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The distribution of elements in sequentially prepared MgB_2 on SiC buffered Si substrate and possible pinning mechanisms

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ABSTRACT

MgB_2 thin films are prepared by sequential evaporation of boron and magnesium bilayers on SiC buffered Si substrates followed by an in situ annealing. Precursor Mg–B bilayers are deposited by electron beam evaporation at room temperature. The amount of B is varied so as to result in different thickness (15 nm and 50 nm) of stoichiometric MgB_2 final film after an in situ reaction with the excess Mg top layer in the vacuum. We show the distribution of the elements through the film.

X-ray photoelectron spectroscopy analyses have shown that carbon is not free in the films (except the surface of the film) and silicon is in the compound form, too. In the case of the 15 nm thick films we see a strong interdiffusion of the elements (C, B) and we observe a suppression of T_C of the film to 20 K. We register different slope of the $H_{C_2}(T)$ dependence – the lowest temperature value of H_{C_2} for the 15 nm thick film exceeds the one for the 50 nm thick film in spite of lower T_C. We suppose that δl pinning mechanism is dominant for the 15 nm thick film.

1. Introduction

The values of transition temperature T_C and upper critical field H_{C_2} in MgB_2 films are often linked with effects of a chemical substitution and incorporation of nanoparticles such as nano SiC, carbon nanotubes, C, Si [1,2]. It has been accepted that increased H_{C_2} is caused by C substitution for B in the crystal lattice as a result of chemical doping (δl pinning). The main free path l, is reduced due to a distortion in the lattice caused by the C substitution. Therefore, the σ band electron scattering is enhanced, leading to an increase in H_{C_2}. However, the large T_C decrease to ∼20 K is observed. On the other hand, the pinning associated with the spatial fluctuation of the transition temperature (δlT pinning), only slightly reduces T_C and it is considered to be the main flux pinning mechanism in pure MgB_2 bulk and thin films [3–5]. However, in general nano-C dopants can contribute to point defects and interboundary pinning, too. We have already shown [6] that a strong diffusion of Si and C into the MgB_2 film occurs in the case of sequentially prepared MgB_2 thin films on SiC buffered Si substrate. We try to answer the questions in what form the elements are incorporated into the film and how they influence its electrical properties. In this work we analyze the composition and electrical properties of the prepared MgB_2 thin films and the possible pinning mechanism.

2. Experimental

MgB_2 films have been prepared similarly to [6,7] by sequential electron beam evaporation of the Mg–B bilayer followed by an in situ annealing. The deposition chamber has been evacuated to a pressure of 10^{-4} Pa. The magnesium and boron layers have been deposited on the SiC buffered Si substrates at room temperature. An excess of about 100% of Mg compared to the stoichiometric composition has been used. The amount of boron corresponds to 50 nm and 15 nm of a stoichiometric MgB_2 film. As-deposited films were in situ heated to 280 °C for 30 min in an argon atmosphere at a pressure of 0.06 Pa. Subsequently, the pressure of Ar was increased up to 16 Pa and the temperature of the heater was increased to the maximum temperature of 750 °C or 830 °C and kept there for 10 min. Finally, the Ar pressure was raised to 10^3 Pa and the samples were cooled down to room temperature.

Superconducting and transport properties of the MgB_2 films were characterized by resistance measurements using the standard DC four-point method.

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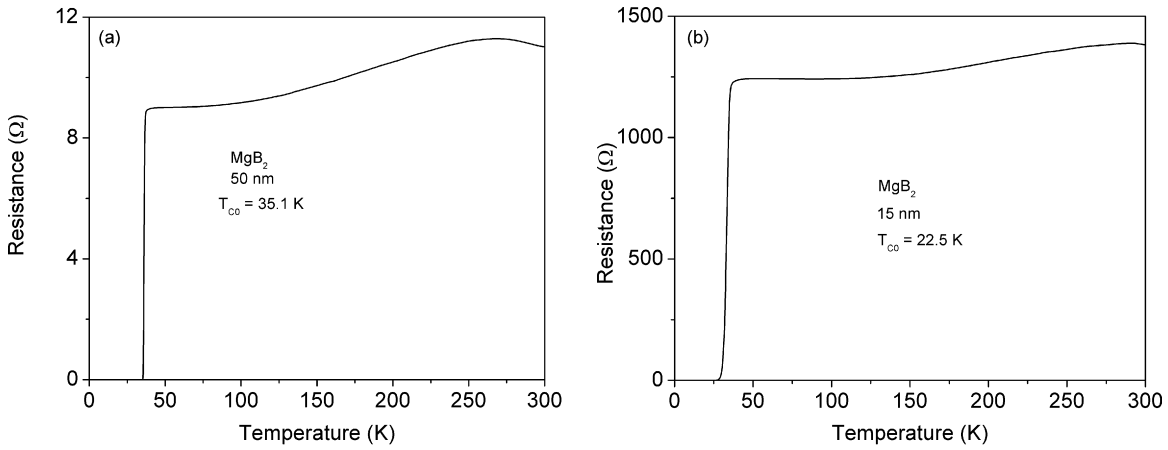

Fig. 1. R–T dependence of the (a) 50 nm and (b) 15 nm thick MgB_2 film.

