Impact of Impurity Mass on Ballistic Phonon Thermal Transport in Silicon Nanowires

Junichi Hattori[1,3] and Shigeyasu Uno[2,3]

[1]The Research Organization of Science and Technology, Ritsumeikan University, Kusatsu, Shiga, 525-8577, Japan
[2]Department of Electrical and Electronic Engineering, Ritsumeikan University, Kusatsu, Shiga, 525-8577, Japan
[3]JST, CREST, Chiyoda-ku, Tokyo 102-0075, Japan
E-mail: jhattori@fc.ritsumei.ac.jp

1. Introduction

Silicon nanowires (Si NWs) have been attracting a lot of interests as thermoelectric (TE) materials, which directly convert temperature gradients to electric fields and vice versa. The conversion efficiency is measured by the figure of merit, ZT. Si NWs have shown a relatively high ZT of about 0.5 at room temperature [1, 2]. Practical TE applications, however, require a higher ZT of at least unity. One way to improve ZT is impurity doping. Impurities behave as scattering sources for phonons and reduce the phonon thermal conductivity κ_p [3]. This reduction leads to an improvement in ZT, because $ZT \propto (\kappa_\mathrm{p} + \kappa_\mathrm{e})^{-1}$, where κ_e is the electron thermal conductivity. Recently, Lee and Hwang have closely examined κ_p in doped bulk Si using molecular dynamics simulations and confirmed its reduction due to doping [4]. They have also revealed that, in bulk Si doped with B or As, the doping-induced disorder of spatial distribution of atomic mass has a much larger contribution to the reduction in κ_p than the change of interatomic potential shape and the lattice strain. However, the mechanism through which the three phenomena directly caused by doping decrease κ_p has not yet been made clear. In this work, we study the doping impact on phonon thermal transport in Si NWs, especially the impact of impurity mass.

2. Dispersion Relations

We considered [001]-oriented ^{28}Si NWs with the unit cell shown in Fig. 1(a), and calculated the phonon dispersion relations in the NWs using the lattice dynamics method [5]. Figure 2 shows the results obtained for a pure and an ^{75}As-doped ^{28}Si NW. In our study, doped NWs differ from undoped ones only in the mass of the atoms substituted by impurity atoms. We assumed impurities not to change the interatomic potential shape and not to cause the lattice strain.

3. Ballistic Thermal Conductance

Phonon thermal conductance is given by

$$K = \sum_n \int_{-a_z/\pi}^{a_z/\pi} \hbar\omega_n v_n(q_z) \frac{\partial f(\omega_n)}{\partial T} \mathcal{T}_n(q_z) H(v_n(q_z)) \frac{\mathrm{d}q_z}{2\pi}, \quad (1)$$

where q_z is the phonon wavevector along the z-axis, ω_n the frequency in the nth dispersion branch, v_n the group velocity, f the Bose–Einstein distribution, $\mathcal{T}_n$ the transmission, and H the Heaviside function [6]. Figure 3 shows K in the ballistic limit ($\mathcal{T}_n(q_z) \to 1$), K_bal, in ^{28}Si NWs randomly doped with ^{11}B, ^{27}Al, ^{31}P, or ^{75}As [see Fig. 1(c)]. Regardless of dopant type, doping decreases K_bal. As can be seen in Fig. 2, on the whole, phonons in the ^{75}As-doped NW have a smaller ω_n than those in the undoped NW. This is attributed to the tendency, $\omega_n \propto \langle m \rangle^{-1/2}$, where $\langle m \rangle$ is the average atomic mass. If a dispersion relation scales down (up) in terms of ω_n, its constituent branches become flat (steep). Note also that some degenerate modes are split in the doped NW. This is because impurities break the orderliness of atomic mass distribution. The mode splitting (MS) flattens dispersion branches more strongly than the change of $\langle m \rangle$, as shown in Fig. 4. The figure shows the change of v_n in ^{75}As-doped ^{28}Si NWs as a function of the doping concentration. The flattening of dispersion branches means a reduction in v_n, which decreases K_bal through (1).

The MS effect on K_bal in a ^{28}Si NW with a unit cell having one impurity atom at the center [see Fig. 1(d)] is shown in Fig. 5 as a function of the impurity mass. Regardless of whether the impurity atom is heavier or lighter than ^{28}Si, the MS effect increases with increasing mass difference between the two atoms. The MS effect is generally evaluated as the mass-difference scattering rate [3], which is proportional to $\Gamma = \sum_i n_i (1 - m_i/\langle m \rangle)^2$. Here, n_i and m_i are the relative concentration and the mass of ith nuclide, respectively. Figure 6 shows the MS effect as a function of Γ. In terms of the scattering rate, even though NWs are doped with various impurities at various concentrations, if they have the same Γ, they are expected to experience the same MS effect. However, the MS effect in NWs heavily doped with impurities having a small mass difference is larger than that in NWs lightly doped with impurities having a large mass difference.

