Extended Abstracts of the 2015 International Conference on Solid State Devices and Materials, Sapporo, 2015, pp900-901

Thermally-Stable High Sn Concentration (~9%) GeSn on Insulator by Ultra-Low Temperature (~180ºC) Solid-Phase Crystallization Triggered by Laser-Anneal Seeding

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Abstract

In order to realize next generation flexible thin-film transistors (TFT), solid-phase crystallization (SPC) of amorphous-GeSn (a-GeSn) films on insulating substrates combined with laser-anneal seeding has been developed. This technique enables crystallization of GeSn with high substitutional Sn concentration (~9%) at low temperatures (~180ºC).

1. Introduction

In order to realize next generation flexible devices, novel functional materials should be crystallized at controlled positions on insulating substrates below the softening temperature of flexible plastic substrates (~200ºC). GeSn is an attractive material for this purpose, since GeSn has higher carrier mobility than Si or Ge, and moreover, GeSn shows direct band structure by introducing substitutional Sn with a concentration more than 7%.

In line with this, we propose an idea to use laser-annealing (LA) to form crystal seeds. In this paper, lateral solid-phase crystallization (SPC) from LA seed has been investigated. As a result, we have realized crystallization of GeSn with high substitutional Sn concentration (~9%) at low temperatures (~180ºC).

2. Experiments

In the experiment, amorphous $Ge_{0.8}Sn_{0.2}$ layers (100 nm thickness) were deposited on quartz substrates by a molecular beam technique. The samples were irradiated with a KrF excimer laser (wavelength: 248 nm, energy density: 160 mJ/cm^2, pulse width: 80 ns) for 100 pulses/sec to form seed regions. Here, irradiation was performed with a rectangle shape (500 μm x 360 μm) beam with a scanning speed of 360 μm/sec, followed by thermal annealing (180-220ºC) for lateral SPC as shown in Fig. 1(a).

3. Results and Discussions

A Nomarski image of sample after LA is shown in Fig. 1(b). The annealed region shows bright contrast. Fig. 1(c) shows sample after subsequent annealing at 200ºC for 4h. We can clearly see a bright region expanding from the LA region. The Raman measurements were carried out at points shown in Figs. 1(b) and 1(c). These results indicate that both LA region (A) and laterally expanding region (B) seen in the thermal annealed sample have been crystallized, and there is no spontaneous nucleation outside of these regions (C).

Lateral growth length from LA seed was investigated after annealing at 180-220ºC. The results are summarized as a function of the annealing time in Fig. 2(a). The growth length linearly increases with increasing annealing time without incubation time for all annealing temperatures. From the slope of these lines, the growth rates were evaluated, which are summarized as Arrhenius plots in Fig. 2(b). Here, the reported data for Si and Ge [1] are also plotted for comparison. It is found that the growth rate of GeSn is about 1 μm/h at 180 ºC, which is nearly 10^6 times as large as that of Ge.

The substitutional Sn concentration profiles obtained by Raman measurements are summarized in Fig. 3 as a function of distance from seed edges for various annealing conditions. The Sn concentrations show higher values in SPC regions (~9%) than LA regions (~6%), which indicate the difference of non-equilibrium conditions. Interestingly, for all samples, substitutional Sn concentrations show very high values exceeding 15% near growth edges. The Sn concentrations decrease with increasing distance from the edges. The decrease in Sn concentration from >15% to ~9% is attributed to the post annealing effect of grown regions. Thus, we analyze the post-annealing effects. The time of post-annealing t_{PA} in grown region at distance x from seed can be expressed by the following equation,
where, L_{grow} is the length of the grown region, and v is growth rate obtained in the previous section.

$$t_{PA} = \frac{L_{grow} - x}{v}$$

Values of substitutional Sn concentrations are summarized as a function of t_{PA} in Fig. 4. For 180ºC samples, decrease of Sn concentration shows a good agreement between samples with different annealing time (4h and 48h), which supports our speculation of post-annealing effect. For all samples, substitutional Sn concentrations stabilize at ~9% with increasing t_{PA}, which is far above thermal equilibrium solid solubility of Sn in Ge (~2%). The detailed physics will be discussed in the presentation.

4. Summary

LA-seeded SPC of $Ge_{0.8}Sn_{0.2}$ has been investigate. By forming seed region by LA, lateral SPC have been initiated from controlled positions at very low temperature of 180ºC. Growth rate is about 1 μm/h at 180 ºC, which is nearly 10^6 times as large as that of Ge. Moreover, by investigating substitutional Sn concentration in grown layer, thermally stable high-Sn concentration (~9%) has been obtained. This technique is very useful to realize next generation flexible thin-film devices.

