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Low-Temperature (~150°C) Solid-Phase Epitaxy of a-GeSn/c-Ge for High Non-Equilibrium Substitutional Sn-Concentration GeSn

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Abstract

To achieve GeSn with a high substitutional Sn concentration (>7%), low-temperature solid-phase epitaxy using a-GeSn/c-Ge stacked structures has been investigated. As a result, epitaxial growth of GeSn with substitutional Sn concentrations of ~8% has been achieved by decreasing the growth temperature to 150°C using a-GeSn (initial Sn concentration: 36%)/c-Ge stacked structures.

1. Introduction

To break through the scaling limit of performance of large-scale integrated circuits (LSIs), functional devices, such as photo devices, should be merged with LSIs. GeSn with a high substitutional Sn concentration (>7%) is an attractive direct band gap material, which can be merged with LSIs.[1]

Among various growth methods, solid-phase techniques are expected to be useful to achieve a high concentration of substitutional Sn, because migration of atoms during growth is suppressed compared with other techniques. To supress migration more effectively, lowering of the growth temperature is essential.

In the present study, solid-phase epitaxy (SPE) at low temperatures below the eutectic point of GeSn is investigated using a-GeSn/c-Ge structures.

2. Experiments and Results

The sample structure is schematically shown in Fig. 1. Amorphous-GeSn films (thickness: 100 nm, initial Sn concentration: 10-36%) were deposited on Ge(100) substrates at room temperature. The samples were annealed at 150-200°C to induce SPE.

Crystal structures of the grown layers of samples were investigated by electron backscattering diffraction (EBSD). Typical EBSD images for samples (initial Sn concentration: 28%, 36%) after annealing at 150 and 200°C are summarized in Fig. 2. For annealing temperature of 200°C, the EBSD images for the samples (initial Sn concentration: 28%) indicate non-epitaxial growth and (100)-oriented growth after annealing for 1 and 2 h, respectively. For the same annealing temperature (200°C), the EBSD images for the samples (initial Sn concentration: 36%) indicate similar results, i.e, non-epitaxial growth and (100)-oriented growth after annealing for 1 and 2 h, respectively. On the other hand, as shown in Fig. 2, the EBSD images for samples (initial Sn concentration: 36%) after annealing at a very low temperature (150°C) indicate non-epitaxial growth and (100)-oriented epitaxial growth after annealing for 50 and 120 h, respectively.

To evaluate the concentrations of substitutional Sn atoms in grown layers, Raman measurements were performed. The results for samples (initial Sn concentration: 28%) after annealing at 200°C for 2 and 120 h are shown in Fig. 3(a), where the spectra for c-Ge are also shown for comparison. For the spectra of GeSn samples, sharp peaks due to Ge-Ge bonding in c-GeSn are clearly observed. From this peak shift, the concentrations of substitutional Sn were evaluated,[2] where we assumed no strain in grown layers. This assumption is confirmed by XRD measurements, described later. The evaluated substitutional Sn concentrations are summarized as a function of the annealing time in Fig. 3(b). For samples (initial Sn concentration: 28% and 36%) annealed at 200°C, the substitutional Sn concentrations are ~5% just after completion of epitaxial growth (annealing time: 2 h). With increasing annealing time, the Sn concentrations converge to a value of ~3%, close to the thermal equilibrium solid-solubility (~2%) of Sn in Ge. Interestingly, for lower annealing temperature (150°C), very high substitutional Sn concentration (~8%) is obtained. These phenomena can be explained on the basis of the realization of very high non-equilibrium concentrations of substitutional Sn just after completion of SPE and subsequent movement of Sn atoms from substitutional sites to interstitial sites, i.e., Sn precipitation. The latter reaction is induced by annealing after growth, i.e., post-annealing. With decreasing temperature, the post-annealing effect becomes weak. Consequently, very high concentration Sn (~8%), significantly higher than the thermal equilibrium solubility, can be frozen in substitutional sites just after completion of SPE at lower temperatures (~150°C) and scarcely migrate to interstitial sites during post-annealing for a long time (~100 h) at the very low temperatures.

XRD measurements were performed to investigate strain in grown layers. The reciprocal space mapping of the sample (initial Sn concentration: 36%) after annealing (150°C, 120 h) is shown in Fig. 4, where Q_x and Q_z axes are along [110] and [001] directions, respectively. In this figure, Ge (224) and GeSn (224) peaks are observed. Though the peak of GeSn (224) is broad, the peak position indicated by the white dotted circle is much closed to the dashed line indicating fully strain relaxed in Fig. 4. By analysing the peak position, a substitutional Sn concentration of 7.6% was obtained. This value shows a good agreement with the value obtained by the Raman analysis.

