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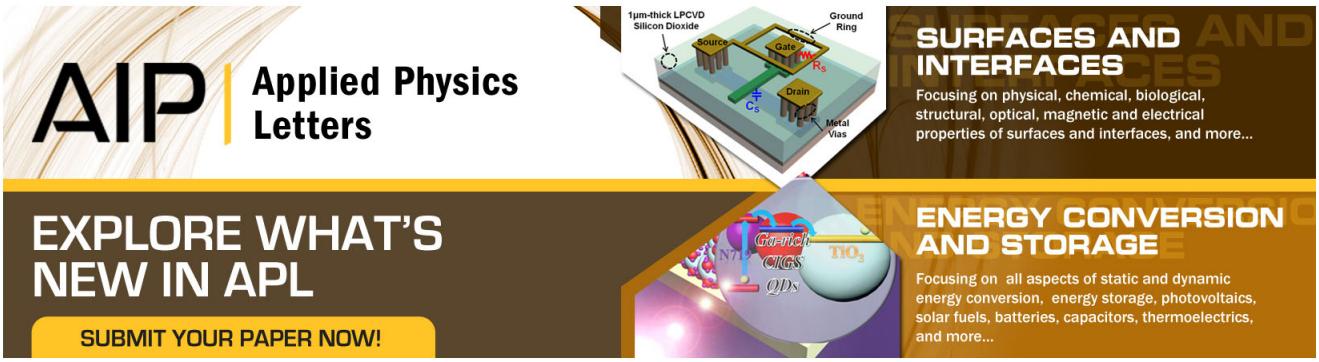

Contribution of optical phonons to thermal boundary conductance

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Thermal boundary conductance (TBC) is a performance determinant for many microsystems due to the numerous interfaces contained within their structure. To assess this transport, theoretical approaches often account for only the acoustic phonons as optical modes are assumed to contribute negligibly due to their low group velocities. To examine this approach, the diffuse mismatch model is reformulated to account for more realistic dispersions containing optical modes. Using this reformulation, it is found that optical phonons contribute to TBC by as much as 80% for a variety of material combinations in the limit of both inelastic and elastic scattering. © *2010 American Institute of Physics*. [doi:10.1063/1.3478844]

The continual miniaturization of microsystems and nanosystems has resulted in system architectures in which there are more interfaces spaced at an ever decreasing pitch. With this progression, the thermal performance of such systems is increasingly determined not by the bulk properties of the material but rather the nature of their intersection. Specifically, the efficiency by which energy may translate from one material to another, the thermal boundary conductance (TBC), has become a significant performance determinant in applications ranging from thermoelectrics to solid state memory.[1]

In parallel with this miniaturization, the materials utilized in these systems have increased in their complexity as well. For example, electronic, thermoelectric, and solid state lighting systems are now typically composed of layered compound semiconductors stacked one atop another. These compound semiconductors, in turn, have a crystal lattice in which at least two, and not uncommonly many more atoms, form the basis. Due to this multiatomic basis, optical phonons (OP) make up a large percentage of the available vibrational modes as there are only three acoustic branches in the phonon dispersion but $3p-3$ branches of the optical variety, where it is assumed that p is the number of atoms forming the basis. Thus, in wurtzite structures where four atoms comprise the basis, 75% of the phonon modes are those of the optical variety. In the same manner, complex lattices such as sapphire (Al_2O_3) or certain polytypes of SiC have greater than 90% of their possible modes made up of OPs.

Despite these large percentages, the contribution of OPs to thermal transport is most often ignored as it is assumed that either these modes have negligible group velocities and thus cannot transport heat or, due to their higher energies, the majority are not available for transport under normal conditions (i.e., they are "frozen out"). Recently, however, it has been shown that these assumptions can lose their validity when considering (1) materials having complex bases[2] and (2) assessing transport at non-cryogenic temperatures.[3,4] In such instances, it becomes necessary to account for the contribution of OPs to accurately assess the thermal transport.

