Diminished thermal conductivity of Si/SiGe multilayers, established through heating current frequency

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ABSTRACT

We report on the measurement of the thermal conductivity of $Si/Si_{0.8}Ge_{0.2}$ multilayers on Si substrates through a variation of the 3ω method. We exploit the frequency dependent variation of the thermal wave, through invoking the thermal penetration depth (TPD), which is inversely proportional to the frequency. Consequently, spectral measurements covering decades of frequency were used to finely probe the substrate and the overlying Si and $Si_{0.8}Ge_{0.2}$ thin film layers. Both in-phase and out-of phase measurements yielded comparable values of the thermal conductivity in the range of 3-5 W/mK, much lower than the reported bulk values. Our results provide proof of the potential of multilayered media to be used for reduced thermal conductance applications such as thermoelectrics, heat insulation etc.

INTRODUCTION

Lower dimensional thermoelectrics, such as superlattices and nanowires have been proposed[1,2] to have a higher thermoelectric figure of merit ZT(= $\frac{S^2\sigma}{\kappa}$), - a prime determinant of the heat conversion efficiency where S is the Seebeck coefficient, σ, the electrical conductivity, and κ, the thermal conductivity. Generally, κ has contributions from both charge carriers and the lattice – in this paper we mainly consider the latter contribution, κ_L, as it typically dominates the total thermal conductivity. The increase in ZT in nanostructures was then predicted to arise from both an increase of the power factor- $S^2\sigma$, due to a net increase in the magnitude[3] of the density of states (DOS) and through a reduction of the κ_L. While a few experiments[4,5] indicate an increase in the power factor most of the understood enhancement in ZT seems to arise from a reduced κ_L. The reduction seems mainly to arise through a reduction of the mean free path, l, as $\kappa_L \sim Cvl$ (C: specific heat, and v: phonon velocity)[6] and is manifested, for example, in bulk nanocomposite thermoelectrics[7,8], Bi_2Te_3/Sb_2Te_3 superlattice devices[9] with the highest recorded ZT (of ~ 2.4), and Si nanowires[10]. It is therefore imperative that methodology for the measurement of κ_L be well developed and understood. In this paper, we focus on planar thin film based $Si/Si_{0.8}Ge_{0.2}$ multilayer structures.

In thin films, the κ_L values are generally anisotropic whichcan be understood through the dependence of κ_L on l and also confirmed through various experimental measurements[11], e.g., on GaAs/AlGaAs thin film superlattice structures, where a smaller l in the superlattice growth direction implies a smaller value of κ_L in that direction compared to the value in the film plane.

However, it was observed that while the κ_L of AlAs/GaAs superlattices[12], comprised of an equal thickness of AlAs and GaAs indeed decreases with a decreasing layer thickness *and* superlattice period (d), the measured κ_L was *greater* than that of $Al_{0.5}Ga_{0.5}As$ bulk alloy. Consequently, the κ_L of the superlattices seemed to be at most lowered to the κ_L of the alloy, from which it can be surmised that alloy scattering is possibly a stronger κ_L suppression mechanism. However, a value below that of the bulk alloy κ_L and above that of amorphous Si was measured[13] for Si_m-Ge_n superlattices (m and n refer to the number of monolayers), where for 3 nm < d < 7 nm, the κ_L decreases with decreasing d. Surprisingly, measurements on structures with d > 13 nm indicated an even smaller κ_L. It is then interesting to consider lowered κ_L in SiGe multilayer/superlattice structures and our work contributes to this goal.

For the measurement of κ_L, a variation of the 3ω method[14] was used. In this method, the periodic heating, through current modulated at a frequency ω, induces a change in the resistance with a 2ω dependence which can then be inferred through measurement of the third harmonic component of the voltage, V (3ω). The κ_L is then inversely related to the temperature rise (ΔT) due to the heating, which is directly proportional to V (3ω). Now, from a solution to the diffusion equation, it can be inferred that the thermal penetration depth (TPD) = $\sqrt{\frac{\kappa_L}{C\rho\omega}}$, which measures the extent to which the heat penetrates into the underlying substrate varies inversely with frequency ($\omega = 2\pi f$). Consequently, in a multilayer sample on a substrate, increasing ω would reduce the TPD and different frequencies could be used to sample, and measure the ΔT drop and thermal conductivity of the different layers.

