In-plane lattice thermal conductivity of a quantum-dot superlattice

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(Received 16 November 1999; accepted for publication 7 April 2000)

We have theoretically investigated the in-plane lattice thermal conductivity of a quantum-dot superlattice. The calculations were carried out for a structure that consists of multiple layers of Si with randomly distributed Ge quantum dots separated by wetting layers and spacers. Our model takes into account scattering of acoustic phonons on spherical quantum dots, and corresponding modification of the phonon dispersion relation. The finite acoustic mismatch between Si and Ge is also taken into account. The obtained results are important for the most recently suggested applications of SiGe quantum-dot superlattices for thermoelectric devices. © *2000 American Institute of Physics.* [S0021-8979(00)01214-7]

I. INTRODUCTION

Continuous progress in fabrication techniques for arrays of semiconductor quantum dots resulted in significant achievements both in understanding the physical processes in quasi zero-dimensional structures and in their applications.[1,2] Until recently, most of the applications envisioned for quantum dot arrays were in the area of optoelectronics.[2–4] The situation has changed with several most recent proposals of utilization of quantum-dot superlattices (QDSs) for thermoelectrics applications.[5–7] Randomly distributed Ge quantum dots in planar layers of Si strongly scatter phonons, while they do not severely deteriorate electron transport. Quantum confinement of carriers in QDS can also add up to the thermoelectric power factor and the figure of merit. Thus, QDS represents a good example of the ''phonon-blocking electron-transmitting'' structure with a great potential for thermoelectric applications.[5,6] An analogous structure with regimented arrays of Bi-doped PbT dots has been examined in Ref. 7. It has been reported that the thermoelectric power factor of the PbSeTe-based QDS structures was higher that that of high quality bulk PbTe. The thermoelectric figure of merit $ZT=S^2\varepsilon/(\kappa+\kappa_e)$ of the regimented QDS has been shown to be as high as 0.8 at 300 K (where S is the Seebeck coefficient, ε is the electric conductivity, κ is the phonon thermal conductivity, and κ_e is the electronic thermal conductivity).

In this article we report the theoretical model of the in-plane thermal conductivity of QDS. Thermal conductivity of semiconductors is the sum of the phonon (lattice) κ and electronic κ_e components. The maximum value of ZT is usually obtained for doped semiconductors, where $\kappa_e/\kappa\sim 1/2$,[8] although this ratio for an unintentionally doped intrinsic SiGe QDS is much lower. Thus, we neglect its electronic part and limit our treatment to the phonon contribution to the thermal conductivity.

II. MODEL

In the relaxation-time approximation the lattice thermal conductivity is written as[9]

$$\kappa=\frac{1}{3}\sum_i dk\,\nu_{g_i}^2(k)\,\tau_{C_i}(k)S_i(k), \tag{1}$$

where i denotes particular phonon polarization branch, ν_{gi} is the phonon group velocity, τ_C is the combined relaxation time, $S_i(k)dk$ is the contribution to the specific heat from modes of the polarization branch i, which is the phonon wave vector interval of kdk. Thermal resistance and thermal conductivity arises due to phonon relaxation in different scattering processes which do not conserve crystal momentum.[10] The scattering processes which are dominant in crystalline Si_xGe_{1-x} include phonon anharmonic interactions (three-phonon Umklapp processes), point defect scattering (isotopes, impurities, etc.), and boundary scattering. They enter Eq. (1) via the combined relaxation rate

$$\frac{1}{\tau_C}=\frac{1}{\tau_U}+\frac{1}{\tau_B}+\frac{1}{\tau_M}, \tag{2}$$

where $1/\tau_U$, $1/\tau_B$, and $1/\tau_M$ are the phonon relaxation rates in the three-phonon Umklapp, boundary, and mass-difference (impurities and isotopes) scattering processes, respectively.

