Thermal Conductivity **23**, 172-182 (1996).

Preprint copy, article presented in:

Proceedings of the Twenty-Third International Thermal Conductivity Conference,
K. E. Wilkes, R. B. Dinwiddie, R. S. Graves, eds. (Technomics, Lancaster, PA).

PHONON KNUDSEN FLOW IN GaAs/AlAs SUPERLATTICES

Per Hyldgaard and Gerald D. Mahan

ABSTRACT

We explain the significant reduction in the in-plane thermal conductivity of GaAs/AlAs superlattices as observed by T. Yao, *Appl. Phys. Lett.* **51**, 1798 (1987.) We estimate that the average bulk-phonon mean free path $l_{mfp} =$ 115 nm is significantly longer than the superlattice layer thickness $d =$ 5–50 nm. The GaAs/AlAs interfaces are generally smooth but does contain a finite density of both microroughness and monolayer island defects which cause an enhanced elastic and anharmonic phonon scattering. We treat this interface scattering as diffusive, i.e., providing a complete phonon thermalization. The resulting phonon Knudsen flow within the individual superlattice layers can account for the observed reduction of the in-plane superlattice thermal conductivity. We also consider the effects of a finite probability for reflection at the interfaces.

INTRODUCTION

Fast phonon transport in semiconductor heterostructures may allow an improved cooling of densely packed devices such as for example phase-array semiconductor lasers.[1] On the other hand, a significant reduction of the thermal transport may make semiconductor heterostructures attractive possibilities for thermoelectric devices. Either way, an increased understanding of the phonon transport in semiconductor heterostructures is important.

For the in-plane heterostructure phonon transport, T. Yao[2] and G. Chen[3] has observed a significant reduction in equal-layer-thickness GaAs/AlAs superlattices compared with the average, $\kappa_{avg} = 67$ W/Km, bulk-GaAs and bulk-AlAs conductivities. For the smallest layer thickness, $d = 5$ nm investigated

Per Hyldgaard and Gerald D. Mahan, Department of Physics and Astronomy, 200 S. College, University of Tennessee, Knoxville, TN 37996; *and* Solid State Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831.

in Ref. 2, the superlattice thermal conductivity is reduced down almost to the value of an actual $Al_{0.5}Ga_{0.5}As$ alloy.

We emphasize, however, that the observed reduction is not due simply to alloy scattering. The molecular-beam-epitaxial grown GaAs/AlAs superlattices are characterized by relatively smooth interfaces and the interdiffusion is limited.[4, 5, 6] However, the interface does contain a finite density of (monolayer) island defects whose in-plane extension can range from below to beyond the exciton Bohr radius,[6] ~ 10 nm. At the interface where an GaAs layer is grown on top of an AlAs layer there is also some atomic-scale microroughness.[6]

In this paper we demonstrate that the interface scattering produced by the island defects and microroughness can account for the observed reduction of the superlattice thermal conductivity. Specifically we describe the GaAs/AlAs interfaces and explain how a monolayer island defect can enhance the elastic and anharmonic phonon interface scattering. The average bulk-phonon mean free path $l_{mfp} = 115$ nm is significantly longer than the superlattice layer thicknesses $d = 5$–50 nm investigated in Ref. 2. Hence, the interface scattering is important.

Assuming that the interface scattering is diffusive, i.e., causes a complete phonon thermalization, we determine the Knudsen flow within the individual superlattice layers adapting a study by K. Fuchs.[7] We demonstrate in particular that the interface scattering can account for the observed reduction of the in-plane superlattice thermal conductivity. Finally, noting that areas without monolayer steps or islands can extend for more than ~ 100 nm at the interface where an AlAs layer is grown on a GaAs layer,[6] we also consider the effect on the thermal conductivity of a finite interface-reflection probability.

THE MEASURED SUPERLATTICE THERMAL CONDUCTIVITY.

In Ref. 2, T. Yao reports the measurements of the thermal transport in a set of superlattices formed by equally thick layers of GaAs and AlAs as illustrated in the top panel of Fig. 1. The thickness d of these individual layers ranges from $d = 5$ nm and to $d = 50$ nm. As also illustrated in the top panel of Fig. 1, the temperature gradient is applied in the plane of these superlattice layers. We may assume[2, 8] the same temperature in all layers of the superlattices which has a total thickness ~ 10 μm.[2] Hence, the resulting heat transport is given directly by in-plane superlattice thermal conductivity.

