Growth, Structural Characterization and Transport Study of Bismuth Thin Films

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Abstract. We report the growth of high quality Bismuth thin films on the (100) oriented Si/SiO_2 substrate. These films are characterized by the x-ray diffraction which shows that films are single phase and have preferred orientation along the <003> direction. The x-ray reflectivity measurements of the thin films give film thickness around 52 nm and roughness of approximately 2 nm. The density of the Bismuth thin film is in agreement with the bulk Bismuth. The temperature dependent resistivity measurements show a non-metallic behavior which is attributed to enhanced grain boundary scattering from the relatively small grains of Bismuth thin films.

INTRODUCTION

Bismuth is an interesting semimetal in the bulk form which has a small band overlap of ~ 38 meV between the conduction and valance band and negligible density of states at the Fermi level. Bismuth has small Fermi surface, small carrier density, small effective mass and long mean free path of charge carriers [1,2]. It exhibits anomalous electronic properties such as large electrical resistivity and low thermal conductivity in comparison to other metals. Bismuth shows large anisotropic thermopower and magnetoresistance and the temperature dependence of thermopower shows strong discrepancy both in bulk and thin film form which is still not completely understood [3-7]. On reducing the dimension Bismuth is expected to show quantum confinement effects [7]. Recently it has been observed that with small antimony doping, the Bismuth can undergo a transition from semimetal to topological insulator state [8]. In bulk crystals the transition from semimetal to topological insulator happens at the antimony doping of around 4%. In the topological insulator phase, the antimony doped Bismuth exhibits interesting magneto-transportproperties such as large linear magnetoresistance and π Berry phase in Shubnikov-de Haas (SdH) oscillations [9]. The transition from trivial to topological state may also occurs on application of pressure and strain, and therefore [10], the study of Bismuth in thin films form is important for investigation of possible topological state in Bismuth.

With the aim of studying the effect of substrate induced strain and size on physical properties of the Bismuth, we have grown high quality thin films of Bismuth by thermal evaporation technique. Here we present the growth, structural characterization, and the transport studies of Bismuth thin films.

EXPERIMENTAL DETAILS

Bismuth thin films were grown on (100) oriented Si/SiO_2substrate using the metal evaporation unit havingbase pressure of approximately 2×10^{-7} m bar. The deposition chamber was brought to high vacuum and flushed with nitrogen gas to remove the trace of any residual atmospheric oxygen. The evaporation was carried at the rate of ~17 angstrom per minute and the substrate temperature of 150 ^{0}C. The thickness of the film was monitored by quartz crystal microbalance. The film was characterized by x-ray diffraction using Cu K_α radiation (wavelength 1.54Å) and the thickness of the film was determined by x-ray reflectivity measurements. The transport measurements on the film were carried by four probe method on 9T physical property measurement system.

RESULTS AND DISCUSSION

The Bismuth thin films on Si/SiO_2 substrate are characterized by the θ-2θ x-ray diffraction**. Figure 1** shows the x-ray diffraction pattern of the film and substrate. The x-ray data shows peaks corresponding to Bismuth and silicon substrates which are indexed accordingly. The presence of the x-ray peaks only corresponding to Bismuth and Si suggest the single phase nature of the film. The major intensity peaks of Bismuth arefor (003), (006), and (009) planes which suggests that majority of the film is highly oriented along the <003> direction. We also observe the peaks corresponding to Cu K_β wavelength. A minor intensity peak of Bismuth (012) plane isalso observed which indicates the presence of tiny<012> oriented regions coexisting along with preferred<003> orientation of the film.

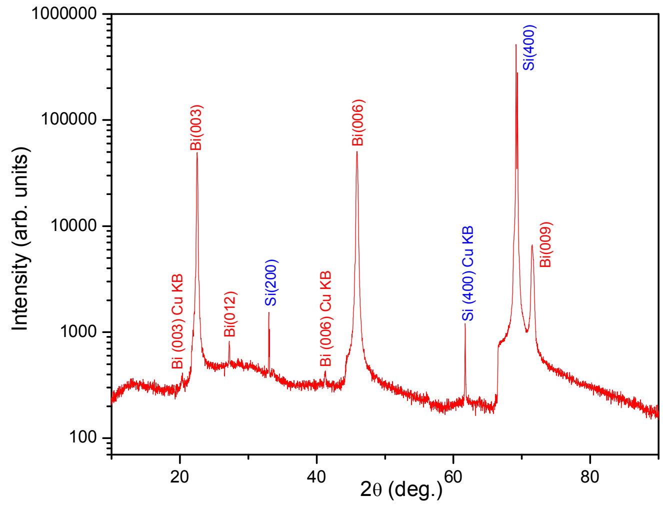

FIGURE 1: Room temperature x-rayθ-2θ diffraction pattern of Bismuth on (100) Si/SiO_2 substrate. Bismuth peaks are indexed by the red text while the silicon substrate peaks are indexed by blue text.

