Phonon Scattering in Doped Silicon by Molecular Dynamics Simulation

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ABSTRACT

Molecular dynamics simulation is used to study the scattering of phonon wave packets of well–defined frequency from a field of point defects in silicon crystal. For simplicity, and to connect as closely as possible with classic theories, the defects differ from the Si only in their mass. The dopant atoms are randomly distributed in the middle region of simulation system along z direction. The phonon wave packet propagation and scattering in the doped region are explicitly determined at the atomic level. We quantitatively analyze the relative amounts of energies in the transmitted and reflected wave packets for different vibrational modes, and determine the main parameters which influence the phonon scattering process. The results show that both the amount of phonon scattering and the modes into which the incident phonons are scattered are strongly dependent on the density of dopant atoms, as pointed out by Klemens's theory. This work sheds light on the effects of point defects on the thermal conductivity of semiconductors.

Keywords: phonon scattering, point defects, molecular dynamics simulation

1 INTRODUCTION

It is well known that phonon-phonon scattering plays a dominant role on thermal transport in electric insulators such as silicon and many oxides. However, when the mean free path associated with phonon-phonon scattering is larger than the dimensions of structure, phonon scattering from surfaces, interfaces, and other structural imperfections can dominate, and phonon-phonon interactions can become negligible[1]. A second regime in which phonon-phonon scattering is less important is when the system is intentionally highly doped; this paper focuses on the phonon scattering from point defects, which is related to the heat dissipation in doped materials

More than 50 years ago, Klemens determined scattering rate of phonons for point defects[2]. The strength of scattering was found to scale linearly with the dopant concentration and the square of the mass difference between the dopant and the host atoms[3]. By comparing thermal conductivity measurements to Klemens's predictions, Cahill and coworkers have recently found experimentally that this classic result, though derived under the assumption of weak scattering is actually well obeyed for Ge-doped Si[4]. However, they suggested that a more sophisticated model is required to develop a predictive theory of the thermal conductivity of semiconductor alloys with better accuracy[4]. These results are our motivation for performing molecular dynamics simulation to develop a molecular view of phonon scattering from point defects.

2 SIMULATION CONDITIONS

As a model system, we choose diamond-structured silicon described by the Stillinger-Weber potential[5,6]. As a simplified model that is expected to capture the basic physics of point defects, we add dopant atoms to substitute for the Si atoms. The dopants only differ from Si in their mass with $M_{dop}=4M_{Si}$ = 112 amu; although this is somewhat larger than the atomic weight of Ge (72.59amu) this mass ratio was chosen both because it was used in our earlier studies of semiconductor superlattices, and because we expect it to increase the effects of dopants, making them easier to observe. In addition to the mass difference between the dopant and host atoms, other parameters such as the force constant and lattice deformation around dopants can also influence the phonon scattering process. This was recently confirmed by Stachowiak et al.[7] by measuring the thermal conductivity of carbon monoxide-doped nitrogen crystals, a system chosen because both the nitrogen molecule and carbon monoxide have the same molecular mass (28 amu). However, the effect is weaker than that caused by the mass difference.

The simulation cell is shown schematically in Fig. 1. The Si crystal is oriented such that the x, y, and z axes correspond to the (100), (010), and (001) directions in the diamond lattice. The unit cell is 6000 unit cells long (L_z =6000, i.e., 3.258 μm for the Si lattice parameter a_0=0.543 nm). The simulation cell is only 2×2 unit cells in the transverse direction. The dopant atoms are randomly

distributed in the doped region from z_1= -100 to z_2 = 100 unit cells. This region contains a total of 6400 atoms. Four different dopant densities were chosen: 0.08% (5 atoms), 0.39% (25 atoms), 0.78% (50 atoms) and 1.56% (100 atoms)

As we will see, our simulation results show that the phonon scattering process is strongly influenced by the density of dopant atoms. In this paper we concentrate on explicitly showing the detailed view of propagation and scattering of phonon wave packet in the doped region, and quantitatively analyzing the energy transmitted, reflected, and left in the doped region.

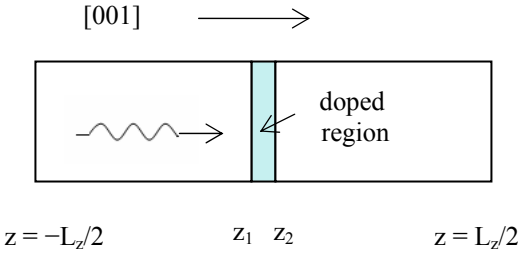

Fig. 1. Schematic of the simulation cell.
L_z=6000 unit cells (3.268μm); z_1= -100, z_2=100

3 SIMULATION PROCEDURE

There are four steps in the simulation procedure: 1) structure creation, 2) initial phonon wave packet generation, 3) MD simulation, and 4) energy analysis.

