Low-Temperature Solid-Phase Crystallization Combined with a-Si Under-Layer for High Sn Concentration GeSn Film without Sn-Segregation

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Abstract

Low-temperature solid-phase crystallization of high Sn concentration GeSn films combined with a-Si under-layers is investigated. By inserting a-Si under-layers, the growth rates of GeSn films are decreased. This phenomenon is attributed to suppression of interface nucleation by a-Si under-layers. Since homogeneous bulk-nucleation becomes dominant, nucleation and subsequent nucleus growth occur uniformly in a-GeSn films, which enables formation of segregation-free high Sn concentration GeSn films on insulator at low temperatures (~200°C).

1. Introduction

Ge based group-IV semiconductors are very attractive materials to realize next generation high-performance devices[1]. Among them, GeSn with high Sn concentration above 8% is attracting much attention due to the direct-transition energy band structure together with very small effective mass of carriers[2]. To realize advanced flexible electronics, where functional GeSn devices are integrated on flexible plastic substrates, low-temperature (\leq 300°C) growth techniques of high Sn concentration (\geq 8%) GeSn on insulator should be developed.

Previously, we investigated solid-phase crystallization (SPC) of high Sn concentration (~20%) GeSn films on insulator, and demonstrated growth of GeSn films at low temperatures ($\leq 250^{\circ}$ C)[3]. This causes an expectation that SPC is useful to realize advanced flexible electronics.

On the other hand, we recently investigated SPC of low Sn concentration (~2%) GeSn films on insulator, and clarified that the carrier mobility of the grown GeSn films is significantly increased by introduction of a-Si under-layers between a-GeSn films and insulating substrates[4]. However, effects of a-Si under-layers on growth characteristics of high Sn concentration GeSn films have not been clarified yet. In the present study, we investigate growth characteristics of high Sn concentration GeSn films on insulator in SPC combined with a-Si under-layers.

2. Experiments and Results

In the experiment, a-Si (thickness: 10 nm) and a-GeSn layers (initial Sn concentration: 15-20%, thickness: 50 nm) were deposited on fused-quartz substrates using a molecularbeam deposition system (base pressure: $\sim 5 \times 10^{-8}$ Torr). For comparison, samples without a-Si under-layers were also prepared. The sample structures are schematically shown in Fig. 1. These samples were annealed (150°C-350°C, 10 min-300 h) to induce SPC in a nitrogen ambient. Nomarski optical micrographs of GeSn/Si and GeSn samples (initial Sn concentration: 20%) after annealing (200°C, 30 min) are shown in Figs. 2(a) and 2(b), respectively. For both samples, bright contrast circular regions are observed The diameters of the circular regions of the GeSn/Si sample are smaller than those of the GeSn sample.

Micro-probe (laser beam spot diameter: ~1 μ m) Raman spectra obtained from the insides (points A and C) and outsides (points B and D) of the bright circular regions in the GeSn/Si and GeSn samples are shown in Figs 2(c) and 2(d), respectively. Raman peaks due to Ge-Ge bonding in c-GeSn are clearly observed from the insides (points A and C), while they are not detected from the outsides (points B and D). These results reveal that the bright contrast regions observed in the Nomarski images are crystallized regions, while the dark contrast regions outside of the bright regions are not crystallized.

We investigate growth characteristics for these samples. The radii of the circular bright contrast regions are summarized as a function of the annealing time (200°C) in Fig. 3(a). This figure indicates that the radii linearly increase with increasing annealing time and can be fitted with straight lines. From the slopes of the fitted lines, growth rates are evaluated.

Similar experiments were performed for samples (initial Sn concentration: 15% and 20%) at various temperatures, and the growth rates were evaluated. Arrhenius plots of the growth rates are summarized in Fig. 3(b), where reported data of growth rates of pure-Ge[5] are also shown. Compared with pure-Ge, the growth rates of GeSn are significantly increased through bond-weakening by Sn doping[3]. On the other hand, the growth rates are decreased by introduction of a-Si under-layers, as shown in Fig. 3(b). This phenomenon is attributed to suppression of interface nucleation, which is faster than bulk nucleation, by introduction of a-Si under-layers[4]. The activation energies of the growth rates are about 1.3-1.7 eV for all samples.

In-depth concentration profiles of Ge and Sn were analyzed by Auger electron spectroscopy. The results of GeSn/Si and GeSn samples (initial Sn concnetration: 20%) after annealing (200°C, 55 min) are shown in Figs. 4(a) and 4(b), respectively. As shown in Fig. 4(b), the Sn concentrations in the surface region become higher than the bottom region for the GeSn sample. This suggests that Sn segregation occurs during growth, which is initiated by nucleation at the interface between the GeSn film and the substrate. On the other hand, interestingly, the Sn concentrations indicate a uniform distribution (~17%) in GeSn/Si sample, as shown in Fig. 4(b). This phenomenon is attributed to suppression of interface nucleation and domination of homogeneous bulk nucleation, which occurs uniformly in a-GeSn films. As a result, segregationfree high Sn concentration GeSn films are obtained by introduction of a-Si under-layers.

3. Conclusions

We have investigated low-temperature SPC of high Sn concentration GeSn combined with a-Si under-layers. By introduction of a-Si under-layers, growth rates are decreased due to suppression of interface nucleation, which dominates homogeneous bulk nucleation in a-GeSn films. This enables formation of high Sn concentration ($\sim 17\%$) GeSn films without Sn segregation at low temperatures (200°C). This technique will be useful to realize advanced flexible electronics, where functional GeSn devices are integrated on flexible plastic substrates.

References

- [1] M. Miyao et al., JJAP 56, 05DA06 (2017).
- [2] S. Gupta, et al., JAP 113, 073707 (2013).
- [3] R. Matsumura, et al, ECS-SSL 4, P95 (2015).
- [4] C. Xu, et al., APL, 115, 042101 (2019).
- [5] K. Toko, et al., APL 94, 192106 (2009).



Fig.2 Nomarski optical micrographs of GeSn/Si (a) and GeSn (b), and Raman spectra of GeSn/Si (c) and GeSn samples (initial Sn: 20%) (d) after annealing (200°C, 30 min).



Fig.3 Annealing time dependence of domain radius (a) and Arrhenius plot of growth rate (b).



Fig.4 In-depth profile of Ge, Sn and Si of GeSn/Si (a) and GeSn samples (initial Sn: 20%) (b) after annealing (200°C, 55 min).