Microsegregation Effects on the Thermal Conductivity of Silicon-Germanium Alloys

by Yongjin Lee

Due to increased need for renewable energy sources in recent years, a significant number of both experimental and theoretical efforts have been undertaken to find effective ways to enhance the performance of thermoelectric (TE) energy conversion. Since the 1960s, SiGe alloys have received much attention as one of the promising candidates for TE materials, due to their low thermal conductivity (κ), as compared to pure Si and Ge. Earlier studies[1,2] demonstrated that the low κ is mainly attributed to phonon scattering as a result of the mass difference between Si and Ge atoms (the so-called alloy scattering). While the strength of alloy scattering is a strong function of the Si/Ge ratio, previous experiments[3] also showed evidence that Si and Ge atoms often remain locally segregated in bulk SiGe samples prepared by mechanical alloying. However, no research has been reported regarding the microsegregation effect on κ. As a part of the ECS summer project, we performed a computational analysis to explore how the local atomic arrangement affects thermal transport in bulk SiGe; some results of this work are presented herein.

To investigate the microsegregation effect, we prepared several $Si_{0.8}Ge_{0.2}$ samples by embedding spherical Ge particles of different sizes (ranging from 5 to 293 atoms) in the Si matrix. As illustrated in Fig. 1, embedded Ge particles were randomly positioned but not allowed to overlap each other. A nonequilibrium MD (NEMD) method[4] with the Stillinger-Weber (SW) potential model[5] was used to calculate the κ of SiGe alloys at 300 K, while the SW parameters were modified using the first-principles-based force-matching method.[6] For each of the $Si_{1-x}Ge_x$ systems considered, five independent NEMD simulations were performed with different atomic arrangements and initial velocity distributions. All NEMD simulations were performed using LAMMPS (Large-Scale Atomic and Molecular Massively Parallel Simulator)[7] with a time step of 1 *fs*; a detailed description of the simulation steps can be found in Ref. 8 and 9.

Figure 2 shows the variation of κ for the $Si_{0.8}Ge_{0.2}$ samples as a function of Ge particle diameter (D_e); here, D_e is approximated by $(6N_{Ge}V_{Ge}/\pi)^{1/3}$, where N_{Ge} is the number of Ge atoms in the particle and V_{Ge} is the volume per atom for Ge (= 0.0238 nm^3 from our DFT-GGA calculation). The κ is predicted to monotonically increase with

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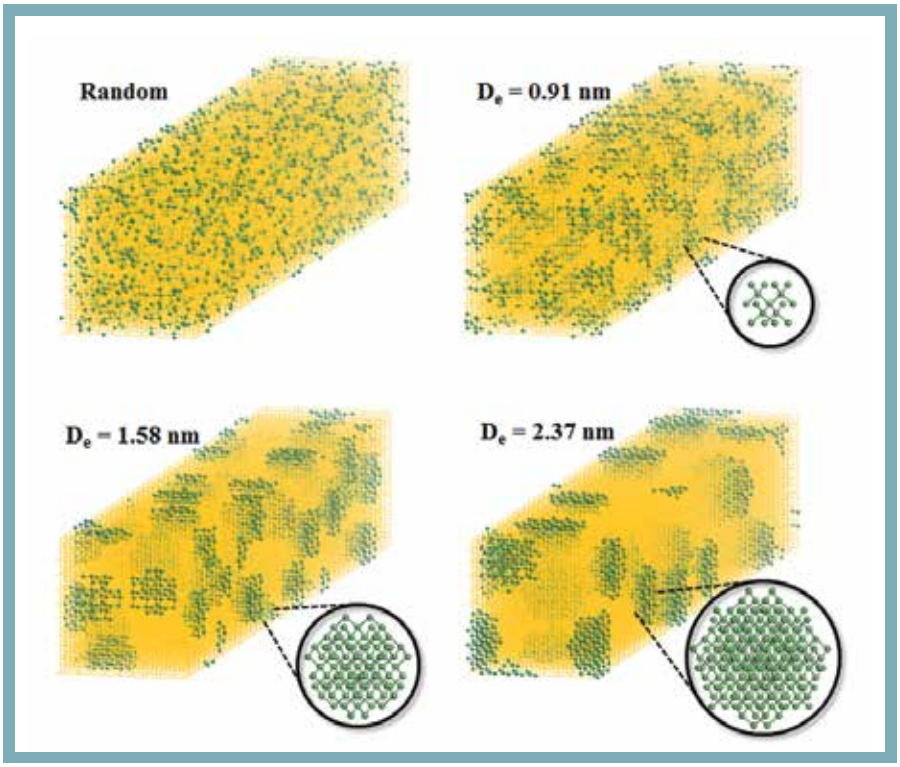