In order to construct a theoretical formalism for QDS, we include a new phonon relaxation mechanism—scattering on quantum dots in this article. Due to the fact that the characteristic feature size of a quantum dot W is smaller than the phonon mean-free-path Λ and approaching the phonon coherency length ($L\sim 2$ nm), phonon relaxation on quantum

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dots has to be considered as a separate scattering process. Thus, we have to calculate phonon scattering rate on quantum dots $1/\tau_D = v_g \sigma_V / V$ (where σ_V is the total phonon scattering cross section in volume V and v_g the phonon group velocity modified by scattering on quantum dots) and to add it to the sum in Eq. (2).

To describe phonon transport in QDS, we use the continuum model approximation and an assumption that the thermal phonon wave can be represented by a sum of plane waves. A phonon wave outside the dot is a superposition of incident plane and scattered waves. We consider semispherical quantum dots of equal size randomly distributed through the plane. The interdot separation distances and spacer layer thickness are assumed to be greater than the characteristic dot size. In this case, the scattering is the incoherent process,[11] and the total scattering cross section of quantum dots simply becomes $\sigma_V \approx N\sigma$, where σ is a scattering cross section of a single dot.

The solution of the scattering problem for a spherical dot is known from acoustics and is not reproduced here.[11] Using this solution, we can write down an expression for the scattering cross section σ of a single dot as

$$\sigma = \frac{\pi}{k^2} \sum_{m=0}^{\infty} (2m+1)|1+R_m|^2. \tag{3}$$

Here R_m is a reflection coefficient

$$R_m = \frac{h_m'^*(ka) + i\beta h_m^*(ka)}{h_m'(ka) + i\beta h_m(ka)}, \tag{4}$$

where

$$\beta_n = i \frac{\rho c}{\rho_e c_e} \left[\frac{j_n'(ka)}{j_m(k_e a)} \right],$$

ρ is density, c is the sound velocity, the subscript e denote the parameter which corresponds to the dot material, $h_m(ka) = j_m(ka) + iy_m(ka)$, j and y are the spherical Bessel functions of the first and second kind, respectively, and h_m^* is the complex conjugate.

Considering absolutely rigid spherical dots ($\beta_n \to 0$), Eq. (3) simplifies to $\sigma \sim 5.6(ka)^4 a^2$ for $ka \ll 1$; and $\sigma \sim 2\pi a^2$ for $ka \gg 1$ in the long-wave and short-wave limits, respectively. It is seen now that phonon scattering on quantum dots is a process intermediate between the point defect and boundary scattering, with the relaxation rate $1/\tau_D$ approaching the well known limits

$$\frac{1}{\tau_D} \sim \frac{\omega^4}{\nu_G^3} \text{ point-defect scattering} \quad (ka \ll 1)$$

and

$$\frac{1}{\tau_D} \sim \nu_G \text{ boundary scattering} \quad (ka \gg 1). \tag{5}$$

Equation (5) implies that a quantum dot acts like as a impurity atom if the dot size is much less than phonon wavelength in one extreme. In the other, when the dot size is much larger than phonon wavelength, a quantum dot acts like an additional boundary. For our numerical study, we use the actual acoustic mismatch between Si and Ge ($\chi = 0.78$), and the general expression for the reflection coefficient given by Eq. (3).

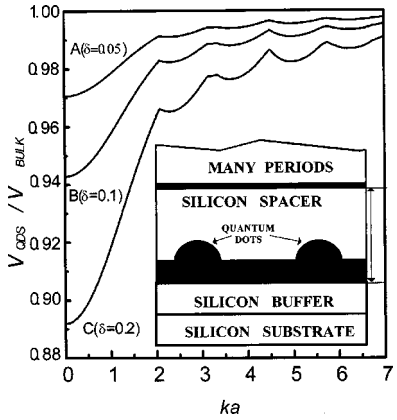

FIG. 1. Phonon group velocity in the quantum dot superlattice (QDS) as a function of the *ka* product (k is the phonon wave vector and a is the dot radius). The velocity is normalized to its bulk value. The results are shown for several volume fractions occupied by quantum dots. The inset shows geometry of QDS. $\delta = 0.05$, 0.1, 0.2 for A,B,C respectively.

