Phonon superlattice transport

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We predict in Si/Ge superlattices a dramatic high-temperature suppression of the perpendicular thermal transport. Total internal reflection confines the superlattice modes and significantly reduces the average group velocity at nonzero in-plane momenta. These consequences of the acoustic mismatch cause at high temperatures an order-of-magnitude reduction in the ratio of superlattice thermal conductivity to phonon relaxation time. [S0163-1829(97)07441-9]

The observed\(^1,2\) reduction of the in-plane thermal conductivity in long-period GaAs/AlAs superlattices implies a potentially attractive application for semiconduc
tor heterostructures in thermoelectric devices.\(^3\) The in-plane superlattice thermal transport has consequently been the focus of theoretical investigations.\(^4,5\) Two recent experiments\(^6,7\) also report a dramatic reduction for the thermal transport perpendicular to the GaAs/AlAs superlattice interfaces. The perpendicular thermal conductivity \(\kappa_{\text{SL}}\) reported in Ref. 7 exhibits an order-of-magnitude room-temperature reduction compared with that of bulk GaAs and cannot be explained as alloy scattering.

A very recent paper\(^8\) investigates this microscopic heat transfer and reports a Boltzmann transport calculation based on a bulk phonon dispersion in each of the superlattice layers. Assuming complete diffusive scattering\(^9,14\) at every interface, that is, a very strong reduction in the effective phonon relaxation time \(\tau_{\text{SL}}\), it is possible to account for the observed\(^6,7\) reduction in the perpendicular superlattice thermal conductivity.

We believe it essential, however, to describe the superlattice thermal transport \(\kappa_{\text{SL}}\) across the increasingly perfect interfaces based instead on the superlattice-phonon spectrum. Total internal reflection results with a finite acoustic mismatch will drive a similar modal confinement and thermal conductivity reduction also in the GaAs/AlAs superlattices.\(^6,7\)

Figure 1 characterizes our simple-cubic model of the double-silicon/double-germanium superlattice phonon dynamics. The upper panel shows one-dimensional model schematic assuming effective silicon/germanium lattice constants \(a_{\text{Si/Ge}}\) and atoms \(M_{\text{Si/Ge}}\) connected by intrasilicon/intergermanium force constants \(F_{\text{Si/Ge}}\) and interlayer coupling constants \(K_p = \sqrt{F_{\text{Si/Ge}} M_{\text{Si/Ge}}}\). This one-dimensional model, however, can only describe the dynamics of superlattice phonons both polarized \((\xi_p \| \hat{z})\) and propagating exactly in the growth or \(\hat{z}\) direction.

The lower panel of Fig. 1 illustrates our simple-cubic model of the general superlattice phonon dynamics. We assume for simplicity a shared in-plane and perpendicular lattice constant, \(a=a_{\text{Si}}\) (identical in all layers), and add in-plane force constants\(^19\) \(F_{\text{Si/Ge}}\) necessary to describe the phonon propagation at a finite in-plane momentum \(\mathbf{q} \neq \mathbf{0}\). We also introduce characteristic frequencies \(\Omega_{p,\text{Si/Ge}}\)

\[
\Omega_p^2 = \sqrt{4F_{p,\text{Si/Ge}} M_{\text{Si/Ge}}}.
\]

The \(\xi_p \| \hat{z}\)-polarized phonon modes at in-plane momenta \(\mathbf{q}\) is then described by a bulk silicon (germanium) model dispersion:

\[
\omega_p^2 = \Omega_{p,\text{Si/Ge}}^2\left[1 - \cos(\mathbf{q}_{\text{Si/Ge}} a)\right]/2 + \Omega_{p,\text{Si/Ge}}^2(\alpha_q^2/2),
\]

\[
\alpha_q = [2\cos(q_x a) - \cos(q_z a)] < 4.
\]
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relating frequency $\omega_q$ and perpendicular momentum $k_{Si(Ge)}$. We describe the bulk silicon (germanium) modes with polarization $\xi_{i,j};\hat{x},\hat{y}$ using the dispersion

$$\omega^2_{q,i,j} = \Omega_{i;Si(Ge)}^2 [1 - \cos(k_{Si(Ge)}a)]/2 + (\Omega^2_{p;Si(Ge)} + \Omega^2_{i;Si(Ge)}) (a_q/4).$$

Silicon is significantly harder than germanium. Our simple model yields $\Omega^2_{p;Si(Ge)} = 40.9$ (22.9) meV and $\Omega^2_{i;Si(Ge)} = 28.3$ (16.5) meV and is not an accurate description of the longitudinal or transverse phonon modes. However, the ratios $\Omega^2_{p;Si(Ge)} / \Omega^2_{i;Si(Ge)}$ are consistent with the actual bulk phonon spectra and our model thus reflects the essential effects of the acoustic mismatch. Below we concentrate our discussion on the $\xi_{p};\hat{z}$-polarized modes.