The x-ray reflectivity (XRR) measurement can be used to study the film thickness, roughness, and the density of the deposited layer. In these measurements film density is obtained by the critical angle for total external reflection and oscillation amplitudes, film thickness depends on the period of oscillations and roughness can be estimated from the rate of fall of intensity and oscillation intensity [12]. **Figure 2** shows the x-ray reflectivity pattern of Bismuth thin film deposited on (100) Si/SiO_2 substrate. The XRR pattern is analyzed by fitting of a model theoretical curve to the experimental data based on Parratt formalism [13]. This uses a non-linear least square fitting algorithm to refine the fitting parameters by minimizing χ^2 and the best theoretically modeled curve for experimental data is shown in the **figure 2**. The fitting parameters which are the thickness of the film, density, and roughness are shown in **Table1**. The thickness of the film comes around 52 nm.

Figure 3 displays the resistivity versus temperature for our Bismuth thin film. The resistivity of the film increases on decreasing the temperature. The behavior of resistivity is different from that observed in bulk Bismuth where the resistivity decreases on increasing the temperature [14]. Similar behavior of resistivity is also observed in previous reports of some Bismuth thin films [3-5]. The non-metallic temperature dependence of resistivity in Bismuth thin films can be due to smaller grain size compared to single crystals. In bulk Bismuth the carrier concentration decreases on decreasing the temperature while the mobility increases. The increase in mobility has a stronger effect in bulk Bismuth giving the decrease in resistivity on lowering the temperature. The smaller grain size of the Bismuth thin films have a stronger grain boundary scattering which in turn weakens the effect of mobility contribution to temperature dependence of resistivity and may be causing the non-metallic resistivity behavior. The effect of this grain boundary scattering needs to be probed in more detail by varying the grain size of thins films and studying their temperature dependence. The bismuth thin films are very sensitive to disorder and the observed small abrupt increase in resistivity around 185K may be due to defects and may vanish on further annealing.

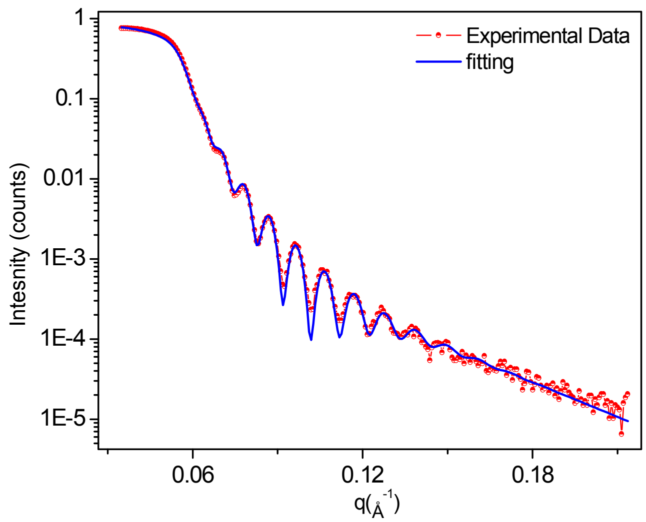

FIGURE 2: X-ray reflectivity pattern (red symbol) for the Bismuth thin film on (100) oriented Si/SiO_2 substrate. The blue solid line shows the best fit curve to the experimental data.

TABLE 1: Parameters obtained from the x-ray reflectivity fitting

Film thickness	**SiO_2 layer thickness**	**Film roughness**	**Film density**
520 Å	3000 Å	21 Å	9.8 g/cm^2

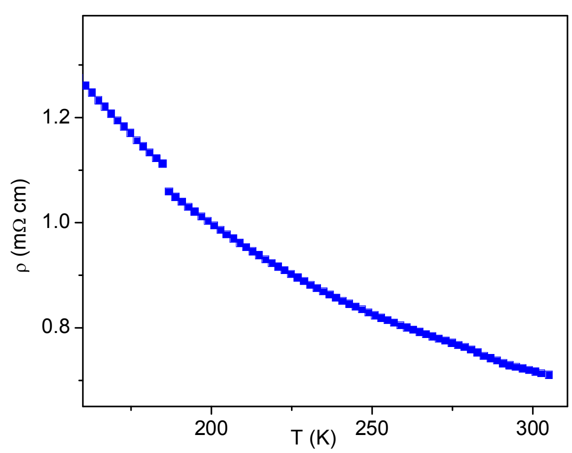

FIGURE 3: Resistivity versus temperature for Bismuth thin film on (100) oriented Si/SiO_2 substrate.

CONCLUSION

We have successfully grown high quality Bismuth thin films on the (100) oriented Si/SiO_2 substrates. The results of x-ray diffraction measurements show that films are single phase and preferentially oriented along <003> direction. The thickness of the films is around 52 nm and roughness around 2 nm and the density of the film layer is same as that of bulk. The films show non- metallic transport behavior which is attributed to decreases in carrier mobility due to enhanced scattering from smaller grain boundaries of the thin film.

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