4. Conclusions

The impact of impurity mass on phonon thermal transport in doped Si NWs was studied in detail with atomistically calculated dispersion relations. The mass disorder induced by impurities reduces the phonon group velocity and thereby thermal transport properties.

978-3-901578-26-7

[1] A. I. Boukai *et al.*, *Silicon nanowires as efficient thermoelectric materials*, Nature **451**, 168 (2008).
[2] A. I. Hochbaum *et al.*, *Enhanced thermoelectric performance of rough silicon nanowires*, Nature **451**, 163 (2008).
[3] M. Asheghi *et al.*, *Thermal conduction in doped single-crystal silicon films*, J. Appl. Phys. **91**, 5079 (2002).
[4] Y. Lee and G. S. Hwang, *Mechanism of thermal conductivity suppression in doped silicon studied with nonequilibrium molecular dynamics* Phys. Rev. B **86**, 075202 (2012).
[5] A. Paul *et al.*, *Modified valence force field approach for phonon dispersion: from zinc-blende bulk to nanowires*, J. Comput. Electron. **9**, 160 (2010).
[6] J. Hattori and S. Uno, *Impact of Isotope Doping on Phonon Thermal Transport in Silicon Nanowires*, Jpn. J. Appl. Phys. (to be published).

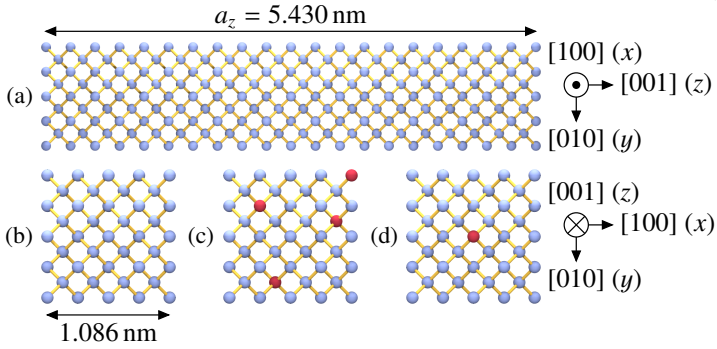

Fig. 1. (a) Side view of a unit cell of the Si NWs considered in this work. The unit cell is repeated periodically along the [001] direction in the NWs. Also, it contains 410 atoms. (b)–(d) Top views of cross sections of the NWs without and with impurities, which are shown by the dark red atoms.

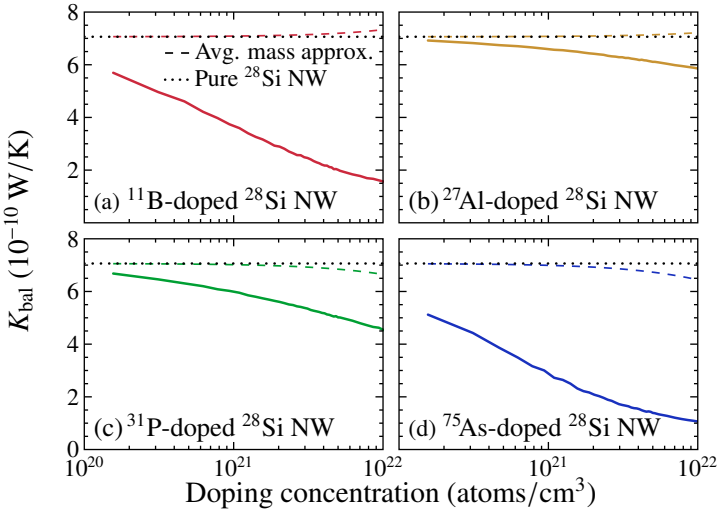

Fig. 3. Ballistic thermal conductance K_{bal} in ^{28}Si NWs randomly doped with (a) ^{11}B, (b) ^{27}Al, (c) ^{31}P, or (d) ^{75}As at 300 K, plotted as a function of the doping concentration N_i. The results for each N_i were averaged over 15 samples to reduce the fluctuation. The dotted lines represent K_{bal} in a pure ^{28}Si NW. Also, the dashed lines show K_{bal} obtained under the assumption that the masses of all the atoms in a NW are equal to their average.