The set of diamonds in the bottom panel of Fig. 1 shows this in-plane thermal conductivity as a function of the superlattice layer thickness d. For comparison the dashed line in the bottom panel of Fig. 1 shows the average value,[2] $\kappa_{avg} = 67$ W/Km, of the bulk-GaAs and bulk-AlAs thermal conductivities. This average conductivity would provide an appropriate description if the GaAs and AlAs layers contributed independently to the thermal transport and if the phonon mean free path was short enough that one simply had bulk thermal conductance in parallel. It is clear that this simple description is invalid even for a layer thickness of $d = 50$ nm.

The superlattice thermal conductivity is, for layer thicknesses $d \sim 10$ nm, reduced almost to the value,[2] $\kappa_{AlGaAs} = 11$ W/Km — indicated by the dotted line in the bottom panel — of an actual $Al_{0.5}Ga_{0.5}As$ alloy. We emphasize, however, that the observed reduction in the superlattice thermal conductivity

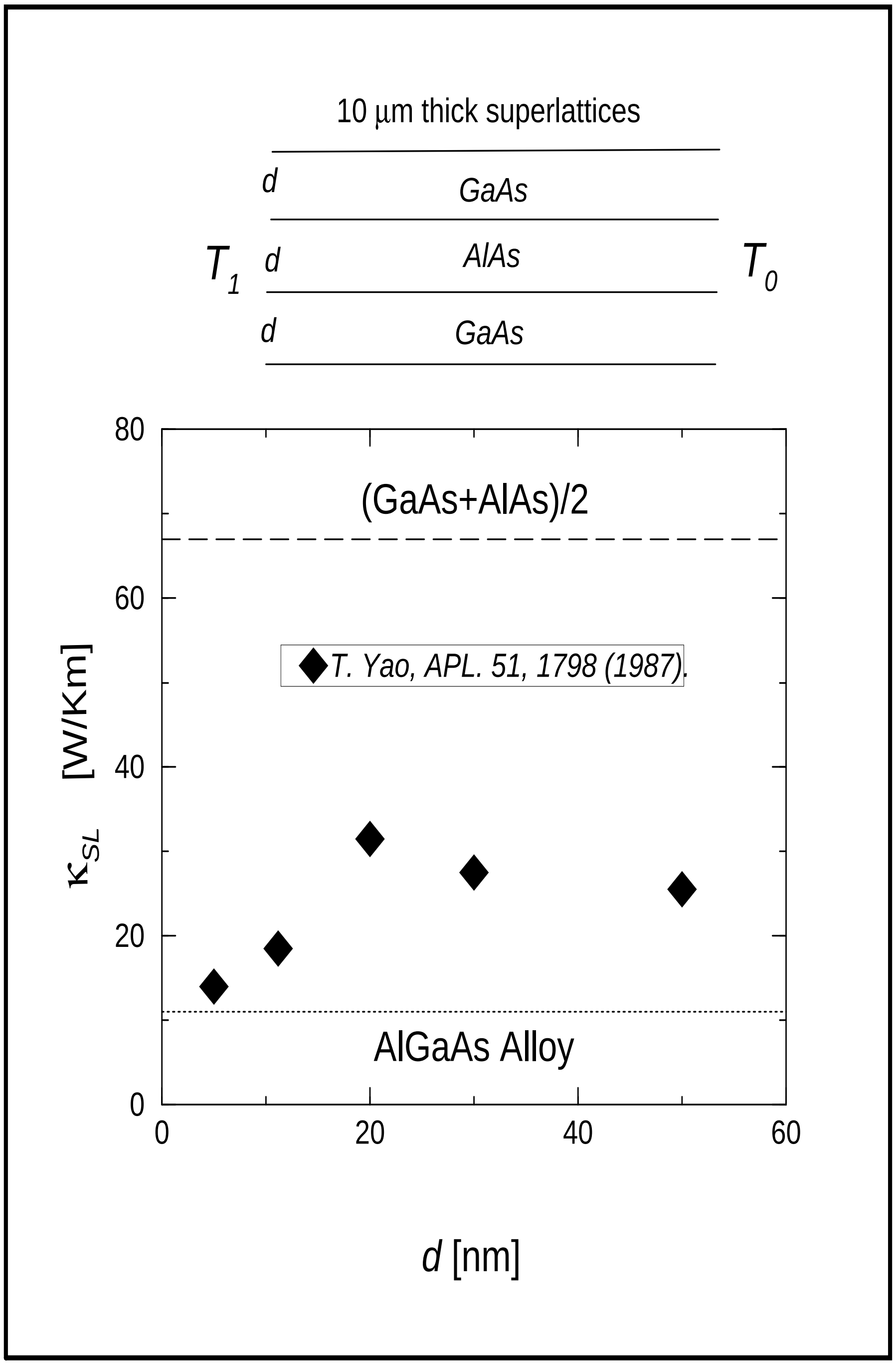

FIGURE 1. *Top* panel shows schematics of experimental determination[2] of in-plane thermal conductivity in 10 μm thick superlattices with GaAs and AlAs layer thicknesses d = 5–50 nm. *Bottom* panel compares measured superlattice thermal conductivities (set of diamonds) with the average bulk-GaAs and bulk-AlAs thermal conductivities (the dashed curve) and with the thermal conductivity of an actual $Al_{0.5}Ga_{0.5}As$ alloy (the dotted curve.)

can not be explained simply as resulting from an increased bulk alloy scattering. Specifically, the observed factor-of-three reduction of the thermal conductivity would — if due to alloy scattering — correspond[9] to a 10–20% bulk concentration of GaAs (AlAs) in the AlAs (GaAs) layers. Such a massive interdiffusion is ruled out by high-resolution transmission-electron-microscopy studies as reported for example in Refs. 5,6.

THE PHONON-INTERFACE SCATTERING

While there is no massive GaAs–AlAs interdiffusion, some disorder does arise at the heterostructure interfaces. As is evident from the high-resolution electron-transmission-microscopy studies in Refs. 5,6 this disorder is confined to within a few monolayers of the interface.

