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Contributions of strain relaxation and interface modes to thermal transport in superlattices

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ABSTRACT

Superlattice structures are widely used in electronic and optoelectronic devices, many of which depend heavily on thermal management for performance and reliability. It has been observed that silicon/germanium superlattices exhibit an enhancement in thermal conductivity at very short period lengths, which has been attributed to the contribution of coherent phonons. Here we investigate additional potential contributions to enhanced thermal conductivity in superlattices as period length is reduced, finding that a reduction in strain relaxation as well as increased contributions of interface modes that have vibrational character intermediate between those of the two constituent materials offer additional mechanisms for increased thermal conductivity.

1. Introduction

Superlattices have shown promise for applications such as thermoelectric devices [1], semiconductor lasers [2,3], and transistors [4]. For these applications, thermal management is often important to performance and reliability; in the case of thermoelectric devices, heat transfer is key to fundamental operation. In general, thermal conductivity has been found to decrease in superlattices as period length decreases, due to increasing interface density [5]. Several studies however have indicated that thermal conductivity increases for very short period lengths, and have attributed this effect to phonon coherence [6,8].

In addition to phonon coherence, other factors may play a role in increasing the thermal conductivity of short-period superlattices, such as a lack of strain relaxation that leads to a more uniform lattice constant. Lattice mismatch in epitaxial structures imposes a residual strain field that has a maximum at the interface and relaxes away from the interface [9]. It is well known that lattice strain can have a significant effect on the optical [11–13] and electrical [10] properties of materials. Because the thermal properties of semiconductors are primarily dictated by phonons, the properties of which are affected by lattice strain, several studies have examined the effect of strain and stress on bulk and interfacial heat transfer [14–16]. Isotropic tensile strain of ~3% was shown [17] to decrease the thermal conductivity of silicon by a factor of ~0.82. Experimental and numerical studies have also shown that compressive stress on a weak interface can increase its thermal conductance [18,19]. As the period length of a superlattice decreases below the strain relaxation length, the lattice constant within the superlattice is expected to fall within a narrower range, potentially leading to reduced scattering as phonons experience less variation in acoustic impedance. Furthermore, as the interface density increases, interfacial vibrational modes (which may have a character intermediate between those of the two constituent materials) are expected to play an increasing role in heat transfer and may create an additional channel for phonon transmission. To assess the contributions of these two factors to thermal transport in short-period superlattices, we apply an approach combining an initial energy minimization process (to accurately account for strain relaxation) followed by MD simulation (to analyze physical properties). This energy minimization method, which provides a means to generate strain profiles in an atomistic simulation that are in agreement with experimental results, can be used to study the physical properties (e.g., electrical, optical, and thermal) of strained interfaces; we use it here to study the effects of strain relaxation on thermal boundary resistance (TBR) and thermal conductivity in Si/Ge heterostructures and superlattices with various period lengths. We also calculate the phonon density of states

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(PDOS) for the materials in the superlattice to examine differences in PDOS that arise in superlattices with different period lengths, particularly the emergence of contributions due to interfacial (silicon–germanium) bonds in short-period superlattices.

2. Simulation technique

Several studies have examined the thermal conductance of a single heterointerface or the thermal conductivity of a multilayer heterostructure in materials systems that have significant lattice mismatch [20–22]. However, these and other previous computational studies have not accounted for strain relaxation when predicting the physical (e.g., optical, electrical, thermal) properties of a junction; instead, piecewise-constant strain have been specified before physical properties are calculated using techniques such as molecular dynamics (MD). This approach neglects the spatial variation of material properties such as phonon dispersion which may significantly affect the junction's properties. We implement a non-equilibrium molecular dynamics (NEMD) technique [23] with a Stillinger–Weber (SW) force field [24] to study thermal transport in Si/Ge heterostructures. An initially unrelaxed piecewise constant strain profile similar to previous studies [20,21] is first implemented in the simulation cells. Non-periodic boundary conditions are applied in all directions to mimic heat transfer in a nanorod and to let the strain relax in the system. For certain simulations a steepest descent (SD) energy minimization technique is applied prior to dynamic simulation in order to relax the strain in the system. The energy minimization stops when either the normalized change of the total potential energy of the system is less than 10^{-12} or the change in the total force vector is less than 10^{-12} (kcal/mol Å). Four layers of atoms on the periphery of the system are fixed in place to avoid the sublimation of atoms into vacuum during MD simulation. A time step of one femtosecond [6] is used to capture the fastest atomic vibrations in the system. A Langevin thermostat [25] is used to thermalize the simulation cell. The temperature of the system is first set to 5 K for 0.1 ns and then set to 50 K for 0.2 ns to relax the initial thermal noise in the system, after which the temperature is set to 500 K for 0.7 ns. The thermostat is then removed and the system evolves in an NVE ensemble (constant number of particles, volume, and energy) in the presence of a constant heat flux that is applied through a heat source and heat sink to impose a temperature gradient. The system evolves in this ensemble for 6.5 ns to let the temperature gradient stabilize, after which the system evolves for one more nanosecond, during which the temperature gradient across the material is recorded to derive the thermal conductivity and/or thermal boundary resistance (TBR). Fig. 1 schematically shows a single junction simulation cell. To achieve a good statistical average, the temperature profile of the system is averaged over the final one nanosecond (which corresponds to one million time steps). The TBR is calculated based on Fourier's law [23]:

$$TBR = \frac{q}{\Delta T} \tag{1}$$

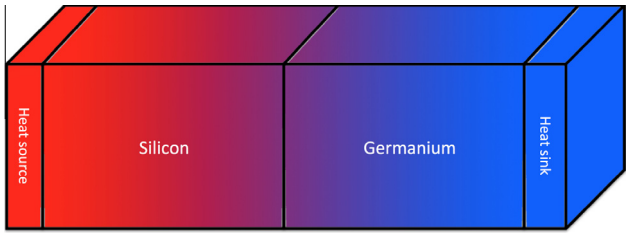

Fig. 1. Single junction heterostructure simulation cell.

where q is the heat flux passing through the interface, and ΔT is the temperature drop at the interface. To calculate the strain in the direction perpendicular to the interface, the position of the center of mass of each atomic layer is calculated at each time step, after which the change in distance between two adjacent layers with respect to the interlayer distance of the pure material at room temperature (one quarter of lattice constant of the pure material) is used to calculate the strain (Eq. (2)).

$$\varepsilon_{xx,ab} = \frac{\left(\frac{\sum(x_{a,i}) - \sum(x_{b,j})}{(y*z*2)}\right)}{SL} \tag{2}$$

where $\varepsilon_{xx,ab}$ is the strain in the xx direction (perpendicular to the interface) between adjacent layers a and b, y and z are the number of cross sectional unit cells, i and j are the atom numbers in the a and b layers respectively, and SL is the distance between the two layers in the unstrained pure materials at room temperature. For strain in the planes parallel to the interface, the average distance between the top and bottom atoms of each layer is used to calculate the strain. To verify the accuracy of our calculations, we first predict the lattice constants and thermal conductivities of pure silicon and germanium, using periodic boundary conditions to represent a bulk crystal. The Green–Kubo technique [23] is used to derive the thermal conductivities based on the averages of 21 calculations. The results are shown in Tables 1 and 2 and are in good agreement with literature values [26,17,27]. All MD simulations have been done using the LAMMPS (http://lammps.sandia.gov) package [28].

3. Results

Starting with a piecewise constant strain profile in the simulation cell and using our strain relaxation technique discussed above, we examine the relaxed strain profile in Si/Ge heterostructures and the effect of this strain relaxation on thermal properties. The simulation cells used here consist of $10 \times 10 \times 288$ unit cells of atoms in the diamond cubic lattice structure (8 atoms in each unit cell).

3.1. Strain relaxation at single Si/Ge heterojunction

For the same simulation cell we run two simulations: one with a conventional piecewise constant strain profile and one with the energy minimization step to relax the strain in the material prior to dynamical simulation. The initial piecewise constant strain profile is similar to those used previously [20,21]. Lattice mismatch between the materials causes strain in the direction parallel to the interface. Axial strain then arises as the system reduces its potential energy. The relaxed strain profile for a single heterojunction in the lateral direction is shown in Fig. 2. The strain profile shows that most of the strain is relaxed in a few layers close to the interface (~2 nm), while a small amount relaxes over a range

Table 1
Derived silicon and germanium lattice constants.

Material	Lattice constant (Å)
Silicon	5.433
Germanium	5.655

Table 2
Derived silicon and germanium thermal conductivities.

Material	Thermal conductivity (W/mK)
Silicon	107 ± 32
Germanium	59 ± 12