yttria (Ceraflex 3Y, MarkeTech International, Inc.). Strips of this material are capable of bending to a radius of 8 mm. Details of the HPCVD technique have been described previously [6], [7]. During the deposition, H_2 was used as the carrier gas and a mixture of 1% B_2H_6 in H_2 was used as the boron source. The flow rate of the B_2H_6 mixture was 25–60 sccm, and the total flow rate was 100–475 sccm. The total pressure in the reactor was 80 Torr, which corresponds to a partial pressure of the diborane from 0.04 to 0.16 Torr. The substrate temperature was held at 670–690 $^\circ$C. At these deposition parameters, the growth rate of the films varied from 10 to 20 Å/s. The film thickness was 250–530 nm. The structure of the MgB_2 films was characterized by four-circle x-ray diffraction with Cu $K\alpha$ radiation.

The temperature dependence of the film resistance was measured by the standard four-probe technique and the resistivity ρ was determined by the Van der Paw method. The critical current density J_c of the films was determined from both transport (close to T_c) and magnetization (at low temperatures) measurements. For the transport measurements, the films were patterned into 20 μm wide bridges by photolithography and ion milling, and J_c was determined using the 1-μV/cm criterion. For the magnetization measurements, the standard Bean model was applied to derive the J_c values from the magnetic hysteresis loops recorded on a Quantum Design Model 6000 Physical Property Measurement System.

The microwave measurements were made at 10 GHz using a parallel plate resonator technique. Two 1 cm × 1 cm MgB_2 films facing each other were separated by a thin Teflon sheet, forming

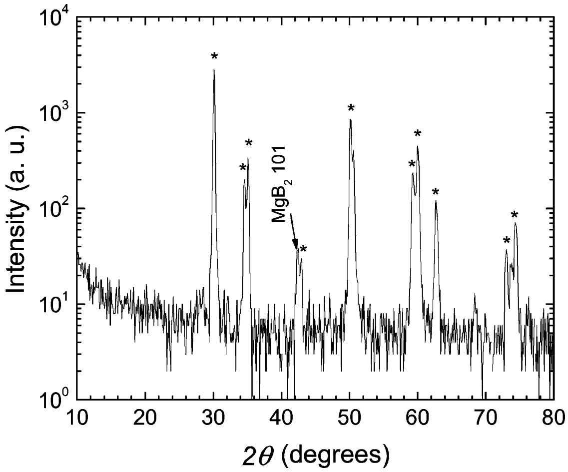

Fig. 1. X-ray diffraction $\theta - 2\theta$ scan of a polycrystalline MgB_2 film on a flexible YSZ substrate. The most intensive 101-reflection peak of MgB_2 is observed in the spectrum. All other peaks, marked by "$*$", are from the polycrystalline $Y_{0.037}Zr_{0.963}O_{1.982}$ substrate.

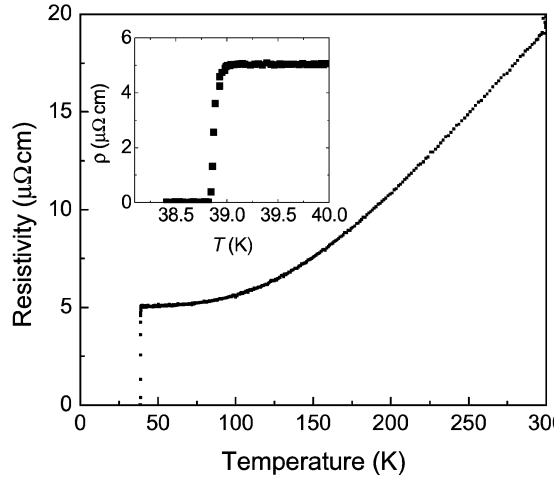

Fig. 2. Resistivity vs. temperature curve for a 400 nm thick MgB_2 film on a flexible YSZ substrate. The inset shows the details of the superconducting transition.

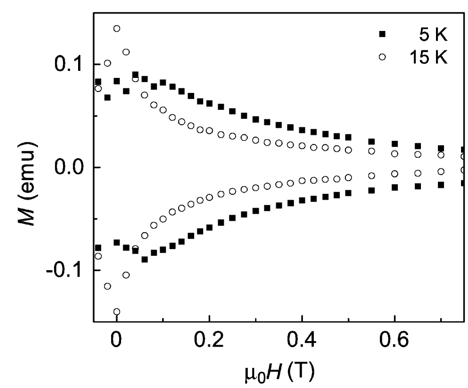

Fig. 3. The $M - H$ loops for a 400 nm thick MgB_2 film, sample 050306b, measured at 5 and 15 K.

a parallel plate resonator. From the measurements of the unloaded quality factor, the surface resistance R_s of the films was determined. Details of the measurement technique have been described previously [8].

