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Structural Characterization of Polycrystalline Ge Thin Films on Insulators Formed by Diffusion-enhanced Al-induced Layer Exchange

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1. Introduction

Formation of polycrystalline Ge (poly-Ge) thin films on plastic substrates is widely studied to achieve low-cost flexible thin-film transistors and thin-film solar cells [1-2]. Because the crystallization temperature of amorphous Ge (a-Ge) is much higher than the softening temperature of plastic substrates (220 ˚C) [3], a special crystallization technique is required.

Aluminum-induced crystallization (AIC) technique has been gathering much attention as a usable method to form polycrystalline Si films on glass substrate, where amorphous Si layers on Al were transformed into crystalline phase via exchange between the Al and Si layers during the annealing [4]. Previously we have investigated Al-induced crystallization of amorphous Ge (a-Ge) thin films on glass substrates [5]. In the results, we obtained highly (111) oriented large-grained poly-Ge layer on glass substrates at 350 ˚C.

In this study, we investigate to reduce the crystallization temperature of a-Ge thin films on SiO_2 glass substrates by promoting the supersaturation on Al with Ge in the AIC technique. We accomplish the poly-Ge layers at a low temperature of 200 ˚C.

2. Experimental Procedures

1-nm-thick a-Ge layers were prepared on SiO_2 glass substrates, and followed by 50-nm-thick Al layer preparation. After that, the samples were exposed to air for 10 min to form native Al oxide layers. Subsequently, 49-nm-thick a-Ge layers were prepared on the Al oxide layers. All the depositions were carried out at room temperature using a radio-frequency (RF) magnetron sputtering method. These samples were annealed in a N_2 ambient at 325-275 ˚C. The sample structure and the growth mechanism are schematically shown in Fig. 1.

3. Results and Discussion

Figure 2 shows a comparison of the growth velocity of the samples with and without a Ge insertion layer. The sample without Ge insertion layer remains unchanged after 15 h annealing. Meanwhile, the layer exchange growth is completed within 15 h for the sample with a Ge insertion layer. These results suggest that the Ge insertion layer enhanced the supersaturation of Al with Ge, which resulted in the growth promotion of the AIC.

The crystal orientation of the sample surface with a Ge insertion layer was evaluated by means of electron backscatter diffraction (EBSD) measurement. Figure 3(a) and 3(b) show the crystal orientation maps along normal direction (ND) and transverse direction (TD) with respect to the sample surface, respectively. Here, annealing was performed at 275 ˚C for 100 h and Al layers were etched away using HF solutions (1.5%) before the measurements. From Fig. 3(a), we found that the crystallized Ge layer is highly oriented to (111) in the whole surface. The area-fraction of the (111) orientation was calculated to be 99%. Here, the (111) fraction by definition contains the planes tilted within 15˚ from the exact (111) plane. Moreover, Fig. 3(b) indicates that the grain size reaches approximately 200-μm diameters. These results indicate that the crystal quality is not degraded by the Ge insertion.

For this sample, the lower limit of the annealing temperature was found to be at 250 ˚C. This is because of the decrease of Ge diffusion rate. Therefore, we changed the interlayer from Al_2O_3 to GeO_2 for enhancing the Ge diffusion rate. The scheme is shown in Fig. 4. Firstly, a 50-nm-thick Al layer was prepared on the glass substrate, followed by a 2-nm-thick a-Ge layer preparation. Then, the samples were dipped in hydrogen peroxide solution for one min to form a GeO_2 membrane. After that, a 48-nm-thick a-Ge layer was prepared. Finally, this sample was annealed at 275-200 ˚C.

Figure 5 exhibits the annealing temperature dependent crystal-orientation maps. The samples prepared with GeO_2 interlayer enabled us to accomplish the AIC at low temperatures below 250 ˚C. This result proves that the Ge diffusion rate was enhanced by changing the interlayer. The ND orientations in Fig. 5(a)-5(e) significantly depend on the annealing temperature: The (111) orientation increases with decreasing annealing temperature. Similarly, the TD orientation maps in Figs. 5(e)-5(h) suggest that the grain sizes increase with decreasing annealing temperature. The grain size of the sample annealed at 200 ˚C is estimated to be more than 30-μm diameters.

4. Conclusion

We investigated the low-temperature AIC of the a-Ge thin film on the glass substrate. The Ge insertion layer promoted the AIC by enhancing the supersaturation of Al with Ge. Moreover, the GeO_2 interlayer strongly reduced the crystallization temperature by promoting the Ge diffusion into the Al layer. As a result, we reduced the AIC temperature to as low as 200 ˚C. The resulted Ge layer has sufficiently large grain and is highly oriented to (111). This low-temperature formation technique of poly-Ge opens up a possibility of low-cost flexible devices.