Since most microsystems are composed of materials having complex bases, operate at temperatures above 300 K, and have a total thermal resistance determined significantly by interfacial transport, it becomes necessary to reconsider the influence of OPs in predictions of TBC. To date, this consideration has been constrained as most tools, including the oft employed diffuse mismatch model (DMM),[5] account for only the acoustic phonon modes. The DMM, for example, is formulated under the assumption of a Debye solid having, by definition, no OPs. In response, we have recently reformulated the DMM to account for a more realistic dispersion, thereby allowing for a consideration of the role of OPs in interfacial thermal transport.[6] Here, we utilize this extension to show that OPs can contribute significantly to TBC for a variety of material systems even if only elastic scattering events are assumed.

To extend beyond Debye solids in this work, the materials on either side of the boundary are described by an isotropic dispersion that is identical to the phonon dispersion along the crystallographic direction perpendicular to the interface. Computationally, this is realized by fitting previously determined dispersion relations using a fourth order polyno-

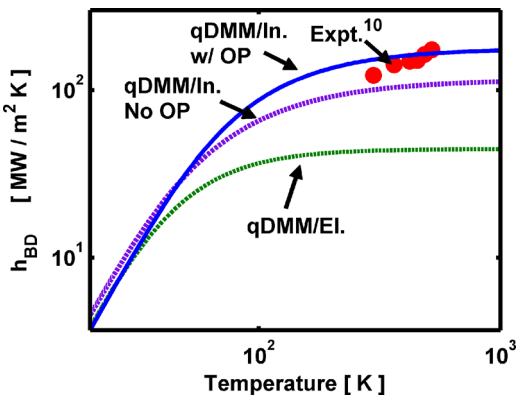

FIG. 1. (Color online) Predictions of TBC for a Pt/AlN interface compared to experiments of Hopkins *et al.* (Ref. 10). Due to the high acoustic mismatch of the materials, inelastic scattering significantly contributes to the transport. Accounting for inelastic scattering of only the acoustic phonons, however, results in a significant under prediction of the experimental results. As OPs contribute 28% of the TBC at room temperature, it is only when accounting for these modes that the qDMM is capable of reflecting the experiment.

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mial to acquire the relationship between frequency, ω, and wave vector, q, for each phonon polarization as has been reported by several groups.[7,8] In addition, such an approach also lends itself to easily quantifying the group velocity for each of the modes by capitalizing upon the fact that the group velocity, v, is defined as $v(q) \equiv d\omega/dq$. Knowing the full dispersion in a discrete fashion, the TBC for energy moving from side 1 to 2, $h_{\mathrm{BD}}^{1\rightarrow 2}$, can then be written entirely within wave vector space as shown in Eq. (1) below:

$$h_{\mathrm{BD}}^{1\rightarrow 2} = \frac{1}{8\pi^2}\sum_j \int_{q_{j,1}} \hbar\omega_{j,1}(q) q_{j,1}^2 |v_{j,1}(q_{j,1})| \frac{df_o}{dT} \zeta^{1\rightarrow 2} dq_{j,1}, \tag{1}$$

where the equation is summed over all phonon branches, j, $\hbar$ is the modified Planck constant, f_o is the equilibrium Bose–Einstein distribution function, and ζ is the transmission coefficient defining the ratio of energy capable of traversing the interface. To calculate this ratio, two different assumptions will be explored as to the nature of the scattering events. First, the fully elastic limit will be considered in which a phonon is permitted to forward scatter only to modes in the complementary material having an equivalent energy regardless of polarization. In this instance, the transmission coefficient takes the following form:

$$\zeta^{1\rightarrow 2}(\omega) = \frac{\sum_j [q_{j,2}(\omega)]^2}{\sum_j [q_{j,2}(\omega)]^2 + \sum_j [q_{j,1}(\omega)]^2}. \tag{2}$$