III. Results and Discussions

Fig. 1 shows an x-ray diffraction $\theta - 2\theta$ scan of an MgB_2 film on a flexible YSZ substrate. The peaks marked by "$*$" match very well the diffraction pattern for $Y_{0.037}Zr_{0.963}O_{1.982}$. The observation of peaks from various diffracting planes indicates that the YSZ substrate is polycrystalline. One diffraction peak from the MgB_2 film is seen, which belongs to the most intensive diffraction from the 101 plane. This is consistent with the result of the polycrystalline MgB_2 film on SiC fiber, where the 101 peak is the strongest among several weak peaks from the film [5]. We can conclude that the MgB_2 film on flexible YSZ substrate is polycrystalline. The atomic force microscopy studies show that the grain size in such films is 0.2–0.5 μm. The YSZ substrate is rough, with a root-mean-squared (RMS) roughness of 96 nm. Consequently, the MgB_2 films on it are also rough. For example, a 500 nm thick MgB_2 film has a RMS roughness of 150 nm.

A resistivity vs. temperature curve for a 400 nm thick MgB_2 film is plotted in Fig. 2. It shows a sharp superconducting transition $(\Delta T_c = 0.1\ \mathrm{K})$ at 38.9 K, which is the same as that of the bulk MgB_2 [1]. The residual resistivity ratio for MgB_2 films on flexible YSZ substrate is typically 3–4, which is similar to those found in polycrystalline films fabricated by other techniques [9]. The value of the resistivity difference between 300 K and 50 K $\Delta\rho_{300-50K} \equiv \rho(300K) - \rho(50K) = 14.9\ \mu\Omega\mathrm{cm}$, which is larger than that of epitaxial HPCVD MgB_2 films on single crystalline substrates ($\Delta\rho_{300-50K} \sim 8\ \mu\Omega\mathrm{cm}$ [10]). This indicate a reduced effective area in the films due to the granular nature of the polycrystalline films, or the large film roughness.

The magnetization M versus applied field H for a 400 nm thick MgB_2 film, sample 050306b, measured at 5 and 15 K, is shown in Fig. 3. At 5 K, the low field portion of the $M - H$ loop suffers from the dendritic flux jumps [11], therefore ΔM near zero field is smaller than that at ~0.1 T. The instability exists for all measurements below 12.5 K. We have shown previously that the dendritic flux instability does not occur in very clean epitaxial MgB_2 films [12]. The polycrystalline MgB_2 films on YSZ substrate reported here have higher residual resistivity ($\sim 5\ \mu\Omega\mathrm{cm}$ for the sample in Fig. 2) than in clean MgB_2 films $(< 1\ \mu\Omega\mathrm{cm})$, enough to cause dendritic flux jumps at low field and low temperature. The critical current density, derived from ΔM, is suppressed rapidly by the magnetic field, consistent with the behavior of undoped MgB_2 films due to low flux pinning [6].

In Fig. 4, the self-field J_c as a function of temperature, determined by both transport (close to T_c) and magnetization (at low temperatures) measurements and collected from three samples, is plotted. The thickness of the films are 400 nm. As can be seen in figure, the polycrystalline MgB_2 films on flexible YSZ substrate have J_c values exceeding $10^6\ \mathrm{A/cm^2}$ at temperatures lower 30 K and reaching $10^7\ \mathrm{A/cm^2}$ at 12.5 K. Because of the dendritic flux instability at low temperatures, we are not able to obtain reliable J_c values below 12.5 K.

For both flexible superconducting interconnect and coated conductor wires, it is important that the MgB_2 films can be bent and they maintain excellent superconducting properties after bending. Chen *et al.* have shown that T_c and resistivity of MgB_2

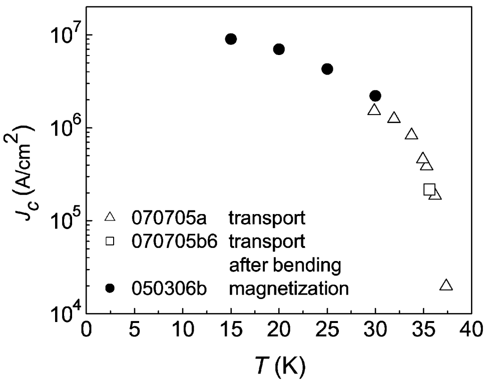

Fig. 4. Self-field critical current density vs. temperature for three MgB_2 films on the flexible YSZ substrates. The result for sample 050306b is from magnetization measurement. The result for sample 070705a is from transport measurements on a 20-μm-wide bridge. The result for sample 070705b6 is from transport measurements on a 1.2 $\times$ 10 mm^2 strip after bending 50 times with a bend radius of 10 mm.

