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Seed Layer Technique Leading to High-photoresponsivity GaAs Films on Glass

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Abstract

We achieved the crystal growth of a pseudo-single-crystal GaAs film on glass using a Ge seed layer formed by Al-induced layer exchange. The resulting GaAs layer exhibited the grain size of 100 μm and the internal quantum efficiency of 70% with a bias voltage of 1.0 V. These values are the largest among the GaAs films grown on amorphous substrates at low temperatures (< 600 °C).

1. Introduction

The III-V compound semiconductors have continued to update the highest conversion efficiency of solar cells. GaAs has a suitable bandgap for a solar cell (E_g = 1.4 eV) and a large absorption coefficient. However, the bulk GaAs substrate is too expensive for consumer application. This motivates researchers to substitute bulk GaAs substrates with GaAs thin films on inexpensive substrates including glass. Conversely, we have formed a large-grained (> 100 μm) Ge layer on glass using Al-induced layer exchange (ALILE) [1]. Here, we applied ALILE-Ge formed on glass to an epitaxial template for a GaAs film.

2. Experimental Procedure

The 50-nm-thick Ge seed layer was prepared using ALILE (Fig. 1(a)). In the ALILE process, 50-nm-thick Al and 70-nm-thick amorphous Ge (a-Ge) thin films were sequentially prepared onto a SiO_2 glass substrate using a radio-frequency magnetron sputtering with Ar plasma. AlO_x layer between Al and a-Ge layers was formed by air exposure (5 min). The sample was annealed at 350 °C for 50 h in a N_2 ambient. After annealing, the upper Al layer was removed by HF (1.5%) treatment for 2 min. The resulting ALILE-Ge seed layer is highly (111)-oriented and large-grained (Figs. 1(b),(c)). Then, the 500-nm-thick GaAs layer was formed by molecular beam epitaxy (MBE). The GaAs layer was grown with heating the substrate at T_g = 200–520 °C. For comparison, the GaAs layers were grown on a singlecrystalline (c-) Ge(111) substrate and a bare glass substrate.

3. Result and Discussion

Raman spectroscopy was used to determine the crystal state of the GaAs films. Figure 2(a) shows that the samples with ALILE-Ge for $T_g \geq$ 370 °C exhibit sharp peaks corresponding to the transverse optical mode (~270 cm^{-1}) and the longitudinal optical mode (~290 cm^{-1}) of crystalline GaAs, while that for T_g = 200 °C exhibits broad peaks corresponding to amorphous GaAs. These results indicate that the GaAs film crystallizes at $T_g \geq$ 370 °C. Figure 2(a) also shows that the GaAs films grown on c-Ge and bare glass substrates are crystalline at T_g = 520 °C. The Raman peaks of the samples with ALILE-Ge are almost as sharp as those of the c-Ge sample, while those of the glass sample are relatively broad. These results indicate that the GaAs film on ALILE-Ge has high crystallinity. Compared to the c-Ge sample, the samples with ALILE-Ge and the glass sample have the GaAs peaks slightly shifted to the lower wavenumber. This is likely due to the stress derived from the difference in the thermal expansion coefficient between GaAs and glass. The EBSD images in Figs. 2(b) and 2(f) show that the T_g = 420 °C sample with ALILE-Ge has lower (111) orientation and smaller grain size than those of the ALILE-Ge layer (Figs. 1(b) and 1(c)), indicating the incomplete epitaxial growth of GaAs. Conversely, Figs. 2(c) and 2(g) show that the (111) orientation fraction and grain size of the T_g = 520 °C sample with ALILE-Ge are the same as the ALILE-Ge layer (Figs. 1(b) and 1(c)). These results suggest that the GaAs film was epitaxially grown from the ALILE-Ge seed layer at T_g = 520 °C. The epitaxial growth of GaAs at T_g = 520 °C is also confirmed for the c-Ge sample by Figs. 2(d) and 2(h). Figures 2(e) and 2(i) show that the GaAs film directly grown on glass

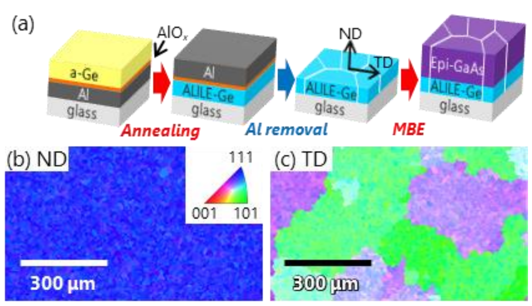

Fig. 1. (a) Schematic of the sample preparation. EBSD images of the ALILE-Ge seed layer in the (b) normal direction (ND) and (c) transvers direction (TD).

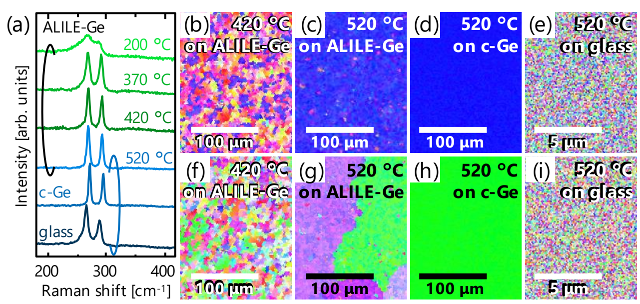

Fig. 2. Characterization of the crystal quality of the GaAs films grown at T_g = 200–520 °C on each substrate. (a) Raman spectra. EBSD images in (b)–(e) normal and (f)–(i) transvers direction.

has very small grains (< 100 nm) which are below the detection limit of the EBSD system. Thus, the pseudo-single-crystal GaAs layer (grain size > 100 μm) was achieved below the heat-proof temperature of general soda-lime glass (~560 °C).

