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June 10, 2013

Dear Professor Phillpot:

Here are the detailed responses to the referee's comments. The changes are included as red text in the copy of the revised manuscript titled "jap_vc_jl_060413_Supplemental_Material".

1) the authors use "phonons" or "phonon-like" to indicate propagating modes: the use of "propagating modes/phonons" would be more precise and understandable.

We removed the one use of the term "phonon-like" and replaced it with "propagating" on page 9. Throughout the manuscript, we have been careful to only use the term "phonon" when referring to the vibrational modes under the virtual crystal approximation.

2) There is a serious problem of size convergence when calculating the thermal conductivity of bulk disordered solids. This is especially serious for silicon/germanium alloys as for example shown in Fig.2 of [He et al. Nano Lett. 11, 3608 (2011)], which suggest that a 1.7million atom cell is not sufficient to get the converged thermal conductivity of SiGe at room temperature. Also Skye and Schelling [Ref.44] admit that their calculations are far from size convergence. In general some consideration of size effects should be made, and possibly it should be pointed out that perturbation methods are an approximated way of circumvented such problems.

The description of how size effects are treated was included in the original manuscript and can now be found on page 25, first full paragraph. We added text and a citation ([71]) to address the referee's comment on page 29 in addition to this previous text:

"This system-size requirement highlights the efficiency of the VC-ALD method compared to VC-NMD, which is necessary when computationally-expensive DFT calculations are used."

3) The description of ALD methods in Sec 2.1 is confusing. Eq. 1 is more general than "Single Mode Relaxation Time Approximation". In fact also when the Boltzmann Transport Equation is solved iteratively, Eq. 1 is used to compute the thermal conductivity, with the SC-BTE lifetimes. The difference between RTA-BTE and SC-BTE is that in the former case lifetimes are computed assuming that all phonons have the equilibrium population, while in the latter the effect of non-equilibrium phonon populations is accounted for by self-consistency.

We have described the ALD method in previous papers and have included the appropriate references. Due to this manuscript already being quite long, we did not want to include these details here.We modified the text on page 4 to clarify that the SC-BTE lifetimes can be used with Eq (1) to predict the thermal conductivity.

Sincerely,

Alan McGaughey