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Non-diffusive thermal transport in GaAs at micron length scales

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We use a transient thermal grating technique in reflection geometry to measure the effective thermal diffusivity in GaAs as a function of heat transfer distance at three temperatures. Utilizing heterodyne detection, we isolate the "amplitude" grating contribution of the transient grating signal, which encodes the thermal transport dynamics. As the thermal grating period decreases, and thus the heat-transfer distance, we observe a reduction in the effective thermal diffusivity, indicating a departure from diffusive behavior. Non-diffusive behavior is observed at room temperature, as well as low temperature (180 K) and high temperature (425 K). At the shortest thermal grating period measured corresponding to a heat transfer distance of approximately 1 μm, the effective diffusivity drops to a value roughly 50% of the bulk thermal diffusivity. These measurements show the utility of the reflection transient thermal grating technique to measure thermal transport properties of opaque materials. © *2015 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4933285]

I. INTRODUCTION

The practical importance of understanding and optimizing heat transfer properties of microelectronics as well as materials for thermoelectric energy conversion has stimulated interest in the fundamentals of phonon-mediated thermal conductivity. Central to the rapidly growing field of micro- and nano-scale thermal transport[1–15] is the relationship between the phonon mean free path (MFP) and a characteristic length scale, be it the grain size in a bulk material or a dimension of a nanostructure. An important development of the past few years has been the realization that the simplistic view that the thermal conductivity of a single crystal material at a given temperature can be characterized by an average MFP has outlived its usefulness.

In dielectrics and semiconductors, all phonons across the Brillouin zone from the lowest frequency acoustic to the highest frequency optical phonons can participate in thermal transport. But due to considerations of phonon mode population, group velocity, heat capacity, MFP, and interactions between phonon modes, certain regions of the Brillouin zone will play a more significant role in energy transport. The interplay between phonon MFPs and a characteristic length scale, potentially from microns to nanometers, determines if the heat flux, at least for part of the phonon spectrum, is better described by diffusive or ballistic transport. When the MFPs are shorter than the characteristic length scale, the diffusion equation holds. When MFPs are much longer than the characteristic length scale, the diffusion equation predicts a heat flux exceeding the radiative, ballistic limit; the actual heat flux will be lower than the diffusion law prediction, resulting in a lower effective thermal conductivity. Thus, comprehension of how each phonon mode, with corresponding wavelength and MFP, contributes to the total thermal conductivity opens new avenues to understand and control transport properties in nanoscale materials and devices.

Useful in this regard is the so-called thermal conductivity accumulation:[17] the percentage of the total thermal conductivity that is attributed to phonons below a particular MFP or above a certain wavelength. The thermal conductivity accumulation has been determined via first principles calculations for a number of materials; in particular, some recent theoretical studies show that low frequency acoustic phonons with long MFPs play a much more important role in room temperature (RT) thermal conductivity of dielectrics and semiconductors than previously thought.[4–7] For example, it has been shown that phonons with MFP exceeding 1 μm contribute almost 40% to the RT thermal conductivity of Si, in stark contrast with the frequently cited average MFP of 43 nm.[18] Furthermore, MFPs of phonons contributing significantly to thermal transport have been shown to vary over orders of magnitude, highlighting the gross shortcoming of the "average MFP" paradigm. Consequently, the Fourier law based on the diffusion model of heat transport is expected to break down at significantly longer distances than the traditional MFP estimates would suggest.

Recently, we proposed the use of the transient thermal grating (TG) technique as a "clean" method for studying non-diffusive heat transport[12] that provides a well-controlled heat transfer distance and avoids complications related to transport across interfaces between different materials.[8–11,13,16] In the TG technique, crossed, short laser pulses set up a spatially periodic temperature profile in the sample, a "thermal grating," and the decay dynamics of the thermal grating are monitored via diffraction of the probe laser beam.[19,20] Moreover, the simplicity of the TG experiment allows us to analyze the non-diffusive transport with the Boltzmann transport equation (BTE).[21]