We have generated simulation cells with the four different dopant concentrations, in which the wave packet will propagate and scatter. Since acoustic phonons are believed to be the main contributors to phonon transport due to their large group velocity[8], in this study we construct phonon wavepackets from longitudinal acoustic (LA) phonons within a narrow frequency range of 2.96THz. Using MD simulation, this wave packet is allowed to propagate towards the doped region (Fig. 1), in which it scatters from the dopants. As a result some of the incident energy is transmitted and some is reflected. The MD simulations give the time-dependence of the displacements of all the atoms, both in the perfect Si regions and in the scattering region (doped region). These data are used to quantitatively analyze the energy transmission and reflection coefficients from the doped region, and to calculate how much of the energy is still left in the doped region at any time. In our energy analysis, we characterize the frequency and branch (LA vs. TA) of the transmitted and reflected wavepackets.

4 RESULTS AND DISCUSSION

4.1 Phonon Wave-packet Propagation and Scattering

Figure 2 shows four snapshots of the atomic displacements for an incident LA wave packet with a frequency of 2.96THz, for the case of 1.56% doping (i.e., 100 atoms in the doped region). The LA wavepacket is launched from z=-250 *a* towards the doped region (Fig. 2, t=0). When it enters the front edge of the doped region (z= -100), the shape of wave changes as a result of scattering (Fig. 2, t=26.3ps). It scatters repeatedly in the doped region, resulting in some of the energy being reflected, and the remainder ultimately being transmitted through the region forwards into the perfect Si structure on the other side of the doped region (Fig.2, t=60.1ps). With increasing time, more and more of the energy emerges from the doped region (either reflected or transmitted). At the end of the simulation only an inconsequential amount remains. (Fig. 2, t=201.3ps)

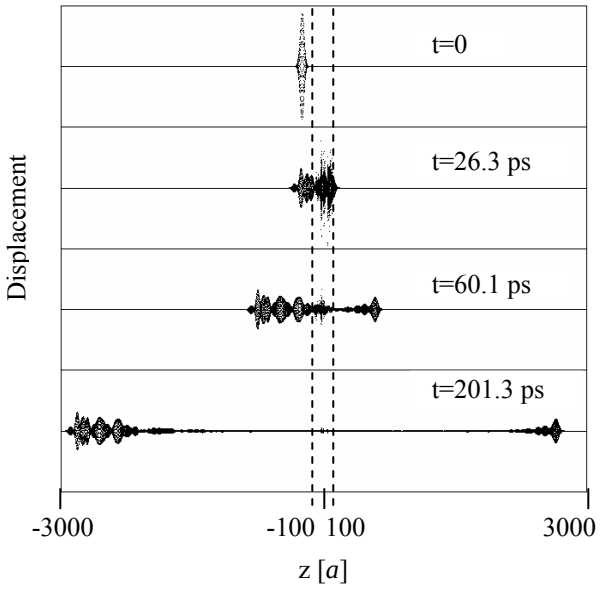

Fig. 2. Snapshots of displacements in the z direction (i.e., the LA component) for scattering and propagation of an incident LA wavepacket with frequency of 2.96THz. The doped region is at -100<z<100, the dopants are randomly distributed at a density of 1.56 atom %.

4.2 Energy Analysis

For the purposes of analysis, we characterize the vibrational energy into three contributions. The fraction of transmitted, reflected energy are defined as the energies in the regions

$z > 100\ a$ and $z < -100\ a$ respectively, with 'trapped energy' being energy in the doped region ($-100\ a < z < 100\ a$). While these three contributions vary individually, their sum is constant over the simulation. In Fig. 3 we see that although the incident wavepacket contained only LA phonons, both LA and TA modes are obtained in scattering. The amount of energy trapped in the doped region decreases with time, reaching almost zero at t=200 ps, at which time the leading edge of the transmitted wave packet approaches to z =3000, as shown in Fig 2. The fraction of the incident LA phonon energy that is scattered from LA to TA in both transmission and reflection direction depends on the density of dopants. Figure 4 explicitly shows the transmitted and reflected LA and TA phonon respectively at t=201.3 ps.

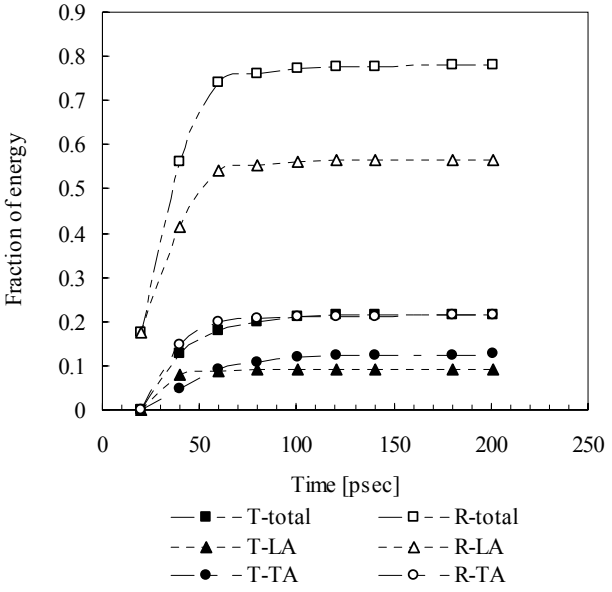

Fig. 3 Time evolution of the fraction of incident phonon energy scattering from the doped region with 1.56% dopants. T is the fraction of energy transmitted (solid symbols), and R is the fraction of energy reflected (open symbols). LA denotes the longitudinal acoustic branch, TA denotes the transverse acoustic branch. T-total is the sum of T-LA and T-TA. R-total is the sum of R-LA and R-TA

