Thickness-dependent magneto-transport of Bi$_2$Se$_3$/SiO$_2$ topological insulator thin films

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ABSTRACT

Topological insulators are immensely investigated for their surface states related properties as these materials can be used for various spintronics, quantum computing, and optoelectronics applications. In this perspective, different thicknesses of bismuth selenide thin films are deposited on the 250 nm SiO$_2$ substrate with the help of thermal deposition. The motive of this study is to investigate the surface and bulk-related behaviour with different thicknesses. The deposited films are characterized through GI-XRD (grazing incidence X-ray diffractometer) and Raman spectroscopy, which ensure the impurity-less deposition. Further, the transport properties are investigated, using Hikami–Larkin–Nagaoka (HLN) model in low field regime (up to 1 Tesla). Also, a quadratic term in field and a constant term added in HLN model to investigate the occurrence of quantum scattering and defects in thin films and this shows thickness dependence of weak anti-localization effect (WAL) in the present Bi$_2$Se$_3$/SiO$_2$ thin films.

1 Introduction

Topological insulators (TIs) are now well-known because of their extensive properties, which show the abilities of this kind of quantum materials to be used in different applications in the field of spintronics, optoelectronics, and quantum computing [1–6]. The quests began with the discovery of TIs in the condensed matter community because of its topological-dependent behaviour, which has its conducting surface states and insulating bulk [3, 6, 7]. This duality in the behaviour is protected with the time-reversal symmetry, and it is present due to its intrinsic spin–orbit coupling (SOC) [6–8]. TIs show numerous properties due to their surface states, which can be probed with highly specialized and complex techniques such as angle-resolved photoelectron spectroscopy (ARPES) [9–12]. Moreover, these particular techniques required a single-crystalline sample, which is hard to synthesize. Another way to probe the surface states is to study TIs behaviour at low temperature and low magnetic field because, at this particular condition, the surface states of the TIs are more prominent than the bulk behaviour. Secondly, it can be possible to decrease the thickness of TIs; thus, there is a high chance of dominance of surface states
at this condition, as the bulk contribution is decreased through decreasing the thickness of TIs [13–15]. However, there is a reasonable possibility of increasing the defects states with thinner films, which can disturb its intrinsic surface states.

In this perspective, the thermal evaporation technique is employed to deposit bismuth selenide thin films on the 250 nm SiO$_2$ substrate for probing surface states related transport properties. The deposition of different thicknesses of the thin film is carried out, and its confirmation is characterized through GI-XRD, while the Raman vibration modes and their dependence with thickness are also investigated. Four different thickness films are deposited, which are 5, 10, 15, and 20 nm. The transport properties of all these thin films are investigated with the help of Quantum Design Physical Property Measurement System (PPMS). It is believed that with decreasing the thickness of the thin films, there is a high probability of an increase in the contribution of surface states. Also, it is observed that with a decrease in thickness, there is an increase in the defect states, which eventually makes 5 nm thin films to be semiconducting, while all other films are metallic in nature. Thus, all these thin films are further investigated for magnetotransport up to ±12 Tesla at different temperatures, in order to see the topological surface states related character. This article is a comprehensive investigation on the thickness-dependent thin films of bismuth selenide, and considering the quantum scattering and defects term in conventional HLN model.

2 Experimental details

The thin films of bismuth selenide are deposited through a thermal evaporation system. The bulk single crystal is used for evaporation where the bulk bismuth selenide is grown under well-optimized heat treatment [16]. The grown crystal is cut down into small pieces, which are put on the molybdenum boat in the thermal evaporation chamber. The SiO$_2$ substrate is also cleaned with IPA (iso-propyl alcohol) and further with nitrogen gas. The cleaned substrate is then placed in the thermal evaporator chamber. Then, after placing the crystal and substrate at respective positions, the chamber is sealed, and a vacuum of $10^{-6}$ bar is attained with the help of rotatory and diffusion pumps. The vacuum is achieved in the chamber in approximately 4–5 h, and then evaporation of crystal is carried out by applying the current. The substrates are just placed on the rotator above the molybdenum boat. And, the rotator is switched on during the evaporation, with a speed of 45 rpm to get a uniform film.

