Effects of deposition temperature and pressure of the surface roughness and the grain size of polycrystalline $Si_{1-x}Ge_x$ films

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The effects of temperature and pressure on the surface roughness and the grain size of poly-$Si_{1-x}Ge_x$ films, and the effect of the initial surface state on the final film surface roughness and grain size, have been investigated. The deposition temperature and pressure were varied from 450 to 600 °C and from 1 to 50 Torr, respectively. The transition temperature from amorphous to polycrystalline during the deposition was about 525 °C for the $Si_{0.46}Ge_{0.54}$ alloy film and the average grain size of the film deposited at 600 °C and 3 Torr was measured approximately as 180 nm. As the temperature increased, the grain size and the rms (root mean square) surface roughness increased at constant pressure, whereas both were decreased with increasing pressure at constant temperature. The initial surface state of $Si_{1-x}Ge_x$ film influenced the final film structure such as the surface roughness and grain size. The smooth surface was obtained at higher pressure and lower temperature.
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1. Introduction

Polycrystalline silicon-germanium(poly-$Si_{1-x}Ge_x$) alloy has been investigated as an alternative to poly-Si in TFT's (thin film transistors) [1, 2]. Poly-$Si_{1-x}Ge_x$ film can be deposited and crystallized at lower temperatures (≤600 °C) than poly-Si film by the addition of Ge to the film, therefore, a lower thermal budget can be used to fabricate poly-$Si_{1-x}Ge_x$ TFT's compared to poly-Si TFT's [3]. A poly-$Si_{1-x}Ge_x$ film with large grain size and smooth surface is needed to achieve high mobility TFT's. Several methods have been explored to enlarge the grain size in poly-$Si_{1-x}Ge_x$ film. These methods, the deposition of amorphous $Si_{1-x}Ge_x$ film with subsequent crystallization and the amorphization of poly-$Si_{1-x}Ge_x$ film by Si^+ implantation with post-recrystallization [4, 5], however, have disadvantages such as the process complexity and a higher thermal budget due to the annealing process after $Si_{1-x}Ge_x$ film deposition. In order to solve these problems, it is required to deposit $Si_{1-x}Ge_x$ film in direct polycrystalline form, thereby simplifying the process. In this case, because the grain size of as-deposited poly-$Si_{1-x}Ge_x$ films is very small (<100 nm), a process with higher deposition temperature and Ge content is required to obtain increased grain size [6, 7]. On the other hand, the surface roughness has been demonstrated to degrade carrier mobility as well as the high-stability of TFT's [8–10]. Violette *et al.* [11] reported that the surface roughness of poly-Si film improved at higher deposition pressure since the initial nucleation density increased with increasing pressure. Therefore, it is reasonable to think that the deposition has to be conducted under the condition of higher deposition pressure to have smooth surface. Consequently, in order to obtain increased grain size and smooth surface in as-deposited poly-$Si_{1-x}Ge_x$ film, it is necessary to deposit film under the conditions of higher temperature and pressure. However, in the previous studies little attention had been paid either to the process of higher deposition pressure condition (>1 torr) or to the initial surface state of $Si_{1-x}Ge_x$ film. For this reason, we have attempted to investigate the variation of the grain size and the surface roughness of poly-$Si_{1-x}Ge_x$ film with the deposition temperature and pressure (>1 torr) at constant Ge content, particularly the effect of the initial surface state on final poly-$Si_{1-x}Ge_x$ film surface roughness and grain size.

2. Experimental

The poly-$Si_{1-x}Ge_x$ films were deposited by the rapid thermal chemical vapor deposition (RTCVD) on thermally oxidized Si (100) wafers. The wafers were cleaned in $H_2O : HCl : H_2O_2$ at 70 °C for 5 minutes followed by deionized water rinse and N_2 drying. The wafers were put into the reaction chamber immediately after cleaning. The chamber was purged with high-purity argon for about 3 minutes. Prior to $Si_{1-x}Ge_x$ film deposition, a thin buffer Si layer was deposited on SiO_2 at 470 °C for 4 min to provide nucleation sites [12]. In sequence, poly-$Si_{1-x}Ge_x$ film was deposited by RTCVD method using SiH_4 (5% SiH_4 in

Ar) and GeH_4 (7% GeH_4 in H_2) as the source gases. The deposition conditions were : deposition temperature range 450–600 °C, deposition pressure range 1–50 Torr, and $SiH_4:GeH_4$ flow ratio range 1 : 1–2 : 1. After the deposition, the films were etched off using KOH and the thickness was determined by the surface profiler. The composition of the $Si_{1-x}Ge_x$ films was measured by Rutherford backscattering spectrometry (RBS) and the phases of the films were determined by using X-ray diffraction (XRD). Atomic force microscopy (AFM) and transmission electron microscopy (TEM) were used to examine the surface morphology and the grain size of the poly-$Si_{1-x}Ge_x$ films.

