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Enhanced Growth of High Sn Concentration (Sn≥10%) GeSn on Insulator by Low Temperature (~170°C) Solid-Phase Crystallization Combined with Weak Laser Irradiation

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Abstract

Low-temperature solid-phase crystallization of GeSn-on-insulator by weak laser irradiation and subsequent annealing has been investigated. For a-GeSn films with high initial Sn concentrations (≥10%), weak laser irradiation (<70 mJ/cm^2) significantly decreases the crystallization temperature of the GeSn films. Moreover, the substitutional Sn concentrations in grown films increase with decreasing film thickness. As a result, crystalline GeSn films (thickness: 50 nm) with a very high substitutional Sn concentration (~10%) are achieved at low temperatures (~170°C).

1. Introduction

Development of a formation technique of crystalline GeSn (substitutional Sn concentration: >8%) films on insulator at low-temperatures (<200°C) is needed to realize advanced flexible electronics, where high-speed thin film transistors (TFTs) and high-efficiency optical devices are integrated on flexible plastic substrates (softening temperature: ~200°C). This is because GeSn (substitutional Sn concentration: >8%) has higher carrier mobility than Si and Ge due to direct-transition energy band structure with smaller effective mass of carriers [1].

Various formation techniques [2], such as solid-phase crystallization (SPC) [3-5] and rapid-melting growth by pulsed laser annealing (PLA) [6,7], have been investigated. However, SPC requires long time annealing, e.g., ~100 h at 180°C, to obtain sufficiently large growth length (~100 μm). On the other hand, PLA enables crystallization in a very short time. However, surface roughening induced by melting in PLA was a big problem for device application.

Recently, we observed enhancement of SPC of a-GeSn films (initial Sn concentration: 20%) by laser irradiation with low fluence [8]. In the present study, we investigate effects of the Sn concentration on the growth enhancement. In addition, effect of film thickness on the substitutional Sn concentration in the grown films is examined.

2. Experiments and Results

Amorphous-GeSn films (Sn concentration: 0−20%, thickness: 50−500 nm) were deposited on quartz substrates by an MBE system and irradiated with a KrF excimer laser (Gigaphoton Inc., wavelength: 248 nm, pulse duration: 80 ns, repetition rate: 100 Hz, irradiation number of pulses: 100) with wide range of fluence per pulse (0−200 mJ/cm^2).

Crystallinity of the grown layers was analyzed by Raman spectroscopy. Typical Raman spectra for samples (initial Sn concentration: 20%, thickness: 100 nm) before and after irradiation (fluence per pulse: 10 and 110 mJ/cm^2) are shown in Fig. 1(a). For the sample irradiated with fluence of 110 mJ/cm^2, a large sharp Raman peak (~300 cm^{-1}) due to Ge-Ge bonding in crystalline GeSn is observed, which indicates laser-induced melting crystallization [7]. On the other hand, for fluence of 10 mJ/cm^2, a very small and broad peak (~280 cm^{-1}) is detected. This peak is due to Ge-Ge bonding in amorphous GeSn and/or nano-crystalline GeSn [9]. Figure 1(b) shows Raman spectra for the samples, which were irradiated (fluence: 0, 10 mJ/cm^2) and subsequently annealed (170°C, 5 h). The Raman peak intensity for the weakly irradiated sample (fluence: 10 mJ/cm^2) is significantly increased after the low temperature annealing for 5 h, which is significantly shorter compared with the conventional SPC without laser irradiation (~100 h) [4]. This shows that low-temperature SPC is enhanced by the weak laser irradiation.

Figure 2 shows the Raman peak intensities for samples (initial Sn concentration: 20%, thickness: 100 nm) as a function of the fluence. Interestingly, in the figure, a flat region is observed in low fluences (10−60 mJ/cm^2), as labeled as weak LA, together with strong LA region (≥110 mJ/cm^2). Atomic force microscopy observations of the samples revealed that the roughness was small (<2 nm) for weak LA; however, it became large (~4 nm) for the intermediate region between weak LA and strong LA, and very large (>10 nm) for strong LA. Thus, irradiation with weak LA is essential to device application.

To reveal the phenomena, growth characteristics at a low temperature (180°C) are investigated for samples (thickness: 100 nm) with various initial Sn concentrations after laser irradiation with various fluences. The results are summarized in Fig. 3, where ×, ▲, and ● represent detection of no Raman peak, detection of small and/or intermediate Raman peak and enhanced SPC, and detection of large Raman peak without enhanced SPC, respectively. In the figure, boundary between no-melt and melt, which were obtained by in-situ reflectance measurements during laser irradiation, is also shown by the solid-line, and the interpolation is shown by the dotted line. For initial Sn concentrations below 2%, no enhancement of SPC is observed, while for initial Sn concentrations above 10%, SPC is enhanced for samples irradiated without melting. Here, the lower limit fluence of the weak LA region (flat region) significantly decreases with increasing initial Sn concentration, resulting in wider process window. This result suggests that SPC enhancement is attributed to nucleation induced by excess Sn in GeSn films during laser irradiation. However, further study is needed to clarify the reason for the flat region of weak LA, as shown in Fig. 2.

