

Magnetoresistance in Semimetallic Bismuth Thin Films

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(Received 28 June 2004)

The magnetotransport properties of electroplated and sputtered Bi thin films have been investigated in the range 4 – 300 K. A marked increase from 5,200 % to 80,000 % in the ordinary magnetoresistance (MR) for the electroplated Bi thin film was observed after thermal anneal at 4 K. The MR ratios for the as-grown and the annealed Bi thin films were found to be 560 % and 590 %, respectively, at 300 K. On the other hand, the MR for the Bi film grown by sputtering was hardly observed at 4 and 300 K, whereas the MR ratios after annealing were found to reach 30,000 % at 4 K and 600 % at 300 K. The room temperature MR in the sputtered films was found to depend on the trigonal-axis oriented microstructures and grain size, in contrast to the electroplated films. Our results support the view that the grain-boundary scattering mechanism is dominant in the MR response at room temperature, whereas the textured grains oriented to the trigonal axis are dominant in the MR response at 4 K for both electroplated and sputtered samples.

PACS numbers: 72.15.Gd, 73.50.Jt, 72.25.Hg, 72.25.Mk

Keywords: Semimetal, Bismuth, Magnetotransport, Ordinary magnetoresistance

I. INTRODUCTION

Over the last decade, semimetallic bismuth (Bi) has been extensively investigated, since it exhibits very intriguing transport properties due to its highly anisotropic Fermi surface, low carrier concentrations, long carrier mean free path l and small effective carrier mass m^* [1–5]. The magnetoresistance (MR) behavior and long carrier mean free path l in Bi thin films are of particular importance, since they can be exploited for spintronic device applications such as magnetic field sensors and spin-injection devices. With respect to “spintronics” [6, 7], it is expected that Bi can be used as a spin channel in a spin-injection device due to the very long spin diffusion length l_{sd} of a few tens of μm [5], following the relation $l_{sd} = (lv_F\tau_{\uparrow\downarrow})^{1/2}$, where v_F is the Fermi velocity and $\tau_{\uparrow\downarrow}$ is the spin relaxation time.

In recent years, comprehensive studies have centered on the MR behaviors of Bi thin films grown by electroplating [2,3,5] and molecular beam epitaxy (MBE) [4], but spintronic devices based on Bi thin films have not yet been studied. In the present work, we report the magnetotransport properties of electroplated and sputtered Bi thin films obtained in the range 4 – 300 K. We discuss the annealing effects on the very large MR at 4 K

and 300 K, respectively, in electroplated and sputtered Bi thin films.

II. EXPERIMENT

Bi thin films in the thickness range 1 – 20 μm were electroplated on 100 Å thick Pt/Si(100) from aqueous solutions of $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$. Bi thin films were also deposited on a thermally oxidized Si(100) substrate in a dc magnetron sputtering system with a base pressure of 4×10^{-8} Torr. The as-electroplated and as-sputtered Bi thin films were heat-treated in a vacuum tube with Sm powder as an oxygen getter at 268 – 270 °C for 10 hrs. The van der Pauw magnetoresistance (MR) measurements were performed by applying a magnetic field up to 9 T in the temperature range 4 – 300 K in order to investigate the magnetotransport properties of the electroplated and sputtered Bi thin films.

III. RESULTS AND DISCUSSION

Figure 1 shows the variation of magnetoresistance ($\Delta R/R$) against magnetic fields for (a) and (b) electroplated, and (c) and (d) sputtered Bi thin films. A

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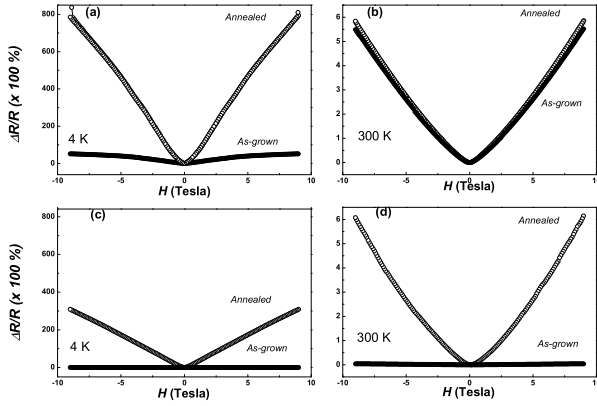


