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Development of Interatomic Potential of $Ge_{(1-x-y)}Si_xSn_y$ Ternary Alloy Semiconductors for Classical Lattice Dynamics Simulation

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Abstract

We have developed the interatomic potential of GeSiSn ternary mixed systems to reproduce the lattice constant, phonon frequency, and phonon dispersion relations by molecular dynamics (MD) simulation. The phonon dispersion relation is derived from the dynamical structure factor which is calculated by the space-time Fourier transform of atomic trajectories in MD simulation. The designed potential parameter set almost reproduces the experimental data of phonon characteristics in $Ge_{(1-x-y)}Si_xSn_y$ alloy. This work enables us to predict the elastic and phonon related properties of bulk group IV alloys, especially including ternary alloys.

1. Introduction

Group IV alloy semiconductor materials are becoming promising for the next-generation devices. One of the most anticipated application is what's called the Internet of Things, which requires to downsize, large scale integrate, or multi-functionalize. The thermal transport related to self-heating in the devices will become complicated and inhomogeneous. Thus, there is a great demand to measure the lattice temperature, which are closely related to the phonon, in such low-dimensional devices.

Computational approaches, such as MD simulation, are useful to calculate the phonon related properties in the nanoscale devices. However, the MD calculation of the group IV ternary system has not been reported. In this study, we try to reproduce the lattice constant, phonon frequency, and phonon dispersion relations in the bulk $Ge_{(1-x-y)}Si_xSn_y$ ternary alloy by classical MD simulation with newly developed the interatomic potential.

2. Simulation Procedure

The models in MD simulation is $Ge_{(1-x-y)}Si_xSn_y$ crystal with periodic boundary condition to reproduce the bulk sample. Periodic boundary condition is adopted in all directions, *i.e.*, the system is replicated throughout space to an infinite lattice. The atomic arrangement in the ternary alloys has been shuffled using random numbers.

The initial lattice constant a_i of sample was set to $5.658 - 0.227x + 0.831y$ Å. The initial model length and the one side width of the regular tetragon were $30a_i$ and $4a_i$ Å, respectively. Fig. 1 shows the unit structure of the bulk $Ge_{0.75}Si_{0.18}Sn_{0.07}$ crystal model.

The Stillinger-Weber (SW) potential is employed as the interatomic potential formula [1]. The parameters for mixed system are determined to reproduce the lattice constant in MD simulation and deformation energies of small cluster models which is estimated by molecular orbital calculation.

3. Results and Discussion

In Table I, we show relevant parameters obtained by the simulation and the previous study [2] for ternary alloys. Fig. 2 shows the (a) Sn and (b) Si concentration dependence of the corrected lattice constant. The corrected values were obtained by subtracting $-0.227x$ or $0.831y$ to compensate the dependence of the Si and Sn atom, respectively. The functions of the red line, which follow the Vegard's law, were shown in Fig. 2. In classical MD simulation, the lattice constant of $Ge_{(1-x-y)}Si_xSn_y$ ternary alloys follow the Vegard's law just like GeSn but not SiGe binary alloys.

Fig. 3 shows the Phonon at the Γ point and the Raman spectrum in the $Ge_{0.75}Si_{0.18}Sn_{0.07}$ obtained by the simulation and the previous study [2], respectively. The spectra are dominated by three modes, which are the Ge-Ge, Si-Ge, and Si-Si vibrations. We could not determine the frequency of the Ge-Sn, Si-Sn., and Sn-Sn peaks, which may be overlapped at approximately 240, 340, and 200 cm^{-1}, because their peaks are weak and broad.

Fig. 4 shows the Sn concentration dependence of the corrected (a) Ge-Ge and (b) Si-Si vibration frequency at the Γ point obtained by the simulation and the previous study [2], respectively. The corrected values were obtained by subtracting $-17.1x$ or $71.2(1-x)$. The functions of the red solid line and the blue dashed line were shown in Fig. 4. The dependence of the Ge-Ge mode was varied small but overestimated than the previous data. On the other hand, that of the Si-Si mode was almost in agreement with the previous data but varied widely. As a result, the classical MD phonon simulation allowed us to almost reproduce the tendency of the Raman measurement.