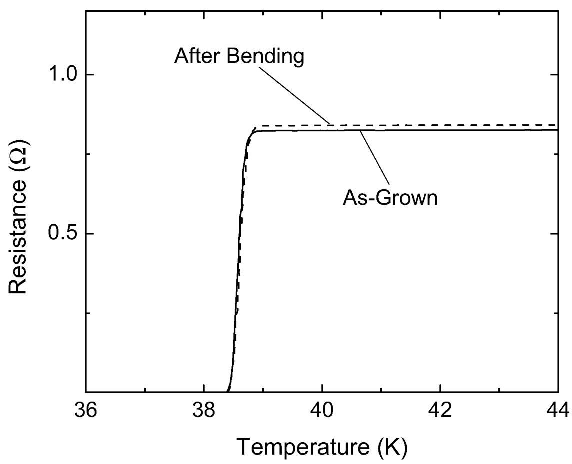

Fig. 5. Resistance vs. temperature curves for an as-grown MgB_2 film (solid curve) as well as a film after bending over a radius of 10 mm for 100 times (dashed curve).

thick films on stainless steel do not change substantially when they are bent [13]. In Fig. 5, we show resistance versus temperature curves of a 1.2 mm $\times$ 10 mm strip of MgB_2 on flexible YSZ substrate before and after bending over a radius of 10 mm (close to the bend radius of the flexible YSZ substrates) for 100 times, measured using the same electrodes. The film demonstrated good adhesion with no delamination after the bending. As the figure shows, there is no change in T_c and ΔT_c of the film after bending test. The resistance increased by 1.5%. Considering various possible changes to the sample between the two measurements, including the possible degradation due to the moisture, the difference is insignificant. Moreover, bending of the films did not change J_c of MgB_2 films on flexible YSZ substrate. In Fig. 4, the samples 070705a and 070705b6 are from the same MgB_2 film, which was cut into several strips of 1.2 mm $\times$ 10 mm in dimension. The J_c for 070705a was measured as grown, and the J_c for 070705b6 was measured after the strip was bent 50 times over a radius of 10 mm and with the strip kept bent during the measurement. At 32 K, J_c of sample 070705b6 is 2.4×10^4 A/cm^2. Given the uncertainty of thickness measurement due to the film roughness, we find that the J_c value of the MgB_2 film was not affected by the bending.

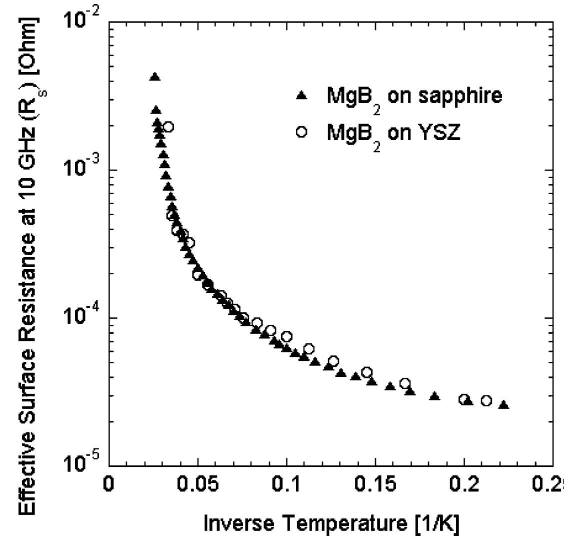

Fig. 6. Surface resistance at 10 GHz as a function of temperature for a polycrystalline MgB_2 film on a flexible YSZ substrate and an epitaxial MgB_2 film on a c-cut sapphire single-crystal, measured by a parallel plate technique.

The microwave surface resistance R_s at 10 GHz as a function of inverse temperature, measured on two 300 nm thick MgB_2 films on flexible YSZ substrate, is shown in Fig. 6. A low R_s value is observed, similar to those reported previously and measured using the same technique on MgB_2 films deposited by reactive co-evaporation on r-plane sapphire [14]. Also shown in Fig. 6 is the result from a pair of epitaxial MgB_2 films on c-cut sapphire substrate. The results from the polycrystalline and epitaxial films are almost identical under the experimental conditions, i.e. the sensitivity limit due to the coupling and radiation losses of the parallel-plate setup and the low microwave power used in the measurement. This low level of microwave surface resistance is adequate for superconducting interconnect in high speed superconducting digital circuits.

IV. Conclusions

In summary, polycrystalline MgB_2 films were deposited on flexible YSZ substrates by HPCVD. The films display sharp superconducting transitions with T_c of 38.9 K, the same as for bulk MgB_2. J_c is high, reaching 10^7 A/cm^2 at 12.5 K. Mechanically, the films are robust: repeated bending over a radius of 10 mm does not change the superconducting properties of the films including T_c and J_c. The microwave surface resistance of the films is low, making them attractive candidates for flexible interconnects in high speed superconducting digital circuits. Further, these good superconducting and mechanical properties are important for MgB_2 coated-conductor wires.

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