Fig. 1. Variation of magnetoresistance ($\Delta R/R$) against magnetic fields for (a) and, (b) electroplated, and (c) and, (d) sputtered Bi thin films.

marked increase from 5,200 % to 80,000 % in positive MR was observed after annealing for the 20- μm -thick Bi film grown by electroplating at 4 K under a magnetic field of 9 T applied perpendicular to the film plane. We found from X-ray diffraction (XRD) measurements that such an increase in the MR is attributable to the trigonal-axis oriented microstructure in the Bi film after annealing. However, the values of the MR ratio for the as-grown and annealed Bi thin films were found to be 560 % and 590 %, respectively, at room temperature, indicating that the MR is hardly dependent upon the trigonal-axis oriented microstructure at 300 K.

Thickness dependence of MR was also found for both the films in the range 1 – 20 μm . This indicates that the carrier mean free path l for the films is of the order of a few tens of μm . It is noted that the carrier mean free path l is comparable to the spin diffusion length l_{sd} . The positive MR effect in the Bi films is the ordinary MR (OMR), caused by the curving of carrier trajectory by the Lorentz force. The MR is proportional to the product of the cyclotron frequency, $\omega_c = eB/m^*$, and the relaxation time τ . In Bi, ω_c is large because of the very small effective mass, $0.002m_e$. The relaxation time τ is related to the carrier mobility $\mu = e\tau/m^*$, which depends on the quality of the sample. The room-temperature MR in the Bi thin films depends mainly on the carrier mean free path l .

On the other hand, the MR response for the 7- μm -thick Bi film grown by sputtering was hardly seen at 4 and 300 K, indicating that the carrier mean free path l for the sputtered sample is much shorter than that for the electroplated sample. However, the MR for the annealed sample was found to reach 30,000 % at 4 K and 600 % at 300 K. Figure 2 displays XRD patterns of (a) as-sputtered Bi film, and those annealed at 270 °C (b) for 5 hrs, and (c) for 10 hrs. The as-sputtered film was found to be polycrystalline, showing not-fully-grown

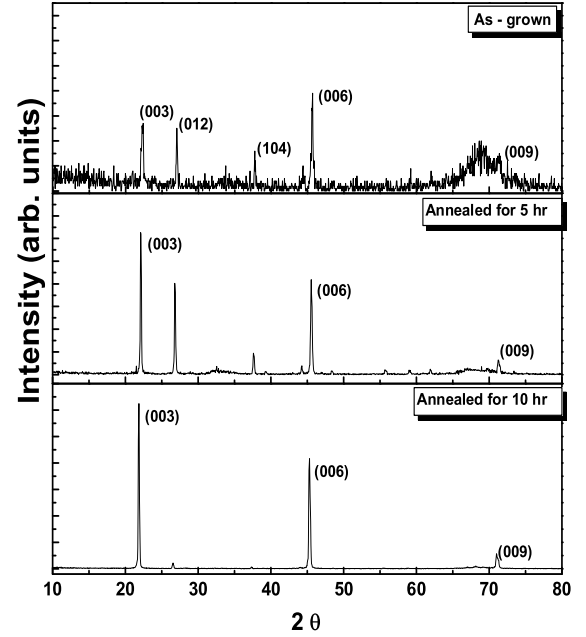


Fig. 2. X-ray diffraction patterns of (a) as-sputtered Bi film, and annealed films at 270 °C (b) for 5 hrs, and (c) for 10 hrs.

rhombohedral structure of Bi. The diffraction pattern of the same sample after annealing at 270 °C for 10 hrs exhibits only the (003), (006), and (009) peaks, revealing that the Bi film is trigonal-axis oriented. Our results demonstrate that the as-grown polycrystalline Bi thin films become trigonal-axis oriented after annealing, giving rise to the enhancement of MR in the sputtered Bi films at 4 K and 300 K.

