Thermal Conductivity of Indium Phosphide Based Superlattices

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Abstract

Semiconductor superlattice structures have shown promise as thermoelectric materials for their high power factor and low thermal conductivity [1,2]. Heat conduction by phonons in semiconductor superlattices is controlled by two mechanisms: interface scattering by defects and/or roughness and phonon filtering by Bragg reflection. The thermal conductivity of three different InP/InGaAs superlattices was measured from 77 - 300 K using the 3ω method. Small "dips" in the measured thermal conductivity were observed near 90 K, which may be indicative of phonon filtering.

Introduction

The thermal conductivity of a thermoelectric material is a key parameter in both the thermoelectric (TE) and thermionic (TI) figure of merit. In both cases, the device performance increases with a decrease in thermal conductivity. Heat transport in semiconductors is dominated by phonons for carrier densities below 10²⁰ cm⁻³. Recent work has shown that the thermal conductivity of semiconductor superlattices is often lower than the value as calculated from their constituent materials using Fourier heat conduction theory [3-8]. Additionally, in some instances, thermal conductivities have been reported that are below the value of a corresponding alloy [5-8]. This reduction of thermal conductivity has mainly been attributed to two mechanisms: (i) phonon scattering at interfaces due to roughness, defects, and/or dislocations, (ii) phonon "filtering" [9]. Previous measurements could not separate out these two effects because the superlattices were not lattice matched. It is well known that interface scattering will reduce the phonon mean free path and, hence, decrease the heat transport. However, an increase in scattering sites also increases the electrical resistance causing an undesirable drop in the power factor

 $S^2\sigma$. Therefore, it seems that the reduction of thermal conductivity through the use of phonon bandgaps and phonon filtering should be explored.

In this work we measured the thermal conductivity of three different lattice matched InGaAs/InP superlattices. The reasons for studying this particular superlattice system are twofold. One is that InGaAs/InP superlattices are currently being utilized as thermionic coolers [10]. Additionally, these superlattices are lattice matched with high quality interfaces, such that the effects of the interface scattering will be greatly diminished. For this reason it may be possible to separate out the effects of phonon filtering and interface scattering.

Analogous to optical filtering by distributed Bragg reflection, phonon filtering takes place when phonons of wavelength λ_0 satisfy the Bragg condition $\lambda_0=2d_0$, where d_0 is the superlattice period. In a very nice set of experiments, Narayanamurti et al. [9] showed that for a GaAs/AlGaAs superlattice, longitudinal phonon transmission near one meV can be reduced by roughly 60 percent over a bandgap of about 0.1 meV. However, that experiment was done with monochromatic phonons, while heat is conducted over a wide range of phonon frequencies. Therefore, it is more difficult to observe the phonon filtering phenomenon directly from thermal conductivity data. However, since the dominant phonon frequency is dependent on temperature it is possible to study spectral filtering through the Planck blackbody distribution by measuring the thermal conductivity over a range of temperatures.

Experimental Details

The thermal conductivity of the InGaAs/InP superlattices was measured using the 3ω technique [7]. The 3ω method is an ac technique that utilizes a narrow metal line that is patterned on the surface of the sample as both a heater and a

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thermometer. A current with angular frequency ω is passed through the metal line, which heats the surface of the sample through Joule heating at a frequency 2ω . The resistance of a pure metal increases with increasing temperature, therefore causing the electrical resistance of the heater to also oscillate at 2ω . This 2ω component times the original driving current at ω produces a small voltage oscillation at 3ω which can be used to determine the thermal response of the sample.

Aluminum Heater / Thermometer (4-20 µm wide, 500-2000 µm long)



Figure 1: Superlattice with aluminum heater patterned on top. The two InGaAs layers are used as electrical contacts for other measurements, while the PECVD oxide is deposited on top for electrical isolation between the aluminum heater and the superlattice.

Figure 1 shows a cross-sectional view of the sample. These samples were also used in a variety of other experiments and the InGaAs layers above and below the superlattice serve as contacts. The lattice-matched InP/InGaAs superlattice was deposited by MetalOrganic Chemical Vapor Deposition (MOCVD). For a dielectric sample the metal heater/thermometer line may be placed directly on the surface. However, since the superlattices are electrically conducting a thin insulating layer must be deposited on the sample first. We used a 130 nm thick SiO₂ layer that is deposited using Plasma Enhanced Chemical Vapor Deposition (PECVD) at 300°C. An aluminum heater with a thickness of ~200 nm was then evaporated and patterned on top of the oxide layer using the lift off technique. The heaters used in this work were typically 5 µm wide and 500 µm long.

The temperature rise at the surface was measured over a wide range of frequencies. This measurement gave the series resistance of the substrate and the films on the surface. The thermal conductivity of the substrate was readily obtained from the slope of the measured temperature rise (ΔT) vs. the natural log of frequency using [11]

$$K_{sub} = \frac{V^3 \ln(f_2/f_1)}{4\pi l R^2 (V_{3\varpi,1} - V_{3\varpi,2})} \frac{dR}{dT}$$
(1)

where K_{sub} is the thermal conductivity of the substrate, V is the voltage across the metal line at ω , R is the resistance of the metal line, I is the length of the metal line, f_2 and f_1 are the two frequencies used to find the slope, $V_{3\omega,2}$ and $V_{3\omega,1}$ are the

in phase 3ω voltages at the two frequencies, and dR/dT is the measured resistance change with temperature of the metal line

Once the thermal conductivity is known, the ΔT of the substrate can be calculated [12]. The thermal conductivity of the InP substrate can be found in Figure 4. Since each film is thin and has little capacitance, it simply adds a frequency independent ΔT to the temperature rise of the substrate.

