C-8-4

Extended Abstracts of the 2014 International Conference on Solid State Devices and Materials, Tsukuba, 2014, pp556-557

Melting-Sn Induced Seeding-Processing for Low-Temperature Lateral-Crystallization of a-GeSn on Insulating Substrate

H. Chikita[1], R. Matsumura[1,2], Y. Kai[1], T. Sadoh[1], and M. Miyao[1]
[1]Department of Electronics, Kyushu University, 744 Motooka, Fukuoka 819-0395, Japan
[2]JSPS Research Fellow, 5-3-1 Kojimachi, Chiyoda-ku, Tokyo, 102-0083, Japan
Tel : +81-92-802-3736 , FAX : +81-92-802-3724, E-mail : h_chikita@nano.ed.kyushu-u.ac.jp

Abstract

A new low-temperature crystallization technique of a-GeSn on insulator has been developed. Here, island-shaped Sn/a-Ge stacked-structures are covered with a-GeSn films and annealed in two-steps. This technique enables lateral solid-phase crystallization of a-GeSn films at a very low temperature (~200ºC), which is useful to realize high-performance devices on flexible plastic substrates.

1. Introduction

Development of low temperature (≤250ºC) formation technique of Ge and GeSn on insulator is essential to realize high-performance electronic and optical devices on flexible plastic substrates (softening temperature: ~300ºC). This is because Ge and GeSn have higher carrier mobility and superior optical properties compared to Si. In line with this, solid-phase crystallization (SPC)[1], metal-induced lateral crystallization (MILC)[2], and Al-Induced layer exchange crystallization (AIC)[3] of a-Ge have been developed. However, SPC and MILC need high temperature annealing (≥350ºC). On the other hand, AIC enables low temperature growth (~200ºC)[4]. However, residual Al may deteriorate electrical properties of grown layers.

To solve these problems, we propose a new seeding crystallization technique using Sn, as shown in Fig.1. Here, island-shaped Sn/a-Ge stacked-structures are covered with a-GeSn films (a) and annealed in two-steps. Since Sn has a very low melting point (~230ºC), melting growth of Sn/a-Ge islands during the first annealing (>230ºC) creates crystallization seed of Sn-rich GeSn (b). Subsequently, lateral SPC of a-GeSn films is generated from the seed during the second annealing at a low temperature (c). We expect that crystallization of a-GeSn in the second step should occur at lower temperatures compared to conventional SPC of a-Ge, owing to seeding and Sn-catalyst[5] effects. In the present work, we examine this idea and investigate growth characteristics of GeSn. As a result, low temperature (~250ºC) crystallization of GeSn on insulating substrates becomes possible.

2. Experiments and Results

In the experiment, Sn and Ge layers (thickness: 10nm/10nm) were deposited on quartz substrates and patterned into island shapes (10μmΦ) by lift-off technique. Then, a-$Ge_{0.8}Sn_{0.2}$ layers (thickness: 100nm) were deposited by molecule beam technique [Fig.1(a)]. In addition to these *seeding structure samples*, *seedless structure samples*, deposited with a-$Ge_{0.8}Sn_{0.2}$ layers (thickness: 100nm) without island patterns, were prepared.

Since spontaneous nucleation in amorphous films suppresses lateral seeding SPC, annealing conditions generating spontaneous nucleation in a-$Ge_{0.8}Sn_{0.2}$ films were examined by micro-probe Raman measurements of the *seedless structure samples*. The Raman spectra of the samples annealed at 200–350ºC (1min) are shown in Fig.2. Sharp peaks due to Ge-Ge bonding in c-GeSn are observed after annealing at 300 and 350ºC. These results indicate that spontaneous nucleation is not generated by annealing below 250ºC. Here, 250ºC is above the melting point of Sn (~230ºC). Thus, the first step annealing conditions for creating seeding regions are selected as 250ºC for 1min.

The Nomarski image of a *seeding structure sample* after the first-annealing (250ºC, 1min) is shown in Fig.3(a). The color of the circular region, indicated by broken circle, around the seeding region, changes after the annealing. Micro-probe Raman measurements revealed that the circular region was crystallized. To examine lateral seeding SPC from the seed, the second-annealing was performed at 200ºC for 4h. Interestingly, the grown area becomes concentrically large. Raman spectra obtained from positions #1 – #4 are shown in Fig.3(c). Here, Ge-Ge peaks are observed from positions #1 – #3, while no peak is observed from #4. These results indicate that the a-$Ge_{0.8}Sn_{0.2}$ layer is laterally crystallized from the seeding region at a very low temperature (200ºC).

To reveal these phenomena, growth characteristics in the second-annealing at 160-250ºC are investigated using the first-annealed *seeding structure samples*. The results are summarized in Fig.4. The growth length linearly increases with increasing annealing time for all annealing temperatures. This clearly indicates that the growth is initiated from the seeding region without any incubation time. The growth velocity drastically increases with increasing annealing temperature. Arrhenius plot of the growth velocity is shown in Fig.5. Here, the reported data for Si [2,6] and Ge [2,7] are also shown for comparison. It is found that the growth velocity of $Ge_{0.8}Sn_{0.2}$ is much faster than that of Ge and Si, e.g., the growth velocity of $Ge_{0.8}Sn_{0.2}$ at 200ºC is about 6μm/h, which is nearly 10^6 times as large as that of Ge. This significant enhancement is attributed to the weakening Ge-Ge bonds by Sn atoms.

3. Conclusion

Melting-Sn induced seeding-processing is proposed for low-temperature growth of GeSn on insulator. As a result, lateral SPC of GeSn has been achieved at low temperatures (~250°C). This technique is useful to realize flexible electronic devices on plastic substrates.