When an AlAs layer is grown on an GaAs layer it is possible to achieve a very smooth interface. There is a finite density of monolayer-island defects[4] but these can have an in-plane extension significantly larger[6] than the exciton Bohr radius $a_B \sim 10$ nm. The top panel of Fig. 2 shows the schematics of such an island defect with a monolayer-region of GaAs protruding into the AlAs layer.

In contrast, the interface where a GaAs layer is grown on top of a AlAs layer is significantly more disordered with so-called microroughness occurring at the atomic scale.[4,5,6] However, with the use of growth interruption it is possible to achieve *additional* longer-range (monolayer) fluctuations with an in-plane extension comparable to the exciton Bohr radius.[6] The GaAs-AlAs interfaces is thus characterized by monolayer defects on several length scales and the interface defects can affect all transport-carrying phonons.

The bottom panel of Fig. 2 illustrates how an interface defect can enhance both the elastic and the anharmonic phonon scattering. Specifically, as shown at the upper interface by the pair of solid arrows, an interface defect breaks the in-plane translational invariance and enhances the elastic scattering. Similarly, since the interface defect in particular softens the stringent momentum-conservation rule limiting the anharmonic phonon scattering,[10] monolayer defects will provide an enhanced inelastic scattering. Such an anharmonic scattering event is illustrated at the lower interface with an incoming energetic phonon (longer arrow) decaying to the pair of low-energy phonons (pair of dashed arrows.)

THE BULK-PHONON MEAN FREE PATH

The interface scattering described above is important because we estimate below a very long average bulk-phonon mean free path, $l_{mfp} \approx 115$ nm. The phonon transport within the heterostructure layers is thus characterized by a finite Knudsen number,[11] $(l_{mfp}/d) \geq 2$.