$$\Delta T_{Total} = \Delta T_{Sub} + \Delta T_{InGaAs} + \Delta T_{SL} + \Delta T_{Oxide}$$
 (2)

The thermal conductivities for the InGaAs and oxide layers were measured separately on other samples using [12]

$$K_{film} = \frac{Pt}{\Delta T_{film} wl} \tag{3}$$

where K_{film} is the film thermal conductivity, P is the heater power, t is the film thickness, ΔT_{film} is the temperature rise of the film, and w and l are the width and length of the heater/thermometer line, respectively. These measurements were performed on separate samples that accompanied the superlattices through the **PECVD** oxidation, photolithography, and lift off processes. These samples were 130 nm of PECVD oxide on a bare silicon wafer and 2 µm of InGaAs on an InP substrate with the 130 nm SiO₂ layer on top. The thermal conductivity of the InGaAs and oxide are shown in Figures 3 and 4, respectively. Once the contributions of the InGaAs and oxide layers were determined, they were subtracted out from the raw data. which left only the ΔT of the superlattice. The thermal conductivity of the superlattice was then determined from eqn (3) in the same manner as for the InGaAs and oxide.

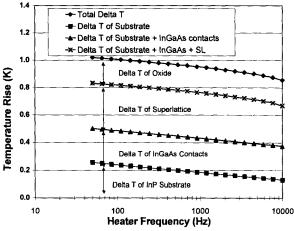


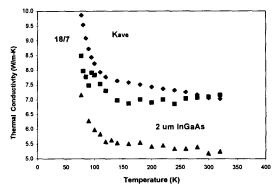
Figure 2: Temperature oscillations as a function of frequency for a superlattice. Note the frequency independent temperature rise associated with each thin film.

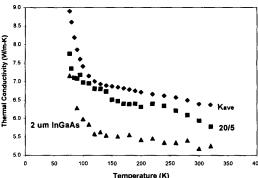
The experiments were conducted in a liquid nitrogen cryostat over a temperature range of 77 - 320 K and the data was taken with a lock in amplifier.

Results and Discussion

Three InGaAs/InP superlattices were measured in this study. All three samples had 80 superlattice periods with each period 25 nm thick to give a total thickness of 2µm. The ratio of the thickness of the InGaAs layer to the InP layer in each period

was different for the three samples. The first superlattice contained 18 nm of InGaAs and 7 nm of InP in each period, while for the 2nd and 3rd samples this ratio was 20:5 and 22:3, respectively. This ratio was changed in order to shift the location of the phonon bandgaps.





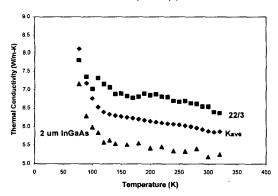


Figure 3: Thermal conductivities for the three superlattices. The labels refer to the ratio of InGaAs to InP in each period (i.e. 18/7 is for the superlattice with a period of 18 nm of InGaAs and 7 nm of InP). All superlattices had 80 periods of 25 nm each giving a total thickness of $2 \, \mu m$.

In this study, only relatively narrow lines (~5 µm) were used. Since the half width of the line is approximately the same as the film thickness, thermal conductivities for the superlattices and the InGaAs film reported here are a combination of the in plane and cross plane values. In the future, wide lines will be used in conjunction with the narrow ones along with a finite difference model of the heat conduction in order to extract the two values [13].

Figure 3 shows the measured thermal conductivities for the three superlattices studied. The 2 μm InGaAs layer is

included for reference. K_{ave} is simply an "average" thermal conductivity that was calculated also for reference. It is calculated from eqn (4) which does not take into account any interface scattering or phonon interactions.

$$K_{ave}=(d_1+d_2)K_1K_2/(K_2d_1+K_1d_2)$$
 (4) where d is the thickness of each layer, K is the thermal conductivity of the layer, and the subscripts 1 and 2 refer to InGaAs and InP, respectively.

The uncertainty in the absolute value of the thermal conductivities is rather high at about 25 percent. This uncertainty was dominated by the uncertainty in the widths of the aluminum lines. Despite the fact that that the widths were measured with an Atomic Force Microscope (AFM), there was some variation in the line width along the length of the heater/thermometer which left us with an uncertainty of approximately $\pm 0.5~\mu m$. This accounted for over half of the total overall uncertainty. The large uncertainty is likely the reason that $K_{\rm ave}$ was above the thermal conductivity for two of the superlattices, but below the third.

However, it is important to note that this uncertainty was constant over the temperature range studied. Therefore, while the absolute value of the thermal conductivity has a high uncertainty, we expect that the general trends of the data are accurate. With that in mind, it is apparent that for all three superlattices measured there is a slight, but noticeable, "dip" in the thermal conductivity curve between 90 and 100 K. The exact reason for this feature is unclear at the present time, although we suggest that it could be indicative of phonon filtering at this temperature.

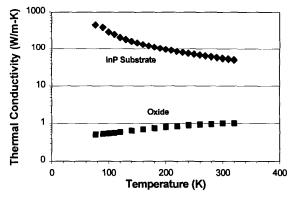


Figure 4: Measured thermal conductivities for the InP substrates and the 130 nm PECVD SiO₂. These values were used to determine the temperature rise in each film for eqn (3).

Future Work

Clearly there is more work to be done. The finite difference model will be completed to extract K_{normal} and K_{parallel} . Additionally, variations in the number of periods will be used in order to gain insight as to the role interface scattering in heat conduction.

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