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In a two-fluid model,[21] we assume that high-frequency phonon modes with significant heat capacity transmit heat in the diffusive regime described by the conventional heat equation, whereas the BTE describes heat transport by low-frequency phonons and captures the transition from diffusive to ballistic regime. As the TG period is reduced, an increasing subset of longer MFP path phonons transmit heat ballistically. The longer MFP ballistic phonons transfer less heat than diffusion model predictions, thus leading to a lower effective thermal diffusivity (see further details in Section III). A *transmission* TG experiment on a thin Si membrane showed a significant deviation from the diffusion model at micron distances, in agreement with the theoretical calculations.[12] While the thin membrane transmission geometry has an advantage in that it ensures one-dimensional heat transport along the membrane, it limits the range of materials that can be investigated. Other techniques proposed to study non-diffusive heat transport at small distances use metal "heaters" deposited on a bulk sample and can be applied to a wide range of materials. However, experiments based on this approach present difficulties for a quantitative analysis.[22] It would be desirable to extend the TG method to study non-diffusive thermal transport in bulk opaque materials.

The TG technique in *reflection* mode has been used to measure thermal conductivity of opaque samples.[23] Recently, we developed a variation of the technique utilizing phase-controlled heterodyned detection to isolate the thermoreflectance signal from that produced by surface displacement.[20] In this work, we use this phase-controlled reflection TG technique to investigate thermal transport in bulk GaAs while systematically varying the heat transfer distance. As the TG length scale is decreased, a departure from diffusive transport is observed. The results are compared with predictions utilizing first-principles calculations of the thermal conductivity accumulation as a function of MFP.[7]

II. EXPERIMENTAL AND DATA ANALYSIS

The reflective transient grating experiment for measuring thermal diffusivity has been described in detail previously.[20] In the current measurements, two 515 nm excitation pulses are crossed with angle θ at the surface of the GaAs sample. A periodic intensity profile results due to optical interference with fringe spacing given by

$$L = \frac{2\pi}{q} = \frac{\lambda_e}{2\sin(\theta/2)}, \quad (1)$$

where λ_e is the excitation wavelength and q is the transient grating wave-vector magnitude. Above bandgap excitation leads to excited carriers that quickly transfer energy to the lattice, leaving a periodic temperature profile. The signal dynamics are measured in a heterodyne geometry by monitoring the intensity of a diffracted probe beam (532 nm) overlapped with a reference beam. Periodic surface displacement together with changes in the imaginary part of the complex reflection coefficient of the sample lead to a transient "phase" grating, whereas thermally induced changes in the real part of the complex reflection coefficient of the sample lead to a transient "amplitude" grating. By utilizing a reference sample to carefully select the heterodyne phase (relative optical phase between signal and reference beam), the transient amplitude grating can be isolated from the phase grating, allowing direct observation of the thermal transport dynamics.[20] As opposed to the case of a transmission TG experiment where diffusive thermal transport can be described by the one-dimensional diffusion equation (such as in the silicon membranes[12]), in a reflection TG measurement, transport takes place from peak to null in the periodic temperature profile parallel to the sample surface as well as into the depth of the material, thus necessitating a two-dimensional diffusion model. In the limit of surface heating, the temperature grating decay at the surface follows the form:[20,23]

$$T(z=0,x,t) = \frac{A}{\zeta\sqrt{\pi}}(\alpha_z t)^{-1/2}\cos(qx)\exp\left(-\alpha_x q^2 t\right), \quad (2)$$

where A is the amplitude, $1/\zeta$ is the optical penetration depth for an intensity decaying exponentially into the depth of the material, α_x and α_z are, respectively, the in- and cross-plane thermal diffusivities, and q is the TG wave-vector from Eq. (1). The $(\alpha_z t)^{-1/2}$ term accounts for diffusion into the depth of the material, and the $\exp(-\alpha_x q^2 t)$ term is the thermal grating decay. In Ref. 20, it was shown that after a waiting time of approximately $\Delta t = (4\alpha_z\zeta^2)^{-1}$, the surface heating approximation of Eq. (2) matches the full solution.

For 1-D transport, upon initially entering the non-diffusive transport regime, the grating decay should remain exponential with a lower "effective" thermal diffusivity reflecting the fact that some heat carrying phonons are traveling ballistically over the heat transfer distance from TG peak to null, which is roughly $L/2$ relative to the TG period.[21,24] To simplify things, we assume that this is the case for the in-plane transport in the 2-D model as well, and fitting the thermal decay to Eq. (2) will allow extraction of the effective in-plane thermal diffusivity for all TG periods.

