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Effects of Aperiodicity and Roughness on Coherent Heat Conduction in Superlattices

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Abstract: Coherent phonon heat conduction has recently been confirmed experimentally in superlattice structures. Such traveling coherent phonon waves in superlattices lead to linear increase of thermal conductivity as the -number of periods increases. For applications such as thermal insulation or thermoelectrics, minimization of phonon coherent effect is desirable. In this work, we use molecular dynamics simulations to study how to control coherent heat conduction in superlattices (SLs). It is found that either aperiodic SLs or SLs with rough interfaces can significantly disrupt coherent heat conduction when the interface densities are high. For sample thickness less than 125 nm, aperiodic SLs with perfect interfaces are found to have lowest thermal conductivity. We further use atomic Green's function method to examine the phonon dynamics. The impact of either aperiodicity or interface roughness is attributed to reduced transmittance. Such impact diminishes as interface density reduces.

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In bulk crystalline materials, phonons carry heat through random Brownian motions. They lose phase information when they scatter with other phonons. As a result, energy is diffusively transferred through numerous phonon collisions and the thermal conductivity is independent of sample size. When phonon mean-free-path (MFP) is longer than the characteristic dimension such as the thickness of a film, phonons transverse the thickness ballistically and maintain their phase information till they scatter at the boundaries or interfaces. Diffusive scattering at the boundaries may randomize the phase of phonon waves, leading to the classical size effect expected at the Casimir limit [1,2]. Due to their static nature, boundary scattering may also lead to wave effects, such as interference or localization. Although localization has not been confirmed, recent experimental studies in superlattices (SLs) have shown that some phonons can travel ballistically through many layers of the SLs without destroying their phases, demonstrating their wave nature of heat conduction [3,4]. Simulations show [3,5–9] that the coherent heat conduction is due to long wavelength phonons that cannot be effectively scattered by interface roughness, while higher frequency phonons are effectively scattered by interface mixing, which is responsible for the low thermal conductivity reported in many superlattice systems [4,10–14]. Reducing the contribution of these long wavelength phonons can lead to even lower thermal conductivity of SLs that is desirable for applications such as thermoelectrics [15–19]~~ [cite Optimization of the random multilayer structure to break the random-alloy limit of thermal conductivity][cite Decomposition of coherent and incoherent phonon conduction in superlattices and random multilayers,]~~.

One way to reduce the contribution of these long wavelength phonons is via aperiodicity. Although the Fibonacci and Thue-Morse SLs or quasicrystals has been investigated in the past for electron and phonon transport and localization [20–25], the impact of aperiodicity on the coherent heat conduction in SLs has not been discussed. In this work, we use molecular dynamics (MD) simulations to study the impact of aperiodicity, interface roughness, and interface density on the coherent heat conduction in symmetrically-strained Si/Ge SLs. We found that aperiodic perfect SLs give the lowest thermal conductivity. Aperiodicity can lead to further decrease of thermal conductivity of rough SLs compared to rough periodic SLs when the average periodicity of SLs is thin (or interface density is high). We also use phonon transmission function through the SLs calculated by atomic Green's function (AGF) method to further understand the MD simulation results.

The MD simulations are carried out using the LAMMPS package [26]. The Tersoff potentials for Si and Ge are adopted in MD simulations and LD calculations [27]. Four types of SLs with effective period thickness of 2 and 20 nm, respectively, are reported: 1) periodic SLs with atomistically perfect interfaces (pSL); 2) periodic SLs with rough interfaces created by intermixing two adjacent atomic layers at the interface (rough pSL); 3) aperiodic SLs with random layer thickness and with atomistically perfect interfaces (apSL); 4) aperiodic SLs with random layer thickness and with rough interfaces (rough apSL). All SLs are prepared by assuming the averaged lattice constant of Si and Ge and relaxing the lattice structure till free of internal pressure [7]. The final lattice constant is 5.54 Å. The resulting superlattice is symmetrically-strained that may lead to lower thermal conductivity [28]~~[cite Origins of thermal conductivity changes in strained crystals]~~. To study the thermal conductivity of SL samples, we use the reverse nonequilibrium molecular dynamics (RNEMD)

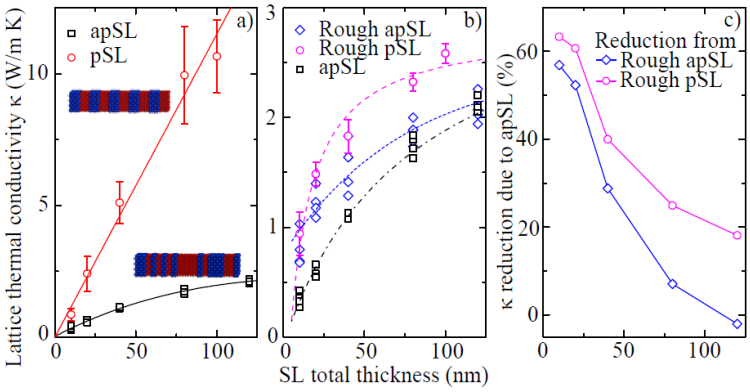

Figure 1. a) Thermal conductivity of pSL and apSL with perfect interfaces and effective period thickness 2 nm. b) Thermal conductivity of apSL (repeat that in a)), rough SL, and rough apSL. c) Reduction of thermal conductivity if one has perfect aperiodic superlattices without interface mixing.

