Study on phonon properties and atomic arrangement of SiGe alloy using synchrotron radiation techniques

Ryo Yokogawa[a,b]

[a]Research Institute for Semiconductor Engineering, Hiroshima Uninversity
[b] Graduate School of Advance Science and Engineering, Hiroshima University,
1-4-2 Kagamiyama, Higashihiroshima, Hiroshima 739-8527, Japan

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Silicon-germanium (SiGe) alloy has a low thermal conductivity due to a significant reduction of the phonon mean free path compared to those of bulk Si and Ge crystals and is one of the promising candidates as a next-generation material for thermoelectric devices. SiGe is also used in nanosheet transistor fabrication processes and attracting attention as a next-generation p-type channel material. As miniaturization progresses toward higher device performance, precise understanding of its thermal transport at nanoscale (phonon properties) and atomic configuration are important from the viewpoint of device design. In particular, the atomic arrangement of SiGe determines its phonon and electronic band structure. Inelastic x-ray scattering (IXS) with synchrotron radiation is a powerful technique to evaluate phonon energy and dispersion nondestructively, but there are no reports of applying this method to epitaxial SiGe thin films, which are commonly used in devices, because the penetration depth of hard x-ray into SiGe. In this study, I demonstrated phonon spectra and dispersion of high-quality bulk SiGe using IXS. Moreover, I tried to observe the atomic arrangement from three-dimensional large-area reciprocal lattice maps of bulk SiGe using synchrotron x-ray diffraction (XRD) with synchrotron radiation.

The single-crystalline SiGe samples for IXS and XRD were prepared by two different growth methods: the Czochralski [1] and traveling liquidus zone methods [2]. The IXS measurements were performed on the BL35XU and BL43LXU beamlines at the SPring-8 synchrotron facility [3]. The incident x-ray energy was set to 17.8 or 21.7 keV, which corresponds to Si (9 9 9) or Si (11 11 11) reflection, respectively. The three-dimensional large-area reciprocal lattice maps of bulk SiGe were obtained using the diffractometer through XRD measurements at the BL02B1 beamline in SPring-8 synchrotron facility. In addition, the reciprocal space was simulated using the free software DISCUS [4].

Figure 1(a) shows the phonon dispersion curves of bulk $Si_{1-x}Ge_x$ with the x value of 0.45 obtained by IXS. The phonon dispersion of bulk SiGe were observed to consist of four phonons in the Γ–X ([00q]) direction: longitudinal optical (LO), transverse optical (TO), longitudinal acoustic (LA), and transverse acoustic (TA) phonon modes. I found that the LO and TO modes are split into three modes (Ge–Ge, Si–Ge, and Si–Si modes). Moreover, an anomalous phonon dispersion on the low-energy side (approximately 13meV), which is different from the optical and acoustic phonon dispersions, was observed [5]. Figure 1(b) shows the phonon dispersion curves obtained by the molecular dynamics (MD) simulation of bulk $Si_{1-x}Ge_x$ with the x value of 0.45. In Fig. 2(b), the intensity of blue, magenta, and red colors shows the density of states (DOS) of the Si–Si, Si–Ge, and Ge–Ge vibration mode, respectively. The results of the optical and acoustic phonon modes in the simulation are in good agreement with the experimental results. Figures 2(a) and 2(b) also show that the experimental anomalous mode including the momentum dependence is well reproduced with the MD simulation. This mode did not appear in the MD simulation with the SiGe compound model (Si–Ge–Si–Ge only configuration). I found that the anomalous mode had no Ge fraction dependence experimentally, indicating that the mode originated from the Ge localized vibration without propagation properties [6-8].

Figure 2(a) shows the reciprocal lattice map of bulk $Si_{1-x}Ge_x$ with the x value of 0.32 (center: 0, 0.5, 0). As a result, the checkerboard-like pattern derived from diffuse scattering due to the atomic arrangement was observed. Figure 2(b) shows the result of the reciprocal lattice space simulation based on the most stable structure obtained from density functional theory (DFT) and genetic algorithm [9]. The reciprocal lattice space based on the most stable structure reproduces checkerboard-like pattern and this profile is generally consistent with the XRD result (see Fig. 2(a)). Figures 2(c) and 2(d) show the simulation results for pure Si and SiGe with random atom position, respectively. In contrast to Fig. 2(b), diffuse scattering intensity was weak in pure Si and scattered diffuse scattering in SiGe with random atom position. From the above, these results suggest that the atomic arrangement of bulk SiGe is not completely random, but rather that there is a tendency for atoms of the same type to bond over a wide area, i.e., percolation.