3. Results and discussion

Fig. 1 shows the Ge composition change of the films deposited at 3 Torr in the $SiH_4:GeH_4$ flow ratio(1 : 1–2 : 1) with various deposition temperatures. At a constant $SiH_4:GeH_4$ flow ratio, the Ge composition decreases with increasing the deposition temperatures, which has previously been explained by the difference of the magnitude of the sticking coefficient between Si precursors and Ge precursors in the deposition of $Si_{1-x}Ge_x$ films using SiH_4 and GeH_4 gases below 600 °C [12]. Fig. 1 also shows that the Ge composition has various values as the deposition temperature and the GeH_4 flow ratio changes, however, the values exceed 0.6 in case of lower temperature and higher GeH_4 flow ratio. It has been reported [13] that when the Ge composition is above 0.6, the films are incompatible with the standard VLSI fabrication processes, so that most experimental work has been carried out under the deposition temperature range of 525 to 600 °C and the $SiH_4:GeH_4$ flow ratio range of 2 : 0.1 to 2 : 1 to obtain the films with Ge composition below 0.6. Fig. 2 shows the deposition rate of the films deposited at 550 and 600 °C as a function of the deposition pressures. It can be seen that the deposition rate increases monotonically

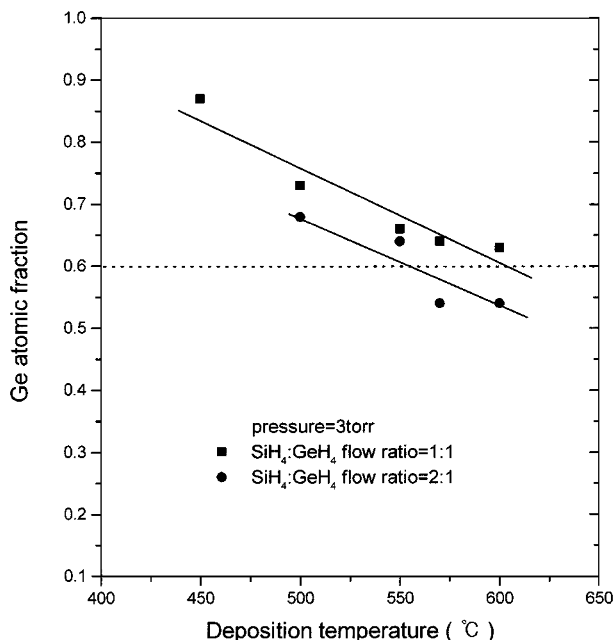


Figure 1 Ge atomic fraction as a function of deposition temperature.

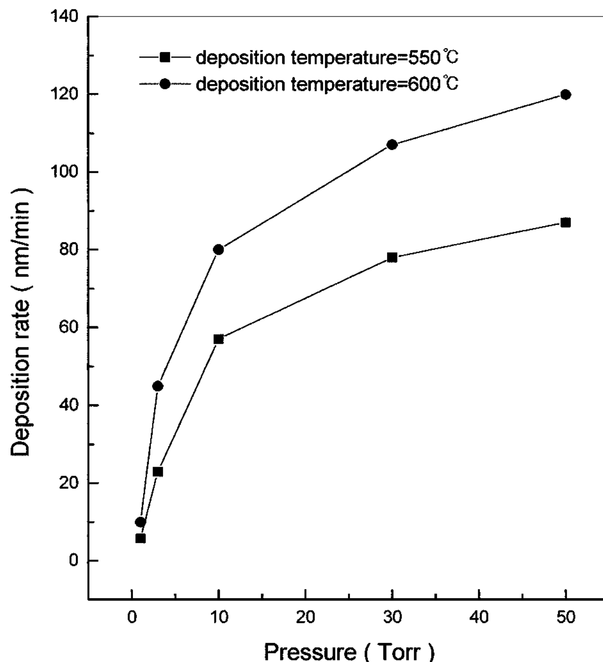


Figure 2 Deposition rate of $Si_{1-x}Ge_x$ films as a function of deposition pressures.

