Towards More Accurate Determination of the Thermoelectric Properties of Bi$_2$Se$_3$ Epifilms by Suspension via Nanomachining Techniques

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Abstract: We report on the characterization of the thermoelectric properties of Bi$_2$Se$_3$ epifilms. MBE-grown Bi$_2$Se$_3$ films on GaAs (111) A are nanomachined with integrated Pt elements serving as local joule heaters, thermometers, and voltage probes. We suspended a 4 $\mu$m $\times$ 120 $\mu$m Bi$_2$Se$_3$ by nanomachining techniques. Specifically, we selectively etched GaAs buffer/substrate layers by citric acid solution followed by a critical point drying method. We found that the self-heating 3$\omega$ method is an appropriate technique for the accurate measurement of the thermal conductivity of suspended Bi$_2$Se$_3$. The measured thermoelectric properties of 200 nm thick Bi$_2$Se$_3$ at room temperature were $\kappa = 1.95$ W/m K, $S = -102.8$ $\mu$V/K, $\sigma = 75,581$ S/m and the figure of merit was $ZT = 0.12$. The study introduces a method to measure thermal conductivity accurately by suspending thin films.

Keywords: Bi$_2$Se$_3$; MBE; thermoelectric; nanostructuring; 3$\omega$ method

1. Introduction

Thermoelectric (TE) devices that create a voltage by heat waste have been under development for a few decades [1–3]. However, their applications are limited to a few industries due to their low efficiency in energy conversion [4,5]. The efficiency of TE devices is determined by the dimensionless figure of merit defined by the Seebeck coefficient ($S$), electrical conductivity ($\sigma$), thermal conductivity ($\kappa$) and the temperature ($T$). A high Seebeck coefficient, high electrical conductivity, and low thermal conductivity are required to maximize the figure of merit [6]. However, the interdependence of the Seebeck coefficient, electrical conductivity, and the electronic contribution to the thermal conductivity makes the optimization of $ZT$ difficult. The electrical conductivity and electrical contribution to thermal conductivity are linked to each other by the Wiedemann–Franz law. According to the simple theory for nearly free electrons, the Seebeck coefficient of metals or degenerate semiconductors [7] decreases when the carrier concentration ($n$) increases. However, when the carrier concentration ($n$) increases, the electrical conductivity ($\sigma$) and electronic thermal conductivity ($\kappa_e$) increase [8].

To overcome these correlations between the quantities, one needs to maximize the power factor ($S^2\sigma$) by optimizing the carrier concentrations ($10^{19}$–$10^{21}$ carriers per cm$^3$) [8] and/or minimize the lattice thermal conductivity ($\kappa_l$) by increasing phonon scattering. The introduction of point defects and doping effectively controls the carrier concentration and the power factor [9–11]. Nanostructuring, alloying, and the application of grain size and strain are used to enhance the phonon scattering and reduce the lattice thermal conductivity [12–15]. Because the mean free path (mfp) of electrons is much shorter than that of phonons in heavily doped semiconductors, nano-structuring effectively reduces the lattice contribution to thermal conductivity while preserving the electrical conductivity [12,16]. Recently, such material...
systems as phonon-glass electron-crystal, where phonon transport is confined in plane, have advanced thermoelectric research [17–19].

Despite the progress in nanostructuring reported for nanowires, superlattices, and nanocomposites in thermoelectric research of nano-films, there have been difficulties associated with heat diffusion to the substrate [20–24]. We present the self-heating $3\omega$ method to measure the fundamental thermal properties by suspending the film [25–27]. Consider a suspended rod-like metal for thermal conductivity measurements [28]. An electrical current of the form $I_0 \sin \omega t$ is sourced from outside probes. This results in Joule heating of the form $0.5 I_0^2 R (1 - \cos 2\omega t)$, where $R$ is the resistance of the metal. This periodic Joule heating results in temperature oscillation at a frequency of $2\omega$, which causes the resistance of the metal to oscillate at $2\omega$. The amplitude of the voltage at $3\omega$ is determined by the thermal properties and described by the 1D heat diffusion equation. With proper boundary conditions and an initial condition, an explicit solution for the 1D heat-conduction equation at the low frequency limit can be found and the thermal conductivity ($\kappa$) can be measured from the excitation current ($I$), electrical resistance ($R$) and its differential respect to temperature ($R' = dR/dT$), geometrical length ($L$) and cross-section area ($A$), and measured $V_{3\omega}$ signal. [29] This solution is obtained from the assumption that the joule’s heat fully dissipates in the rod and not in its surroundings, particularly the underlying substrate. A freely suspended structure best meets these important criteria. The $3\omega$ methods can be applied to materials with a linear Ohmic current-voltage behavior.

