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Temperature dependent Al-induced crystallization of amorphous Ge thin films on glass substrates

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1. Introduction

Formation of high-quality polycrystalline Ge (poly-Ge) films on low-cost glass substrates is widely studied because of its versatile applications such as thin-film transistors [1] and multijunction solar cells [2]. In particular, the Ge (111) plane provides the highest carrier mobility of Ge transistors [3] and acts as the epitaxial templates for Ge nanowires [4] and spintronics materials [5].

Aluminum-induced crystallization (AIC) technique has been gathering much attention as a usable method to form polycrystalline Si (poly-Si) films on glass substrates, where amorphous Si (a-Si) layers on Al were transformed into crystalline phase via exchange between the Al and Si layers during the annealing [6]. Previously we found that the orientation of the poly-Si film strongly depended on the annealing temperature, and obtained highly (111) oriented poly-Si films on glass substrates by reducing the annealing temperature [7].

In this study, we investigate Al-induced crystallization of amorphous Ge thin films on glass substrates focusing on the annealing temperature dependent orientations of grown Ge films, which results in the formation of the highly (111) oriented Ge thin film on a glass substrate.

2. Experimental Procedures

50-nm-thick Al films were deposited on quartz glass substrates. Subsequently, the samples were exposed to air for 5 min to form native AlO_x layers as diffusion barrier layers, followed by 50-nm-thick a-Ge films deposited. All the depositions were carried out at a room temperature using a radio-frequency (RF) magnetron sputtering method. Finally, those samples were annealed in a N_2 atmosphere at 400, 375, 350, and 325°C, where the annealing times were 1, 10, 30, and 100 hours, respectively.

3. Results and Discussion

Figure 1(a) shows the expected schematic diagrams of respective crystallization stages corresponding to the Nomarski optical micrographs of the back surface of the sample annealed at 375^{o}C shown in Figs. 1(b). We can see that Ge crystals come to the back surface and grow laterally, which results in the layer exchange of Ge/Al layers. Figure 1(c) shows a Raman spectrum observed from the Al-induced crystallized Ge (AIC-Ge) sample after 10 hours annealing. The large peak at about 300 cm^{-1} originating at the Ge-Ge bonding confirms the formation of the crystalline Ge. Comparing the data with that obtained from a single-crystal Ge bulk wafer, the peak position shifts to the lower wavenumber. This result suggests that a tensile strain is induced in the AIC-Ge film, which probably originates from the difference of thermal expansion coefficients between the Ge and the quartz glass.

Figures 2(a)-(d) display crystal orientation mappings of AIC-Ge surfaces evaluated by means of electron backscattering diffraction (EBSD) measurements, where the annealing temperatures are 400, 375, 350, and 325°C. Here, Al layers were etched away using HF solutions before the measurements. These results clearly exhibit that the (111) orientation fraction increases with decreasing the annealing temperature, and strongly (111) oriented Ge film is achieved at 325^{o}C. The same behavior was observed in AIC for Si [7]. The reason for this preferred (111) orientation and the dependence on the annealing temperature have not been completely understood; however, the minimization of the interfacial energies between Ge nuclei and SiO_2 or AlO_x will be a possible origin.

The cross-section crystal structure of the highly (111) oriented AIC-Ge was observed by means of the transmission electron microscopy (TEM). The blight-field TEM image shown in Fig. 3(a) and the energy dispersive X-ray (EDX) analysis proved the layer exchange of Ge/Al layers and the uniformly formed Ge layer on the glass substrate. The selected area electron diffraction (SAED) pattern shown in Fig. 3(b) indicates (111) oriented Ge, which agrees with the result obtained from the EBSD measurement. The magnified dark-field TEM image exhibits a stacking fault in the Ge layer as show in Fig. 3(c). This stacking fault is almost parallel to the substrate, supposing a (111) plane defect. Any other defects except such (111) plane defects were not detected in the TEM-observed regions (width: 5 μm). Since those (111) plane defects parallel to the substrates do not appear at the surface of the AIC-Ge films, high-quality epitaxial grown layers are expected on this Ge film. In conclusion, highly (111) oriented Ge thin films are developed on glass substrate, which will be useful as the epitaxial template for multi-functional materials.