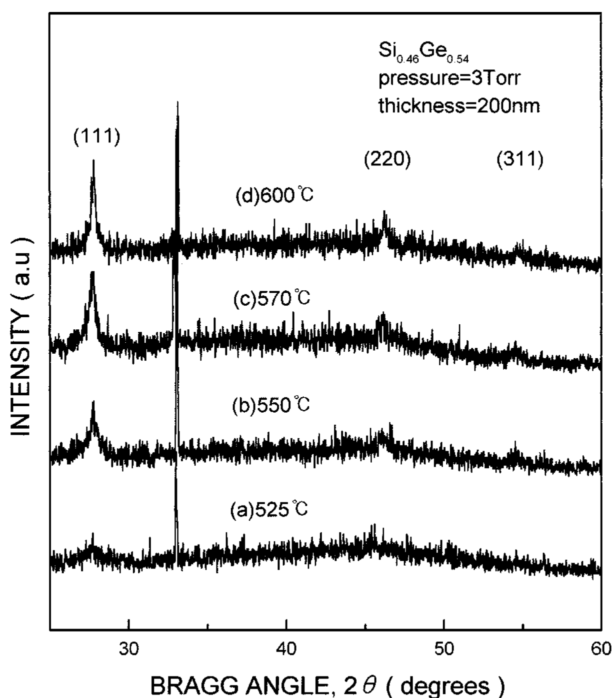


Figure 3 X-ray diffraction patterns of the $Si_{0.46}Ge_{0.54}$ films deposited at different deposition temperature.

with deposition pressure from 1 to 10 Torr and gradually increases thereafter. This deposition behavior for pressures greater than 10 Torr may be caused by limiting surface reaction rate. Fig. 3 shows the X-ray diffraction (XRD) patterns of the $Si_{0.46}Ge_{0.54}$ films deposited at various deposition temperatures. The diffraction peaks around $2\theta = 28°$, $47°$, and $55°$ represent the diffraction from the (111), (220), and (311) planes, respectively. From XRD data, it is known that the polycrystalline diffraction peaks appear in the films deposited at 550, 570, and 600 °C, whereas amorphous and crystalline

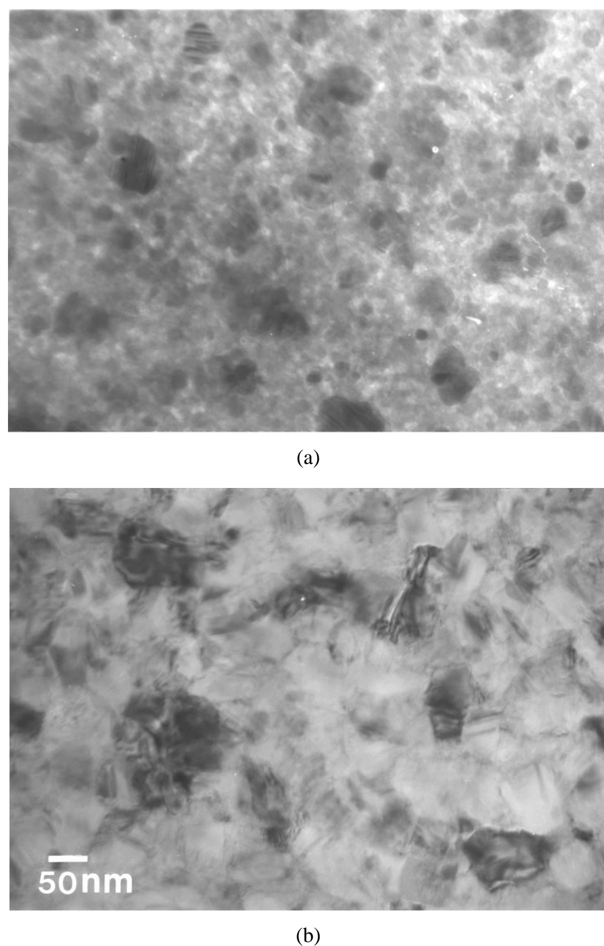


Figure 4 Plan-view TEM micrographs of the $Si_{0.46}Ge_{0.54}$ films deposited at (a) 525 °C and (b) 550 °C.