simulations [29,30]. Periodic boundary conditions are applied in the in-plane direction of the SLs and a constant heat flux is introduced by periodically injecting and extracting energy to and from the hot and cold baths at the two ends of the SL simulation domain. The outmost atomic layers are fixed at both ends to prevent sublimation of atoms during the simulation. The apSLs are prepared by randomizing the Si/Ge layer thickness while maintaining the ratio between the number of Si and Ge atoms to be 1:1. About five randomly uncorrelated configurations are generated for each apSL and rough SL sample. Five independent simulations with different initial conditions are run for each pSL sample to minimize statistical fluctuations. The thermal conductance of SL samples is obtained as the ratio between the heat flux and the resulting temperature difference across the sample at steady-state:

$$G = q / \Delta T \tag{1}$$

and the effective thermal conductivity is obtained as

$$k = Gd \tag{2}$$

where d is the total SL thickness. Convergence of extracted effective thermal conductivity is achieved with cross-sectional area of 2.2×2.2 nm^2, two thermal baths of 1 nm width each, and heat current of 0.1 eV/ps. Depending on the sample size, the temperature difference across the sample can be up to 60 K. All simulations are done at average temperature of 300 K without quantum corrections. At higher temperatures, the anharmonic scattering will dominate and the system will be in diffusive-dominant regime and the effect of superlattice structuring will diminish. It should be emphasized that most of previous works consider SLs of infinite thickness in the cross-plane direction [5–8,31,32] while this work reports thermal conductivity of finite thickness samples.

The thermal conductivity as a function of total SL thickness for SLs with effective period thickness of 2 nm is shown in Fig. 1. As in Fig. 1 a), for SLs with perfect interfaces, the thermal conductivity of pSL increases linearly with SL total thickness up to 100 nm, which is consistent with the coherent heat conduction reported experimentally in GaAs/AlAs SLs [3]. Although the potential used include anharmonicity, the linear dependence of thermal conductivity indicates that phonon MFPs in such superlattices are longer than the total thickness of the SLs. In contrast, the thermal conductivity of apSL is much lower and increases slowly as sample becomes thicker, implying that the phonon waves may have been partially localized by the one-dimensional (1D) aperiodicity, which is consistent with previous studies that localization occurs in 1D structures. However, since the transmittance is a sum over all the different modes at the same frequency, we do not observe the exponential decay in the transmittance, which could be caused by different localization length of each specific mode or not all modes are localized. In Fig. 1 b), we plot thermal conductivity of pSL and apSL including interface mixing. Including both aperiodicity and interface mixing can reduce thermal conductivity from that of rough pSLs. However, interface mixing reduces the effectiveness of aperiodicity, and the lowest thermal conductivity is found in apSL without interface mixing. This is likely due to the density of states (DOS) overlapping introduced by interface mixing, which actually leads to enhanced heat conduction [8]. Meanwhile, it may be more difficult to form complete destructive interference, in other words, localization, through 3D disorder in rough SLs compared to 1D disorder introduced by aperiodicity only. The localization of phonon waves in disordered 1D geometry and the transition of localization into higher-dimensional scatterings have been reported in the works by Sheng and colleagues [33–36] although localization is not directly observed in this work. . In Fig. 1 c), we plot the ratio of thermal conductivity reduction of apSL to that of rough apSL and that of rough pSL to show the reduction of thermal conductivity if one has perfect aperiodic superlattices without interface mixing. As seen, if only aperiodicity is present, the reduction from rough SLs can be up to 60% in thin samples.

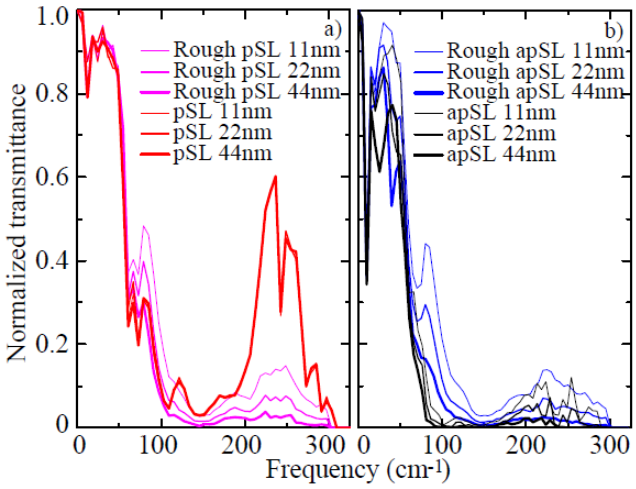

Figure 2. Normalized phonon transmittance from AGF for a) pSL and rough pSL, b) apSL and rough apSL. Each effective layer thickness is 1.1 nm.