We assume for simplicity an isotropic phonon dispersion with a maximum wave vector, $q_{max} \approx 0.11$ nm^{-1}, given by

$$\frac{4\pi}{3} q_{max}^3 = 4(2\pi)^3/a^3, \tag{1}$$

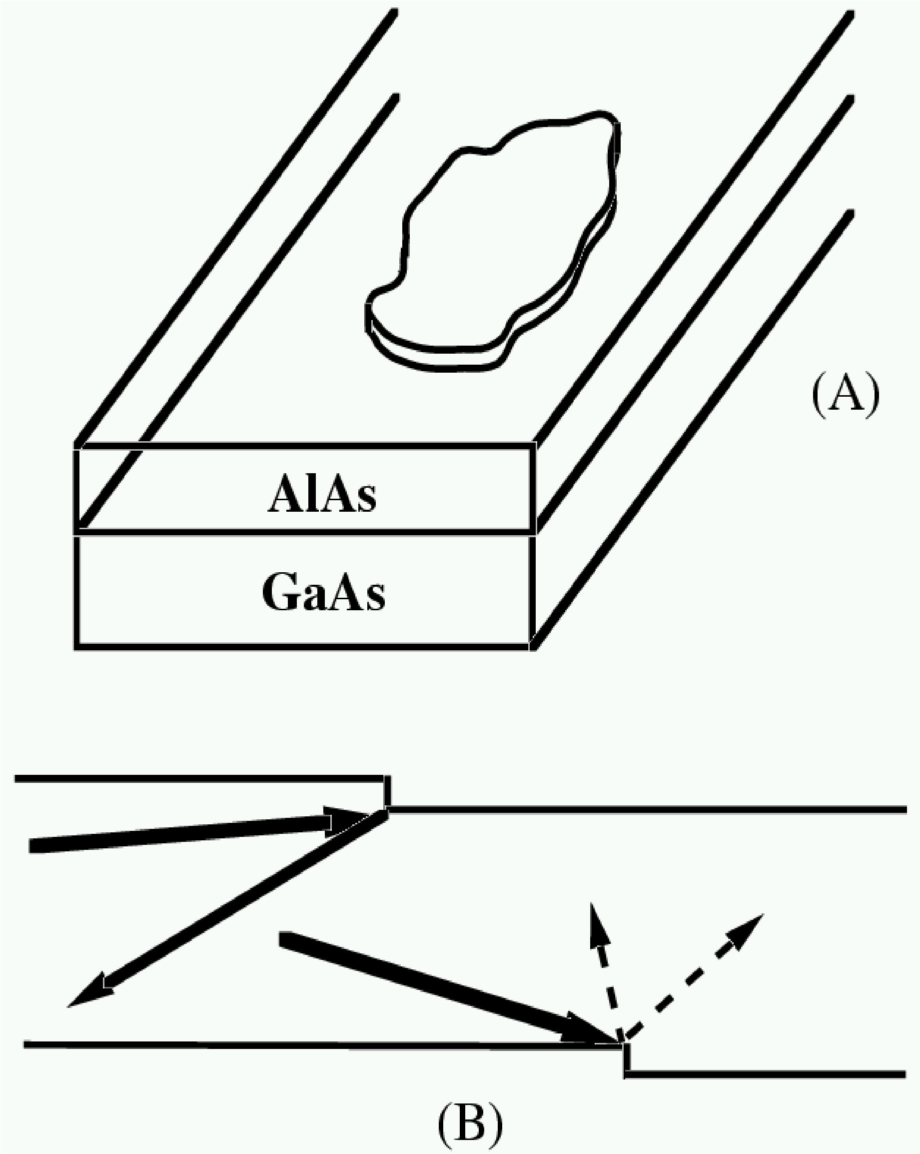

FIGURE 2. *Top* panel shows schematics of the interface where an AlAs layer is grown on a GaAs layer. Although this interface is smooth there is still as indicated a finite density of monolayer island defects. The *bottom* panel illustrates how such interface defects enhances both the elastic and anharmonic or inelastic electron scattering as the defect breaks the in-plane translational invariance.

where $a = 0.565$ nm denotes the GaAs lattice constant. For the sound velocities of the transverse and longitudinal modes we use the values,[12] $c_t = 3340$ m/s and $c_l = 4730$ m/s.