EXPERIMENTAL DETAILS

$Si/Si_{0.8}Ge_{0.2}$ films on Si substrates (~ 0.5 mm thick) with a varying number of periods, and crystallinity, were deposited using a high throughput DC magnetron sputtering system. In this paper, we report studies on multilayer samples corresponding to thicknesses of 0.4 μm, 1.0 μm, and 5.6 μm constituted of 20, 50, and 280 periods, respectively where each period comprises Si (10 nm)/$Si_{0.8}Ge_{0.2}$ (10 nm). We then probe the κ_L using standard measurement techniques[14,15] through placing electrodes using photolithography.

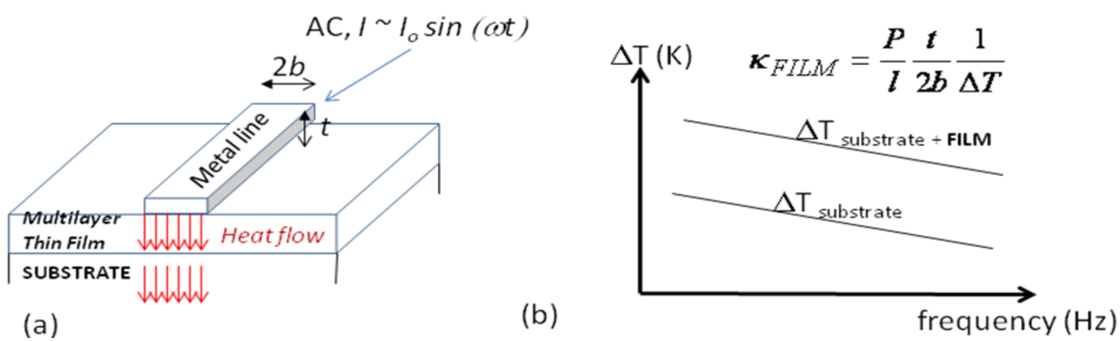
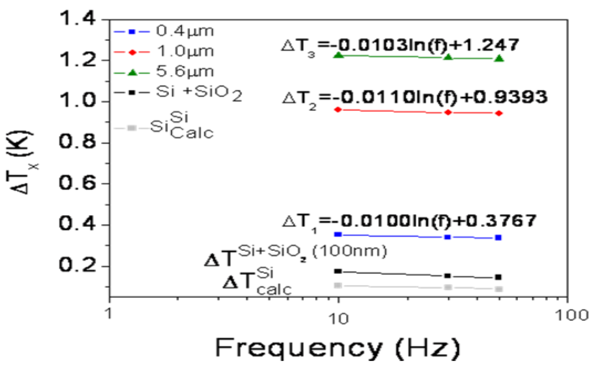


Figure 1 (a) One-dimensional heat flow is ensured for measuring cross-plane κ_L of $Si/Si_{0.8}Ge_{0.2}$ multilayered films, with $2b$ ~ 30 μm, t = 0.4 μm, 1.0 μm, and 5.6 μm, and a power density (P/l) corresponding to 10 W/m^2. **(b)** The total temperature drop, ΔT, across the multilayer/substrate system is the sum of the individual ΔT across the multilayer and substrate. The κ_L of $Si/Si_{0.8}Ge_{0.2}$ multilayered films ($\equiv \kappa_{FILM}$) can be estimated through a one-dimensional heat equation model.

One-dimensional heat flow is ensured through using 35 μm wide (=2*b*) heaters, which are large in comparison to the multilayer thickness – **Figure 1**. Under the assumption that the κ_L of the multilayers is small relative to the substrate $\kappa_{L,}$ the multilayer module would act as a thermal resistance, whereby the net temperature drop (ΔT) due to the heating is the algebraic sum of the temperature drops on the film (ΔT_{FILM}) and the substrate ($\Delta T_{SUBSTRATE}$). It should noted that this condition would be achieved when the heating current frequency is in the range where the thermal wave penetrates through to the substrate, e.g., at ~ 100 Hz, the TPD is ~ 180 μm for κ_L (~ 68 W/mK), C (624 J/kg·K), and ρ (2.67 g/cm^3) calculated for the Si//$Si_{0.8}Ge_{0.2}$ system. Such a method was initially used to estimate the κ_L ($\equiv \kappa_{FILM}$) of the multilayers.