The detailed crystal structures and composition profiles of the sample (initial Sn concentration: 36%)

after annealing (150°C, 120 h) were investigated by TEM. A scanning TEM (STEM) high-angle annular dark field (HAADF) image is shown in Fig. 5. In addition, Sn and Ge concentration profiles obtained by energy dispersive x-ray spectroscopy (EDX) along the arrow in the HAADF image are shown. These indicate that Sn concentration profile is almost uniform in bulk, though Sn segregation occurs in the surface region.

A TEM bright field image is also shown in Fig. 5, together with an electron diffraction pattern obtained from the grown layer. From the analysis of the pattern, lattice constant was obtained as 0.572 nm. The lattice constant corresponds to GeSn with a Sn concentration of 8.8%, which shows a good agreement with the results of Raman and XRD measurements. These results indicate that epitaxial growth of GeSn having a high substitutional Sn concentration (~8%) becomes possible by SPE at a low temperature (150°C).

3. Conclusion

Solid-phase epitaxial growth of GeSn with substitutional Sn concentrations of ~8% has been achieved by decreasing the growth temperature to 150°C using a-GeSn (Sn concentration: 36%)/c-Ge stacked structures. This technique is expected to be useful to realize multi-function LSIs, where high-efficiency photo devices are integrated with transistors.

References

[1] Nakatsuka et al., Jpn. J. Appl. Phys. **49**, 04DA10 (2010).
[2] D'Costa et al., Solid State Communications **144**, 240 (2007).

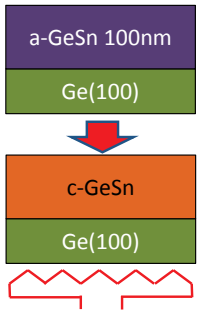


Fig. 1. Sample structures.

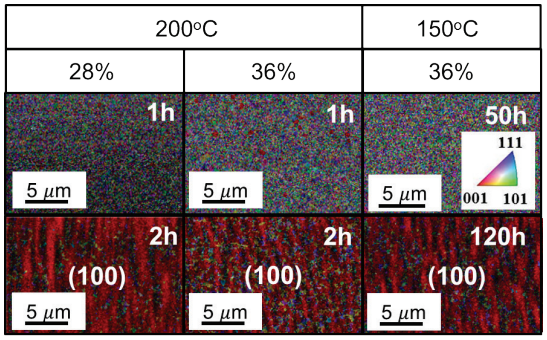
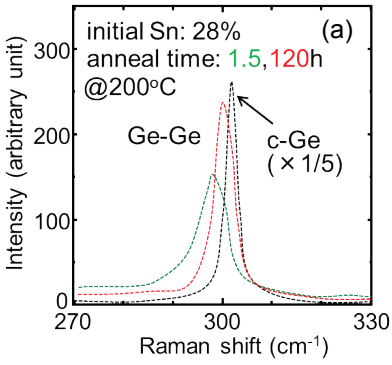


Fig. 2. EBSD images of grown samples.



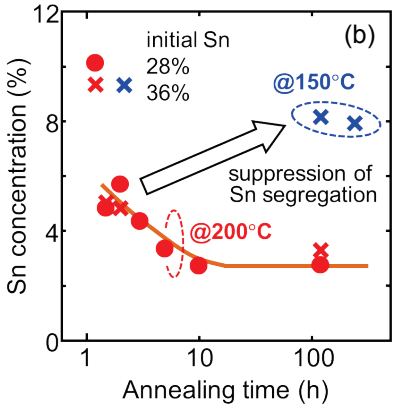


Fig. 3. (a) Raman spectra of samples after annealing (200°C) and (b) annealing time dependence of substitutional Sn concentration (150 and 200°C).

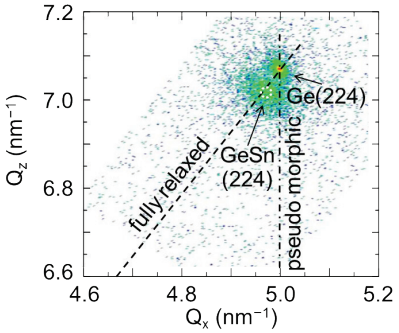


Fig. 4. XRD reciprocal space mapping of sample (initial Sn concentration: 36%) after annealing at 150°C for 120 h.

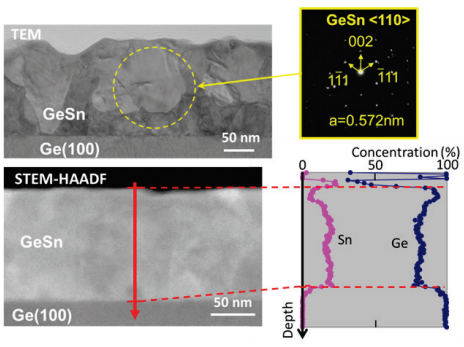


Fig. 5. HAADF image together with in-depth composition profiles, and TEM image together with ED pattern for sample (initial Sn concentration: 36%) after annealing at 150°C for 120 h.