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Effects of flexible substrate thickness on Al-induced crystallization of amorphous Ge thin films

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ABSTRACT

Amorphous germanium (a-Ge) thin films were directly crystallized on flexible plastic substrates at 325 °C using Al-induced crystallization. The thickness of the plastic substrate strongly influenced the crystal quality of the resulting polycrystalline Ge layers. Using a thicker substrate lowered the stress on the a-Ge layer during annealing, which increased the grain size and fraction of (111)-oriented grains within the Ge layer. Employing a 125-μm-thick substrate led to 95% (111)-oriented Ge with grains having an average size of 100 μm. Transmission electron microscopy demonstrated that the Ge grains had a low-defect density. Production of high-quality Ge films on plastic substrates allows for the possibility for developing Ge-based electronic and optical devices on inexpensive flexible substrates.

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1. Introduction

Germanium (Ge) is a promising candidate for next-generation electro-optical devices because of its high carrier mobility, large absorption coefficient, and narrow band gap, which is suitable for optical communication [1]. (111)-oriented Ge is particularly desirable because it provides a high carrier mobility for metal-oxide-semiconductor transistors [2,3] and acts as an epitaxial template for groups III–V compound semiconductors, aligned nanowires, and spintronic materials [4–6]. However, the high cost of bulk Ge substrates prevents widespread application of Ge-based devices. One promising approach for reducing the fabrication cost is substituting the bulk Ge substrate with a Ge thin film grown on an inexpensive substrate.

Recently, there has been interest in studying Ge on plastic substrate. Transferring single-crystal Ge membranes to plastic substrates has been widely investigated [7–9]. However, there have been difficulties in lowering the processing cost and fabricating large-area devices such as displays and solar cells. This motivated researchers to synthesize high-quality Ge directly onto plastic substrates below softening temperatures of the plastic (e.g., below 400 °C for polyimide). Because the solid-phase crystallization of amorphous Ge (a-Ge) requires temperatures higher than 400 °C [10,11], metal-induced crystallization (MIC) has received attention as a way to lower the crystallization temperature of a-Ge [12–21]. Researchers have produced polycrystalline Ge (poly-Ge) on plastic substrates using MIC [19–21]. In these cases, the crystal quality of the Ge layers was poor or the methods relied on the expensive catalytic metals.

Layer exchange between an amorphous semiconductor film and a catalytic metal is important to form a large-grained semiconductor film using MIC [22–27]. We investigated the Al-induced crystallization (AIC) of a-Ge and achieved a large-grained, (111)-oriented poly-Ge on glass at low temperatures (180–375 °C) by controlling the layer exchange velocity [28–30]. In the present study, we developed a method to apply the AIC technique to flexible plastic substrates, which has many advantages compared with glass, including lower weight, ease of handling, and reduced cost. We found that the substrate thickness strongly influenced the crystal quality of AIC-Ge. Our fabrication process yields a large-grained, highly (111)-oriented Ge thin films.

2. Experimental details

Fig. 1 presents a schematic of the sample preparation process. We employed polyimide films (thickness: 12–125 μm) as substrates. The softening temperature of the polyimide is approximately 400 °C, which is sufficiently higher than the temperature required for the AIC of a-Ge [29]. Because the crystal quality of AIC-Ge depends on the substrate materials, the polyimides were coated with a 100-nm-thick SiO_2 film, which has been shown to yield high-quality Ge [30]. Then, 50-nm-thick Al layers were deposited onto the SiO_2 films, and then exposed to air for 10 min to form native AlO_x membranes as diffusion-limiting layers [28,29]. Finally, 45 nm of a-Ge was deposited on the AlO_x. All of the depositions were performed at room temperature using radio frequency magnetron sputtering (base pressure: 3.0×10^{-4} Pa)

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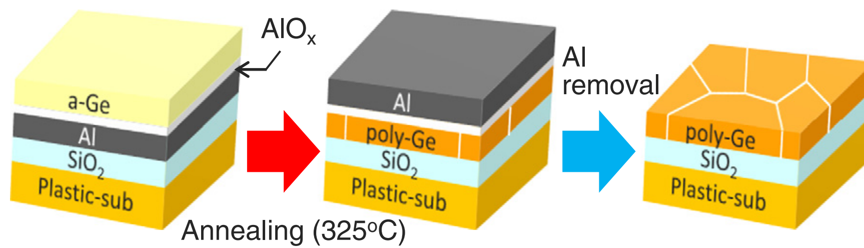

Fig. 1. Illustration of the process of AIC of a-Ge on a polyimide substrate coated with SiO_2.

with an Ar plasma. The deposition rate was 28 nm min^{-1} for Ge and 31 nm min^{-1} for Al. Pieces of Si substrates were used to fix the samples to the vacuum chamber stage and prevent the samples from moving during deposition. Following Ge deposition, the samples were annealed at 325 °C in N_2 for 100 h to induce layer exchange between Ge and Al.