EXPERIMENTAL RESULTS AND DISCUSSION

From measurements carried out on the various multilayers, we obtained values of the κ_{FILM}, as illustrated in Figure 2. However, a large uncertainty in the measurements was noted. Moreover, no systematic variation in κ_{FILM} was observed.

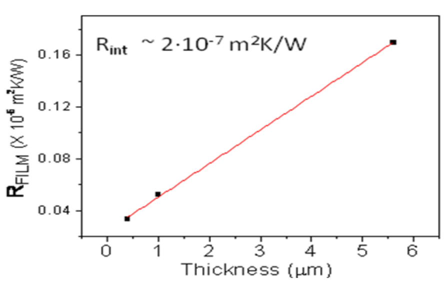


Determined Values

Film Thickness	κ_{FILM} (W/mK)
0.4 μm	~ 1.2 ?
1.0 μm	~ 1.9 ?
5.6 μm	3.3

Figure 2 The measured in-phase temperature fluctuation (ΔT_x) vs. frequency for $Si/Si_{0.8}Ge_{0.2}$ multilayers (left) and the determined values for κ_{FILM}.

We also corrected for the interfacial thermal resistance, R_{int}, where the total thermal resistance, using $R_{FILM}(=\frac{t}{\kappa_{meas}}) = R_{\text{int}} + \frac{t}{\kappa_{FILM}}$ and obtained the corrected values of κ_{FILM} through extrapolation to $t=0$, as represented in the table of **Figure 3.**

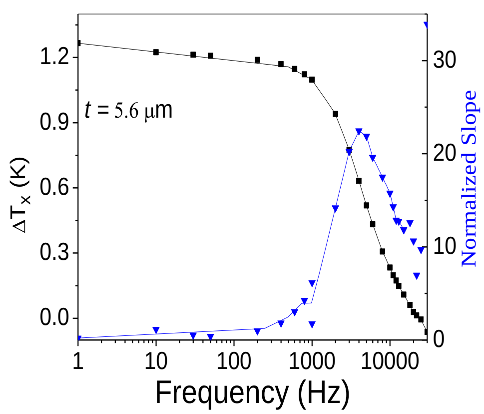


Film Thickness	κ_{FILM} (W/mK) *corrected*
0.4 μm	4.7
1.0 μm	3.6
5.6 μm	3.8

Figure 3 The interfacial thermal resistance, R_{int} was estimated to be ~ $2 \cdot 10^{-7}$ m^{2}K/W and yields corrected values of κ_{FILM} which illustrate that the thermal conductivity values may decrease with increasing multilayer thickness.

However, it was seen that even small variations (±10%) in R_{int} can yield κ_{FILM} values that are quite different. Such a variation can occur due to subtle differences in the ambient conditions. Considering the difficulties in using the slope offsets, as represented in Figures 1 and 2, we then decided to probe the thermal conductivity through an alternate method.

As previously mentioned the TPD can be used as a variable to gauge the extent to which the thermal wave penetrates the sample and we calculated that at frequencies around 10-30 KHz, the TPD would be adequate to sample the multilayer film and hence determine its thermal conductivity. Frequency dependent measurements on a 5.6 μm multilayer film then yield a graph as illustrated in Figure 4, which can be divided into distinct frequency regimes corresponding to whether the substrate is being sampled/measured (low frequency) or whether the multilayer is being sampled/measured (high frequency). It has been found[16] that the plotting of the normalized slope, i.e., $\dfrac{d(\Delta T)}{d(\ln\omega)} / \left|\dfrac{P_l}{\pi\kappa_n}\right|$ can give a better idea of the frequency at which a thermal conductivity change occurs. We use a corrected expression[16] for ΔT which considers the thermal capacitance and resistance of the heater, along with R_{int} to determine the values of κ_{FILM} which are listed in the table of Figure 4, and which have been found to be reproducible and reliable. It was seen that the κ_L determined from both the in-phase (ΔT_x) and out-of-phase (ΔT_y) temperature drops, through $\Delta T = \dfrac{2}{\alpha}\dfrac{V_{3\omega}}{V_w}$, where α is the temperature coefficient of resistance of the heater line, was comparable.