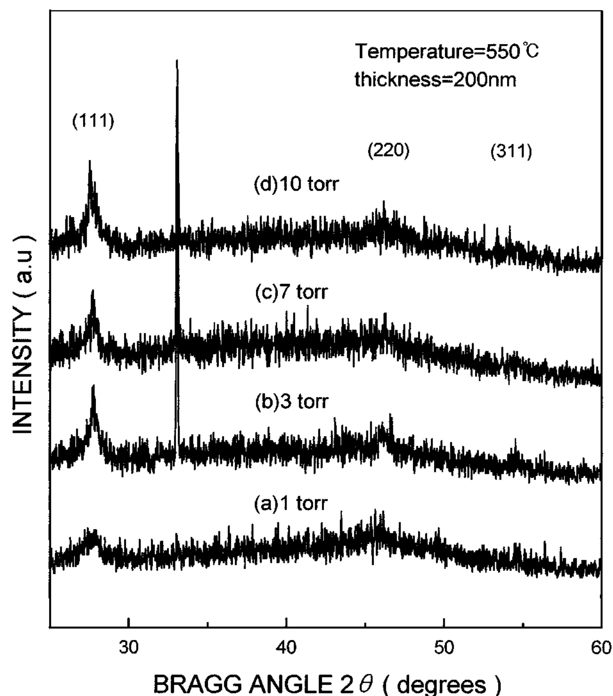
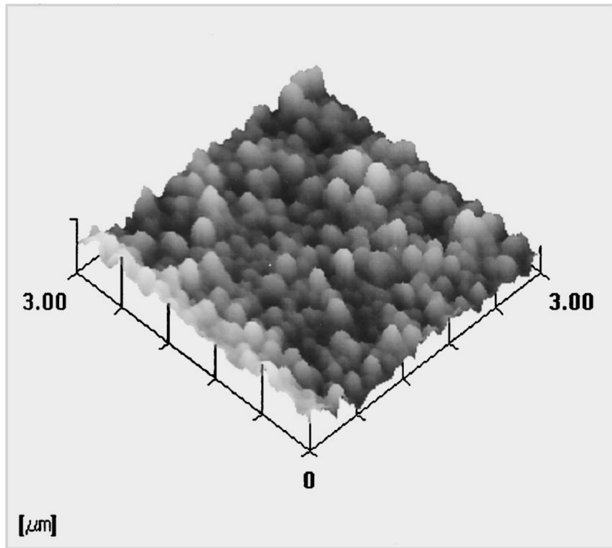


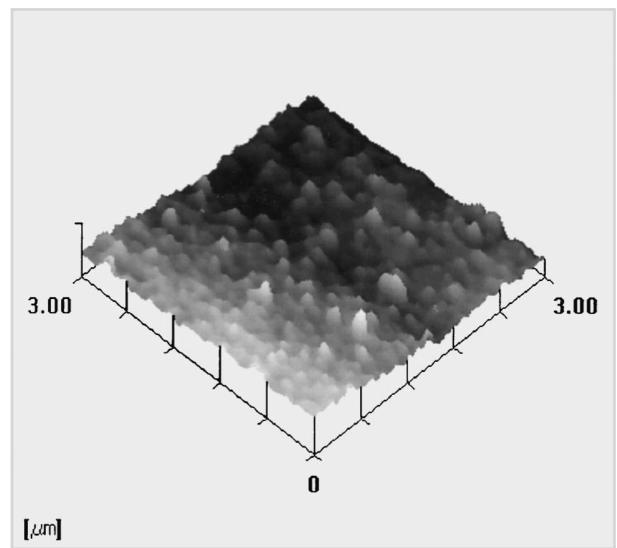
Figure 5 X-ray diffraction patterns of the $Si_{1-x}Ge_x$ films deposited at different deposition pressure.

