Phonon Scattering by Ultrafine Particulates in SiGe Alloys at High Temperatures

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1. Introduction and Theory

Silicon-Germanium (SiGe) alloys are used as thermoelectric materials in space power applications at high temperatures. The material conversion efficiency is unfortunately low so needs to be improved. The most promising approach to improving the figure of merit, Z, and hence the efficiency is to reduce the thermal conductivity of the SiGe alloy. A theoretical model has been developed that predicts that the thermal conductivity of these alloys can be reduced by adding appropriately sized particles (40-200Å in diameter) to act as scattering centers for those phonons that conduct most of the heat through the material [1, 2]. The mean-free-path of the high frequency phonons is reduced by point-defect (mass difference) scattering as a result of the use of a SiGe alloy as the basic material, whereas that of the low frequency phonons is reduced by a combination of grain boundary scattering and the interaction of phonons with electrons (or holes). This leaves unaffected a range of intermediate phonon frequencies to carry most of the heat. Figure 1 shows the calculated thermal conductivity as a function of inclusion concentration. A reduction of up to 40 percent is possible in the case of a 40Å diameter particulate that conducts no heat and has a concentration of about 7 volume percent.

To test this prediction, ultra-fine particles of BN and Si_3N_4 were made by eroding single crystal boron and silicon in a nitrogen atmosphere in an apparatus described elsewhere [3]. The particulates were introduced into ultra-fine p-type doped SiGe powder, uniformly dispersed, and hot pressed at 1525K into the alloy. Samples were heat treated at 1550K to grow the grains from submicron to greater than a micron in size.

2. Results and Discussion

The results of thermal conductivity measurements on a sample with Si_3N_4 particulates (TPF-766) and on one with BN particulates (TPF-790) are shown in Figure 2. Also shown is the thermal conductivity of a p-type SiGe sample made using standard fabrication techniques. The grain size of the standard material is several microns which is similar to the heat treated samples made from ultra-fine powders. The reduction in thermal conductivity (from 15 to 30 percent) of the two samples with the fine particulates can clearly be seen.

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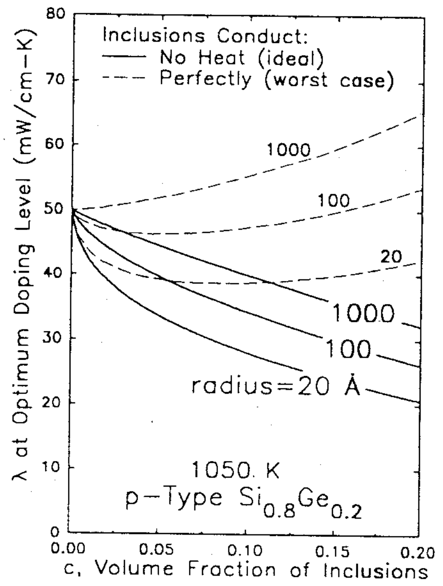

Figure 1: Thermal conductivity of SiGe alloy at 1050K as a function of inclusion concentration

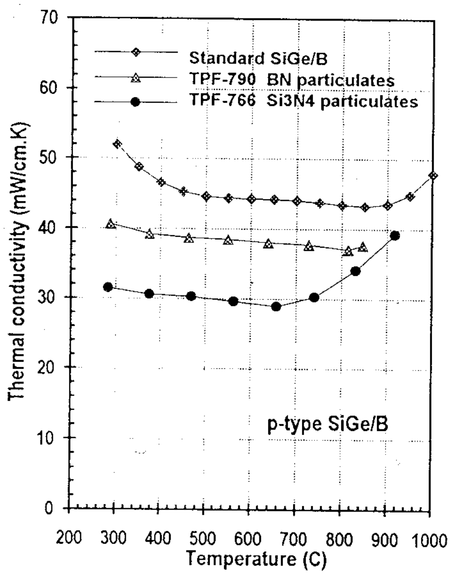

Figure 2: Thermal conductivity of samples with BN and Si_3N_4 particulates compared to a sample of standard p-type SiGe

The carrier concentrations of the samples varied by up to one order of magnitude but calculations showed that the effect of this on the total conductivity is at most a few percent. Sample TPF-790 was examined in the TEM. The grains were found to be around 2 μm in size and BN particulates of 50-200Å in diameter were found inside the grains. The concentration of BN particulates appeared to be at most a few percent which is probably the reason for only a 15 percent reduction in thermal conductivity.

These TEM observations and the measured conductivities are consistent with the theoretical predictions of increased scattering of intermediate frequency phonons, which carry most of the heat in SiGe alloys.

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3. References

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