In order to see how aperiodicity and interfacial roughness lead to similarly low thermal conductivity, we carry out AGF calculations [8,35], using force constants from first-principles [36]. The normalized transmittance for four types of SLs with different total sample thicknesses are shown in Fig. 2. In these cases, the effective period thickness is 1.1 nm. It can be seen that the transmittance for pSL saturates when the sample thickness is greater than 11 nm, leading to constant thermal conductance or linear thermal conductivity in sample thickness, which indicates coherent heat conduction. For SLs with either rough interfaces or aperiodicity, the transmittance monotonically decreases as more layers are added into the SLs over most frequency range except at very low frequencies, leading to a decreasing thermal conductance. Between 200 and 300 cm^{-1}, either interface mixing or aperiodicity significantly reduces the phonon transmittance from the pSLs. Below 60 cm^{-1}, however, only aperiodicity is effective in reducing phonon transmission. As seen in Fig. 2 b), if only aperiodicity is present, the transmittance is the lowest among all cases, especially in the intermediate frequency of 60 – 100 cm^{-1}, leading to lowest thermal conductivity, consistent with Fig. 1 b) and c). With interface mixing, the transmittance is increased. This can be understood as the interface mixing scatters phonons into all directions, reducing the effectiveness of phase cancellation and localization in purely 1D structures [34–36].

The thermal conductivity as a function of total sample thickness for SLs with effective period thickness of 20 nm is shown in Fig. 3. In contrast to previous cases where the interface densities are ten times higher, the thermal conductivities of these SLs are about linear in total sample thickness and the values are not significantly different, indicating coherent heat conduction in all the thick SLs investigated in this study. Both roughness and aperiodicity barely destroy the coherence. This finding is surprising. Our understanding is that at low interface densities, the disorder scattering is so weak that the majority of phonons, especially dominant heat-carrying modes with low frequencies, are not affected by the disorder scatterings and transverse the relatively thin SLs (up to 150nm) ballistically, regardless of aperiodicity or interface roughness. It should be noted that, due to the large effective period in the present system, the apSL is not

expected to be fully random up to total length of 150 nm. At very long length, the anharmonic scattering will likely dominate and the system will be in diffusive-dominant regime~~-~~, shadowing the effects of superlattice structuring~~and the effect of superlattice structuring will diminish~~.

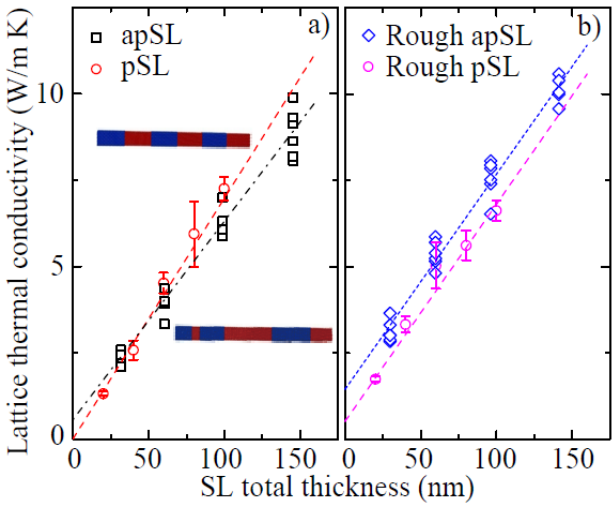

Figure 3. Thermal conductivity of pSL and apSL with perfect and rough interfaces. The effective period thickness is 20 nm.

In summary, we have used MD simulations to study the coherent heat conduction in Si/Ge SLs with periodic and aperiodic layers as well as with interfacial roughness. For SLs with low interface densities with sample thicknesses up to 150 nm, the thermal conductivity appears to be linear in total sample thickness, regardless of periodicity and interface roughness. With high interface densities, we found either aperiodicity or interface roughness leads to incoherent phonon transport with lower thermal conductivities than perfect periodic SLs. Perfect apSLs are found to give the lowest thermal conductivity. This indicates that localization may be achieved through 1D disorder which transits to higher-dimensional scattering with the inclusion of interfacial roughness. We also found apSL with rough interface leads to slightly lower thermal conductivity than that of rough pSL due to combined disorder scatterings. From the normalized transmittance computed from AGF, it was seen aperiodicity and interfacial roughness reduces thermal conductivity to the comparable extent.

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