References

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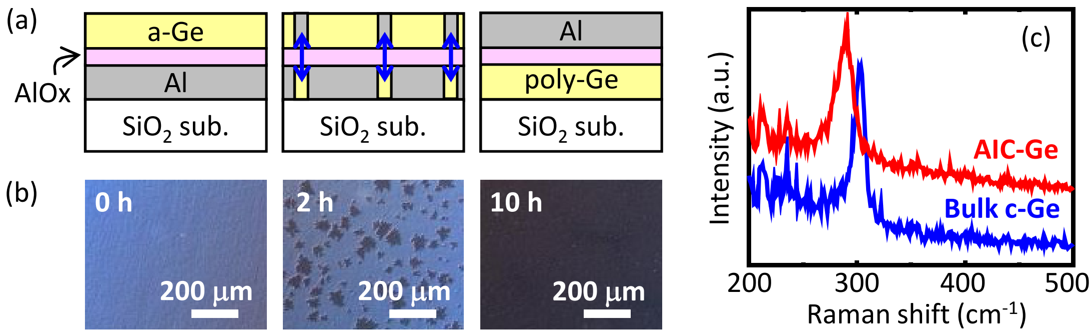

Fig. 1. (a) Schematic cross-sectional diagrams of respective crystallization stages. (b) Nomarski optical micrographs showing the annealing (375°C) time evolutions of the underside surface morphologies of the sample. (c) Raman spectra obtained from the samples of AIC-Ge and bulk c-Ge.

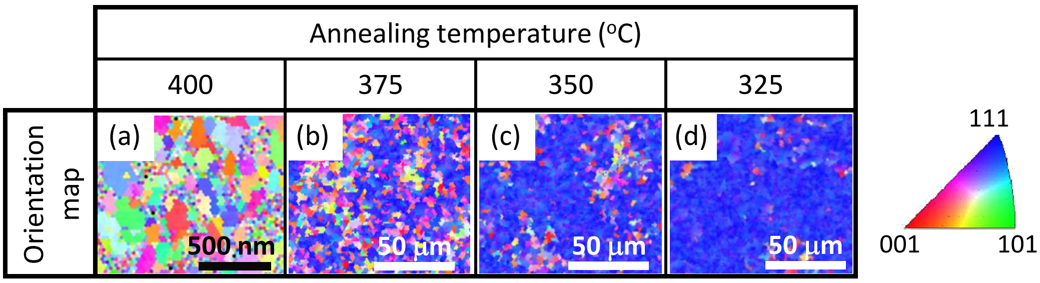

Fig. 2. (a) EBSD images of the samples annealed at 400°C (a), 375°C (b), 350°C (c), and 325°C (d). A color key corresponding to the crystal orientations is inserted.

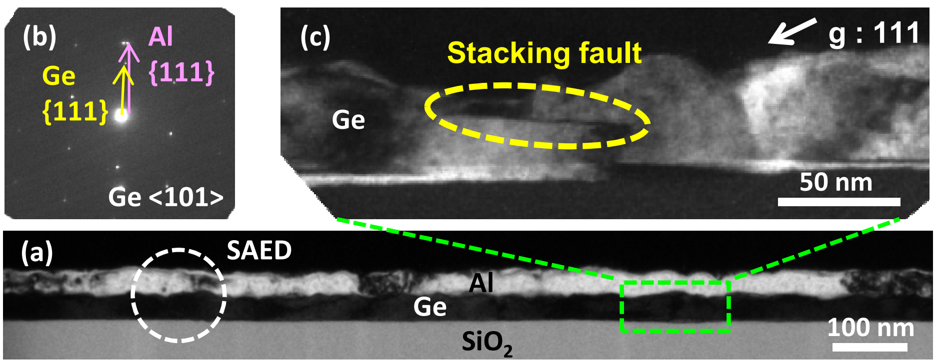

Fig. 3. A cross-sectional blight-field TEM image (a), a SAED pattern (b), and a magnified dark-filed TEM image (c) for the AIC-Ge sample.