Thermoelectric Properties of Nanoscale-Controlled Silicon

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Bulk Si exhibits good electrical properties. The power factor of heavily doped single-crystal Si is 4.1 $mWm^{-1}K^{-1}$ at room temperature, which is comparable to a typical thermoelectric material such as Bi_2Te_3 (for example, *p*-type BiSbTe has a power factor of 4.3 $mWm^{-1}K^{-1}$ and *ZT* of 1.2 at room temperature). Nevertheless, bulk Si has a high thermal conductivity (κ) (46 $Wm^{-1}K^{-1}$ at room temperature for heavily doped Si), giving *ZT* value of around 0.03 at room temperature.

Recent result for phonon transport in bulk silicon shows that 90% of the heat is transported by phonons with a mean free path longer than 50 nm. Thus, taking into consideration the fact that the carrier mean free path is only a few nanometers at room temperature in heavily doped Si, large reductions of the thermal conductivity can be expected to be achieved without much impact on the electron mobility by nanostructuring Si. Indeed, in recent studies several groups have reported *ZT* enhancement in bulk Si assembled from nanocrystals (NCs). For example, Bux and coworkers have obtained Si NC powder by ball-milling.[1] The lattice thermal conductivity (κ_L) was reduced to 6.3 W/mK at room temperature, leading to a maximum *ZT* value of 0.7. To obtain additional reduction in κ_L, additional nanosize control of Si NCs without oxidation or the incorporation of impurities is necessary.

We investigated a new approach for reducing the grain size of Si NC. We synthesized semiconducting composite films of Si and Mo-silicide nanocrystals (NCs) by phase separation from amorphous Mo–Si alloy (Si:Mo = 12:1).[2] Transmission electron microscope images (Fig. 1) show that Si and Mo silicide NCs were grown to average diameters of 8 nm and 11 nm, respectively, by annealing at 800°C. The thermal conductivity was reduced to 2.2 W/mK by enhancement of phonon scattering at NC interfaces.

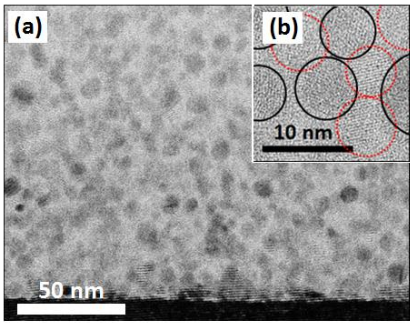

Fig. 1. (a) Cross-sectional bright-field TEM image of $MoSi_{12}$ film and (b) magnified view. The solid circles show the Mo-Si NCs and the dotted circles show the Si NCs.

We also investigate a new method for obtaining low thermal conductivity in bulk Si. In this method, which we call "HF-etching nanosize-controlling process for powder" (HNPP), self-limiting oxidation coupled with HF etching is applied to nanopowder Si. The application of HNPP to non-doped nanopowder Si reduces the average diameter from 58 nm 35 nm. The thermal conductivity is reduced from 25.7 $Wm^{-1}K^{-1}$ to 13.5 $Wm^{-1}K^{-1}$ at 300 K. Theoretical calculation including grain boundary transmission and frequency-dependent grain boundary scattering shows that these thermal conductivity reductions can be attributed to phonon scattering at grain boundaries.

In conclusion, we have synthesized semiconducting composite films of nanocrystalline Si and Mo-Si from by a phase separation technique. The films have thermal conductivity of 2.2 $Wm^{-1}K^{-1}$, which is substantially lower than that of Si NCs prepared by ball milling. Carrier doping into the film is being conducted. In order to achieve such a low thermal conductivity in bulk Si, we have investigated a new method to reduce the grain size using HF-etching.

References

1. S. K. Bux and R. G. Blair et al.: Adv. Mater. 19 (2009) 2445.
2. N. Uchida and T. Kanayama, Japanese Patent No. 2010-207987 (16 September 2010).