mixed diffraction peak appears at 525 °C. Also, as the deposition temperature increases, the diffraction peak intensity of the films increases. Fig. 4 shows the plan-view TEM microphotographs of the films deposited at 525 and 550 °C. The TEM observation reveals that amorphous and crystalline phases coexist in the film deposited at 525°C and are fully crystallized at 550 °C, which is in agreement with XRD results. These results imply that the amorphous to polycrystalline transition temperature of $Si_{0.46}Ge_{0.54}$ films during the deposition is ∼525 °C. However, Lin *et al.* [14] reported that the fully crystallized poly-$Si_{0.56}Ge_{0.44}$ film was obtained at 500 °C using ultrahigh vacuum (UHV) CVD system. This difference in transition temperature may be caused by the difference between the Ge atomic concentration and the deposition pressure. Fig. 5 shows the X-ray diffraction patterns of the $Si_{1-x}Ge_x$ films deposited at 550 °C as a function of the pressure in the range from 1 to 10 Torr. As can be seen in the figure, the apparent polycrystalline diffraction peaks of as-deposited $Si_{1-x}Ge_x$ films appear above 1 Torr. From these XRD and TEM results, it can be supposed that the $Si_{1-x}Ge_x$ films are deposited perfectly in the polycrystalline form at the deposition temperatures range of 550 to 600 °C and the pressures range of 3 to 10 Torr. Therefore, we have performed all experiments under these conditions in order to investigate the effects of pressure and temperature on the surface roughness and the grain size of poly-$Si_{1-x}Ge_x$ films.

The surface morphology and roughness were investigated by AFM on the 200 nm thick poly-$Si_{0.4}Ge_{0.6}$ layers. Fig. 6 shows AFM images of the films deposited at two different temperatures and pressures, 550 °C and 600 °C, 3 Torr and 10 Torr, respectively. As shown in this figure, each sample reveals different cluster sizes and as the pressure is increased from 3 to 10 Torr, the surface morphology becomes smoother. Figs 7 and 8 show the cluster size and the rms surface roughness of the samples in Fig. 6 a–c respectively, which were measured by AFM. As shown in these figures, both the cluster size and the rms surface roughness values follow a trend that increases with increasing temperature and decreases with increasing pressure. In addition, the rms roughness of the sample obtained at 600 °C and 3 Torr shows the highest value. Following the above AFM results, it can be observed that the final film surface morphology is affected by temperatures and pressures. In general, the temperature and pressure strongly influence the film growth caused by the initial nucleation and the subsequent film surface morphology. According to Violette *et al.* [11] they pointed out that the film continuously grew in initial surface state which influence the final film surface structures such as the grain size and surface roughness. They also indicated that the initial surface state is depending on nucleation behavior which is affected by pressure and temperature. Therefore, it is necessary to analysis the initial surface state at an early stage of deposition to reveal the cause of as-mentioned AFM results, which is shown in Fig. 9. This figure shows the initial surface states at different temperatures and pressures. This observation was obtained by AFM after exposure of the input reactants for short time. Fig. 9a shows the surface image of the thin buffer Si layer, and Fig. 9b–d show initial surface states of the $Si_{0.4}Ge_{0.6}$ deposited on buffer layer. As



(a)



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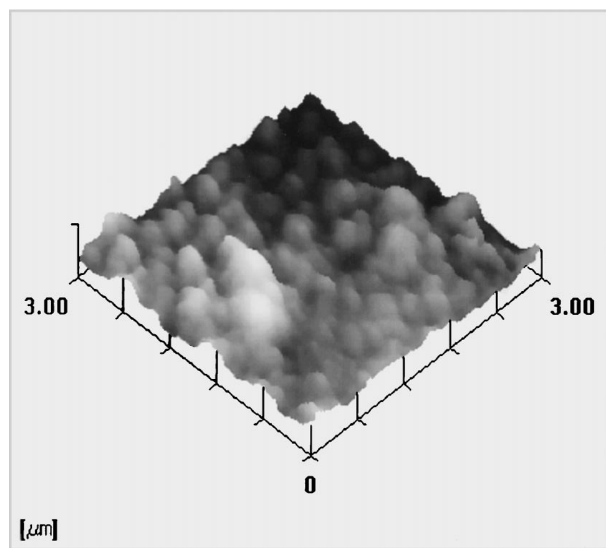

