Phonon-hopping thermal conduction in quantum dot superlattices

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We investigated the thermal conductivity in Ge/Si quantum dot superlattices both theoretically and experimentally. It is proposed that thermal conduction through the quantum dot superlattices can be described by the phonon-hopping model with the interface transparency obtained from the experiment. Thermal conductivity has been measured as a function of temperature T from 10 K to 400 K. We have observed an order of magnitude decrease in thermal conductivity value compared to bulk and shift of its peak position to higher temperatures. The thermal conductivity manifests $T^{0.7}$-$T^{0.9}$ dependence for $T \leqslant 200$ K. The phonon-hopping model describes the measured thermal conductivity surprisingly well over the wide range of T from ~40 K to 400 K. The model allows one to include the specifics of thermal conduction in quantum dot superlattices, such as the dot size, disorder, and interface quality. Our results suggest that the examined quantum dot superlattices are closer to the disordered or polycrystalline materials in terms of thermal transport. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2130711]

Quantum dot superlattices (QDS) continue to attract significant attention of the physics and device research communities due to their proposed applications in electronics, optoelectronics, and thermoelectrics.[1–4] Different device applications impose different requirements on the thermal transport properties of QDS. For example, it is desirable that QDS used in optoelectronic devices have high thermal conductivity for better thermal management. At the same time, thermoelectric device applications of QDS require low thermal conductivity.

Despite practical importance, there have been few studies of thermal conduction in QDS.[5,6] Heat in semiconductor materials is mostly carried by acoustic phonons. Since the feature sizes in QDS are smaller than the phonon mean-free path (MFP) in constituent bulk materials, one has to deal with both phonon scattering inside the dots and at the dots boundaries. Earlier attempts to describe thermal conductivity of QDS used a Callaway–Klemens type of approach within the relaxation time approximation by adding to the bulk relaxation time the extra term induced by phonon scattering on quantum dots.[5] The validity of such model is restricted to very high quality QDS, i.e., crystalline material with small dot size variation, low defects, sharp heterointerface, etc. The dot-host interface effects on phonon transport have been neglected. Taking into account that most of the molecular-beam epitaxy (MBE)-grown QDS are far from perfect, in this letter we suggest another treatment of thermal conduction in realistic disordered QDS. In the proposed approach, the phonon hopping from dot to dot, which strongly depends on interface conditions, is considered as a major thermal resistive mechanism. The phonon-hopping model has been successfully applied to a variety of disordered systems.[7] The validity of the application of this model to QDS is demonstrated by comparison with the experimental data.

The example material system investigated is a Ge/Si QDS, which consists of MBE-grown multiple arrays of Ge quantum dots separated by Si barrier layers. The schematic structure of QDS is shown in Fig. 1(a). To carry heat across the superlattice, the acoustic phonons have to go through the host material, quantum dots, and the dot-host interfaces. Phonon transport inside the barrier layers and dot material can be

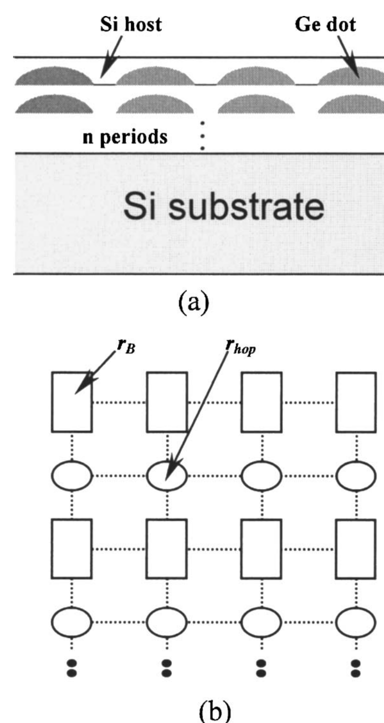

FIG. 1. (a) Schematic picture of Ge quantum dot arrays embedded in Si host material. (b) Corresponding phonon transport resistances network for the QDS structure.

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modeled using the Callaway–Klemens approach.[5] In view of the structural similarity between QDS and granular materials, we propose treating the additional thermal resistance—associated with the dot-host interfaces—by using the phonon hopping model developed by Braginsky *et al.*[8] for granular materials. We modified the theoretical approach of Ref. 8 by calculating the relevant phonon scattering rates to determine internal phonon MFP and by taking into account the finite dimensions of the dots.