Due to scattering on quantum dots, the phonon dispersion and corresponding phonon group velocity in QDS will be changed. This change can be taken into account using the following relation:[12,13]

$$k'^2 = k^2 \left(\left[1 + \frac{2\pi NF(0)}{Vk^2} \right]^2 - \left[\frac{2\pi NF(\pi)}{Vk^2} \right]^2 \right), \tag{6}$$

where k' is the wave number in the presence of quantum dots, k is the wave number when the dots are absent, and F is the scattering function.

$$F(\vartheta) = \frac{i}{2k} \sum_{n}^{\infty} (2n+1)(1+R_n) P_n(\cos\vartheta), \tag{7}$$

where $P_n(\cos\vartheta)$ are Legandre polynominals, and $F(0)$ and $F(\pi)$ correspond to the forward and backward scattering, respectively. When the concentration of quantum dots (N/V) is low, the influence of the backward scattering, which is proportional to the $(N/V)^2$, can be ignored. An additional modification of the phonon group velocity may come from spatial confinement of phonon modes inside the two-dimensional spacer and wetting layers of QDS.[14]

III. RESULTS OF COMPUTATIONS

Numerical calculations have been carried out for QDS with the spacer layer of 100 nm. First, we have calculated a reflection coefficient and a scattering cross section of a single dot were. Second, using Eq. (6), the phonon dispersion and modified group velocity were found. In Fig. 1 the phonon group velocity in QDS was shown as a function of the *ka* product. The results were shown for several volume fractions of quantum dots δ, which were approximated by semispheres with radius $a = 4.0$ nm ($\delta = 0.05, 0.1, 0.2$ A,B,C, respectively). One can see a noticeable drop in the phonon group velocity for wave vectors comparable to or less than the size of a quantum dot. The drop is particularly strong for a large volume fraction of dots ($\delta = 0.2$). One should mention here, that

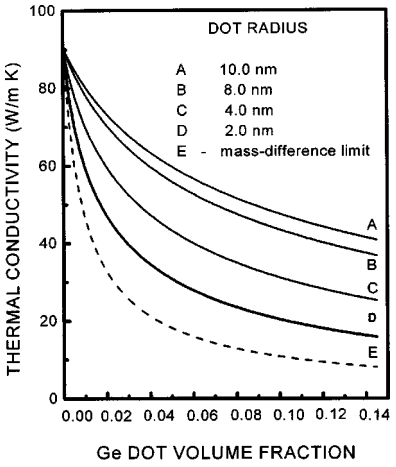

FIG. 2. In-plane lattice thermal conductivity as a function of dot volume fraction. For each dot volume fraction δ the results are shown for several dots sizes (10.0, 8.0, 4.0, 2.0 nm A,B,C,D respectively). The B curve corresponds to the lattice thermal conductivity of $Si_{1-\delta}Ge_{\delta}$ alloy. Note a significant decrease of the QDS thermal conductivity as the dot radius decreases.

this result cannot be extrapolated for even larger fractions of quantum dots, since our formalism is based on the assumption that scattering from each dot is an independent process. The latter holds only for relatively small fractions of quantum dots ($\delta < 0.3$).

