Extended Abstracts of the 2020 International Conference on Solid State Devices and Materials, VIRTUAL conference, 2020, pp367-368

Inelastic X-ray Scattering Measurement on SiGeSn Polycrystalline Alloy

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Abstract

Inelastic X-ray scattering measurements for $Si_{1-x}Ge_x$ and $Si_{1-x-y}Ge_xSn_y$ polycrystals were carried out at SPring-8 to investigate the impact of the introduction of Sn on phonon vibrations. Energy peak positions of IXS peaks corresponding to the local vibration mode were shifted by Sn toward lower energy. This result indicates that the low thermal conductivity obtained in Sn-containing group-IV alloys is due to the modification of the local structure producing the local vibration of the phonon.

1. Introduction

The thermoelectric generator consisting of group-IV semiconductor materials, such as $Si_{1-x}Ge_x$, has been realized and implemented into space probes. It is crucial to reduce the thermal conductivity of the material used in the thermoelectric generator to enhance its conversion efficiency. To date, we reported that introduction of Sn into Ge can reduce its thermal conductivity [1], however, impact of Sn atoms on phonons which carries heat has not been clarified.

On the other hand, an anomalous phonon dispersion with no dependence on wavenumber has been observed by inelastic X-ray scattering (IXS) measurements for $Si_{1-x}Ge_x$ alloy [2]. The anomalous phonon dispersion was observed in neither pure-Si nor pure-Ge, and did not appear in SiGe compound having only -Si-Ge-Si-Ge- configuration modeled via molecular dynamics simulation. This means that clusters spontaneously formed in the $Si_{1-x}Ge_x$ alloy play an important role for the anomalous phonon dispersion expression. In this study, IXS measurements were performed on $Si_{1-x}Ge_x$ and $Si_{1-x-y}Ge_xSn_y$ polycrystalline samples to investigate the impact of Sn atoms on the anomalous phonon dispersion which contributes to reduce the thermal conductivity in $Si_{1-x-y}Ge_xSn_y$ alloys.

2. Experimental

$Si_{1-x}Ge_x$ and $Si_{1-x-y}Ge_xSn_y$ polycrystalline powders were synthesized by ball-milling method. X-ray diffraction revealed their compositions as Si : Ge=92.5 : 7.5 and Si : Ge : Sn=90.7 : 7.4 : 1.9, respectively. The powders were pressed into small pellets at RT for the IXS measurement. The IXS measurement was performed on BL35XU beamline at the SPring-8 synchrotron facility. The used X-ray having incident energy of 21.7 keV realizes the energy resolution around 1.5 meV. The measurement ranges in the unit of wavenumber, k, were k=3.78~7.92 nm^{-1} and k=46.15~50.11 nm^{-1}.

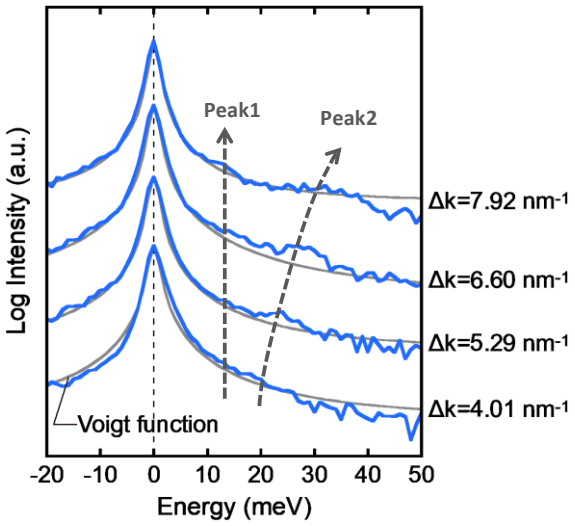

Fig. 1. IXS spectra and Voigt function fitted to the Rayleigh scattering peaks for the $Si_{1-x}Ge_x$ polycrystalline sample measured at different wavenumbers.

