



High (111) orientation poly-Ge film fabricated by Al induced crystallization without the introduction of AlO_x interlayer

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ARTICLE INFO

Article history:

Received 16 March 2015

Received in revised form 29 June 2015

Accepted 24 July 2015

Available online 26 July 2015

Keywords:

A. Amorphous materials

A. Semiconductor

B. Epitaxial growth

B. Phase equilibria

ABSTRACT

High (111) orientation poly-Ge film was fabricated by Al induced crystallization (AIC), where Al and amorphous Ge (a-Ge) layers were continuously deposited by magnetron sputtering, avoiding the deliberate introduction of an AlO_x interlayer. To improve the quality of poly-Ge film, the ratio of thicknesses of Al and a-Ge was adjusted. Electron backscattered diffraction (EBSD) results revealed that the (111) fraction of poly-Ge film reached 97% and the average crystal grain size surpassed 100 μm .

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1. Introduction

High-quality polycrystalline germanium (poly-Ge) films on insulating substrates are essential materials for high-speed thin-film-transistors and high-efficiency solar cells [1,2]. (111)-oriented Ge film provides the highest carrier mobility for Ge transistors and acts as an epitaxial template for Ge material [3,4]. To produce these films, many advanced growth techniques have been proposed, like solid-phase crystallization (SPC), laser annealing [5], and chemical vapor deposition (CVD). However, the Ge films from these methods tend to have small grain sizes and random orientations. Another, more promising fabrication approach is Al induced crystallization (AIC), which has been widely investigated in the past several decades [6–10]. Using this technique, high (111) orientation and large crystal grains can be acquired, with a lower operational temperature. When fabricating poly-Ge film, the main focus is on three aspects: (111) fraction, grain size, and Al residue in the Ge layer. Poly-Ge film with a 97–99% (111) fraction and an average crystal grain size of above 100 μm has been successfully fabricated in the recent years [11–14]. These materials resulted from the use of the molecular beam epitaxy (MBE) technique together with the introduction of AlO_x interlayer, which was regarded as essential for controlling crystal orientation. However, this increases the

complexity of the fabrication process and the employment of MBE makes this experiment costly. In addition, the issue of Al residue in the Ge film after corrosion is avoided.

In this paper, two further contributions are made. The fabrication steps mentioned above are simplified, reducing the influence of random variables which may be introduced by air exposure. The employment of magnetron sputtering leads to lower production costs, making it more feasible to manufacture the material on a large scale. The issue of Al residue after corrosion is also discussed and the source is analyzed.

2. Experimental

2.1. Materials fabrication and Al removal process

Al film was first deposited at room temperature onto a degreased quartz glass (SiO_2) substrate using the magnetron sputtering method. The a-Ge film was subsequently prepared on the Al film without breaking the vacuum, which was kept at lower than 10^{-3} Pa. Samples were then annealed at 330 °C for 100 h in a N_2 atmosphere. To study the influence of the ratio of Ge and Al thicknesses on AIC, the thickness was changed by adjusting the sputtering time. Sample labels are shown in Table 1. Samples C and D were added to improve the results after the initial experiment. Acid corrosion proceeded by soaking samples in an acidic solution ($\text{H}_3\text{PO}_4:\text{HNO}_3:\text{CH}_3\text{COOH}:\text{H}_2\text{O}=9:1:1:1$) for 15 min in order to remove the Al in the top layer.

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Table 1
Sample definitions.

| Sample no. | Thicknesses of a-Ge and Al | Ratios |
|------------|----------------------------|--------|
| A | 40 nm and 80 nm | 0.5 |
| B | 60 nm and 60 nm | 1.0 |
| C | 60 nm and 55 nm | 1.1 |
| D | 65 nm and 55 nm | 1.2 |
| E | 80 nm and 40 nm | 2.0 |

2.2. Characterization

PHI Quantum 2000 X-ray photoelectron spectroscopy (XPS) was used to study the depth distribution of Al and Ge atoms in the film. Morphology imagery was collected by an OLYMPUS laser scanning confocal microscope. Since the quartz substrate was transparent, it was feasible to observe the morphologies from the bottom of the sample. X-ray diffraction (XRD) measurements were performed using a Rigaku system with a wavelength of 1.54 Å (Cu, K-alpha radiation). The grain size and crystal orientation were evaluated by an Oxford HIKARI XP2 electron backscattered diffraction (EBSD) measurement system with samples being previously processed by an Ion Beam Technology PG-11A ion miller.