The best TE materials typically degenerate semiconductors that have high carrier mobilities and a low electronic contribution to thermal conductivity [14]. $\text{Bi}_2\text{Se}_3$ is a semiconductor with a narrow bandgap of about 0.3 eV, which meets the conditions for a large Seebeck coefficient [9]. Naturally occurring Se-vacancies that act as electron donors result in high electrical conductivity, thus $\text{Bi}_2\text{Se}_3$ is known as one of the best TE materials [8]. A recent theoretical prediction of a topologically protected surface states in $\text{Bi}_2\text{Se}_3$ has rekindled interest in the development of high-quality materials [30,31]. In this study, we epitaxially grew 200 nm thick $\text{Bi}_2\text{Se}_3$ films on a GaAs (111) A substrate by molecular beam epitaxy (MBE). Nanostructuring techniques enable the accurate measurement of the thermal conductivity by freely suspending the $\text{Bi}_2\text{Se}_3$ from the GaAs substrate. Additionally, nanostructuring effectively suppresses the lattice’s contribution to thermal conductivity due to the low dimension [9].

2. Materials and Methods

In this study, $\text{Bi}_2\text{Se}_3$ epifilms were grown on a semi-insulating GaAs (111) A substrate using effusion cells charged with Bi (99.9999%), Se (99.999+%), Ga (99.999%), and As (99.99999+%). In the growth chamber, after standard deoxidation, a substrate temperature of 700 °C, a 40 nm thick GaAs buffer layer was grown at a deposition rate of 1 nm/min under As-rich conditions. Then, 200 nm $\text{Bi}_2\text{Se}_3$ epifilms were grown at a substrate temperature of 300 °C with a growth rate of 2 nm/min under Se-rich conditions with the flux ratio ($\Phi_{\text{Se}} / \Phi_{\text{Bi}}$) of ~20 [32,33]. The growth rate and surface morphology were monitored by RHEED using oscillations and typical surface reconstructions (Figure 1a,b) [34,35]. Finally, a Se-capping layer was deposited below 150 °C to minimize the Se-vacancies. The resulting morphologies and crystal structures (Figure 1) are characterized by a commercially available non-contact mode AFM and from high resolution XRD measurements. Epifilms for this study all showed characteristic triangular pyramid morphologies with sizes and densities being dependent on the stoichiometry and other growth conditions. Thickness was measured by SEM and a profilometer.
After epitaxy deposition, the samples were readied for nanofabrication by mild thermal treatment at 200 °C to remove the Se-capping layer. All nanopatterning was conducted by a modified commercially available SEM. The first alignment marks were realized after lift-off of DC sputter deposition of 20 nm thick Pt [36] after a negative resist process using an image reversal of AZ520X. Then, Bi$_2$Se$_3$ was patterned into a nanobeam structure (0.2 μm × 4 μm × 120 μm) by a similar image reversal process and mildly etched in a commercially available RIE in Ar plasma (0.015 mTorr and 150 W) with an etch rate of 100 nm/min. Then, necessary electrical probes and resistive thermometer in contact with a Bi$_2$Se$_3$ nanobeam as well as in-situ joule heaters situated on the GaAs substrate in proximity to the Bi$_2$Se$_3$ nanobeam were used again after the lift-off process of DC sputtered 200 nm Pt. To minimize substrate effects in thermal-transport measurements, the Bi$_2$Se$_3$ nanobeam was freely suspended by first patterning an etch window and then selectively etching the underlying GaAs using a mild citric acid solution (volumetric 6:1 ratio of citric acid and hydrogen peroxide) [37] at an etch rate of 100 nm/min. After suspending the Bi$_2$Se$_3$ nanobeam, the sample is dried in a commercially available critical point dryer (CPD). The final structure is shown in Figure 2f.

After nanofabrication, electrical contacts were formed by Au wire-bonding. Electrical probes on Bi$_2$Se$_3$ were verified as being Ohmic from $I$–$V$ measurements. Then, the fabricated Pt thermometer in proximity to the Bi$_2$Se$_3$ nanobeam was calibrated by measuring its resistivity while monitoring the sample temperature in a vacuum cryostat (<0.1 mTorr) using a commercially available temperature controller (LakeShore, Westerville, OH, USA, LS330). As expected, the Pt thermometer’s resistivity showed a linear dependence to the temperature range reported in this study (60–300 K) (Figure 3b). As the Pt thermometer was in physical contact with the Bi$_2$Se$_3$ nanobeam, the joule heater was placed in proximity to the nanobeam on the semi-insulating GaAs substrate. Accordingly, the thermometer and heater were electrically isolated.