Figure 1 shows the results of measurements performed on undoped GaAs. The heterodyne phase was controlled to record both amplitude and phase grating signals (see the inset of Fig. 1(a)) for TG periods ranging from 2.05 to 10 μm at 180 K, 295 K, and 425 K. Note that the phase grating signal includes temperature and surface displacement dynamics, as evidenced by the oscillating surface acoustic wave signal, which complicate the analysis; therefore here we only analyze the amplitude grating dynamics that encode the temperature decay.[20] Figure 1(a) shows representative amplitude grating decay traces recorded at room temperature along with fits to Eq. (2). To exclude initial times that can be influenced by the finite laser penetration depth as well as excited carrier dynamics, we fit all traces starting from 10 ns. We also use an isotropic version of Eq. (2), where the in- and cross-plane diffusivities are equivalent. We admit that this isotropic simplification neglects the fact that in- and cross-plane transport could be coupled in the non-equilibrium transport regime and would as well be influenced by different transport length scales, but we expect these factors to affect mainly the early signal times that are excluded from the fits.

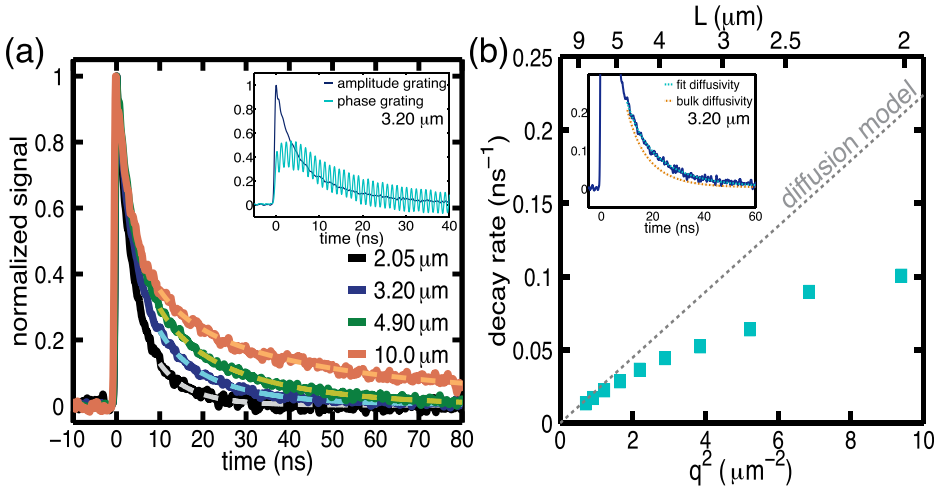

FIG. 1. (a) Thermal decay (amplitude grating) traces in GaAs at 295 K. The inset shows the amplitude and phase grating signals for the 3.20 μm TG period. (b) Comparison of the in-plane decay rate to q^2; the dashed line shows the diffusion model predicted behavior calculated using the literature GaAs diffusivity.[25] The inset shows the 3.20 μm TG period trace with the comparison between the fit and predicted behavior using the bulk diffusivity.

According to the diffusion model, the in-plane thermal decay rate should increase linearly with q^2. The dashed line in Fig. 1(b) shows the expected decay rate using the literature GaAs diffusivity. The inset of Fig. 1(b) shows a comparison of the data with dashed lines using the diffusivity extracted from the fit compared to the bulk diffusivity for 3.20 μm TG period. As seen in Fig. 1(b), as q^2 increases, the measured decay rate shows an increasingly larger departure from the diffusion model; as the length scale decreases, the transport is increasingly non-diffusive. The decay rate always increases with q; as the thermal transport length scale decreases, it will always take heat less time to traverse the distance. But for ballistic phonons, the traversal rate is linear with distance rather than quadratic as in the diffusion model.