Moreover, the thickness of the films is observed by thickness monitor of telemark model 851. The deposited films are then annealed at 300°C in the nitrogen environment to have oxidation-free bismuth selenide thin films. Different thicknesses of 5, 10, 15, and 20 nm thin films are deposited using the same process. These deposited thin films are then characterized using GI-XRD and Raman spectroscopy techniques, where the GI-XRD is taken from Panalytical X’pert Pro with Cu-K$_{\alpha1}$ X-ray radiation which is operating at 40 kV and 40 mA. While the table-top Renishaw Raman spectrometer is used for investigating Raman vibration modes of the thin films with a laser of 514 nm. Moreover, the transport properties are explored using Quantum Design Physical Property Measurement System (PPMS) down to 2 K, where the chromium-gold pads are deposited using masking in the thermal evaporation technique, and contact on the PPMS puck is made by wire-bounder.

3 Results and discussion

The GI-XRD of 5, 10, 15, and 20 nm thin films of bismuth selenide is shown in Fig. 1a. The GI-XRD peaks signify different planes of bismuth selenide, which are $(0,1,5)$, $(0,1,8)$, $(0,0,12)$, $(1,0,10)$, $(0,1,11)$, $(0,0,15)$, $(1,0,13)$, and $(0,2,4)$. All these planes are matched with bulk crystals, which ensure the deposition of bismuth selenide in the purest form. However, the sharpness of the observed peaks of all these films are found to be dependent on thickness. Basically, with a decrease in thickness, the peaks become more broader, which may be due to the increase of defect states in the system. These thin films are further investigated through Raman spectroscopy to investigate various Raman vibration modes. There are three Raman modes observed in all these films, which are $A^{1g}$, $E^{2g}$, and $A^{2g}$ [17, 18], as shown in Fig. 1b. These modes are the intrinsic modes of bismuth selenide, which ensure impurity-free deposition. Moreover, there is a Raman peak at 520 cm$^{-1}$ due to SiO$_2$ [19], as shown in the inset of Fig. 1b. It is essential to highlight that the sharpness of Raman peak is found to be dependent on thickness in such a
way that with decreasing thickness, there is a
decrease in sharpness is observed. This particular
phenomenon supports the earlier argument that with
a decrease in the thickness of the films, there is an
increase in the defect states in the system. Thus, the
GI-XRD and Raman spectroscopy ensure the depo-
sition of pure bismuth selenide, where defects states
are induced with decreasing thickness.

The transport properties of all these films are
investigated using a Quantum Design-PPMS system.
Figure 2 shows the normalized resistivity vs. tem-
perature curve of all the four thin films down to 2 K.
The 10, 15, and 20 nm films show metallic character
as their resistance decreases with a decrease in tem-
perature to 20 K, but it starts increasing thereafter
down to 2 K. The increase in resistance below 20 K
signifies the metal to semiconductor transition. It is
possible that the low thickness of thin films and the
technique employed to deposit these films have
induced defects, which may be the reason for such a
transition. It is observed that the 10 and 15 nm thin
films almost have the same upward increase from 20
to 2 K, while the 20 nm film shows only a slight push.
Apart from 10, 15, and 20 nm film, the 5 nm film
shows an increase in resistance with a lowering of
temperature down to 2 K and this increase in resis-
tance ensures the semiconducting nature of 5 nm thin
films.

The RT curve shows the semiconducting to
metallic character transition as the thickness of the
films go to 5 to 10 nm, while metal to semiconductor
transition as the thinner films goes down to 2 K.
These films are then analysed for magnetic field
dependency on resistance through RH (resistance vs.
magnetic field) curves to find the magnetoresistance
(MR %) at ± 12 Tesla, which is given as

\[
\text{MR} \% = \frac{\rho_H - \rho_0}{\rho_0} \times 100\% 
\]

where \(\rho_H\) and \(\rho_0\) are resistivity at \(H\) filed and zero
filed, respectively. Figure 3 shows the MR% of all the
thin films, where the 5 nm film has \(~ 12\%\) MR at ±
12 Tesla and 2 K, which decreases with increasing
thickness. Basically, the 10 and 15 nm film has nearly
similar \(~ 10\%\) MR which further decreased to \(~ 8\%\)
MR for 20 nm at ± 12 Tesla. This decrease symbol-
izes the increment of bulk contribution in the thin
films, while the V-shaped character at the low field in
all the thin films signifies the presence of the WAL
effect, which is an evident property of TIs. Moreover,
the V-shaped character has different MR% for dif-
ferent thickness films. As in 5 nm films, the V-shaped