We also observe that both the transverse and longitudinal modes are relatively soft and is at the zone-boundary (i.e., $q \approx q_{max}$) described by energies $\Theta_t = 100$ K and $\Theta_l = 270$ K.[13] The low value, $\Theta_t = 100$ K, reflects a significant nonlinear dispersion for the transverse modes.[13] There is also a smaller nonlinearity in the longitudinal-mode dispersion.

The nonlinear dispersion reduces the group velocity, $v_g^{t,l} \equiv (\partial\omega_{t,l}/\partial q)$, of both the transverse and longitudinal phonon modes near the zone-boundary and causes a smaller contribution,

$$\kappa_{i=t,l} = \tfrac{1}{3} k_B T^2 \int_0^{q_{max}} q^2 dq \frac{e^{\hbar\omega_i(q)/k_B T}}{(e^{\hbar\omega_i(q)/k_B T} - 1)^2} \times v_g^i(q)[v_g^i(q)\tau_i], \tag{2}$$

to the bulk thermal conductivity. This transverse- and longitudinal-phonon contribution, Eq. (2), is derived within a phonon-relaxation-time approximation (given by τ_i.) For simplicity we assume a cubic relation,

$$q = \omega(1 + \alpha_i\omega^2)/c_i \tag{3}$$

between the phonon momentum and frequency within mode $i = t, l$. We determine the value of $\alpha_{i=t,l}$ by the condition, $\hbar\omega_i(q_{max}) = k_B\Theta_i$.

The contribution from phonon mode (i, q) to the bulk thermal conductivity, Eq. (2), is proportional to the mean free path, $[v_g^i(q)\tau_i]$. Assuming for simplicity a mode-independent and constant bulk-phonon mean free path, l_{mfp}, we find

$$\kappa_{GaAs} = l_{mfp} \times \frac{k_B^4 T^3}{6\pi^2\hbar^3}(2\frac{R_t}{c_t^2} + \frac{R_l}{c_l^2}) \tag{4}$$

where we have introduced the dimensional integrals $(i = t, l)$

$$R_i(T) = \int_0^{\Theta_i/T} dx \frac{x^4 e^x}{(e^x - 1)^2}(1 + \alpha_i(k_B T x/\hbar)^2)^2. \tag{5}$$

Inserting the *measured* value, $\kappa_{GaAs} = 45$ W/Km, of the bulk-GaAs thermal conductivity in Eq. (4) we obtain an estimate for the averaged bulk-phonon mean-free path, $l_{mfp} \approx 115$ nm. We predict in particular a significant value of the Knudsen number, $l_{mfp}/d \geq 2$, for the phonon transport within the semiconductor layers.

THE INTERFACE-LIMITED TRANSPORT

The *top* panel of Fig. 3 illustrates how the interface scattering significantly affects the in-plane thermal transport when the average bulk-phonon mean free path l_{mfp} *exceeds* the superlattice layer thickness d. The phonon scattering off the interface defects then becomes the dominant source of scattering and causes a reduction in the in-plane thermal conductivity.

The *bottom* panel of Fig. 3 shows as a function of the layer thickness d the estimated resulting superlattice thermal conductivity reduction. The three curves correspond to a set of different assumptions for this scattering as described below.

The simplest possible treatment of interface-scattering results by replacing the relaxation time τ in Eq. (2) by the layer-traversal time, $\tau_B = d/v_g$, for a phonon moving at right angle to the interfaces. The resulting estimate for the superlattice thermal conductivity is

$$\kappa_{SL}^{B} \equiv (d/l_{mfp})\kappa_{avg}, \tag{6}$$

expressed relative to the average bulk thermal conductivity, $\kappa_{avg} = 67$ W/Km of GaAs and AlAs. This simple estimate is shown by the dotted curve in the bottom panel of Fig. 3 and is smaller than the measured thermal conductivities (set of diamonds.)

The *solid* curve in Fig. 3 shows the superlattice-thermal conductivity obtained in the phonon-Knudsen-flow[7,11] estimate described further below. We assume in this estimate again a constant bulk-phonon mean free path[14] and that the interface defects causes a diffusive phonon scattering, i.e., causes a complete thermalization of the phonon distribution.