3. Results and discussion

To carry out a preliminary study, Samples A, B, and E were first deposited and annealed. The ratios of Ge to Al were 0.5, 1, and 2 respectively. Fig. 1 is the laser scanning confocal microscope images of morphologies in top and bottom views of samples with different ratios of Ge and Al. In the as grown sample, Al was first deposited on a quartz substrate (visible as a white color in Fig. 1(f)) and then a-Ge was fabricated on the Al (visible as a dark color in Fig. 1(f')). After annealing, Sample A had a white surface in the top view, indicating that Al moved to the top layer (Fig. 1(a)). During this process, Al dissolved into the Ge and then separated out from the Ge when the concentration of Al atoms dissolving into the Ge reached a certain threshold. Ge diffused to the back surface and laterally grew in discrete snow-like patterns rather than into a continuous film (Fig. 1(a')). With a thickness ratio of 1:1 [6,13], Sample B had a visibly continuous morphology from the bottom (Fig. 1(b')). Both Al and Ge could be seen in the top view (Fig. 1(b)). However, for Sample E, a Ge film was not fabricated on the bottom (Fig. 1(c')). In the subsequent measurements, as described later, it was found that the Ge content at the bottom was low and so the sample could be easily removed by the acidic solution. Hence, additional Samples C and D were added to improve the results, where the Ge to Al thickness ratios were increased to 1.1 and 1.2 respectively. After AIC, compared with Sample B in Fig. 1, it

could still be seen that Ge films were fabricated at the bottom of Samples C and D (Fig. 1(d') and (e')), and the content of Ge in the top layer (Fig. 1(d) and (e)) was higher than that in Fig. 1(b), indicating that the phenomenon was stable.

Since for Samples B–D, the Ge film on the bottom was almost entirely intact from the bottom morphology, XPS measurements of these areas were collected in order to further characterize their atomic depth distributions. Sample A was chosen to serve as a contrast. Fig. 2 is Atomic composition from XPS measurements of samples with different thicknesses ratios after AIC. The Ge content of Sample A was only 36%, although Ge in sample A mainly gathered at the bottom. As the thickness ratio of Ge and Al increased, so did the Ge content in this region: for Samples B–D, the content of Ge at the bottom layer comes to 57%, 64%, and 61% respectively after annealing. However, the content of Ge in the top layer also increased, which corresponds to visible trends in Fig. 1(c)–(e). The distribution of Ge was divided into two parts for samples with increased relative Ge content.

To check how continuous the Ge film fabricated on the bottom layer was, an acid corrosion test was carried out. Fig. 3 is the atomic depth distributions after the corrosion and the corresponding morphologies. Samples A and B, shown in Fig. 3(a') and (b') respectively, were not continuous. The white background under the transparent quartz substrate was exposed. However, for Samples C and D in Fig. 3(c') and (d'), the Ge film was shown to be unbroken. To get quantitative data corresponding to morphologies, XPS measurements were taken again to indicate the amount of Al residue. As seen in Fig. 3(a) and (b), Samples A and B had a high Si content from the quartz substrate visible from the beginning of testing. The detection of Si stemmed from the fact that the Ge film was not continuous and the quartz substrate was exposed. However, for Samples C and D, the content of Si was zero until the fourth data point, indicating that the Ge film was intact.

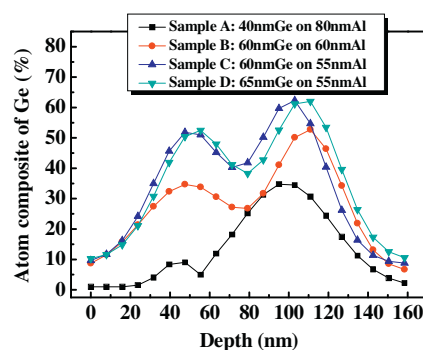


Fig. 2. Atomic composition from XPS measurements of samples with different thicknesses ratios after AIC.

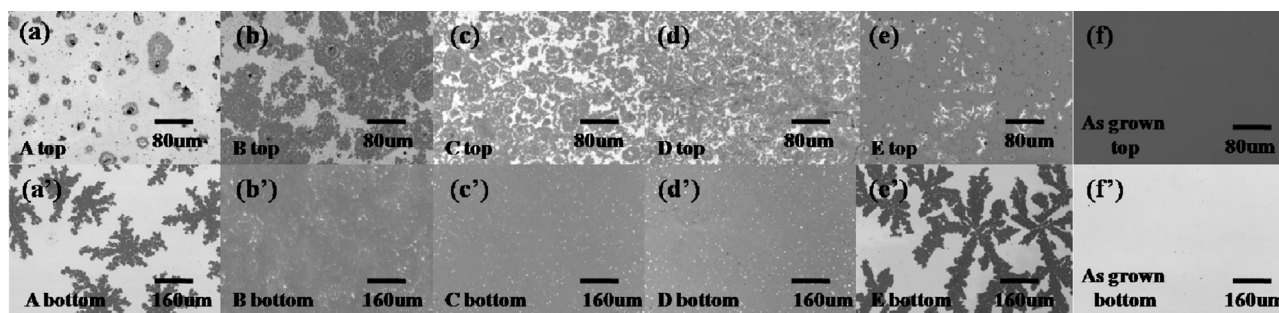


Fig. 1. Laser scanning confocal microscope images of morphologies in top and bottom views of samples with different ratios of Ge and Al thicknesses after annealing. (a)–(e) are the top views of Samples A–E, respectively. (a')–(e') are their corresponding bottom views. (f) and (f') correspond to samples as grown (without annealing) respectively, in which Al and a-Ge were repeatedly deposited on the quartz substrate in order.

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