FIG 1. *The various $Si_{0.8}Ge_{0.2}$ configurations show a random distribution of Si and Ge atoms (random) and embedded Ge particles with different diameters (D_e = 0.91, 1.58, and 2.37 nm) in the Si matrix. Green (black) balls and yellow lattices represent Ge and Si atoms, respectively.*

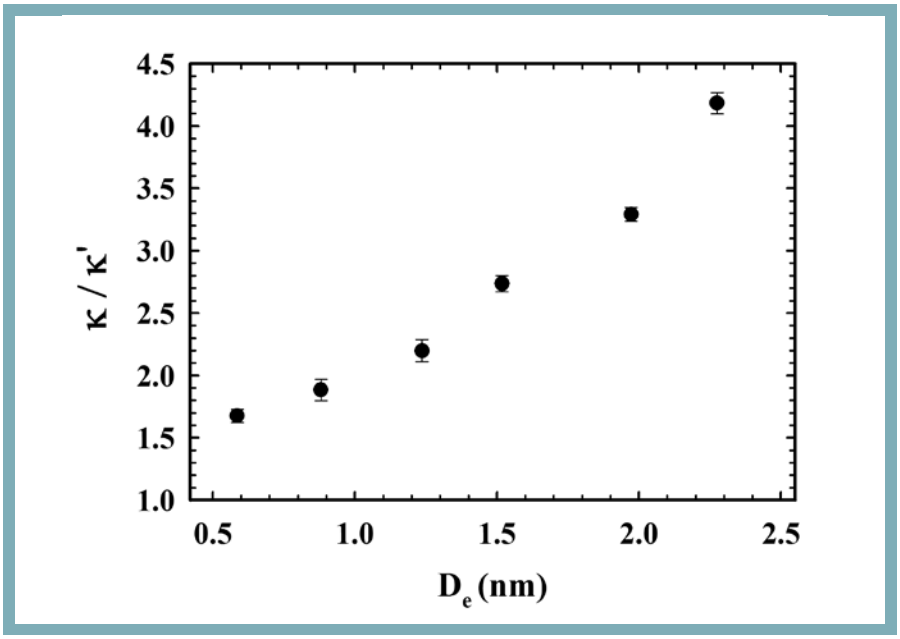

FIG 2. *Predicted variation of the relative thermal conductivity with respect to the random alloy (κ/κ′) as a function of the diameter (D_e) of Ge particles embedded in $Si_{0.8}Ge_{0.2}$ (see Fig. 1); note that the predicted κ′ for the randomly distributed $Si_{0.8}Ge_{0.2}$ sample is about 1.25 $Wm^{-1}K^{-1}$.*

D_e; note that κ = 4.18 $Wm^{-1}K^{-1}$ at D_e = 2.37 nm is about 3.3 times greater than κ = 1.25 $Wm^{-1}K^{-1}$ for the random alloy. Given that mass disorder is mainly responsible for the reduction of κ in the SiGe alloy, single and paired Ge atoms may act mainly as scattering centers when they are atomically dispersed. On the other hand, when Ge atoms remain locally segregated, scattering by the mass difference would occur at Ge particle-Si matrix interfaces. Therefore, such Ge segregation will reduce the number of scattering centers, thereby increasing phonon transport, compared to when Ge atoms are homogeneously distributed in the $Si_{0.8}Ge_{0.2}$ matrix.

Our results clearly highlight that the local segregation (microsegregation) of alloying elements, along with composition, can be a critical factor in determining the κ of alloys. The increase of κ with microsegregation suggests that the minimum κ would be achieved when Si and Ge atoms are randomly distributed.

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