Atomistic Calculations of Heat Transport in a Silicon Crystal*

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We have performed atomistic calculations of phonon transport properties. The dynamical matrix is obtained from ab initio calculations, and phonon transport properties are analyzed by the nonequilibrium Green's function method. As an example of the applications of the method, we present the temperature dependence of heat transport properties and the phonon lifetime due to phonon-phonon scatterings by the anharmonic terms of interatomic forces for a silicon crystal. [DOI: 10.1380/ejsnt.2014.154]

Keywords: Density functional calculations; Green's function methods; Phonons; Silicon

I. INTRODUCTION

Heat transport of nanoscale systems has attracted considerable attention for applications in nanometer-scale electron devices and thermoelectric devices. Heat transport properties are sensitive to atomic-scale crystal structures, and thus, the development of theoretical approaches from atomistic levels is indispensable. In semiconductor and insulator materials, heat energy is transported primarily through lattice vibrations, i.e., phonons, and phonon thermal transport has been studied by various theoretical methods such as molecular dynamics [1–5],

Boltzmann equation method [6, 7], and nonequilibrium Green's function method [8–12].

In this paper, we report on atomistic calculations of phonon transport properties using ab initio calculations and the nonequilibrium Green's function method. The dynamical matrix is calculated on the basis of the density functional theory, and the transmission function is obtained using the Green's function, which is useful for the analysis of atomic-scale structures and the inclusion of quantum effects. Furthermore, phonon lifetime due to the phonon-phonon scatterings by the anharmonic terms of interatomic forces is presented and discussed.

II. METHOD

We take the Hamiltonian H of the system for phonon transport as

$$H = \sum_{\substack{i \in sys \\ \alpha=x,y,z}} \frac{1}{2M_i} p_{i\alpha}^2(t) + \frac{1}{2} \sum_{\substack{i,j \in sys \\ \alpha,\beta=x,y,z}} u_{i\alpha}(t) K_{i\alpha,j\beta} u_{j\beta}(t). \quad (1)$$

M_i is the mass of atom i and $u_{i\alpha}(t)$ is an operator for the displacement of atom i along the α direction from the equilibrium position. The total Hamiltonian is split into four parts: $H = H_L + H_S + H_R + H_{int}$, where $H_{L(R)}$ is the Hamiltonian for the left (right) lead, H_S is for the scattering region, and H_{int} is for the interaction between the scattering region and the left (right) lead. The thermal current J_{th} and thermal conductance $G(T)$ from the left lead to the scattering region are expressed as

$$\begin{aligned} J_{th} &= -\langle \dot{H}_L \rangle \\ &= \int_0^\infty \frac{d\omega}{2\pi} \hbar \omega [n(\omega, T_L) - n(\omega, T_R)] \zeta(\omega) \\ G(T) &= \frac{k_B}{2\pi} \int_0^\infty d\omega \frac{\hbar \omega}{k_B} \frac{\partial n}{\partial T} \zeta(\omega). \end{aligned} \quad (2)$$

Here, the bracket $\langle \cdots \rangle$ denotes the nonequilibrium statistical average of the physical observable, and $n(\omega, T_{L(R)})$

FIG. 1: Phonon dispersion of silicon crystal calculated using dynamical matrix from ab initio calculations.

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FIG. 2: Transmission function for silicon crystal.

FIG. 3: Temperature dependence of heat conductance of silicon crystal.

is the Bose-Einstein distribution function of equilibrium phonons with an energy of $\hbar\omega$ in the left (right) lead at temperature T . The transmission function $\zeta(\omega)$ is expressed using the retarded (advanced) Green's function $G^{r(a)}(\omega)$ and the coupling constant $\Gamma_{L(R)}(\omega)$ as

$$\zeta(\omega) = \text{Tr}[\Gamma_L(\omega)G^r(\omega)\Gamma_R(\omega)G^a(\omega)]. \quad (3)$$

The retarded (advanced) Green's function for the scattering region is calculated as

$$G^{r(a)}(\omega) = [\omega^2 M - K - \Sigma_L^{r(a)} - \Sigma_R^{r(a)}]^{-1}, \quad (4)$$

where M is the diagonal matrix whose element is the mass of an atom and $\Sigma_{L(R)}^{r(a)}(\omega)$ is the retarded (advanced) self-energy due to the coupling to the left (right) semi-infinite lead with the scattering region. The dynamical matrix K is constructed from ab initio calculations [13]. To confirm the validity of the present method, we show in Fig. 1 the phonon dispersion relations calculated using the dynamical matrix K , which is in good agreement with the experimental data [14].

III. RESULTS

Figures 2 and 3 show the transmission function integrated in the Brillouin zone and the temperature depen-

FIG. 4: Temperature dependence of lifetime of TA phonon at X point.

dence of the heat conductance of a silicon crystal, respectively, where the heat conductance per face of a unit cell for the fcc lattice in the [100] direction is shown. At low-energy regimes, a small number of excited phonons contribute to the heat transport, and the transmission function and conductance are thus small. As temperature increases, various phonon modes begin to contribute to the transport and the conductance increases. Since the phonon energy has an upper limit, as shown in Fig. 1, the transmission function becomes zero for high energy regimes and the conductance saturates at a high temperature.

On the other hand, as temperature increases, phonon-phonon scatterings become enhanced and the conductance should show a decrease at the high-temperature regime. For bulk silicon, the scattering effect is seen above 30 K [15]. To discuss this effect, we investigate the phonon-phonon interaction. Note that, in the above calculations, phonons are treated as noninteracting with an infinite lifetime in the harmonic approximation. Here, we deal with the anharmonic terms of the interatomic force to analyze the phonon-phonon scatterings.

Force F_i acting on an atom i is expanded using the displacement x_j of an atom j as [16]

$$F_i = \sum_j \Psi_{i,j} \cdot x_j - \frac{1}{2} \sum_{j,k} \Phi_{i,j,k} \cdot x_j x_k. \quad (5)$$

We obtained the third term of the potential $\Phi_{i,j,k}$ using the Lennard-Jones potential for simplicity in this work. It is possible to calculate the third and higher terms using ab initio calculations in the same manner. The phonon lifetime τ_{qs} is calculated as [17]

$$\frac{1}{\tau_{qs}} = \pi \sum_{q's'} \sum_{q''s''} |V_3(qs, q's', q''s'')|^2 \quad (6)$$

$$[(n_{q's'} + n_{q''s''} + 1)\delta(\omega(qs) - \omega(q's') - \omega(q''s'')) + 2(n_{q's'} - n_{q''s''})\delta(\omega(qs) + \omega(q's') - \omega(q''s''))],$$

where q is a phonon wave vector and s is a phonon branch.

Figure 4 shows the temperature dependence of phonon lifetime for the transverse acoustic phonon mode. It is calculated using the primitive cell for a periodic system. With increasing temperature, the phonon lifetime becomes short, which causes the decrease in thermal conductance at a high temperature. Note that the calculation

of the phonon lifetime enables us to analyze the transport properties using the nonequilibrium Green's function, including the effects of phonon-phonon scatterings. The self-energy for phonon-phonon scatterings is obtained using the Kramers-Kronig relation with coupling constant, and then the conductance is analyzed, including the anharmonic effects.

IV. SUMMARY

In summary, we reported the calculations of heat transport using *ab initio* calculations and the Green's function method and phonon lifetime including anharmonic effects from atomistic viewpoints. As an example of the method,

we show the temperature dependences of the conductance and phonon lifetime for a bulk silicon crystal. The present theoretical approach is useful for the analysis of not only heat transport but also thermoelectric properties by the combination with electron transport calculations.

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