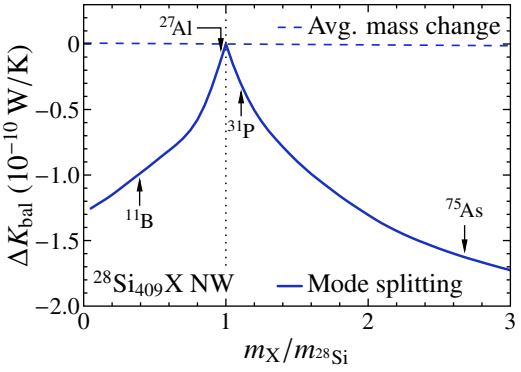

Fig. 5. Variations of K_{bal} due to the mode splitting (solid line) and the change of average atomic mass (dashed line). The calculation was done for ^{28}Si NWs having one impurity atom per unit cell. Each impurity atom was located at the center of each unit cell, as shown in Fig. 1(d). The horizontal axis represents the atomic mass of the impurity divided by that of ^{28}Si.

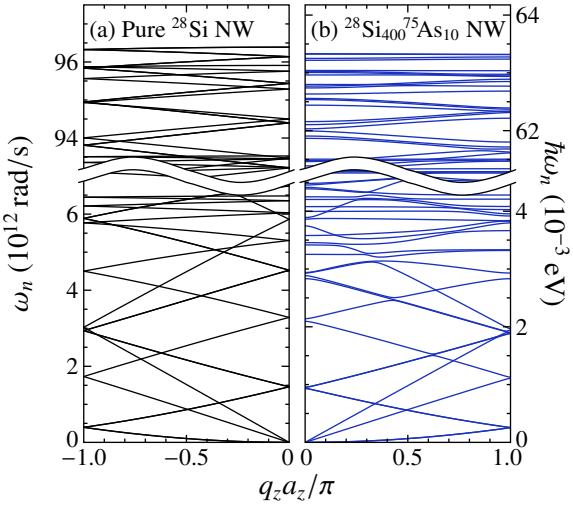

Fig. 2. Dispersion relations of phonons in (a) a pure ^{28}Si NW and (b) a ^{28}Si$_{400}$^{75}As$_{10}$ NW. The horizontal axes represent the phonon wavevector along the z-axis, q_z, and the vertical axes the phonon frequency, ω_n.

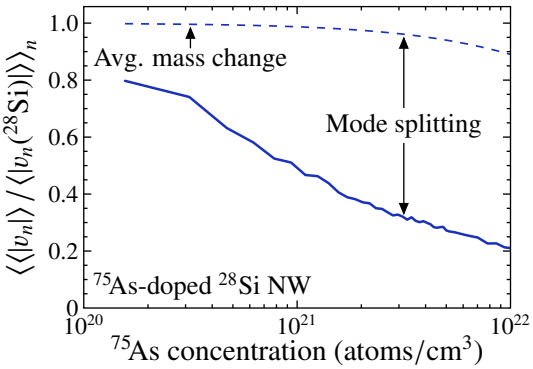

Fig. 4. Ratio of average phonon speed in a dispersion branch in an ^{75}As-doped ^{28}Si NW to that in the corresponding dispersion branch in a pure ^{28}Si NW, averaged over all the branches. The horizontal axis represents ^{75}As concentration.

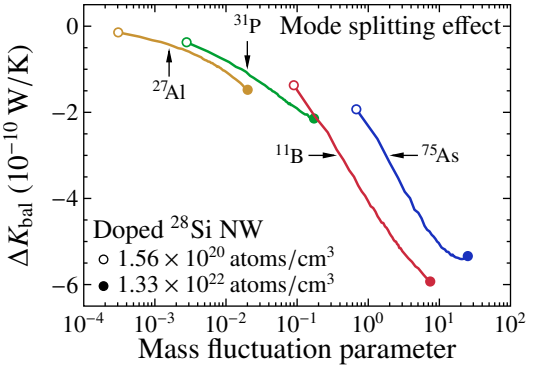

Fig. 6. Mode splitting effects on K_{bal} in ^{11}B-, ^{27}Al-, ^{31}P-, and ^{75}As-doped ^{28}Si NWs, plotted as a function of the mass fluctuation parameter Γ. In the calculation, the doping concentration was varied from 1.56 × 10^{20} (one) to 1.33 × 10^{22} atoms/cm^3 (85 atoms per unit cell).