To calculate the thermal conductivity of QDS in the framework of the phonon-hopping model, we draw the equivalent thermal resistances network for the phonon current flow in a QDS [see Fig. 1(b)]. For a specific phonon mode, the internal phonon resistance r_B is

$$r_B=\left(\frac{\omega^2}{2\pi^2 v}\frac{(\hbar\omega)^2}{\kappa_B T^2}\frac{e^{\hbar\omega/\kappa_B T}}{(e^{\hbar\omega/\kappa_B T}-1)^2}\tau_B\right)^{-1}, \quad (1)$$

where v is phonon group velocity, ω is phonon frequency, $\hbar$ is Plank constant, κ_B is Boltzmann constant, and τ_B is bulk relaxation time. The reverse space integration and branch summation of Eq. (1) will give the relevant bulk thermal conductivity. The hopping resistance, r_{hop}, can be written as[8]

$$r_{hop}=\left(\frac{9}{2}\frac{\kappa_B^2}{\hbar}\frac{T^5}{\Theta^4}\frac{\left(\frac{\hbar\omega}{\kappa_B T}\right)^4 e^{\hbar\omega/\kappa_B T}}{(e^{\hbar\omega/\kappa_B T}-1)^2}\left(\frac{\Theta}{T}-\frac{\hbar\omega}{\kappa_B T}\right)^2\frac{t\bar{S}}{a^2 d}\right)^{-1}, \quad (2)$$

where $\Theta=\hbar\omega_D/\kappa_B$ is the Debye temperature, d is the characteristic dimension of the quantum dot, $\bar{S}$ ($\approx d^2$) is the mean area of the hopping boundary, a is lattice constant, and t is the hopping transparency parameter. In the ideal case, the t parameter, which depends on the dot and host materials constants, can be calculated from the first principles. Using a simple one-dimensional model, we have estimated t to be $\sim$0.15 for a Ge/Si material system (atomic mass of Si and Ge are taken as 28 and 72, respectively). In the case of rough boundaries, t is a semiempirical parameter defined by the boundary quality. We further treat it as a fitting parameter determined from comparison with the experiment.

For the envisioned thermal resistance network, one can assume that the phonon scattering at the dot boundary is independent of the phonon bulk scattering mechanisms, i.e., phonon-phonon scattering, phonon-impurity scattering, etc. In this case, the effective MFP, Λ, satisfies: $1/\Lambda=1/\Lambda_B+1/\Lambda_{hop}$, where Λ_B is the internal scattering MFP and Λ_{hop} is the hopping distance. The total thermal resistance is therefore $r_{tot}=r_B+r_{hop}$. After integration over the inverse space for three phonon branches, we obtain the thermal conductivity as

$$k=\int_0^{\Theta/T}\frac{(9/2\pi^2 v)\tau_B\Theta(T/\Theta)^5\kappa_B(\kappa_B T/\hbar)^2x^8e^{2x}(e^x-1)^{-4}(x-\Theta/T)^2t\bar{S}\Phi}{(1/\pi^2 v)\hbar(\kappa_B T/\hbar)^2\tau_B d x^4e^x(e^x-1)^{-2}+9\Theta(T/\Theta)^5x^4e^x(e^x-1)^{-2}(x-\Theta/T)^2t\bar{S}\Phi}dx, \quad (3)$$

where $x=\hbar\omega/\kappa_B T$, Φ is the dot size fluctuation factor accommodating the variation of the individual hopping resistances in the network. For a network with more than 100×100 nodes, we take the numerically calculated value $\Phi=0.87$.[8] According to the Eq. (2), $r_{hop}\propto(td)^{-1}$. Therefore, k approaches the bulk value in two cases: (i) t is large enough at a constant d, and (ii) d is large enough at a constant t. Over a wide range of parameters in this model, the hopping resistance is dominant, i.e., $r_B\ll r_{hop}$. We prove *a posteriori* that for a typical MBE-grown Ge/Si QDS structure, the phonon hopping through the dot interface is the main resistive mechanism.

We apply the developed model to characterize the thermal conduction in a batch of MBE-grown Ge/Si QDS. The investigated samples consisted of ten layers of self-assembled disk-shaped Ge QDSs grown on a Si substrate. In one bilayer period, the nominal thickness of Si layer is 20 nm and that of Ge layer, in three different samples, they are 1.2 nm, 1.5 nm and 1.8 nm, respectively. Details of the growth procedure have been described elsewhere.[9,10] The base diameter of the dots estimated from atomic force microscopy topography is on the order of 100 nm with the aspect ratio of about 10. The density of Ge quantum dots is about 3.6×10^9 cm^{-2}. The characteristic space scale ($\sim$20 nm) is suitable for application of the phonon hopping model.[7,8]

The thermal conductivity of the QDS was measured in the temperature range from 10 K to 400 K using a home-built 3ω experimental setup.[11] To improve the measurement accuracy, the two-mask 3ω fabrication procedure[12] is adopted in this study. First, on the top surface of the samples, 250 nm of SiO_2 was deposited by plasma enhanced chemical vapor deposition. A 2 mm$\times$1 mm window was then opened by photolithography and buffered oxide etching. On the sample surface with an oxide window, we deposited a 30 nm thick SiN insulation layer. A Cr (100 Å)/Au (1000 Å) metallic 3ω heater thermometer, with the wire width of 5 μm and 10 μm, was patterned on the insulation layer and fabricated by electron-beam evaporation and lift-off technique. The pattern of heater thermometer was aligned with that of the oxide window so that the heater wire is inside the window while the contact pads are outside. The 3ω measurements were conducted inside a vacuum cryostat in the temperature range from 10 K to 400 K. An SR850 lock-in amplifier was used to provide first-harmonic input power and collect the third-harmonic temperature-rise signals from the sample. The thermal resistance of QDS was sufficiently larger than that of the Si substrate, which ensured that most of the temperature drop happens across the QDS layer. A numerical program based on the analytical solution of the 3ω heat conduction model was developed to fit the experimental data and obtain the thermal conductivity.