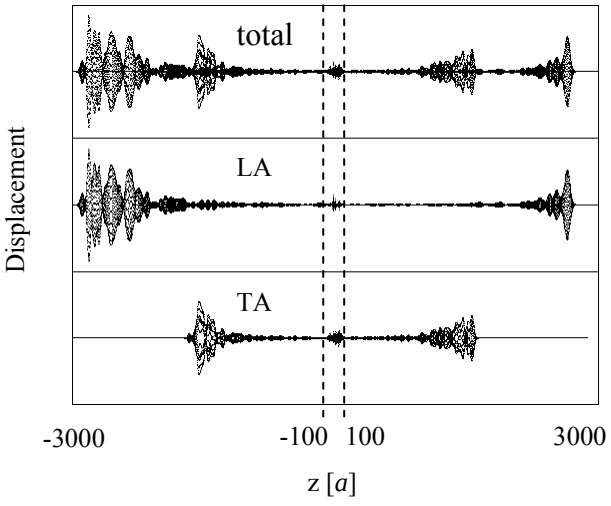

Fig. 4. Snapshot of transmitted and reflected wave packet with LA and TA mode separated for t =201.3 ps in Fig. 2.

4.3 The influence of the dopant density on phonon scattering

The phonon scattering is strongly influenced by the density of doped atoms. Figure 5 shows that the reflection coefficient increases approximately linearly with the concentration of dopants. While there is scattering from the LA to TA modes at a dopant density of 1.56%, for dopant densities below 0.78%, there is no LA→TA scattering.

To characterize the amount of time it takes for the entire scattering process to take place, we have monitored the time dependence of the fraction of energy trapped in the doped region. As one would expect, the entire scattering process takes longer as the dopant density increases. It is evident from Fig. 6 that the higher the concentration of dopants, the longer it takes for all of the energy to emerge from the dopant region: a physically very reasonable result.

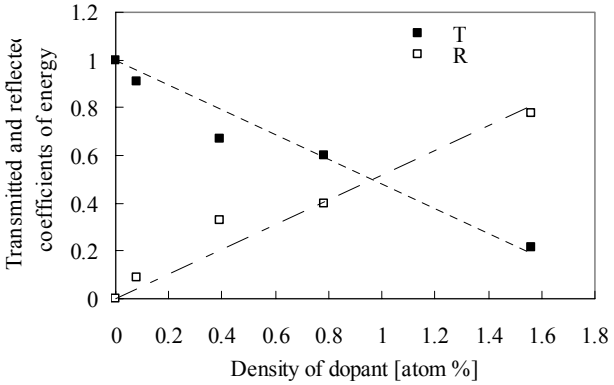

Fig. 5. The effect of dopant density on the transmitted and reflected coefficients of energy from the doped region. The frequency of the incident LA wave packet is 2.96THz. T and R are calculated at the end of the simulation, at which time the trapped energy in the doped region is negligible.

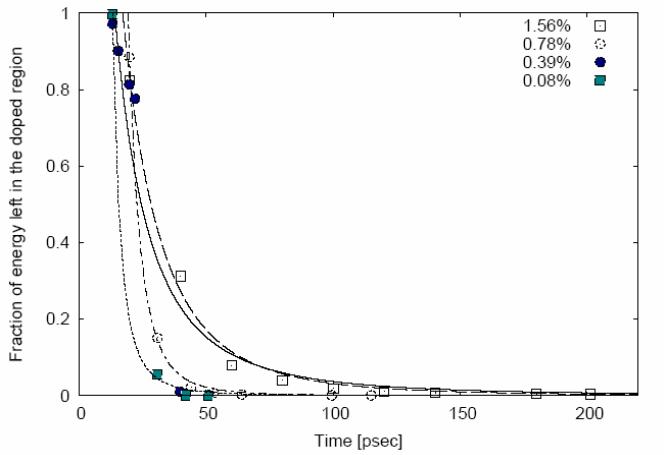

Fig. 6. The fraction of energy trapped in the doped region vs. time as a function of the dopant concentration.

5 CONCLUDING REMARKS

We have shown here that phonon wave-packet dynamics can be used to give atomic-level information on the scattering of phonons from point defect. Showing results for a single phonon LA wave packet with a narrow range of frequencies, we have seen an approximately linear dependence on the dopant density, both in the energy transmission and reflection coefficients, and in the amount of mode mixing. Ongoing and future work in this area will explicitly map put the dependence on frequency and on phonon branch - optical vs. acoustic, longitudinal vs. transverse. Moreover, armed with such an extensive database, we will be able to compare with the classic results of Klemens.[2] Finally, we will extend our work from isotopic defects to defects in which the atomic radius and strength of interatomic bonding are modified.

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