Additionally, the limit of combined inelastic and elastic scattering—termed here for simplicity the inelastic case—will also be examined where it is assumed that a phonon in material 1 is capable of forward scattering into *any* available mode within material 2. Under these constraints, the transmission ratio may be written as follows:[9]

$$\zeta^{1\rightarrow 2}(T) = \frac{\sum_j \int_{q_{j,2}} \hbar\omega_{j,2}(q) q_{j,2}^2 |v_{j,2}(q_{j,2})| f_o dq_{j,2}}{\sum_j \int_{q_{j,2}} \hbar\omega_{j,2}(q) q_{j,2}^2 |v_{j,2}(q_{j,2})| f_o dq_{j,2} + \sum_j \int_{q_{j,1}} \hbar\omega_{j,1}(q) q_{j,1}^2 |v_{j,1}(q_{j,1})| f_o dq_{j,1}}. \tag{3}$$

Implementing these transmission coefficients into Eq. (1) then allows for the prediction of TBC. Although these equations are equivalent to those utilized in the traditional DMM, this form is modified in the sense that it uses a computationally discrete dispersion that is more realistic, which in turn makes calculation more direct if the integration is performed over the wave vector, q, rather than the frequency. For this reason and to easily differentiate previous utilizations of the DMM from the current approach, this implementation will be termed the qDMM. Full details of its derivation and implementation are given in Ref. 6.

The qDMM was utilized to examine a series of material systems in which at least one of the materials on either side of the interface had a minimum of a two point basis and thus OPs. Figure 1 displays one of these calculations in which the TBC of a platinum/aluminum nitride (Pt/AlN) interface is calculated from 25 to 1000 K and compared to previously reported experimental measurements performed from 300–500 K.[10] While platinum contains no OPs, AlN takes on the wurtzite form and thus 9 of its 12 phonon branches are of the optical kind. Additionally, platinum and AlN are highly mismatched as the ratio of their Debye temperatures is 4.8 indicating that inelastic scattering can play a large role in the transport.[10] However, even when fully accounting for inelastic scattering, the qDMM (inelastic/no OP) significantly under-predicts the experiment when only acoustic phonons are considered. It is only when both the acoustic and OPs are considered in the inelastic limit that the qDMM (inelastic/OP) is capable of approaching the experimental results. In this case, the contribution of OPs, $h_{\mathrm{BD}}^{\mathrm{Optical}}/(h_{\mathrm{BD}}^{\mathrm{Acoustic}} + h_{\mathrm{BD}}^{\mathrm{Optical}})$, is 28% of the total TBC at room temperature indicating the necessity of accounting for these types of modes in assessments of interfacial transport. For the elastic case, since even the lowest energy optical modes in AlN have a higher energy than the highest energy acoustic phonon mode in platinum, OPs do not contribute under this limiting assumption.

To assess the contributions of OPs in the limit of elastic scattering, an interface of 6H–SiC on sapphire (Al_2O_3) was investigated subsequently. 6H–SiC has a 12 point basis whereas sapphire has a 10 point basis meaning that 60 out of the possible 66 polarizations of the combined system will be OP branches. Combined with the fact that the materials have a Debye ratio close to 1, the system is expected to be dominated by elastic scattering and thus provide insight into the consequence of OPs elastically scattering at the interface. To that end, shown in Fig. 2 is each material's phonon dispersion along the [0001] direction along with the elastic transmission ratio calculated from Eq. (2). For the entire system, the maximum acoustic frequency is $\sim 6\times 10^{13}$ rad/s and thus all transport above this energy level takes place solely due to OPs. Directly above this cut-off, the OPs in the 6H–SiC have a non-negligible velocity as evidenced by their significant slope while also easily being transmitted into the sapphire ($\zeta \approx 1$) due to the comparative dearth of modes within this spectral region as evidenced by the resulting high transmission coefficient. Such a high transmission coefficient within this spectral region in turn, results in a significant OP contribution to TBC over nearly the entire temperature range. For example, at 300 K, the OP contribution is 77% and 74% for the elastic and inelastic cases, respectively.[11] Most simply, the great majority of the interfacial conductance within this system arises from the optical, rather than the acoustic, phonon modes.