In Figure 2, we plot the effective diffusivity as a function of TG period; again, we admit our simplification of using our measured effective diffusivity as representative of the 1-D bulk effective diffusivity. We see that at the largest grating periods measured, the experimentally determined effective thermal diffusivity approaches the literature reported values of 0.53 cm^2/s at 180 K, 0.25 cm^2/s at 295 K, and 0.16 cm^2/s at 425 K (respectively, the labeled blue, teal, and orange lines[25]). Note that values from Adachi[25] are within 4% of the often cited values of Holland[26] and Inyushkin *et al.*,[27] but the values from Adachi[25] covered our entire temperature range so no extrapolation was needed.

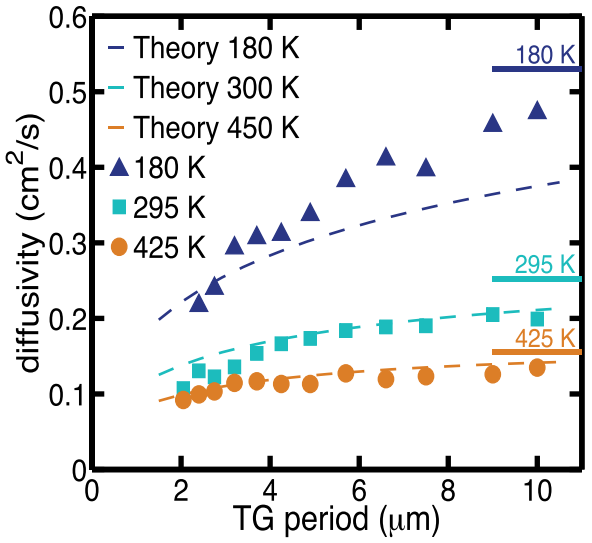

FIG. 2. The measured effective thermal diffusivity as a function of TG period. The dark blue triangles are for 180 K, the teal squares are for 295 K, and the orange circles are for 425 K. The lines on the right give the bulk GaAs diffusivity from the literature.[25] The dashed lines show the comparison between the measured effective diffusivities and theoretical predictions for 180 K, 300 K, and 450 K.[7]

For the recorded TG periods longer than 10 μm, reliable in-plane decay rates could unfortunately not be extracted from the data. As the in-plane decay gets longer and longer, the overall decay begins to follow more closely the $(\alpha_z t)^{-1/2}$ cross-plane decay, and sensitivity to the in-plane dynamics disappears.[20] But it is clear, that as we probe shorter and shorter length scales, the effective in-plane diffusivity clearly becomes lower and lower. As the temperature decreases, MFPs increase, therefore increasing the fraction of the ballistic transport, and lowering the effective diffusivity; at the smallest TG periods, we measure corresponding to a heat transfer distance of approximately 1 μm, the effective diffusivity ranges from approximately 60% of the bulk value at 425 K down to 40% of the bulk value at 180 K.

III. COMPARISON TO THEORY

The calculated thermal conductivity accumulation function is central to the comparison of experimental data with theoretical simulations; recent first principles calculations of the thermal conductivity accumulation in GaAs, calculated according to the methods of Luo *et al.*[7] for temperatures of 180 K, 300 K, and 450 K, make it possible to compare our measured GaAs values to the theoretical simulations. We use the calculated accumulation function together with the transient grating suppression function to generate an effective conductivity as a function of TG period,[21] and then scale this by the literature bulk diffusivity to compare with our measured data. The transient grating suppression function, determined using a two-fluid BTE model, modifies the integrated thermal conductivity by accounting for the fact that ballistic phonons (phonons with MFP on the order of the heat transfer length scale) contribute differently to the total thermal conductivity than predicted by the diffusion equation. We therefore calculate the effective thermal conductivity as a function of TG wavevector magnitude q and phonon MFP Λ using

$$k_{eff} = \int_0^\infty A(q\Lambda) k_\Lambda d\Lambda$$
$$A(q\Lambda) = \frac{3}{q^2\Lambda^2}\left(1 - \frac{arctan(q\Lambda)}{q\Lambda}\right), \quad (3)$$

where k_Λ is the differential thermal conductivity accumulation function from the simulations and $A(q\Lambda)$ is the TG suppression function.[21] It should be noted that Eq. (3) has been derived for a one-dimensional thermal grating in a bulk material; our conjecture is that this equation can still be used to describe the in-plane thermal transport in our more complicated 2D geometry. Efficient methods of solving BTE for multidimensional problems are currently being developed[28,29] and we hope that a more rigorous solution of the 2D thermal transport problem accurately representing our experiment will soon become available.