Fig. 5 shows the phonon dispersion relations in (a) pure Ge, (b) $Ge_{0.7}Si_{0.3}$, (c) $Ge_{0.75}Si_{0.18}Sn_{0.07}$, and (d) $Ge_{0.34}Si_{0.33}Sn_{0.33}$ alloys obtained by the MD simulation. The phonon branches in the alloys were broadened than that in the pure Ge. The phonon dispersion relations of $Ge_{0.75}Si_{0.18}Sn_{0.07}$ was similar to that of $Ge_{0.7}Si_{0.3}$, in other words, the Sn related phonons were minor than other phonons. However, in $Ge_{0.34}Si_{0.33}Sn_{0.33}$, the Sn related phonons are overlapped with Si-Ge and Ge-Ge phonon branches. Thus, their branches were drastically broadened. Moreover, the acoustic phonon is closely related to the thermal transport. This study predicted the broad acoustic phonon branches. The classical MD simulation with SW potential allowed us to calculate

the phonon properties in $Ge_{(1-x-y)}Si_xSn_y$ ternary alloys.

4. Conclusions

This is the first report of the classical MD simulation with newly developed potential parameter for GeSiSn ternary system. The result of simulation almost reproduced the phonon property tendency obtained in the previous study. The classical MD simulation with the SW potential is necessary for the thermal transport simulation in $Ge_{(1-x-y)}Si_xSn_y$ nanostructure which has the quasi-ballistic phonon conduction. Our approach of parametrization is expected to be applicable to other group IV mixed systems ternary and quaternary alloys in future.

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References

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Table I Summary of parameters in GeSiSn ternary alloys obtained by simulation and previous study.

				Phonon frequency in MD (cm^{-1})			Raman shift [2] (cm^{-1})		
x (%)	y (%)	a_{Vegard} (Å)	a_{MD} (Å)	$\omega_{Si\text{-}Si}$	$\omega_{Si\text{-}Ge}$	$\omega_{Ge\text{-}Ge}$	$\omega_{Si\text{-}Si}$	$\omega_{Si\text{-}Ge}$	$\omega_{Ge\text{-}Ge}$
0	0	5.658	5.658	N/A	N/A	300.9	N/A	N/A	301.0
18	10	5.700	5.701	439.4	373.8	284.3	446.1	389.8	288.8
20	8	5.679	5.677	448.2	376.7	286.4	451.8	393.4	289.9
18	7	5.675	5.673	446.6	376.8	286.4	452.9	394.9	290.9
13	3	5.653	5.652	446.9	375.3	291.5	454.8	395.7	296.7
20	2	5.692	5.626	448.0	380.7	289.6	458.3	400.6	295.6
33	33	5.857	5.858	433.4	356.7	257.7	N/A	N/A	N/A

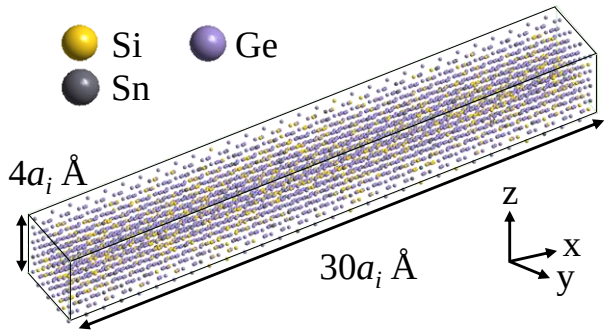

Fig. 1 Unit structure of bulk $Ge_{0.75}Si_{0.18}Sn_{0.07}$ crystal model.