We denote by $\hat{z}$ the direction perpendicular to the superlattice layers (the growth direction) and assume a thermal gradient in direction $\hat{x}$ which induces a change in the thermal distribution,

$$N_{\vec{q}}(z) \equiv N^0(q) + g_{\vec{q}}(z). \tag{7}$$

Because of the interface scattering (at $z = \pm d/2$) this induced distribution, $g_{\vec{q}}(z)$, depends as indicated on the coordinate z. Assuming a constant bulk-phonon mean free path this z-variation is described by the linearized Boltzmann equation

$$\begin{aligned} \Big\{ l_{mfp}(q_z/q)(\partial/\partial z) + 1 \Big\} g_{\vec{q}}(z) &= (-\partial T/\partial x) Q(q_x/q) \\ Q(q_x/q) &= l_{mfp}(q_x/q)(\partial N^0(q)/\partial T). \end{aligned} \tag{8}$$

We solve this linearized Boltzmann equation Eq. (8) subject to the boundary conditions mandated by the assumed diffusive interface scattering. Specifically, because phonons *leaving* the interface at $z = d/2$ (and thus traveling with $q_z < 0$) are assumed to be in equilibrium with the interface we must have

$$g_{q_z<0}(z = d/2) \equiv 0. \tag{9}$$

A similar aguement yields the boundary condition, $g_{q_z>0}(z = -d/2) \equiv 0$, at the upper interface, $z = d/2$.

As a function of the inverse Knudsen number $\eta \equiv d/l_{mfp}$ we then obtain the relative reduction of the Knudsen-flow superlattice thermal conductivity,

$$\frac{\kappa_{SL}}{\kappa_{avg}}(\eta \equiv d/l_{mfp}) = 1 - \frac{3}{8\eta}\left[1 + 4D(\eta)\right] \tag{10}$$