After the modified phonon group velocity has been found, we calculated combined phonon relaxation rate [Eq. (2)] and the lattice thermal conductivity κ [Eq. (1)]. Figure 2 shows the in-plane lattice thermal conductivity of QDS as a function of Ge dot volume fraction. Since the same volume fraction δ can be filled by dots of different sizes, we have calculated dependence of κ on δ for different dot sizes. The difference in κ values is caused by variation of scattering cross sections for dots of different sizes. As the dot size decreases, the scattering cross section attains maximum at the lower values of the phonon wave vector, so that smaller dots scatter phonons better than big ones. In the limiting case of $ka \ll 1$, the phonon relaxation on quantum dots is approaching the regular mass-difference scattering (curve E). For maximizing *ZT*, it is better not to approach this limit since it would also significantly deteriorate electron transport. In Fig. 3 we present the lattice thermal conductivity as a function of temperature for Si/Ge QDS in comparison to bulk Si. The QDS results are shown for $\delta = 0.05$ and $a = 4.0$ nm. It is important to note that even at this small volume fraction and relatively large dot radius, the decrease of the thermal conductivity of QDS is significant ($\sim$30% of its bulk value).[15] In Fig. 4 we present the evaluation of electron transport coefficients (electron conductivity, Seebeck coefficient, and thermopower (εS^2) of SiGe QDS as the functions of quantum dot volume fraction obtained by the model developed in Ref. 16. The transport coefficients at fixed dot volume fraction (subscribed by *D*) are normalized by their value obtained in the absence of dots. As it is shown in Fig. 4 there is a nonsignificant change in thermopower at small quantum dot volume fractions (dot's radius 4.0 nm).

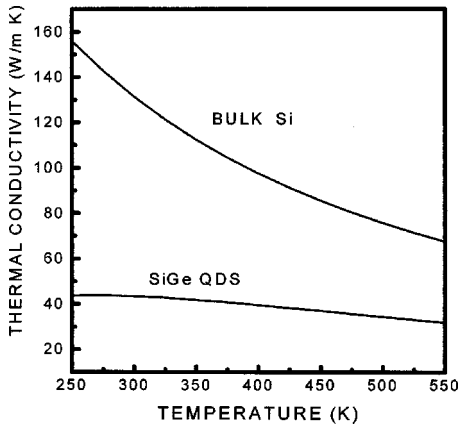

FIG. 3. In-plane lattice thermal conductivity as a function of temperature for the Si/Ge QDS for $\delta = 0.05$ and $a = 4.0$ nm and bulk Si. The bulk Si result is after Balandin and Wang (Ref. 15). The decrease is a result of the additional phonon scattering on quantum dots.

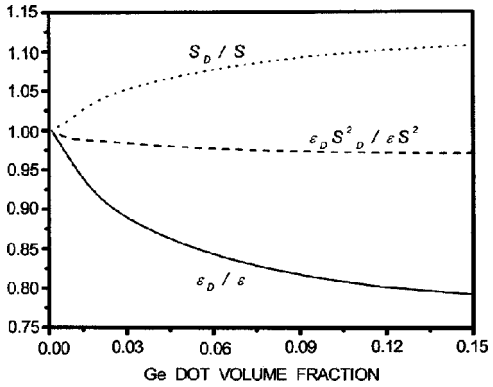

FIG. 4. Electron transport coefficients: electron conductivity (solid line), Seebeck coefficient (dotted line), and thermopower εS^2 (dashed line) as the functions of Ge dot volume fraction. All coefficients are normalized on their value in the absence of dots. Dot size 4.0 nm, $T = 300$ K.

IV. CONCLUSION

In conclusion, we have theoretically investigated the in-plane lattice thermal conductivity κ of a QDS. The developed model is applicable while phonon transport inside a quantum dot can be described in a continuum approximation. In this article we treated quantum dots as equal semispheres. In a more realistic case, the whole ensemble of quantum dots may be described by a set of fractions for dots of different shape and size. It was found that κ of the SiGe QDS decreases by several times relative to the κ of its constituent materials. The decrease is caused by the scattering of acoustic phonons on quantum dots. The strength of this effect depends on the volume fraction occupied by quantum dots and their characteristic size. The decrease of κ remains significant in a wide temperature range. The obtained results are important for high-temperature thermoelectric applications of the quantum-dot superlattices.

ACKNOWLEDGMENT

This work was supported by the DoD MURI-ONR program on Thermoelectrics (Dr. John Pazik).

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