Figure 1. RHEED patterns during epitaxial growth of (a) the GaAs buffer layer and (b) Bi$_2$Se$_3$, (c) 1 μm × 1 μm AFM scan image, (d) high-resolution X-ray diffraction of Bi$_2$Se$_3$. 
were determined at substrate temperature. Then, a DC current was allowed to flow in one
whole temperature range (inset) when the Joule heater was ‘on’ (red square) and ‘off’ (blue circle).

After mild thermal treatment to remove the Se capping layer, the voltage across the Bi
2Se3 was measured to determine the temperature gradient, which was found to be in the range 0.1 K/µm
at 300 K–0.01 K/µm at 60 K. Considering that GaAs thermal conductivity greatly increases
with a rise in temperature (for the whole temperature range (inset)) when the Joule heater is ‘on’ (red square) and ‘off’ (blue circle).

Figure 2 schematically depicts AC thermal-transport measurements (i.e., thermal
Seebeck effect). First, to measure the thermal gradient, the resistivities of the two
thermometers were measured using an ac-lock-in technique (17 Hz and 27 Hz with IAC 5 µA) to determine the
temperature gradient, which was found to be in the range 0.1 K/µm at 300–0.01 K/µm at
60 K. Considering that GaAs thermal conductivity greatly increases at lower temperatures
and is an order of magnitude higher than that of Bi2Se3, this result (lower thermal gradient
at lower temperature) indicates that the effect of the substrate (i.e., increase in thermal
conductivity of the GaAs substrate at low temperature) cannot be easily ignored. After a
time delay of 10 s after the heater is turned on to ensure thermal equilibrium, the DC
voltage across the Bi2Se3 nanobeam was averaged and recorded after the probes, as the
thermometers are disconnected by relay signals. The process was repeated from room
temperature to 60 K while the substrate temperature was monitored and maintained.
Figure 4 schematically depicts AC thermal-transport measurements (i.e., thermal conductivity). We utilized two phased-locked ac lock-in amplifiers. To validate the $3\omega$ method, we swept the ac current ($I_{AC}$) at $f = 13$ Hz. One ac lock-in at $f$ was used to measure the electrical ac conductivity of the Bi$_2$Se$_3$ nanobeam, while the other lock-in at $3f$ was recorded as $V_{3\omega}$. Figure 5b shows the expected cubic dependence of $V_{3\omega}$ to the injected current $I_{AC}$. Similar to the above Seebeck effect measurements, the sample temperature was varied from room temperature to 60 K. From these measurements, we recorded the electrical conductivity $\sigma$, $V_{3\omega}$, and determined $R' = [R(T_1) - R(T_2)]/[T_1 - T_2]$.
3. Results

From the Bi$_2$Se$_3$ epifilms, we successfully fashioned a versatile thermal-transport measurement platform in which the epifilm to be characterized is freely suspended to minimize thermal transport through the substrate. From in-situ RHEED monitoring during growth as well as post-growth morphologies characterized by non-contact mode AFM and high-resolution XRD, the resulting Bi$_2$Se$_3$ epifilms (Table 1) were found to compare favorably to those reported elsewhere [28,38–40]. Then, the resulting suspended Bi$_2$Se$_3$ with integrated electrical probes as thermometry in its proximity were then characterized in a cryogenic cryostat. First, the temperature dependence of the electrical conductivity of the as-grown Bi$_2$Se$_3$ epifilm and Bi$_2$Se$_3$ nanobeam is depicted in (Figure 6b). Characteristic temperature dependence indicate that the nanomachining processes had a minimal effect. At room temperature, for the 200 nm Bi$_2$Se$_3$ nanobeam, the electrical conductivity (σ) was 75,580 Ω$^{-1}$·m$^{-1}$. Again, this value is favorable compared with that of similarly sized bulk nanoribbon samples reported elsewhere [38].

Table 1. Summary of selected thermoelectric measurements of Bi$_2$Se$_3$.

| Type of Bi$_2$Se$_3$                      | κ (W/mK) | σ (10$^4$ S/m) | S (µV/K) | ZT |
|------------------------------------------|----------|----------------|----------|----|
| mechanically exfoliated Bi$_2$Se$_3$ [28]| ~2.1     | N/A            | N/A      | N/A|
| nanoribbon synthesized via VLS (S2) [38]| ~1.7     | ~7             | ~120     | 0.17|
| 30 nm epifilm [39]                      | N/A      | ~6             | ~100     | N/A|
| 77 nm vapor solid grown Bi$_2$Se$_3$ [40]| N/A      | 15.9           | ~99.9    | N/A|
| 200 nm epifilm (this report)            | ~1.9     | ~7.6           | ~103     | 0.12|
| bulk [41]                               | 4        | 16             | ~70      | 0.05|