3. Results

IXS spectra clearly contain 2 inelastic scattering peaks in addition to intense Rayleigh scattering signal at the energy, E=0 (Fig. 1). The Rayleigh scattering signal was fitted by Voigt function and subtracted to perform subsequent peak fitting only on the inelastic scattering signals. The evaluated energy peak positions for $Si_{1-x}Ge_x$ and $Si_{1-x-y}Ge_xSn_y$ samples were summarized in Fig. 2 as a function of wavenumber. Peak energy positions for peaks appeared in the relatively higher energy region surely consistent with longitudinal acoustic (LA) phonon dispersion curves simulated by molecular dynamics simulation. Note that the measured samples are polycrystal, thus the observed trend corresponds to overlapping phonon dispersion curves along several direction in the polycrystalline samples. In contrast, the fact that another peak energy positions at the relatively lower energy region have no dependence on wavenumber indicates the local vibration mode [2] was detected both in $Si_{1-x}Ge_x$ and $Si_{1-x-y}Ge_xSn_y$ polycrystalline materials. In this lower wavenumber region, no

significant effect of introduction of Sn on phonon dispersion curves was observed.

In the case of higher wavenumber region (k=46.14~50.11 nm^{-1}), IXS peaks which correspond to LA or transverse acoustic (TA) phonon dispersion were not observed because of different periodicity of Brillouin zone along different direction in the polycrystalline materials. However, the vibrations with no dependence on wavenumber were clearly observed (Fig. 3) as same as the case of measurement performed in the range of lower wavenumber. In contrast to the local vibration mode shown in Fig. 2, energy shift of the energy peak position of the local vibration mode by the introduction of Sn toward lower energy was confirmed.

In $Si_{1-x}Ge_x$ alloy, the local vibration mode is produced at the Ge-Ge pairs or Ge atom clusters, which are surrounded by Si atoms [2]. On the other hand, we previously reported that the extended X-ray absorption fine structure measurement on a $Si_{1-x-y}Ge_xSn_y$ alloy revealed a Sn atom locates at the 2nd nearest neighbor of a Si atom to form Si-Ge-Sn local structure [3]. Addition of a Sn atom to the Si-Ge local structure to form the Si-Ge-Sn local structure is the possible reason of the energy shift of local vibration mode observed via the IXS measurement. In other words, the introduction of Sn modifies the local structure producing the local vibration mode which contributes to reduce the thermal conductivity.

Acknowledgements

This work was supported by JSPS KAKENHI Grant Number JP18K13786 from the Japan Society for the Promotion of Science. The IXS measurements were performed at SPring-8 with the approval of the JASRI (Proposal No. 2019B1286).

References

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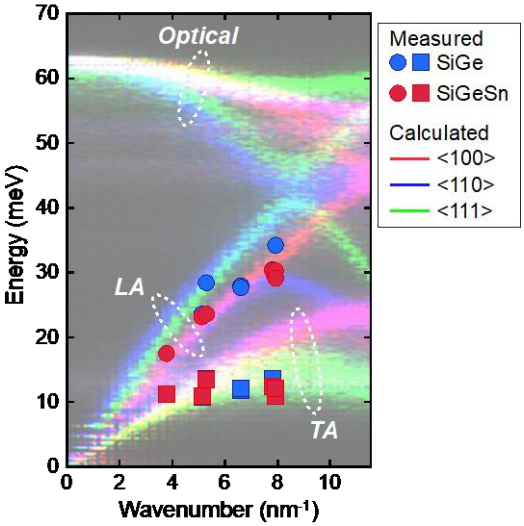

Fig. 2. Energy peak positions of IXS signals for $Si_{1-x}Ge_x$ and $Si_{1-x-y}Ge_xSn_y$ polycrystals measured up to 50 meV at k=3.78~7.92 nm^{-1}. Background is the calculated phonon dispersion for $Si_{0.925}Ge_{0.075}$.

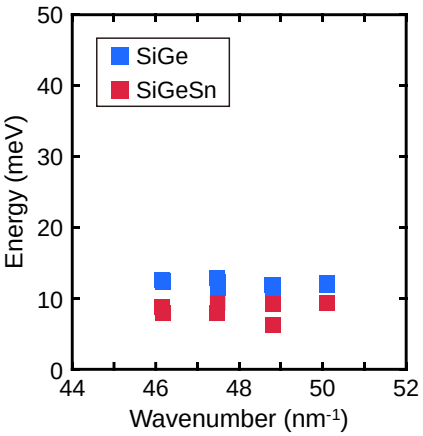

Fig. 3. Energy peak positions of IXS signals for $Si_{1-x}Ge_x$ and $Si_{1-x-y}Ge_xSn_y$ polycrystals measured at k=46.15~50.11 nm^{-1}.