Figure 3 presents scanning electron microscopy (SEM) images of (a) the as-electroplated, and (c) the as-sputtered Bi thin films. The (b) and (d) samples correspond to (a) and (c), respectively, after annealing at 270 °C for 10 hrs. The grains in the as-electroplated sample (a) are spatially distributed on a few μm and are, in size, almost identical to those in the annealed sample (b). We thus infer that the increase in MR ratio at low temperature for the electroplated sample after annealing is due to trigonal-axis oriented grains [see Fig. 1 (a)], while no increase in MR ratio at room temperature after annealing is due to the same grain size [see Fig. 1 (b)]. However, the as-sputtered sample shows submicron-sized grains, which are much smaller than the trigonal-axis-oriented grains in the annealed sample. Our results support the view that the grain-boundary scattering mechanism is dominant in the MR response at room temperature, whereas the textured grains oriented to the trigonal axis are dominant in the MR response at very low temperature for both electroplated and sputtered samples.

Our results demonstrate that the electroplated and sputtered Bi films exhibit very large MR, 600 %, at room temperature. This implies that the Bi films can be used

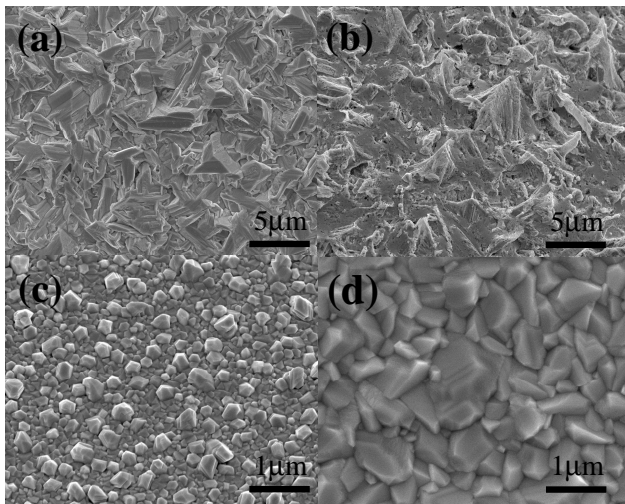


Fig. 3. SEM images of (a) as-electroplated, and (c) as-sputtered Bi thin films. (b) and (d) correspond to (a) and (c), respectively, after annealing at 270 °C for 10 hrs.

as a spin channel in a spin-injection device due to the very long spin diffusion length l_{sd} of a few tens of μm . This is expected to demonstrate a “spin-valve” effect in the spin-injection device consisting of the Bi film and two ferromagnetic contacts: an injector and a detector. However, it was found that the surface of the electroplated films is so rough, as seen in Fig. 3 (a) and (b) that it is unlikely to utilize a conventional photolithography process for making a spin-injection device based on the electroplated Bi film. By contrast, the surface of the sputtered films was found to be smooth enough for a photolithography process. Very recently, we demonstrated the spin transport in a spin-valve device incorporating Bi thin film and two ferromagnetic (FM) contacts [5]. We found that the non-local output voltage in the device depended upon the relative magnetization state of the two FM electrodes, indicating that the spin-polarized electrons are injected from the first FM (injector) into Bi and are detected by the second FM (detector) due to spin accumulation. The observed Hanle effect supports the spin injection and detection in our device [5].

IV. CONCLUSION

We have investigated the magnetotransport properties of electroplated and sputtered Bi thin films in the temperature range 4 – 300 K. We found that the enhanced MR (magnetoresistance) response after annealing at 4 K is attributed to the trigonal-axis oriented microstructure in the electroplated and sputtered Bi films. We also found that the room temperature MR for the electroplated and sputtered films largely depends on grain size, indicating that the grain-boundary scattering mechanism is dominant in the MR response at room temperature. Our results show that very large room temperature MR can be obtained in both the electroplated and the sputtered Bi thin films, which can be exploited for spintronic device applications

ACKNOWLEDGMENTS

This work was supported by Korea Research Foundation Grant (KRF-2003-003-C00047).

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