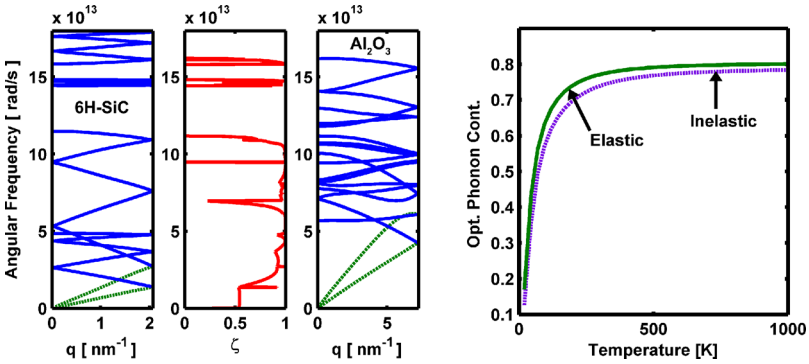

FIG. 2. (Color online) Dispersion curves of 6H–SiC and sapphire in the [0001] direction compared to the frequency dependent transmission coefficient. A portion of the OP modes (solid) in the 6H–SiC have a substantial group velocity and easily scatter into the sapphire due to the large number of modes in the region between 6×10^{13} and 12×10^{13} rad/s. As a consequence, the OPs contribute significantly (right) to the system's TBC for almost all temperatures. The TBC at 300 K for this system was calculated to be 46 and 39 MW/m^2K for the inelastic and elastic cases, respectively. The dispersion curves were adapted from Refs. 12 and 13.

OPs may also contribute significantly in a variety of other systems even when the number of OP branches does not excessively outnumber that of the acoustic variety. Figure 3 exhibits this fact as the OP contribution at 300 K is displayed for systems containing a total number of optical branches ranging from 3 to 60. In most instances, the room temperature contribution of the optical modes is between 20% and 45% of the entire TBC. As a general rule, this contribution increases with the number of optical branches for both the consideration of elastic and inelastic scattering. Perhaps counter to intuition, however, in many instances the OP contribution is seen to be greater in the limit of elastic, rather than inelastic, scattering. This can be attributed to the fact that for inelastic scattering the more thermally efficient acoustic modes are more capable of interacting with one another and thus marginally mitigate the relative effect of the OPs. Regardless, the optical modes play a significant role for a host of systems and thus may oftentimes be considered a key component of the system's thermal response.

To summarize, a reformulation of the commonly employed DMM that is capable of utilizing more realistic lattice dispersions has been utilized to examine the contribution of OPs to TBC. This contribution was found to be significant for a variety of systems under both the assumptions of inelastic and elastic scattering. As more complex materials are employed in microsystems, consideration of the OPs to interfacial resistance should be considered in a full quantification of a system's thermal response.

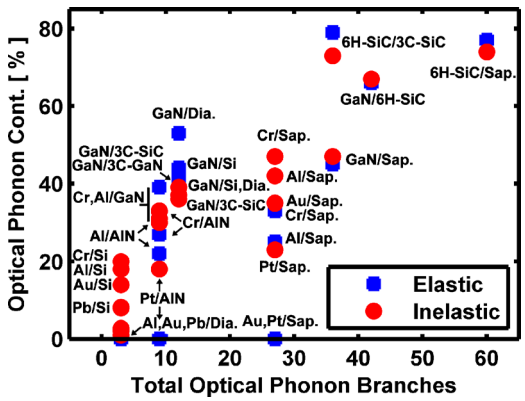

FIG. 3. (Color online) Contribution of OPs to a system's TBC as a function the total number of OP branches taking part. In general, more OP branches lead to a greater contribution. For nearly all material combinations, the OPs contribute between 20%–40% of the total TBC.

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