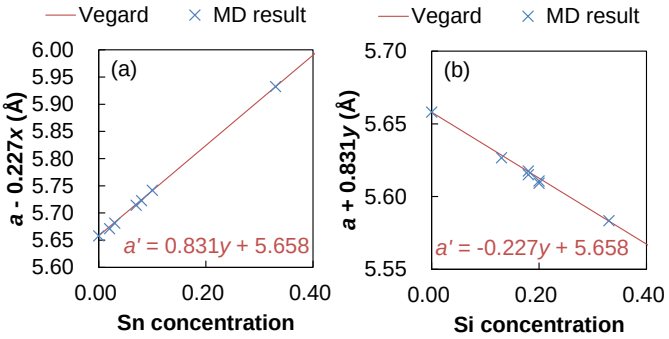

Fig. 2 Atom concentration dependence of corrected lattice constant.

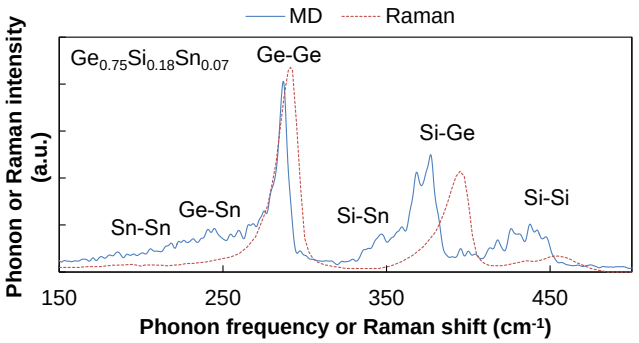

Fig. 3 Phonon at Γ point and Raman spectrum [2] in $Ge_{0.75}Si_{0.18}Sn_{0.07}$.

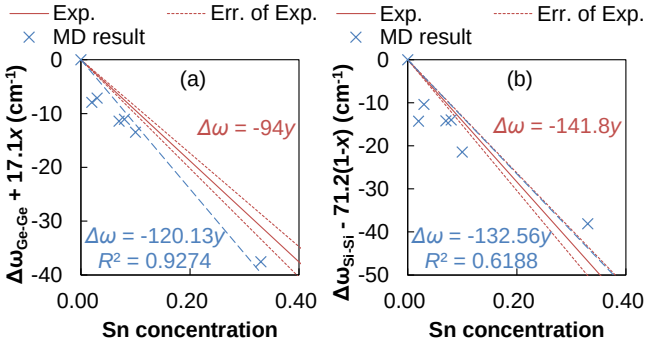

Fig. 4 Sn concentration dependence of corrected phonon frequency.

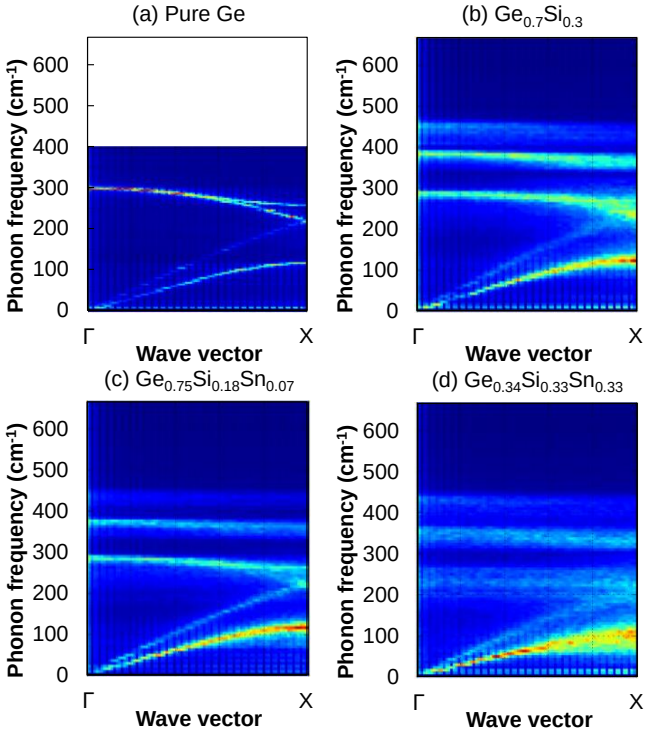

Fig. 5 Phonon dispersion relations in pure Ge and GeSiSn alloys.