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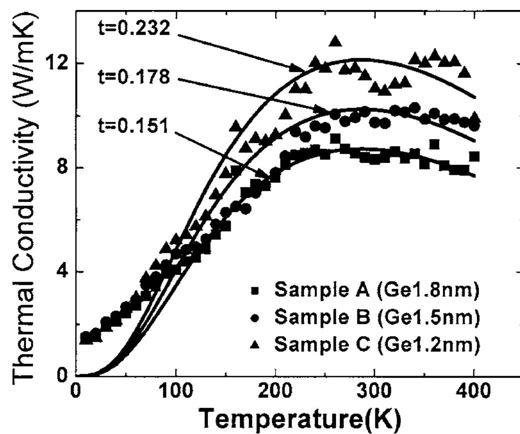

FIG. 2. Measured and calculated thermal conductivity as a function of temperature for three Ge/Si QDSs. Note the good agreement between the experimental and simulation results over a wide temperature range.

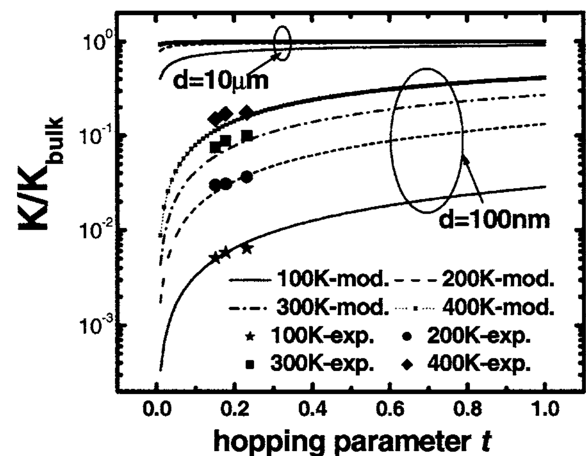

FIG. 3. Calculated normalized thermal conductivity as a function of the hopping parameter t at 100 K (solid curve), 200 K (dashed curve), 300 K (dashed-dotted curve) and 400 K (dotted curve with small solid squares) for the dot size $d=100$ nm (circled) and polycrystalline size $d=10$ μm (circled). For comparison, experimental data obtained for three Ge/Si QDSs is also shown (large solid symbols).

Figure 2 shows the measured thermal conductivity as a function of temperature for three samples, as well as simulation results obtained from the phonon-hopping model. The measured data indicate a significant reduction in the cross-plane thermal conductivity compared to that of the bulk constituents. The average thermal conductivity of the three QDS samples is about 10 W/mK at 300 K, while the corresponding value for bulk Si is about 130–160 W/mK. The thermal conductivity peak values in all examined QDS are shifted to higher temperatures and positioned at nearly 250 K–300 K, while for bulk Si or Ge the maximum value is achieved at around $T=20$ K. In the simulations, the quantum dots base diameter, 100 nm, is taken as the characteristic dimension. The simulation results reproduce the measured data very well over the whole temperature range for three samples with different hopping parameter t, as shown in Fig. 2. It is obtained that the hopping parameter goes reversely with the variation of the Ge layer thickness: The largest $t(=0.23)$ is associated with the data from the sample having the smallest Ge nominal thickness. This complies with the observation that the interface becomes rougher as Ge thickness in QDS increases.[13]

To further illustrate the combined effect of the hopping parameter and the quantum dot size, in Fig. 3, we plotted the calculated thermal conductivity as a function of t at $T=100$ K, 200 K, 300 K, and 400 K with the dot size $d=100$ nm and $d=10$ μm, respectively. The calculated thermal conductivity values are normalized to the bulk value at each given temperature. The shown experimental data for three samples are normalized to the bulk thermal conductivity obtained from Fourier's law for the effective Si (200 nm)+Ge (12, 15, and 18 nm) medium. One can see that the considered model provides a smooth transition to the bulk value when the "dot" size becomes very large or the hopping parameter t increases. This finding is in line with the recently reported phonon-hopping treatment of thermal conductivity in granular materials with a rather large grain size (from 100 nm to 500 nm).[14] The values of t obtained from the fitting to the experimental data are close to $t\sim 0.15$ estimated from the difference in the materials properties of Si and Ge.

In conclusion, we propose treating heat conduction in QDS as the phonon hopping transport rather than conduction in the bulk crystals with extra scattering centers (quantum dots). Our experimental and simulation results indicate that the phonon-hopping model describes the thermal conduction very well in a wide temperature range. In the framework of the phonon-hopping model, the thermal conductivity of QDS naturally transforms to the bulk limit for either very large hopping parameters (low thermal interface resistance) or large dot sizes (a dot becomes a granular). It also allows one to take into account the size variation and other disorder in QDS. The results and the modeling approach can be used for nanostructure optimization for electronic or thermoelectric applications.[15,16]

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