The crystal states of the resulting poly-Ge layers were evaluated by using a θ–2θ X-ray diffraction (XRD) measurement (spot size: 10 mm). The surface morphology was observed using scanning electron microscopy (SEM). The crystal orientations and the grain sizes were characterized using electron backscatter diffraction (EBSD) analysis. The fraction of (111)-oriented grains and average grain size of AIC-Ge were calculated using the EBSD analysis software, TSL OIM Analysis 7. By definition, the (111)-oriented grains included those with surface planes having orientations up to 15° from the exact (111) plane orientation. A grain was considered to be an area surrounded by random grain boundaries. Before XRD and EBSD, the surface Al layers were removed by dipping the samples into a diluted HF solution (1.5% HF) for 1 min to expose the poly-Ge layers. The cross-sectional structure and elemental composition were investigated using analytical transmission electron microscopy (TEM) and high-angle annular dark-field scanning TEM (HAADF-STEM), performed on FEI Tecnai Osiris operated at 200 kV and equipped with an energy dispersive X-ray (EDX) detector. A probe diameter of approximately 1 nm was used for HAADF-STEM. Cross-sectional TEM samples were prepared using conventional focused ion beam method.

3. Results and discussion

Fig. 2 shows the θ–2θ XRD patterns of the samples after annealing. A sharp peak at approximately 27°, corresponding to the (111) plane in crystalline Ge, was observed in all of the samples. This indicates that crystallization of a-Ge was achieved at 325 °C. The small peaks at approximately 45° and 54°, shown in the inset, correspond to Ge(220) and Ge(311), respectively. Some broad peaks, especially at approximately 22° and 26°, correspond to polyimide substrates, and therefore these peaks are more intense for thicker substrates. Based on reference data from Ge powder diffraction patterns (JCPDS 04-0545), the XRD patterns indicate that the Ge layers are preferentially (111)-oriented for all samples. We note that samples with the thicker substrates have stronger Ge(111) peaks, indicating increased occurrence of the (111) orientation.

Fig. 3(a)–(f) shows the cross-sectional elemental maps obtained by STEM–EDX analysis of the annealed sample with a 125-μm-thick polyimide substrate, which show the stacked Al/Ge/SiO_2/polyimide structure that results from the layer exchange between the Al and Ge

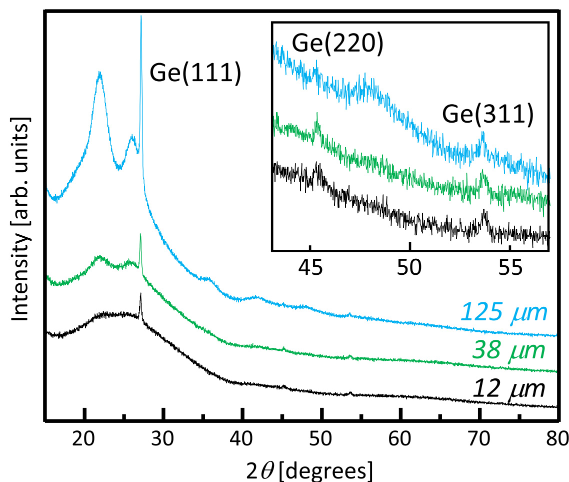

Fig. 2. θ–2θ XRD patterns taken from the samples after annealing and removing the Al layer using 1.5% HF. The thicknesses of the polyimide substrates are 12 μm, 38 μm, and 125 μm. The insertion shows magnified patterns around the Ge(220) and Ge(311) peaks.

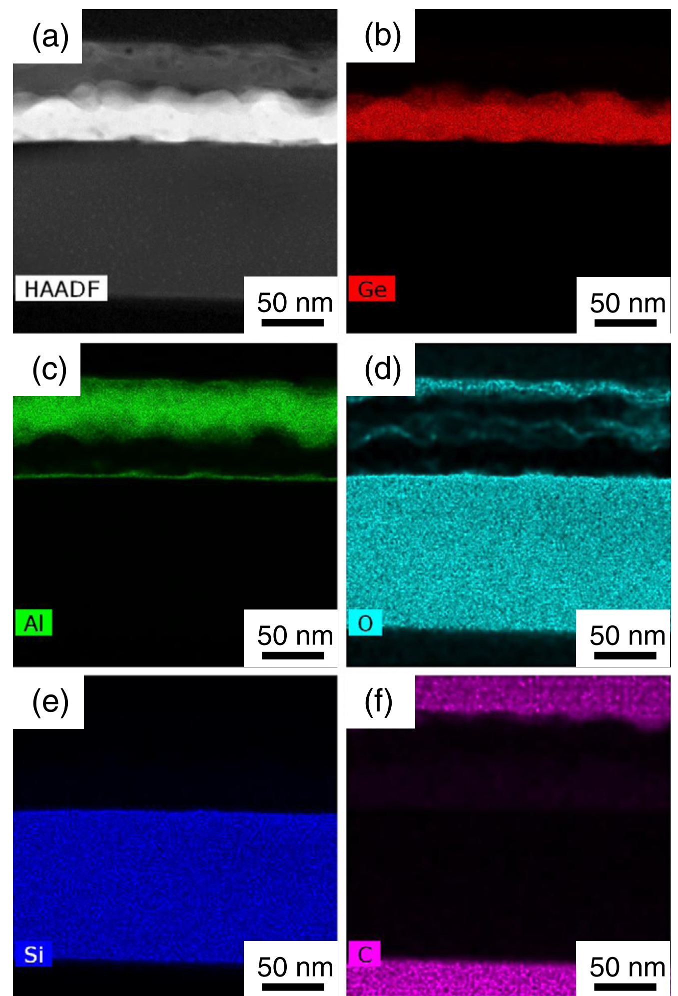

Fig. 3. Cross-sectional HAADF-STEM and EDX images of AIC-Ge grown on a 125-μm-thick polyimide substrate coated with SiO_2. (a) HAADF-STEM image. Elemental maps of (b) Ge, (c) Al, (d) O, (e) Si, and (f) C from the same region obtained using EDX.

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