In Fig. 3, we plot the calculated effective thermal diffusivity as a function of TG period (the dashed lines with labeled temperature for the calculation), where the maximum thermal conductivity value is scaled by the literature thermal diffusivity value[25] for comparison with our recorded data also shown (triangles, squares, and circles for 180, 295, and 425 K, respectively). The simulated effective diffusivity values are also plotted in Fig. 2 to facilitate comparison to the recorded data.

Calculations involving silicon[4–6] show similar behavior as to that seen with GaAs,[7] with some contributions to the bulk thermal conductivity coming from phonons with MFP $> 1\ \mu$m. Here, we observe qualitative agreement with the predicted behavior for all temperatures measured. At longer TG periods, there exist larger discrepancies for the lowest temperature (180 K). This is perhaps due to the fact that the relaxation time approximation (RTA) used in the calculations becomes less accurate at lower temperatures: the RTA does not distinguish between normal and umklapp scattering, which makes it inadequate at low temperatures.[30] Overall, there is a relatively good agreement indicating the validity of the simulations. Currently, the simulations are the main methods that offer details on which regions of the Brillouin zone are of importance in heat transfer. In future efforts to customize thermal transport properties in practical materials, such details will be valuable.

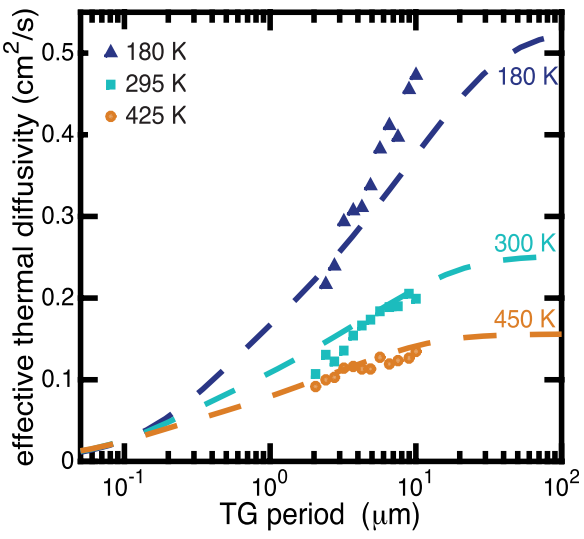

FIG. 3. Comparison of the calculated (dashed lines) and measured (points) effective diffusivities as a function of TG period for three temperatures.

IV. CONCLUSIONS

Utilizing the reflective transient thermal grating technique, we have investigated the departure from diffusive thermal transport in gallium arsenide at three temperatures. At TG periods below 10 μm, we see deviations from behavior predicted using the bulk thermal diffusivity. The measured effective in-plane diffusivity decreases as the TG period is reduced, indicating that the experimentally imposed heat transfer distance becomes shorter than the MFPs of some heat carrying phonons, with the lowest temperature data showing the greatest departure from the diffusive model. These measurements show an effective in-plane thermal diffusivity reduced to ~40%–60% of the bulk value at a heat transfer distance of $L/2 = \sim 1\ \mu$m, showing that at all measured temperatures, a large portion of the heat in GaAs is carried by phonons with relatively long MFP phonons, with good agreement to the theoretical calculations. With the theoretical calculations, we can determine a "median thermal conductivity MFP" Λ_m, such that phonons with mean free path $\Lambda > \Lambda_m$ contribute 50% to the bulk thermal conductivity. For GaAs, the theoretical calculations here give $\Lambda_m = 0.75\ \mu$m, 0.36 μm, and 0.23 μm for 180 K, 300 K, and 450 K, respectively.

We have previously shown the utility of measuring diffusive thermal transport in reflection TG experiments on thin films[20] and superlattices,[31] and here we show the capability to investigate the departure from diffusive transport in an opaque material where long MFP phonons contribute significantly to the thermal transport over micron distances. This opens avenues to deepen our understanding of heat transfer on micron length scales in opaque materials such as single crystal semiconductors, alloys, and heterostructures.

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