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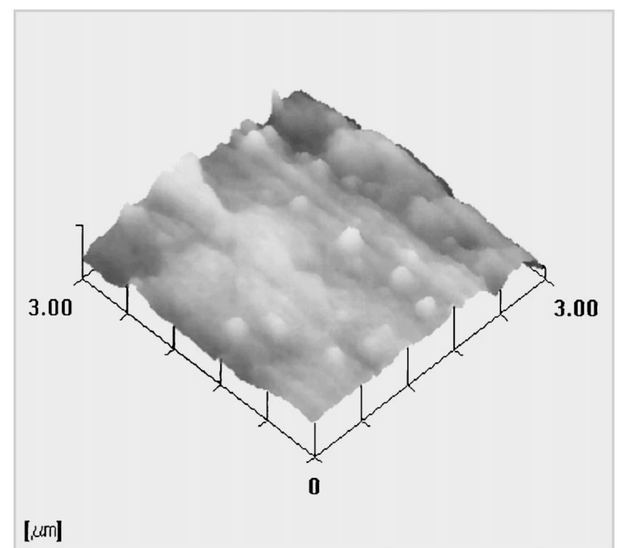

(d)

Figure 6 AFM image of the poly-$Si_{0.4}Ge_{0.6}$ films deposited at (a) 550 °C, 3 Torr (b) 550 °C, 10 Torr (c) 600 °C, 3 Torr (d) 600 °C, 10 Torr.

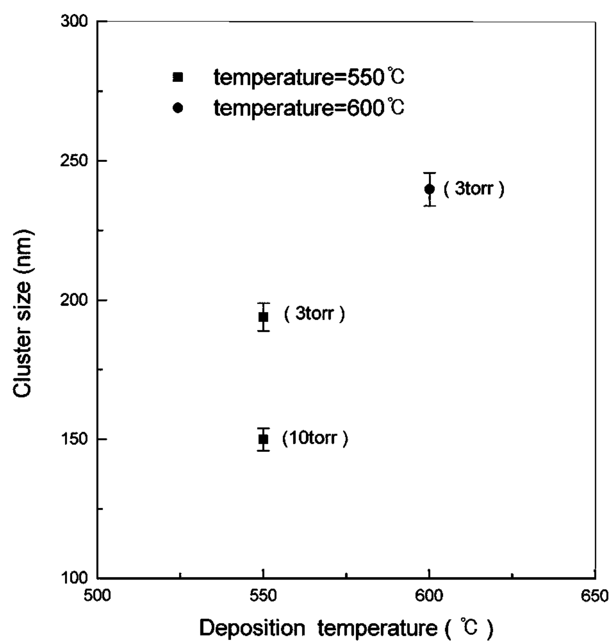

Figure 7 Variation of cluster size of poly-$Si_{0.4}Ge_{0.6}$ films deposited at 550 °C (3, 10 Torr) and 600 °C, 3 Torr.

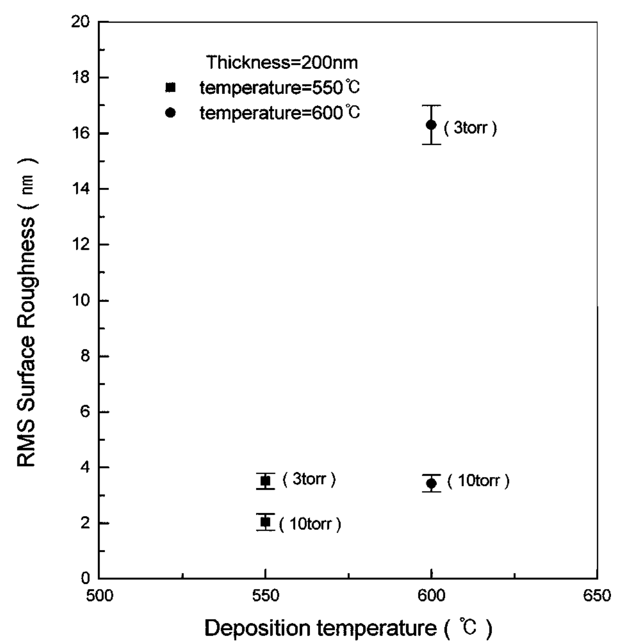

Figure 8 Rms Surface roughness of poly-$Si_{0.4}Ge_{0.6}$ films deposited at 550 °C (3, 10 Torr) and 600 °C (3, 10 Torr).