Assuming perfect interfaces the in-plane momenta $q$ is conserved and thus characterizes also the superlattice modes together with the squared frequency value $\omega^2_q$. The perpendicular dynamics of such a mode is described by the individual-layer model dispersion

$$\omega^2_{q;Si(Ge)} = \Omega^2_{p;Si(Ge)} [1 - \cos(k_{Si(Ge)}a)]/2,$$  

where $\omega^2_{q;Si(Ge)} = \omega^2_q - \Omega^2_{p;Si(Ge)} (a_q/2)$ defines the effective squared frequency available for the perpendicular motion within the silicon/germanium.

A superlattice mode $(q, \omega^2_q)$ will propagate (i.e., be characterized by a real wave vector $k_{Si(Ge)}$ through a silicon/germanium layer if and only if $0 \leq \omega^2_q - \Omega^2_{p;Si(Ge)} \leq \Omega^2_{p;Si(Ge)}$. In contrast, superlattice modes with $\omega^2_{q;Si(Ge)} < 0 (> \Omega^2_{p;Si(Ge)})$ are germanium (silicon) confined by total internal reflection. For example, a high-energy mode with $\omega^2_{q;Si} - \Omega^2_{p;Si}$ yields a sub-monolayer decay length $1/\gamma_{Si}$ as may be estimated from

$$\cosh(\gamma_{Si}d) = (2 \omega^2_{q;Si}/\Omega^2_{p;Si}) - 1.$$ 

We describe the general phonon displacement field at $\hat{z}$ coordinate $j$ in layer $l$ and in-plane momentum $q$ by

$$\eta_{q,l,j}(j) = A_l \exp(ik_ja) + B_l \exp(-ik_ja),$$

using either real or complex wave vectors $k_{l;Si(Ge)}$ for modes which in the silicon/germanium are propagating or evanescent, respectively. For the $\xi_{p};\hat{z}$-polarized modes the individual-layer displacement fields, Eq. (6), couple at the heterostructure interfaces through force constants $F_{p;Si(Ge)}$. The equation of motion for the two atoms bordering the $l/l+1$ interface establishes the relation, $(A_l,B_l) \leftrightarrow (A_{l+1},B_{l+1})$.

Our four-atom superlattice has period $d = 4a$ and is described by the perpendicular superlattice phonon momentum $k \in [-\pi/d, \pi/d]$. We solve the equations linking $(A_l,B_l) \leftrightarrow (A_{l+2},B_{l+2})$ subject to the Bloch ansatz at $k$:

$$\eta_{q,l,j}(j+4) = \eta_{q,l,j}(j)e^{ikd},$$

to obtain the frequency-squared values, $\omega^2_{q,l;\xi,p}\xi,\lambda = 1 - 4$, and thus determine the (zone-folded) superlattice modal spectra (for $\xi_{p};\hat{z}$- as well as for $\xi_{\hat{x}};\hat{x},\hat{y}$-polarized modes).

We assume a frequency-independent phonon relaxation time $\tau_{Sl}$ and denote by $N(T)$ the Bose-Einstein distribution function at temperature $T$. Generalizing the Boltzmann equation result for bulk phonon transport, we evaluate the ratio of the perpendicular superlattice thermal conductivity $\kappa_{Sl}$ to phonon relaxation time $\tau_{Sl}$ using

$$\frac{\kappa_{Sl}}{\tau_{Sl}}(T) = \frac{h}{2 \pi} \sum_{m} \left[ \int \frac{d^2q}{(2\pi)^2} \int_{-\pi/d}^{\pi/d} \frac{2\pi}{d\pi} \right] \left[ \sum_{\lambda} \omega^2_{q,l;\xi,p;\lambda} \left( \frac{d\omega_{q,l;\xi,p;\lambda}}{dk} \right)^2 \left( \frac{dN}{dT} \right) \right].$$

Here the index $m$ denotes a sum over polarization $(\xi_{p},\xi_{\hat{x}},\lambda)$ and $\lambda$ identifies the set of superlattice modes at $m$, $q$, and (zone-folded) perpendicular momentum $k$. The corresponding estimates, $\kappa_{Si(Ge)}/\tau_{Si(Ge)}$, for bulk silicon/germanium result by replacing in Eq. (8) $d$ by $a_{Si(Ge)}$ and of course restricting $\lambda$ to the single acoustic band described by our effective simple-cubic lattice model. Note that we divide out the very different phonon relaxation times, $\tau_{Si(Ge)}$, to obtain a fair comparison of the thermal conductivities and illustrate the superlattice effects.

Figure 2 compares the temperature variation of superlattice ratio $\kappa_{Sl}/\tau_{Sl}$, solid curve, with the corresponding estimates for bulk silicon, dashed curve, and for bulk germanium.
nium, dotted curve. To interpret this temperature variation of $k/\tau$ both for the superlattice and in the bulk we approximate $\kappa$ as a product of the average group velocity, $\langle \partial \omega / \partial k \rangle$, average mean free path, $\tau(\partial \omega / \partial k)$, and of the heat capacity, $C$:

$$\kappa \sim \tau(\partial \omega / \partial k)^2 C/3.$$  

(9)

In the low-temperature limit we may approximate the bulk silicon/germanium heat capacity, $C_{SiGe} \propto (T/\bar{v}_{SiGe})^3$ where $\bar{v}_{SiGe}$ denotes the average long-wavelength sound velocity. Thus we have ratios $\kappa_{SiGe}/\tau_{SiGe} \propto T^3/\bar{v}_{SiGe}$ yielding a cubic temperature dependence for the bulk materials. The low-temperature ratio $\kappa_{SL}/\tau_{SL}$ is an average of $\kappa_{SiGe}/\tau_{SiGe}$ because the long-wavelength superlattice velocity is an average of the Si/Ge velocities as documented in the inset of Fig. 2.