Auger electron spectroscopy (AES) depth profiling was carried out by a spectrometer equipped with a cylindrical mirror analyzer (CMA) and EX 05 VG ion gun. A primary electron beam was used with the energy of 3 keV and the incident angle of 20° with respect to the surface normal. The sputtering was achieved by scanned Xe^+ ion beams with the energy of 1 keV and 60° of incidence with respect to the surface normal. The energy resolution of the CMA was $\Delta E/E = 0.3\%$.

X-ray photoelectron spectroscopy (XPS) spectra were obtained for the 50 nm thick MgB_2 films in an ESCALAB 210 spectrometer (Vacuum Generators), with a base pressure in the range of 10^{-8} Pa. A hemispherical electron energy analyzer working in the pass energy constant mode at a value of 50 eV was used. Unmonochromatized Al-Kα radiation (1486.6 eV) was used as an excitation source. Successive spectra were recorded after sputtering treatments with Ar^+ ions of 3.5 keV of kinetic energy, impinging the surface sample at normal incidence.

XPS measurements were carried out on the 15 nm thick MgB_2 films with Omicron NanoTechnology Multiprobe instrument using monochromatized Al-Kα radiation with energy of 1486.7 eV and Sphera hemispherical electron spectrometer. The base pressure of the chamber was 8×10^{-9} Pa and the XPS data were collected at 2×10^{-7} Pa. The sputtering of the sample was achieved by Ar bombardment with the energy of the ions 2 keV for 30 min for one sputtering cycle.

3. Results and discussion

Typical R–T dependences of the 50 and 15 nm thick MgB_2 films are presented in Fig. 1a and b, respectively. Fig. 1b shows suppressed maximum zero resistance critical temperature T_{C_0} to ~20 K.

However, when we studied H_{C_2} as a function of the temperature in perpendicular and parallel magnetic field we found different slope of $H_{C_2}(T)$ dependence where the lowest temperature value of H_{C_2} for the 15 nm thick film even exceeds the one for the 50 nm film in spite of lower T_C (Fig. 2). We suppose a strong pinning mechanism.

The distribution of the elements in 50 nm and 15 nm thick MgB_2 film is characterized by AES spectroscopy (Fig. 3a–c). It can be seen that in the case of the 50 nm thick films the maximum heater temperature −830 °C (Fig. 3b) increases the diffusion of B and Mg into the SiC buffer layer comparing to the film prepared at 750 °C (Fig. 3a). In the case of the 15 nm thick MgB_2 film we observe a strong interdiffusion of the elements at the interface (Fig. 3c). The maximum concentration of B is practically at the interface. We register a high concentration of C and Si, too. In all cases the trace of nitrogen is probably a consequence of the addition of NH_3 during the preparation of the SiC buffer films.

XPS analyses confirm that most elements are bound in various form of compounds. Fig. 4a and b show XPS spectra of the sample surfaces prepared by postannealing at a substrate temperature of 830 °C and 750 °C taken after different sputtering times. The XPS spectra have shown that the thermal treatment of the sample at 830 °C has degraded the purity of MgB_2, and it has induced the reaction between MgB_2 and MgO, giving rise to BO_x species. We can see the decrease of the peak belonging to B–Mg compound and the increase of the peak belonging to BO_x at a temperature of 830 °C (Fig. 4a).

It was interesting to study the presence of any carbon substitution for boron in our MgB_2 films. We registered, especially for the 15 nm thick film, that T_C decreased to ~20 K (Fig. 1b). We also observed an enhanced H_{C_2} (Fig. 2) and the AES depth profile showed an increased concentration of carbon. All these three aspects support this idea. To investigate the bonding character of C atoms in the composites we performed high resolution XPS analysis using the monochromatic X-ray, especially on the 15 nm thick MgB_2 film.

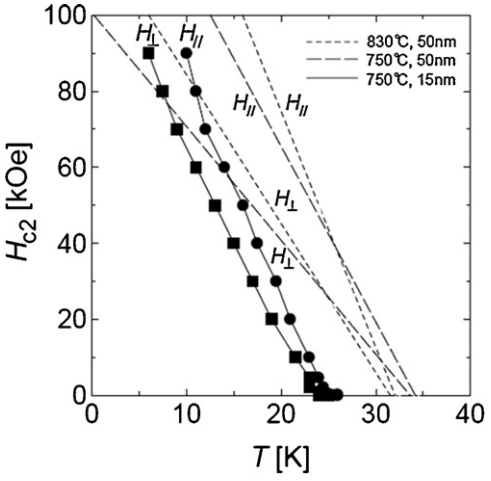

Fig. 2. H_{C_2} as a function of the temperature T for the 15 nm thick film. Solid squares for perpendicular and solid circles for parallel magnetic fields. Dashed lines are for 50 nm thick films.