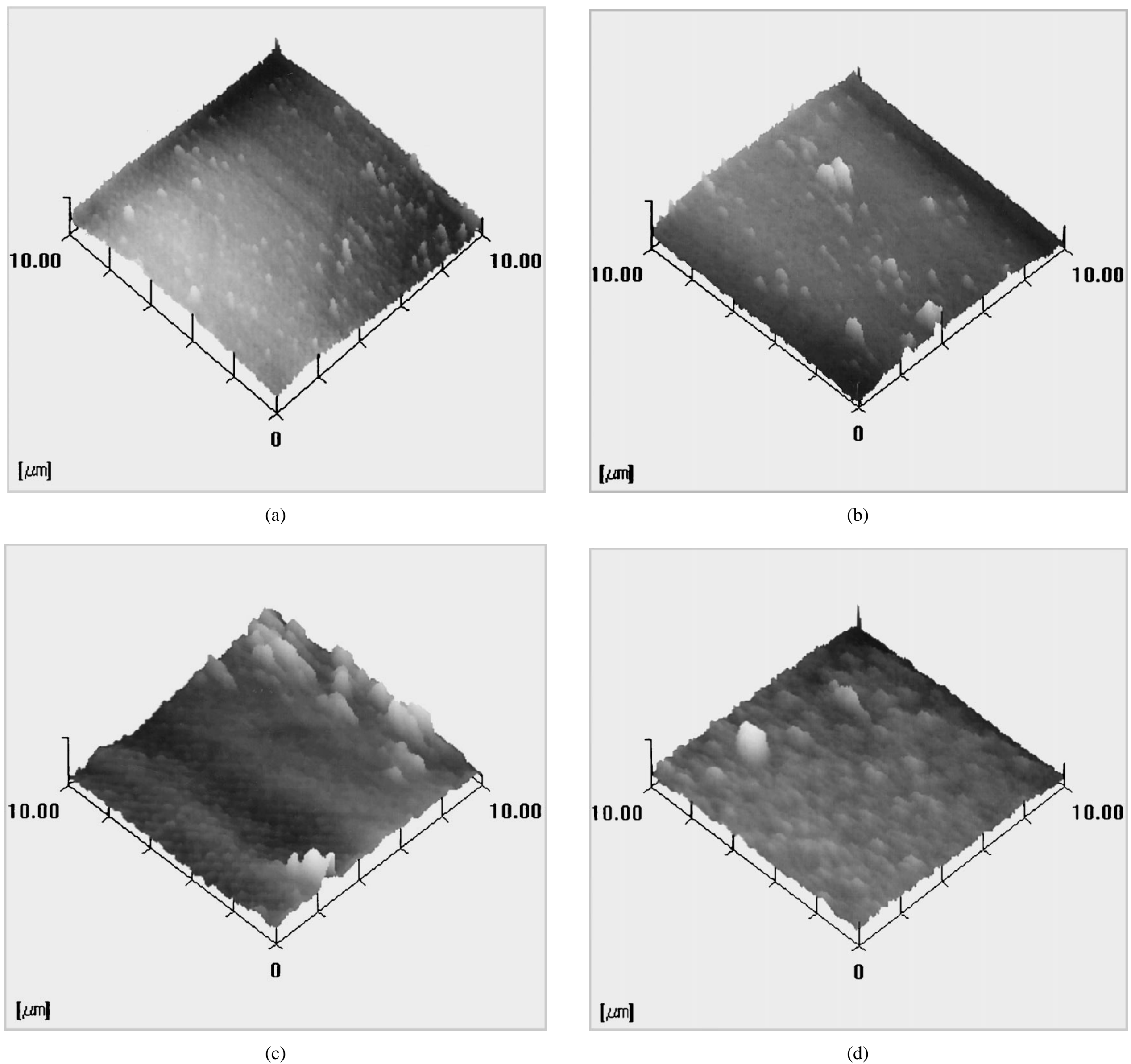

Figure 9 AFM image of the (a) buffer Si layer and initial surface states of the poly-$Si_{0.4}Ge_{0.6}$ films deposited at (b) 550 °C, 3 Torr (c) 600 °C, 3 Torr (d) 600 °C, 10 Torr.