Fig. 1 a Grazing incidence X-ray diffraction pattern of Bi2Se3
thin films with various thicknesses. b The Raman spectrum of
Bi2Se3 thin films at different thicknesses and the inset shows the
Raman mode for SiO2

Fig. 2 Variation of normalized resistivity ($\rho/\rho_{300K}$) as a function
of the temperature of Bi2Se3 thin films
character shows $\sim 10\%$ MR, while it is $\sim 8\%$ in 10 nm and $\sim 4\%$ in 15 and 20 nm film. These changes in MR% again ensure the bulk character increase with thickness in the bismuth selenide thin films. In 5 and 10 nm films, the MR% is found to be almost saturated above $\pm 4$ Tesla, while, in 15 and 20 nm film, it shows a dependence on the magnetic field. It again supports the argument that with an increase in the thickness of thin films, there is an increase in the bulk character of TIs, and there is suppression of surface states related dependence. Further, the HLN (Hikami–Larkin–Nagaoka) is employed to probe the MR response of all these films to find various other conduction channels. The HLN equation [20–23] is for change in conductivity of a 2D electron system in the presence of a magnetic field, and it is given by

$$\Delta \sigma(H) = -\frac{e^2}{\pi h} \left[ \ln\left(\frac{B_\varphi}{H}\right) - \psi\left(\frac{1}{2} + \frac{B_\varphi}{H}\right) \right]$$

(2)

where $h$ is Plank’s constant, $\psi$ is digamma function, $e$ is the electronic charge, $H$ is applied magnetic field and $B_\varphi$ is the characteristic field, and it is given by

$$B_\varphi = \frac{h}{8\pi e L_\varphi^2}$$

(3)

where $L_\varphi$ is phase coherence length. For theoretically fitting the change in conductivity using HLN equation, the change in magneto-conductivity (MC) is calculated by the difference between conductivity at field $H$ and zero field ($\Delta\sigma(H) = \sigma(H) - \sigma(0)$). The HLN parameters have different symbols as $\alpha$ and $L_\varphi$ are used to determine the 2D coherent conducting channel and different types of localization or scattering. Moreover, the value of $\alpha$ can resemble different cases, such as if $\alpha$ is 1, then it is referred to as an orthogonal case, while it is $-1/2$, then it is a symplectic case. Moreover, if $\alpha$ is 0, it is a unitary case. The $L_\varphi$ predicts the coupling between electron–electron or electron–phonon by inelastic scattering. Figure 4 shows the HLN fitting of MC of all the thin films, while Table 1 shows the fitting parameters.
extracted from HLN fitting. For the thickness of 5 nm film, the extracted value of $a$ at 2 K is close to $-0.5$ which signifies the contribution from single conduction channel. Also, with increasing temperature it lies in between 0 to $-0.5$ range, ensuring the presence of a single conduction channel and, most probably, surface states conduction.

Further, the 10 nm films show a similar range of extracted parameter values as in 5 nm, which confirms the presence of a single conduction channel. While its resistivity is also shown metallic behaviour along with V-shaped behaviour in MR% due to WAL effect, which confirms the dominance of surface states in 10 nm film. However, in case of 15 and 20 nm film thickness, the $a$ value lies in $-0.5$ to $-1.0$, which shows the presence of two conduction channels. The possible conduction channels in 15 and 20 nm films would be surface and bulk conduction channels, respectively. Hence, HLN fitting ensures the presence of a prominent surface conduction channel in 5 and 10 nm film, while the bulk contribution comes into the picture with 15 and 20 nm thin films. However, it is also observed that the HLN model fitting diverges at a high magnetic field of ±1 Tesla. Thus, the intrinsic characteristics of MC at high fields cannot be explained by HLN model. In order to improvise the HLN fitting, two different parameters are added along with HLN, and the modified equation of theoretical fit of MC is given by

![Fig. 4](image)