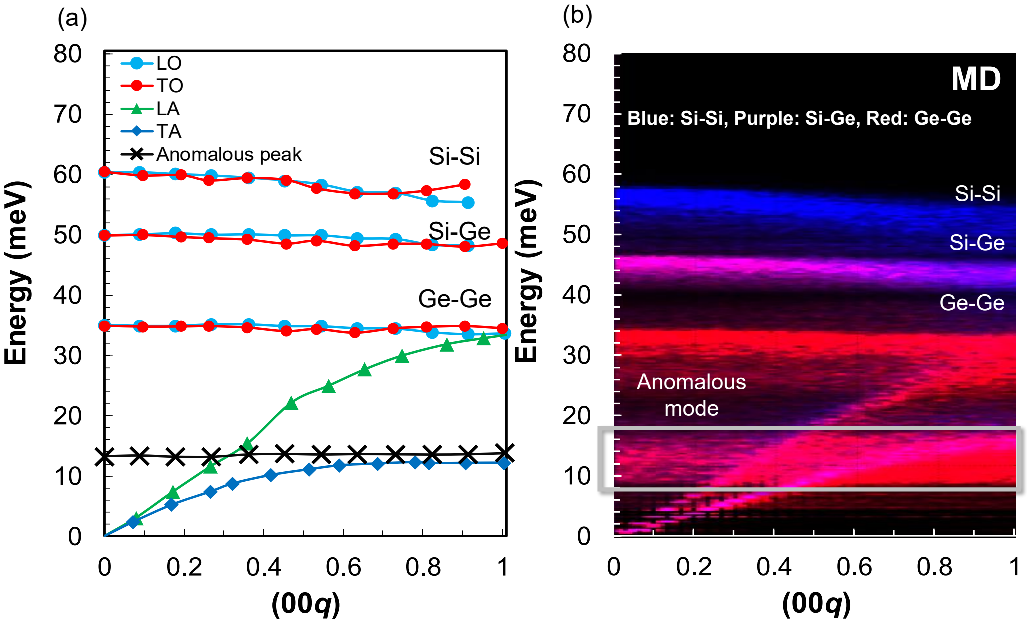

FIGURE 1. Phonon dispersion relations of bulk $Si_{1-x}Ge_x$ with the x value of 0.45 [6]. (a) Phonon dispersion curves including the anomalous peaks obtained by peak positions of IXS spectra. (b) Corresponding phonon dispersion curves simulated by the MD calculations. The blue, magenta, and red colors show the Si–Si, Si–Ge, and Ge–Ge vibration modes, respectively. The phonon dispersion of the anomalous mode is marked with a gray rectangle.

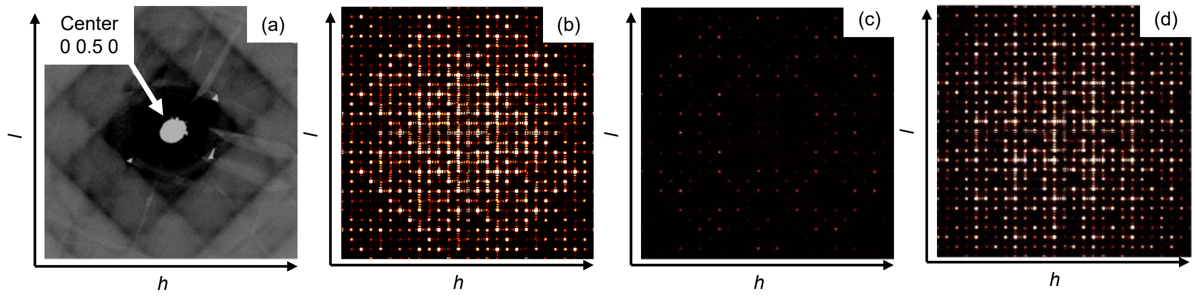

FIGURE 2. (a) Reciprocal space mapping of bulk SiGe bulk $Si_{1-x}Ge_x$ with the x value of 0.32 (center: 0 0.5 0). Reciprocal space simulation results of (b) most stable SiGe (Ge: 30%) using DFT and genetic algorithm, (c) Si, and (d) SiGe with random atom position (Ge: 30%).

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