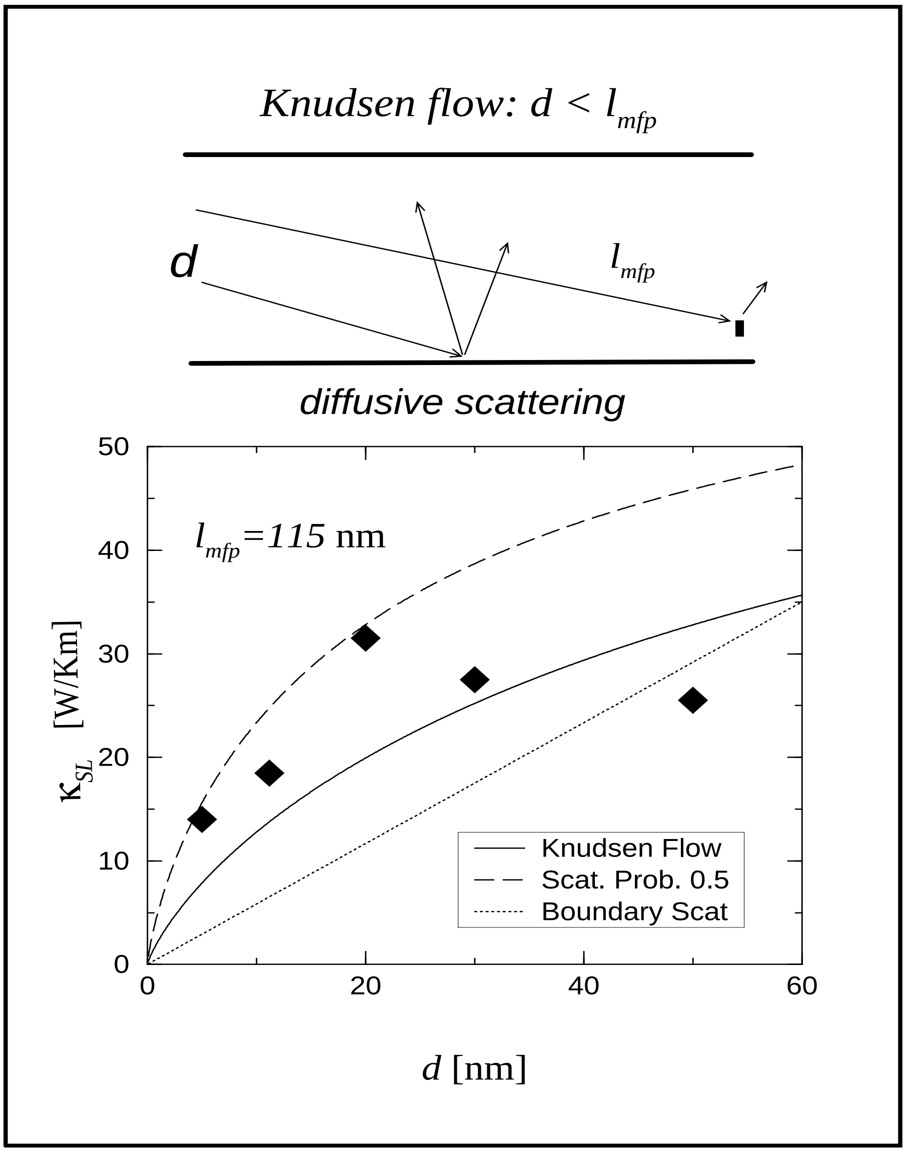

FIGURE 3. *Top* panel illustrates how interface scattering restricts the in-plane thermal conductivity because the layer thickness d is shorter than the average bulk-phonon mean free path l_{mfp}. *Bottom* panel shows estimates for resulting interface-limited in-plane thermal conductivity. Specifically, the dotted curve shows the boundary-scattering estimate where the layer thickness d simply replaces the average mean free path l_{mfp} in the expression for the bulk thermal conductivity. The solid curve shows the Knudsen-flow estimate where we assume the phonon-interface scattering is diffusive, i.e., provides a complete thermalization. Finally, the dashed curve shows the estimate assuming a finite interface-reflection probability $p = 0.5$.

where

$$D(\eta) \equiv \left(-\frac{\eta^3}{24} + \frac{\eta^2}{24} + \frac{5\eta}{12} - \frac{1}{4}\right) e^{-\eta} + \left(\frac{\eta^4}{24} - \frac{\eta^2}{2}\right) \int_{\eta}^{\infty} d\xi \frac{e^{-\xi}}{\xi}. \qquad (11)$$

Equation (10) is evaluated with $\kappa_{avg} = 67$ W/Km for the average bulk-GaAs and bulk-AlAs thermal conductivities which yields the phonon Knudsen flow shown by the solid curve in Fig. 3.

Compared with the measured superlattice thermal conductivities (set of diamonds) we find that both the simple boundary-scattering evaluation, Eq. (6) (dotted curve,) and the phonon-Knudsen-flow calculation, Eq. (10) (solid curve,) *overestimates* the observed conductivity reduction from the average bulk conductivity. We conclude that the phonon-interface scattering *can* account for the superlattice conductivity.

A PARTIAL INTERFACE-REFLECTION

It is no surprise that the Knudsen-flow calculation overestimates the in-plane superlattice thermal conductivity since we have assumed a complete phonon thermalization at both interfaces. At the interface where a AlAs layer is grown on a GaAs layer, however, it is possible to both avoid the microroughness and achieve monolayer islands with a very large an in-plane extension > 100 nm.[6] It is thus likely that the majority of the incoming phonons are simply reflected rather than experiencing an elastic or anharmonic scattering event at this interface. We conclude this paper by a simple estimate of the effect of a finite interface-reflection probability.

In one case we may assume that all phonons approaching the GaAs-to-AlAs interface are simply reflected while all phonons at the AlAs-to-GaAs interface experience a complete thermalization. We can solve the Boltzmann equation in the same manner as for the phonon Knudsen flow but now subject to a reflection boundary condition at the upper ($z = d/2$) interface, i.e., using

$$g_{q_z<0}(z = d/2) \equiv g_{q_z>0}(z = d/2) \qquad (12)$$

rather than the condition given in Eq. (9).

A related problem arises with the assumption of an *equal* finite reflection probability, $p = 0.5$, at both interfaces. In this estimate we may simply adapt the calculation provided in Ref. 7 and obtain for the in-plane superlattice thermal conductivity shown by the dashed curve in Fig. 3. We note that this estimate is larger than the measured superlattice thermal conductivity.