**Fig. 4** Temperature dependence of magneto-conductivity in a transverse magnetic field is fitted with HLN equation of Bi$_2$Se$_3$ thin films with the thickness. a 5 nm, b 10 nm, c 15 nm, and d 20 nm

| Temperature (K) | Thickness (nm) | $a$  | $L_o$ (nm) |
|-----------------|----------------|------|------------|
| 2               | 5              | $-0.51$ | 207.06     |
|                 | 10             | $-0.54$ | 185.62     |
|                 | 15             | $-0.62$ | 147.99     |
|                 | 20             | $-0.68$ | 133.91     |
| 5               | 5              | $-0.37$ | 188.02     |
|                 | 10             | $-0.46$ | 131.87     |
|                 | 15             | $-0.52$ | 117.30     |
|                 | 20             | $-0.56$ | 110.36     |
| 20              | 5              | $-0.32$ | 70.82      |
|                 | 10             | $-0.41$ | 54.79      |
|                 | 15             | $-0.58$ | 51.99      |
|                 | 20             | $-0.75$ | 49.32      |
\[ \Delta \sigma(H) = - \frac{\alpha e^2}{n h} \left[ \ln \left( \frac{B_\sigma}{H} \right) - \Psi \left( \frac{1}{2} + \frac{B_\sigma}{H} \right) \right] + \beta H^2 + c \]

(4)

where the first part is again the HLN part, as it best describes the V-shaped contribution due to WAL effect and surface conduction at low field, while at high field, \( \beta H^2 \) term comes into the picture. It combines both classical and quantum terms, which correspond to elastic and spin–orbit scattering lengths and also cyclotronic MR [24, 25]. At last, the \( 'c' \) is added to represent the defect states related contributions. Figure 5 shows the fitting of MC using Eq. 5, and it is well evident from the fitting that there is no divergence of theoretical fit with experimental results. Table 2 shows the fitting parameters for Bi₂Se₃ thin films obtained from fitting the HLN + \( \beta H^2 + c \) equation at different temperatures. The value of \( \alpha \) lies in the same regime as in the HLN case, while the \( \beta \) is found to be decreasing with temperature in all the films, which shows an increase in the bulk contribution. Moreover, the value of \( \beta \) is also decreased with an increase in thickness, which also supports the argument of an increase in the bulk contribution with increasing thickness. At last, the value of \( 'c' \) represents the defects state, which shows that there are more defects in 5 nm as compared to the 20 nm thin film of bismuth selenide. In all, this study shows the dependence of the presence of surface and bulk conduction channel with the thickness of the thin films in 5, 10, 15, and 20 nm thin films of bismuth selenide, which open up a wide area of applications in the field of spintronics, quantum computing and as a thermistor device.

4 Conclusion

This article shows the successful deposition of different thicknesses of thin films using the thermal evaporation technique under a well-optimized annealing treatment. The GI-XRD and Raman spectroscopy ensure the impurity-free deposition of all the thin films. While, the transport properties show

![Figure 5](image-url)
the metal to semiconductor transition in 10, 15, and 20 nm thin films whereas, 5 nm film is semiconducting in full temperature range. Moreover, the MC studies ensure the presence of the WAL effect in all the films along with surface and bulk conduction channel dominance at different thicknesses. The modified HLN fit helps in concluding the defects and bulk contribution in the thin films.

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**Author contributions**

All the authors contributed equally to the research work and its planning.

**Data availability**

All data generated or analysed during this study are included in this published article.

| Temperature (K) | Thickness (nm) | $\alpha$ | $L_\varphi$ (nm) | $\beta$ | $c$ |
|-----------------|----------------|--------|-----------------|-------|-----|
| 2               | 5              | -0.347 | 474.55          | 0.00074 | 0.0406 |
|                 | 10             | -0.443 | 289.01          | 0.00070 | 0.0397 |
|                 | 15             | -0.564 | 175.39          | -0.00274 | 0.0182 |
|                 | 20             | -0.680 | 142.86          | -0.00325 | 0.0238 |
| 5               | 5              | -0.257 | 356.05          | 0.00012 | 0.0033 |
|                 | 10             | -0.387 | 170.10          | 0.00024 | 0.0068 |
|                 | 15             | -0.481 | 131.89          | -0.00284 | 0.0045 |
|                 | 20             | -0.678 | 84.11           | -0.00344 | -0.0229 |
| 20              | 5              | -0.208 | 115.29          | -0.00012 | 0.0107 |
|                 | 10             | -0.323 | 68.52           | -0.00007 | 0.0082 |
|                 | 15             | -0.462 | 62.54           | -0.00293 | 0.0085 |
|                 | 20             | -0.761 | 46.03           | -0.00355 | -0.0101 |

**Declarations**

**Conflict of interest** This is to certify, that the authors of the MS below submitted to Journal of Materials Science: Materials in Electronics have NO competing interest and the research is shared equally by all the authors.

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