In contrast, the high-temperature ratio $\kappa_{SL}/\tau_{SL}$ shows a dramatic suppression compared with the average bulk silicon/germanium values. This reduction reflects a suppression of the perpendicular average group velocity:

$$k/\tau \propto (\partial \omega / \partial k)^2; \quad T \lesssim \Omega_{p,SL}.$$

(10)

as the high-temperature bulk silicon/germanium and superlattice heat capacities saturate at the Boltzmann constant times the density of phonon modes. To understand the dramatic reduction of the high-temperature ratio $\kappa_{SL}/\tau_{SL}$ we investigate below the zone-folded superlattice phonon spectra at general in-plane momenta $q$ and illustrate how the acoustic mismatch forces a significant reduction of the average phonon group velocity.

Figure 3 compares squared frequency values, $\omega_q^2$, for zone-folded superlattice modes, $(0 \leq k_{SL} \leq \pi)$ central column, and for bulk-silicon/germanium propagating modes, $(0 \leq k_{SiGe} \leq \pi)$ left/right column, at increasing in-plane momentum $q$. The bottom row of panels compares these phonon spectra at $\alpha_q=0$, the middle row at $\alpha_q=0.9$, and the top row at $\alpha_q=2$. All panels shows squared frequencies relative to $\Omega_{p,SL}^2(\alpha_q^2/2)$ (dashed lines)—minimal value necessary for silicon propagation at $\alpha_q$. The dotted lines identifies squared frequencies which at $\alpha_q$ allow germanium propagation.

FIG. 2. Dramatic suppression of perpendicular superlattice thermal transport. The solid curve shows order-of-magnitude high-temperature reduction for calculated superlattice ratio of thermal conductivity ($\kappa_{SL}$) to phonon relaxation time ($\tau_{SL}$) when compared to average of bulk silicon/germanium ratios, dashed/dotted curve. The conductivity suppression results from the nonzero in-plane momenta $q \neq 0$ ensuring an effective confinement of the superlattice modes (Fig. 3). The inset verifies that no such confinement effect arises in the long-wavelength limit (relevant for the low-temperature transport): lowest $q=0$ superlattice spectrum, solid curve, is an average of corresponding Si/Ge spectra, dashed/dotted curves.
and two modes located above the range of germanium-
propagating modes. The top superlattice mode is almost en-
trirely siliconlike. It is much too energetic to penetrate the
germanium, and is consequently characterized by a vanish-
ing group velocity.\textsuperscript{13}

The intermediate value $\alpha_q = 0.9$, central middle panel,
forces a finite offset between silicon- and germanium-
propagating modes as identified by the set of dashed and
dotted lines. The offset quantifies the traditional total internal
effect: germanium phonons at a given $q \neq 0$ must have also a
finite perpendicular momentum—that is, they must have more
than the minimum energy set by $q \neq 0$—to enter the
harder silicon.

In turn this offset between the silicon- and germanium-
propagating phonons causes a partial germanium confine-
ment of the lowest superlattice mode and an increased silicon
confinement of both upper two modes. Already at $\alpha_q = 0.9$
we observe a significantly reduced contribution to the av-
erage superlattice phonon group velocity.

The central top panel illustrates how in-plane momenta
with $\alpha_q \approx 2$ ensures a partial confinement of all superlattice
phonon modes. The range of squared frequency values for
modes propagating in the silicon (germanium) layers is at
$\alpha_q \approx 2$ situated entirely above (below) the dashed-dotted line.
The top (bottom) two superlattice modes are thus at $\alpha_q \geq 2$
increasingly silicon- (germanium-) confined even by the thin
adjacent germanium (silicon) layers.

The value $\alpha_q = 2$ corresponds roughly to a minimal pho-
on energy of one half the germanium Debye temperature. At
such temperatures and above the average modal confine-
ment causes a dramatic suppression of the ratio $k_{\text{sil}}/k_{\text{ge}}$
as documented in Fig. 2.

We have theoretically investigated the thermal transport
perpendicular to the interfaces in a double-silicon/double-
 germanium superlattice assuming a simple-cubic model for
the lattice vibrations. We predict an order-of-magnitude
high-temperature suppression in the ratio of the perpendicu-
lar thermal conductivity to the phonon relaxation time. This
dramatic suppression occurs at temperatures above one half
the germanium Debye temperature when the acoustic mis-
match forces an effective modal confinement at the finite
in-plane momenta $q$ characterizing this transport. Upon
completion of this work we have learned of recent Si/Ge-
superlattice measurements reporting perpendicular thermal
conductivities smaller than that of an actual Si/Ge alloy.\textsuperscript{22}

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