CONCLUSION

We have theoretically investigated the in-plane thermal conductivity in GaAs-AlAs superlattices with layer thicknesses d smaller than the estimated average bulk-phonon mean free path, $l_{mfp} \approx 115$ nm. We have described how the defects at the superlattice interfaces enhances both the elastic and anharmonic phonon scattering as they break the in-plane translational invariance. Furthermore, we have demonstrated how this interface scattering can account for the significant reduction of the in-plane thermal conductivity observed by T. Yao[2] in GaAs/AlAs superlattice with layer thicknesses ranging from $d = 5$ nm and to $d = 50$ nm. Finally, we have discusses the effect of a finite reflection probability at the interfaces.

ACKNOWLEDGEMENTS

The authors appreciate useful discussions with M. Huberman and G. Chen. This work was supported by the University of Tennessee and the Division of Material Science, U. S. Department of Energy under Contract No. DE-AC05-84OR21400 with Lockheed Martin Energy Systems.

REFERENCES

1. H.-J. Yoo, A. Scherer, J. P. Harbison, L. T. Florez, E. G. Paek, B. P. Van der Gaag, J. R. Hayes, A. von Lehmen, E. Kapon, Y.-S. Kwon, *Appl. Phys. Lett.* **56**, 1198 (1990); P. L. Gourley, M. E. Warren, G. R. Hadley, G. A. Vawter, T. M. Brennan, and B. E. Hammons, *Appl. Phys. Lett.* **58**, 890 (1991).
2. T. Yao, *Appl. Phys. Lett.* **51**, 1798 (1987).
3. G. Chen, "Size and Interface effects on thermal conductivity of superlattices and periodic thin-film structures," preprint.
4. See for example D. Bimberg, D. Mars, J. N. Miller, R. Bauer, and D. Oertel, J. Vac. Sci. Technol. B **4**, 1014 (1986).
5. A. Ourmazd, D. W. Taylor, J. Cunningham, and C. W. Tu, *Phys. Rev. Lett.* **62**, 933 (1989).
6. D. Bimberg, F. Heinrichsdorff, R. K. Bauer, D. Gerthsen, D. Stenkamp, D. E. Mars, and J. N. Miller, J. Vac. Sci. Technol. B **10**, 1793 (1992).
7. K. Fuchs, *Proc. Cambridge Philos. Soc.* **34**, 100 (1938); see also H. B. G. Casimir, *Physica* **V**, 495 (1938).
8. I. Hatta, Y. Sasuga, R. Kato, and A. Maesono, *Rev. Sci. Instrum.* **56**, 1643 (1985).
9. See Fig. 3 in M. A. Afromowitz, *J. Appl. Phys.* **44**, 1292 (1973) showing calculated $Al_xGa_{1-x}As$-alloy thermal conductivity as a function of Aluminum concentration x.
10. J. M. Ziman, *Electrons and Phonons* (Oxford University Press, Oxford, 1979,) pp. 128–145.

11. Henrik Smith and H. Højgaard Jensen, *Transport Phenomena* (Clarendon press, Oxford, 1989,) pp. 246–257.

12. H. H. Landolt and R. Börstien, *Numerical Data and Functional Relationships in Science and Technology,* New Series, **17e**, pp. 218–258.

13. See calculated GaAs-phonon dispersion in H. H. Landolt and R. Börstien, *Numerical Data and Functional Relationships in Science and Technology,* New Series, **17e**, p. 528.

14. It is of course straight forward to generalize this Knudsen flow estimate to the case of a q- (or frequency-) dependent bulk-phonon mean free path, $l_{mfp}(q)$. The formal results corresponding to Eqs. (10) and (11) — with $\eta(q) \equiv d/l_{mfp}(q)$ — then express the Knudsen-flow reduction from the (average) bulk-conductivity contribution, $\kappa_{avg}(q)$, at phonon momentum q. The in-plane superlattice thermal conductivity obtains by a weighted integration of the estimated superlattice contributions, $\kappa_{SL}(q)$, over q.