shown in Fig. 9b and c, as the temperature is increased from 550 to 600 °C at constant pressure, sparse and large initial clusters are observed. This may be caused by the adatoms which attach more likely to an existing initial clusters than to form a new nucleus because the increased temperature provides enhanced surface mobility and diffusion length. On the other hand, as shown in Fig. 9c and d, as the pressure is increased from 3 to 10 Torr at fixed temperature, a large number of fine clusters appear. Bloem [15] pointed out that nucleation can occur more readily leading to higher nucleation densities at higher pressure. It may, therefore, be concluded that the increase in number of clusters at high pressure is caused by higher nucleation densities which lead to the formation of a large number of initial clusters. Relating this analysis of the initial surface states to the results shown in Figs 7 and 8, the effect of the temperature and pressure on the surface roughness can be explained as follows. The increase in surface roughness at higher deposition temperature is due to the sparse and large initial clusters and the decrease in surface roughness at high pressure is due to the formation of initial cluster with small and dense population as a result of the rise in the nucleation density. Consequently, it is necessary to deposit dense population of primary nuclei achieved under higher pressures to obtain the final smooth poly-$Si_{1-x}Ge_x$ film surface. Fig. 10 shows the grain size of the samples shown in Fig. 6. Similarly to the case of surface roughness, the grain size increases with increasing the temperature and decreases with increasing the pressure. The average grain size of the film deposited at 600 °C and 3 Torr is around 180 nm and is much larger than that of other samples. The nucleation sites act as seed crystallites for the diffusing adatoms and form the initial grains in a polycrystalline film [16]. As shown in Fig. 10, larger grains are more easily formed at 600 °C and 3 Torr due to larger initial clusters at high temperature and low pressure (see Fig. 9c). It is, therefore, believed that the initial surface state play an important role in the final film surface roughness and grain size, both of which are affected strongly by the deposition temperature and pressure.

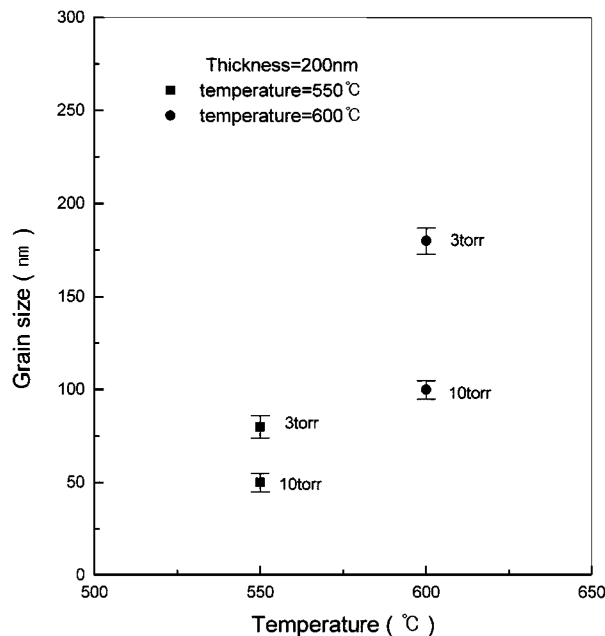

Figure 10 Average grain sizes of poly-$Si_{0.4}Ge_{0.6}$ films deposited at 550 °C (3, 10 Torr) and 600 °C (3, 10 Torr).

4. Conclusion

In this work, we have investigated the effects of pressure and temperature on the surface roughness and grain size of poly-$Si_{1-x}Ge_x$ films. Also, we have discussed that the surface roughness and grain size of poly-$Si_{1-x}Ge_x$ films are strongly related to the nucleation and initial surface state. As the temperature increased, the grain size and the rms surface roughness increased at constant pressure, whereas both decreased with increasing pressure at constant temperature. Smooth surface morphology was obtained at 550 °C and 3 Torr. At 600 °C and 3 Torr observed grain size was around 180 nm and was much larger than that of other cases. The initial surface state was thought to influence the final film surface roughness and grain size, both of which were affected by pressure and temperature. At higher deposition pressure (>1 Torr), smooth surface morphology was obtained, however, large grain size was not obtained.

Acknowledgement

This study was supported by the academic research fund of Ministry of Education, Republic of Korea, through Inter-University Semiconductor Research Center(ISRC 97-E-1028) in Seoul National University

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Received 10 November 1999
and accepted 2 March 2000