After preparing ITO electrodes, we evaluated the detailed cross-sectional structure of the sample with ALILE-Ge for T_g = 520 °C using a transmission electron microscopy (TEM) equipped with an energy-dispersive X-ray spectrometer (EDX). Figures 3(a) and 3(b) show the stacked structure of ITO/GaAs/Ge/glass as intended. The surface of the GaAs film is flatter than that of the ALILE-Ge layer likely due to the appearance of a (111) facet during MBE. The selected-area electron diffraction (SAED) pattern in Fig. 3(c) indicates that the GaAs film is epitaxially (111) oriented and single crystalline in this area (800 nm in diameter). Figures 3(d) and 3(e) show that the GaAs film contains dislocations and stacking faults, whereas the Ge seed layer is free from extended defects. Such defects, mostly extended from the epitaxial interface, were also found in the Ge film homoepitaxially grown on ALILE-Ge at 500 °C [2]. The cause of these defects is presumed to be the rough surface of the ALILE-Ge layer and/or the low growth temperature of GaAs. Figures 3(f)–3(h) show that (111) planes are in an orderly line up from the Ge/glass interface to the GaAs film. The GaAs/Ge interface is so continuous that it is difficult to identify from the lattice image (Fig. 3(g)).

Photoresponsivity was measured for the sample with ALILE-Ge for T_g = 520 °C under standard AM1.5, 100 mW/cm^2 illumination at approximately 25 °C [3]. The resistivity of ALILE-Ge is low (< 10^{-3} Ω·cm) enough to be used as a bottom electrode due to highly Al doping. Figure 4(a) shows clear photoresponse spectra rising near a wavelength of 900 nm corresponding to the GaAs bandgap with each bias voltage. Figure 4(b) shows that internal quantum efficiency (*IQE*) reaches up to 70% with a bias voltage of 1.0 V. Figures 4(c) and 4(d) show that the sample with ALILE-Ge exhibits photoresponsivity which is comparable to that of the c-Ge sample, while the glass sample exhibits no photoresponsivity [4]. The high photoresponsivity of the sample with ALILE-Ge is owing to the GaAs film being pseudo-single-crystal. Compared with the c-Ge sample, the photoresponsivity of the sample with ALILE-Ge begins to decrease at the short wavelength side (< 850 nm) likely because of the defects in the GaAs film. Therefore, further investigation of low-temperature growth of GaAs will be able to improve photoresponsivity. However, to the best of our knowledge, the photoresponsivity is the largest currently among the GaAs film grown on a glass substrate.

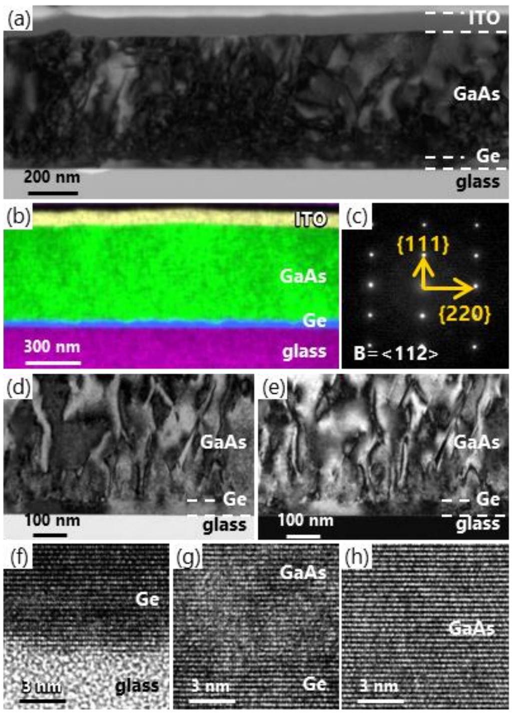

Fig. 3. (a) Low-magnification bright-field TEM image. (b) EDX elemental mapping. (c) SAED pattern. (d) High-magnification bright-field TEM image. (e) Dark-field TEM image using the (111) plane reflection. High-resolution lattice images showing the (f) Ge/glass interface, (g) GaAs/Ge interface, and (h) GaAs film.

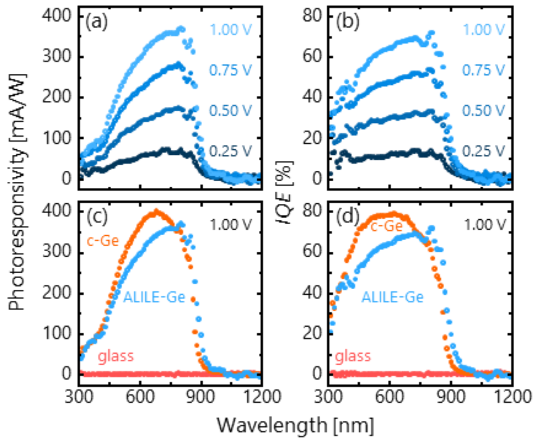

Fig. 4. (a) Photoresponsivity and (b) internal quantum efficiency of the GaAs layer with the ALILE-Ge layer for T_g = 520 °C. Comparison of (c) Photoresponsivity and (d) internal quantum efficiency among the samples with each substrate.

4. Conclusions

We demonstrated a great potential of ALILE-Ge as a seed layer for GaAs thin film solar cells. The GaAs film grown epitaxially from ALILE-Ge on glass at 520 °C became a pseudo-single-crystal (grain size > 100 μm) with high (111) orientation. Reflecting the large grain size, the photoresponsivity reached up to 70% under a bias voltage of 1.0 V. This value approached the simultaneously formed GaAs film on c-Ge and is the highest value as a GaAs film synthesized on glass. The achievements will open up the possibility of developing high-efficiency thin-film solar cells with III-V compound semiconductors based on low-cost substrates.

References

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