Using the patterned thermometry elements near the Bi$_2$Se$_3$ nanobeam, Pt resistive thermometers in contact with Bi$_2$Se$_3$, and the Pt joule heater in proximity, we showed that a steady temperature gradient (ΔT ranging from 1 K to 10 K) can be maintained across the Bi$_2$Se$_3$ nanobeam for the temperature range presented here (60–300 K). Interestingly, for the equal heater power, higher temperature gradients were achieved at higher temperatures. This suggests that at lower temperatures, a significant amount of heat flows underneath the suspended Bi$_2$Se$_3$ nanobeam across the GaAs substrate. With a controllable temperature gradient and electrical probes to measure the electrical potential across the nanobeam, the Seebeck voltage can be directly measured. Figure 6a plots the Seebeck voltage for the 200 nm Bi$_2$Se$_3$ nanobeam structure. The sign of the voltage as well as the temperature dependence agree with the expected dominant n-type carriers from Se vacancies. Again, the Seebeck voltage of $-102.8$ µV/K for our 200 nm Bi$_2$Se$_3$ nanobeam compares favorably to values obtained from Bi$_2$Se$_3$ film grown by MBE [39], vapor-solid method [40] and Bi$_2$Se$_3$ nanoribbon synthesized by the vapor liquid solid method, which are also suspended from the substrate [38]. As the Seebeck voltage is expected to be dominated by thermal carrier diffusion, the fact that the absolute measured values of the Seebeck voltage for the nanobeam were slightly smaller than those of the nanoribbon possibly indicated a higher quality Bi$_2$Se$_3$ epifilm.
introduction, we applied the 3ω method, specifically the self-heating method derived from the 1D heat diffusion equation [28]. As the heat was transferred directly to the nanobeam, the resulting measurements were robust with minimal substrate contributions.

Starting from the 1D heat diffusion model, thermal conductivity (κ) can be quantified by the self-heating 3ω method. An injected current of ω is used to “self-heat” the sample, and by trigonometric identity and joule heating, we can measure the $V_ω$ to determine the electrical conductivity (σ) and $V_3ω$. Then, along with σ (T), dR/dT, and $V_ω$, a temperature dependence of $\kappa$ ($\sim [4RRL^3][\pi^4AV_3ω]^{-1}$) can be seen as depicted in Figure 6c. At 300 K, the Bi$_2$Se$_3$ nanobeam $\kappa$ was found to be ~1.95 Wm$^{-1}$K$^{-1}$ (see Supplementary Materials). Similar to the nanoribbon sample [28,38], the measured value of $\kappa$ and its temperature dependence compared favorably. The monotonic increase in $\kappa$ with a decrease in temperature is consistent with metallic-like behavior found in Se-vacancy dominated Bi$_2$Se$_3$ materials. Along with the temperature dependent measurement of the Seebeck voltage, electrical conductance, and thermal conductance, the thermoelectric figure of merit (ZT) temperature dependence is plotted in Figure 6d. The uncertainty in our measurements is limited from thickness measurements, which we found from RHEED oscillations as well as stylus profilometer measurements and is typically within 5%. However, in determining the ZT values, the geometrical factors are cancelled out. With suppressed thermal conduc-
tivity from the reduced dimensionality of the nanobeam, we found the $ZT$ values to be significantly larger than the reported bulk values at room temperature.

4. Summary

To more accurately determine the thermoelectric properties of Bi$_2$Se$_3$ epifilms, we utilized nanomachining techniques to freely suspend the structure. We also integrated thermometric and electrical probes. We applied the self-heating $3\omega$ method to more accurately measure thermal conductivity. This method further minimizes heat transfer into the substrate and allows for more accurate determination as an ac measurement technique. From the characterization of the surface morphology and electrical measurements, we found our Bi$_2$Se$_3$ epifilms to be comparable to other reported epifilms. Electrical measurements conducted after nanofabrication processing showed minimal effects on the quality of the Bi$_2$Se$_3$. A reduction in the dimensionality of the epifilms resulted in a significant decrease in thermal conductivity compared to the bulk [41], with minimal decreases in the electrical conductivity and an increase in the Seebeck coefficient. Such changes in the thermal electric properties resulted in an increase in the figure of merit ($ZT$) compared with the bulk and reported studies, as in [41,42].

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/s22208042/s1, Figure S1: Replot of Figure 5b in main text; Figure S2: Temperature dependence of 200 nm Bi$_2$Se$_3$ nanobeam.

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