References

1. K. Toko, et al., Appl. Phys. Lett. **94** 192106 (2009).

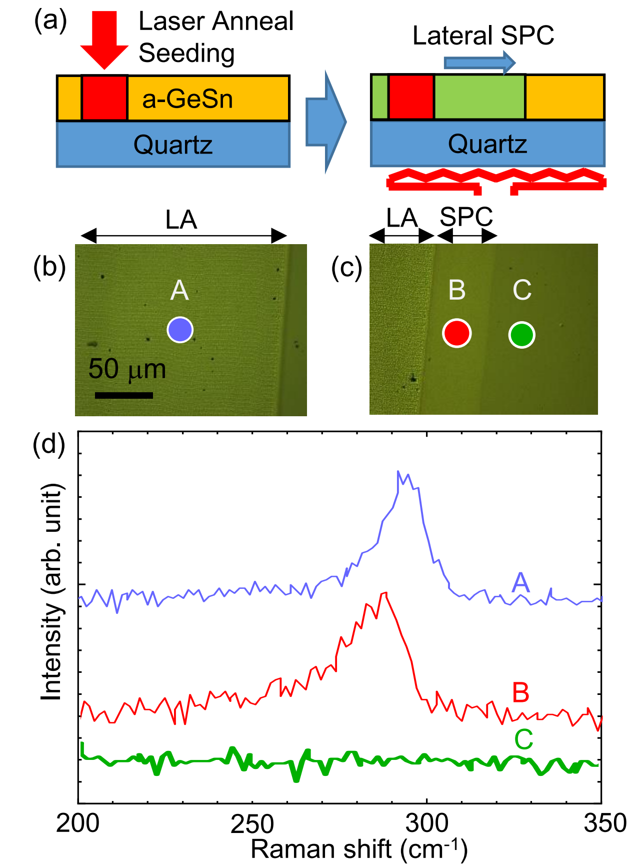

Fig. 1(a) Experimental procedure.
Nomarski images (b) just after LA and (c) after annealed at 200ºC for 4h. Raman spectra obtained at A-C are shown in (c).

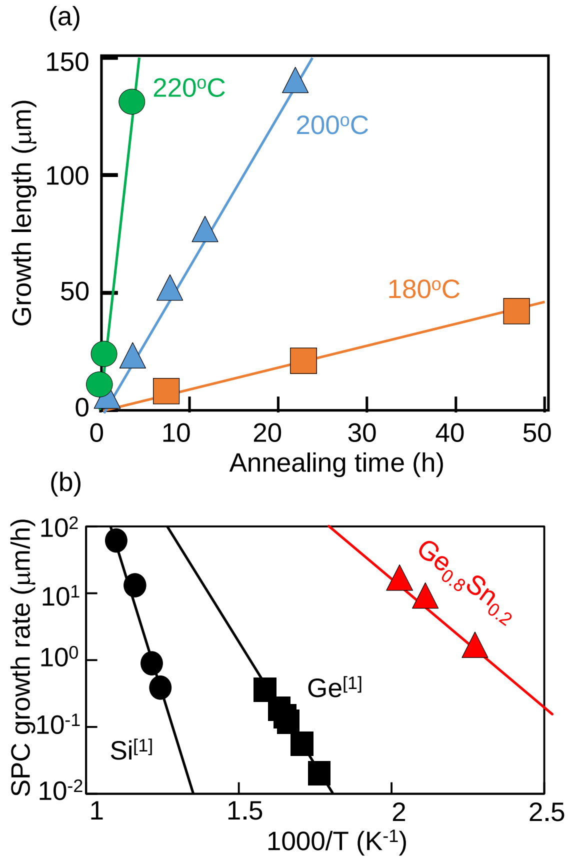

Fig. 2(a) Growth length of $Ge_{0.8}Sn_{0.2}$ with various annealing temperatures.
(b) Arrhenius plots of $Ge_{0.8}Sn_{0.2}$ at various temperatures. Growth rates of Si and Ge[1] are also shown for comparison.

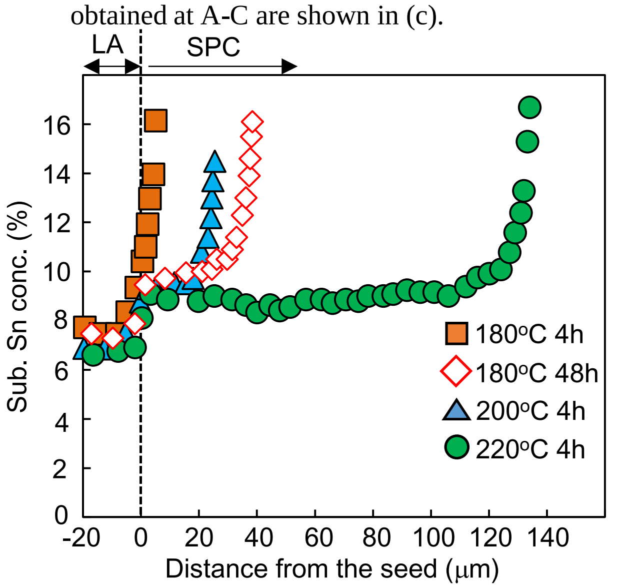

Fig. 3 Substitutional Sn concentration in GeSn after growth at various temperatures.

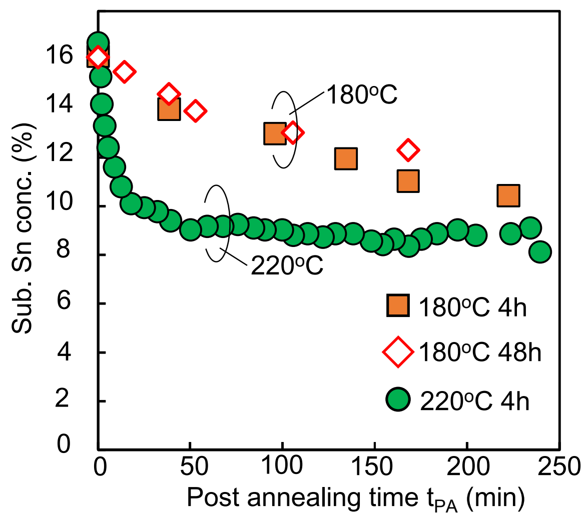

Fig. 4 Post annealing effect of substitutional Sn concentration evaluated from lateral distribution.