References

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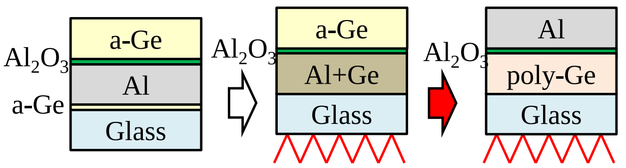

Fig. 1. Layer exchange growth of the sample with the Ge insertion layer.

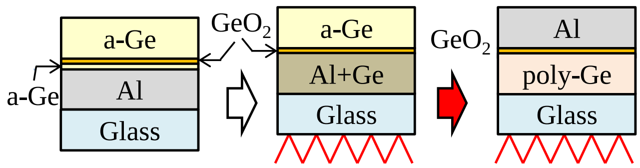

Fig. 4. Layer exchange growth of the sample with the GeO_2 interlayer.

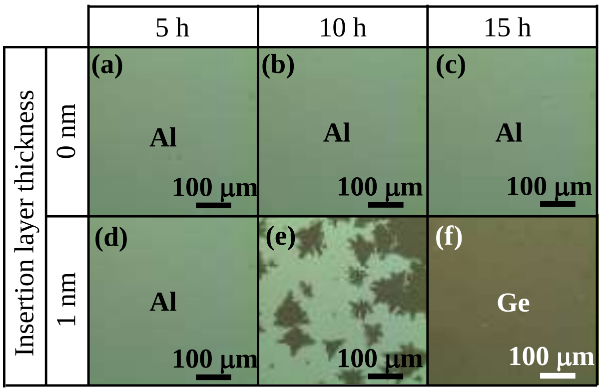

Fig. 2. Normarski optical micrographs of the back surface of the 300 ºC annealed samples (a)-(c) without and (d)-(f)with Ge insertion layer.

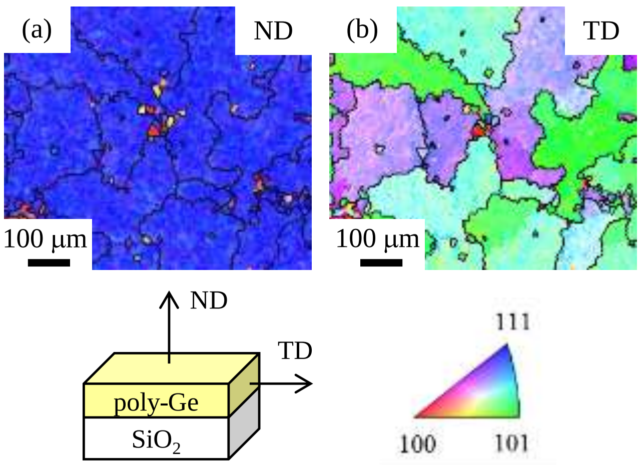

Fig. 3. Crystal orientation of the sample surface with the Ge insertion layer annealed at 275 ºC for 100 h: EBSD images along (a) ND and (b) TD.

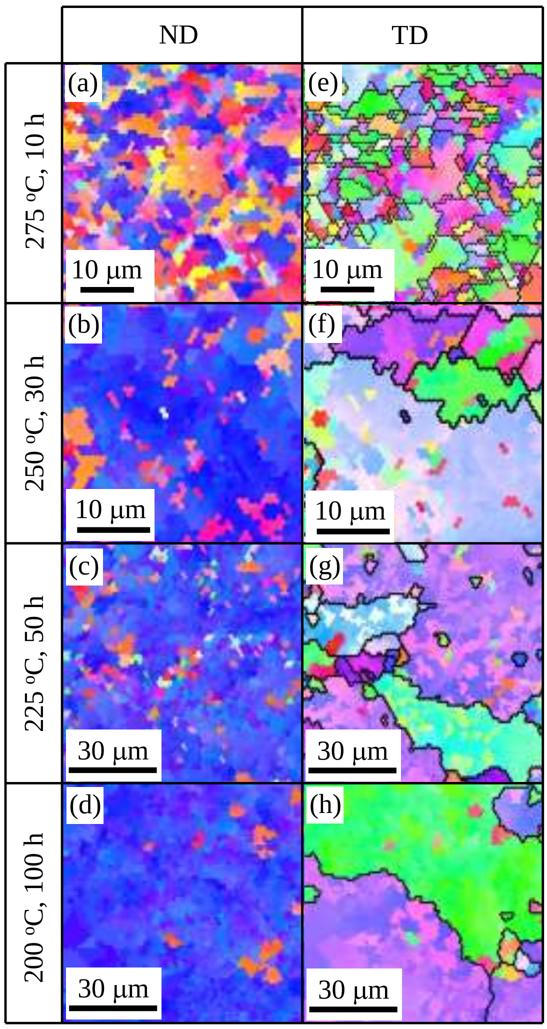

Fig. 5. EBSD images of the sample surfaces with GeO_2 interlayers along (a)-(d) ND and (e)-(h) TD.