Substitutional Sn concentrations in grown layers were investigated as a function of the film thickness from the Raman peak position. The results for samples (initial Sn concentration: 20%) after weak laser irradiation and subsequent annealing (180°C, 5 h) are summarized in Fig. 4. The substitutional Sn concentrations increase with decreasing film-thickness, and a very high value (~10%) is achieved for 50-nm-thickness.

3. Conclusion

A novel growth method of GeSn-on-insulator by melting-free weak laser irradiation and subsequent low-temperature annealing has been developed. For samples with initial Sn concentration above 10%, SPC is significantly enhanced by weak laser irradiation, enabling growth in a short time (~5 h) at 170°C. Substitutional Sn concentrations in grown layers increase with decreasing film thickness. As a result, a very high substitutional Sn concentration of ~10% has been achieved for film-thickness of 50 nm. This technique will facilitate realization of advanced flexible electronics.

References

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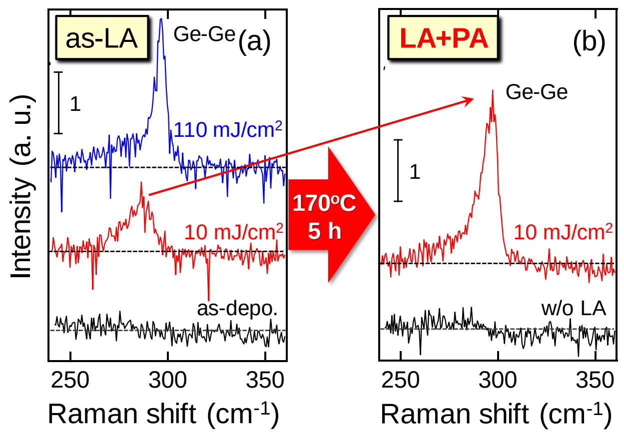

Fig.1. Raman spectra obtained from samples (initial Sn: 20%, thickness: 100 nm) (a) before and after laser irradiation (fluence: 10, 110 mJ/cm^2) and (b) those after subsequent annealing (170°C, 5 h).

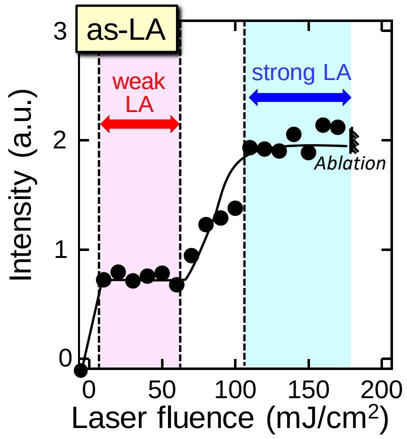

Fig.2. Laser fluence dependence of Raman peak intensity for samples (initial Sn: 20%, thickness: 100 nm). Two flat regions, i.e., weak LA (10−60 mJ/cm^2) and strong LA regions (≥110 mJ/cm^2), are observed.

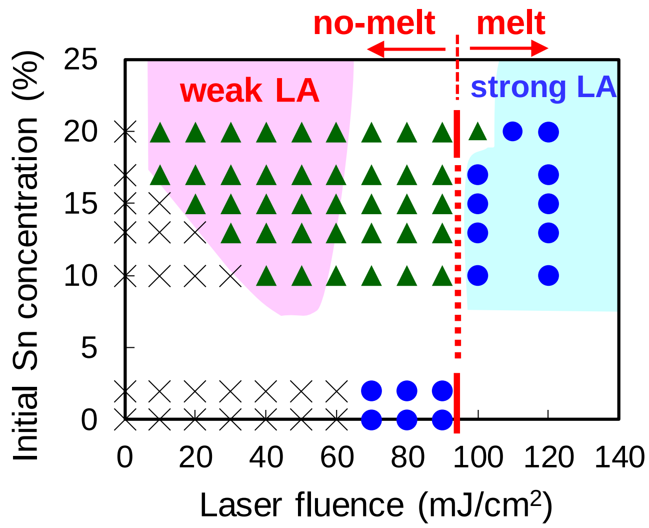

Fig.3. Summary of growth features for samples (thickness: 100 nm). ×, ▲, and ● represent detection of no Raman peak, detection of small and/or intermediate Raman peak and enhanced SPC, and detection of large Raman peak without enhanced SPC, respectively. The boundary between no-melt and melt is shown by the straight line.

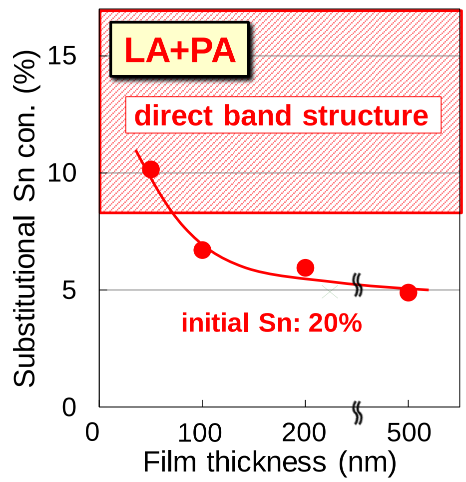

Fig.4. Film thickness dependent substitutional Sn concentration for samples (initial Sn: 20%) after weak laser irradiation (50 mJ/cm^2) and subsequent annealing (180°C, 5h). Direct band structure is expected for sub. Sn concentration above 8% [1].