Film Thickness	$\kappa_{FILM}^{in\text{-}phase}$ W/mK	$\kappa_{FILM}^{out\text{-}phase}$ W/mK
0.4μm	~4.6 ± 0.2	~ 4.9 ± 0.2
1.0μm	4.3 ± 0.1	4.5 ± 0.1
5.6μm	3.5 ± 0.1	4.4 ± 0.1

Figure 4 The variation of ΔT_x vs, frequency can be used to determine the individual κ_L of the substrate (low frequency, high TPD, small slope) and the overlaid multilayer (high frequency, low TPD, larger slope). The table on the right yields the measured values of κ_L for the multilayer films of various thicknesses, determined from both the in-phase (ΔT_x) and out-of-phase (ΔT_y) values and indicates that the κ_L decreases with increasing multilayer thickness.

Similar results (not shown) were observed for the 0.4 μm and 1.0 μm multilayer samples. It would also be possible (work in progress) to probe the individual layers of the multilayer samples and obtain a clear signal of the evolution of the thermal conductivity. However, much larger frequencies, of the order of 10 MHz and above would be required and this places stringent demands on the electronic circuitry. For example, the third harmonic voltages produced from the power supply would be comparable in magnitude to the measured $V(3\omega)$.

Two aspects of the measured κ_L are to be noted: (1) A decrease of κ_L with increasing multilayer thickness, and (2) a drastic reduction below the bulk value. The increased number of interfaces in the larger thickness samples, which causes a greater degree of phonon scattering, is arguably a major contributor for the decreased κ_L. In this context, it is to be mentioned that a κ_L value of ~ 9.5 W/mK has been obtained for bulk $Si_{0.9}Ge_{0.1}$ and the obtained values in this work are indeed smaller. It is noted that the average composition of our $Si/Si_{0.8}Ge_{0.2}$ films has been approximated to be $Si_{0.9}Ge_{0.1}$, as also done by Huxtable *et al*[17] . Our obtained κ_L values also correspond favorably to other studies of multilayers/superlattices, as shown in Figure 5.

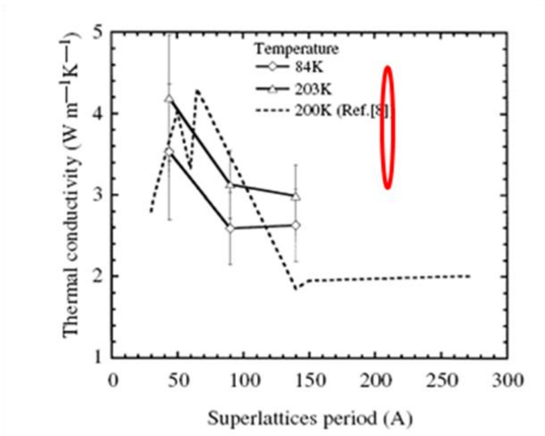


Figure 5 A comparison of the obtained thermal conductivity (κ_L) values (in red) with literature. The main graph was taken from the paper by Borca-Tasciuc *et al*[18], where Reference 8 refers to the paper by Lee *et al*[13]. The comparison is obtained with our samples where the total period is 20 nm, comprising Si (10 nm)/$Si_{0.8}Ge_{0.2}$ (10 nm).

The adoption of the frequency dependent extension of the 3ω method seems to yield values of κ_L that are more in accordance with previous measurements, while the method based on slope offsets has yielded much more variation. We have shown that subtle variations in the interfacial thermal resistance are probably responsible for the latter. The reduction of κ_L while being ascribed to the decreased mean free path, also needs to be analyzed with respect to the effects of frequency[19] and as a function of the crystallinity of the sputter deposited materials.

CONCLUSIONS

We have determined the values of cross-plane κ_L of sputter-deposited $Si/Si_{0.8}Ge_{0.2}$ films on Si substrates through (a) both the traditional slope-offset method, by considering the thin film as an added thermal resistance, and (b) through a variation of the heating current frequency. The uncertainties in the interfacial thermal resistances preclude accurate determination of κ_L in the former method. Using the frequency as an experimental variable has enabled more accurate and repeatable determination of κ_L. It has also been ascertained that the κ_L is reduced to below the bulk value of $Si_{0.9}Ge_{0.1}$ alloy which has been tentatively ascribed to a decreased mean free path due to a greater number of interfaces. The reduced κ_L when more interfaces are involved could possibly be due to an increasing number of interfacial defects.

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