References

[1] K. Toko et al., SSE **53**, 1159(2009).
[2] H. Kannno et al., APL **89** 182120(2006).
[3] M. Kurosawa et al., ECS JSSST **1**, 144 (2012).
[4] K. Toko et al., APL **104**, 022106 (2014).
[5] A. Ooato et al., TSF **557**, 155 (2014).
[6] L. Csepregi et al., JAP **49**, 3906(1978).
[7] L. Csepregi et al., SSC **21**, 1019(1977).

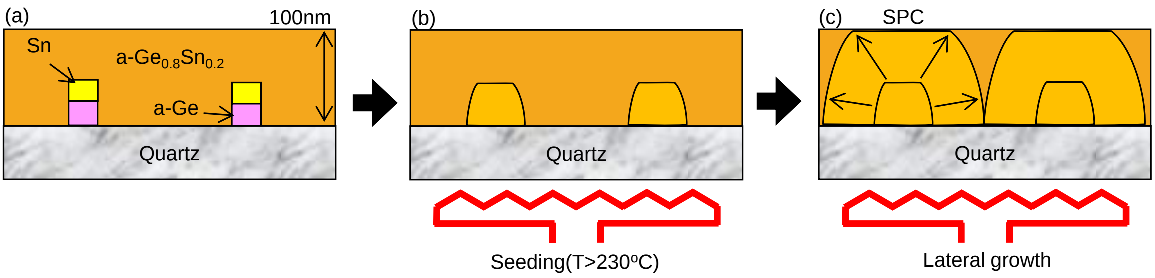

Fig.1. Initial sample structure (a), seed formation (T>230°C) (b), and lateral-solid phase crystallization (c).

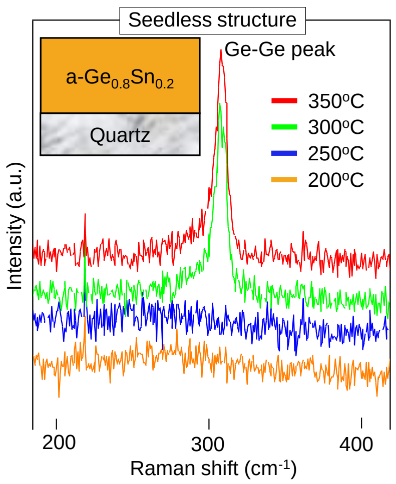

Fig.2. Raman spectra of seedless structure after annealing at 200-350°C (1min).

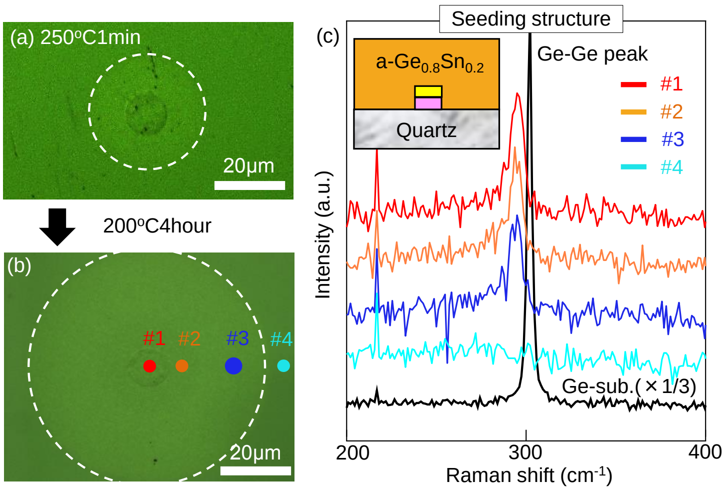

Fig.3. The Nomarski images of seeding structure after first-annealing (250°C,1min) (a) and after second-annealing (200°C,4h) (b), and Raman spectra were obtained from #1 - #4 of (b) (c).

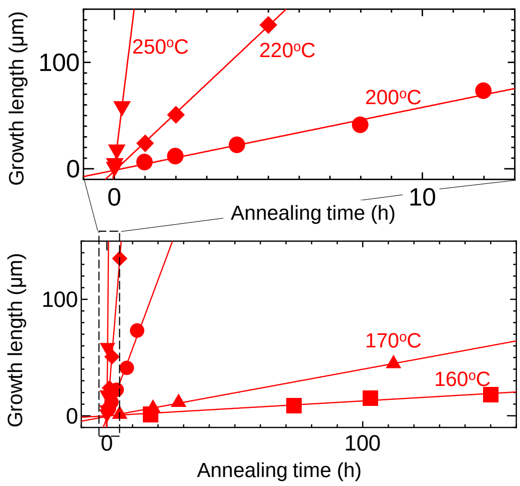

Fig.4. Growth length as a function of annealing time (annealing temperature: 160 – 250°C).

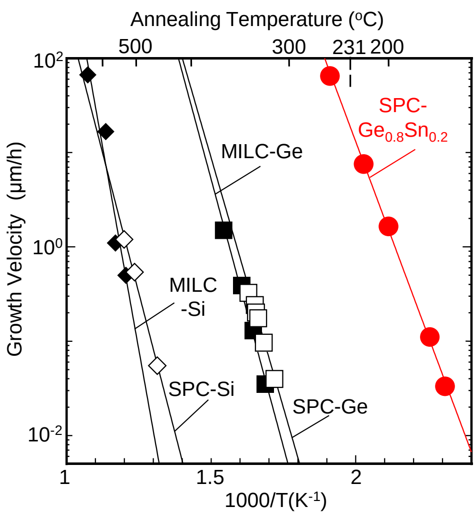

Fig.5. Arrhenius plots of growth velocity of $Ge_{0.8}Sn_{0.2}$. The data of SPC-Si [6], MILC-Si [2], SPC-Ge [7] and MILC-Ge [2] are also shown.