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Enlargement of Compositionally Gradient Area in Si-Ge Thin Films Prepared by Ion-Beam Evaporation

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Compositionally gradient Si-Ge thin films have been successfully prepared by irradiating an ion beam on Si and Ge plates without mask control. The compositionally gradient area in a Si-Ge thin film was approximately 33 mm to control the deposition conditions as the incident angle (θ) of ion beam or the distance (d_{TS}) between target and substrate. This result was approximately 20 mm larger than that of our previous work. One of the reasons to explain a larger compositionally gradient area was given that the thickness distribution having a gentler slope is achieved by changing θ and d_{TS}.

Key words: intense ion-beam evaporation, compositionally gradient thin films, ablation plasma

1. INTRODUCTION

Various functional materials have recently been developed. In the development of the materials, it is necessary to optimize the composition of such materials. For this purpose, many samples with various compositions have to be synthesized and an optimum composition should be determined. However, in this process, much time and effort are spent to obtain the materials having excellent properties.

For determining the optimum composition, quick material synthesis methods as "combinatorial chemistry" have been developed [1,2]. In these methods using a sputtering or a pulsed laser deposition (PLD), a thin film which has various compositions, i.e., a compositionally gradient thin film, is produced on a substrate by controlling a mask. The optimum composition is determined by characterizing this thin film. By using these methods, much time and effort can be saved for the development of novel materials. However, in order to prepare compositionally gradient thin films by those methods, the number of atoms from the sources must be varied at each position on the substrate. For this purpose, masks, which were placed between the sources and the substrate, should be precisely controlled.

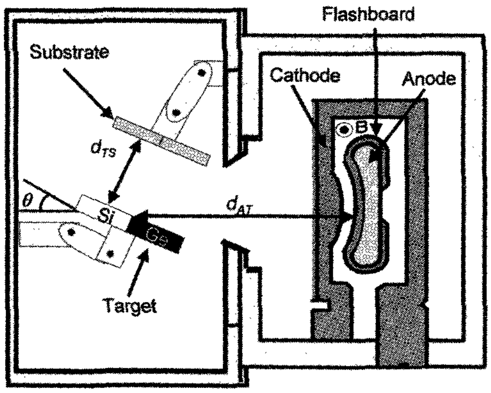
We proposed a new method for preparing compositionally gradient thin films without mask control [3]. Intense ion-beam evaporation (IBE) [4] is used for the method. An intense pulsed ion beam with energy of 1 MeV, energy density of 20~120 J/cm^2 and pulse width of 50 ns, is irradiated on a segmented target consisting of different materials. High temperature and high density (~10^{19} cm^{-3}) plasma are formed from the segmented target. A compositionally gradient thin film was deposited on a substrate.

We have previously reported compositionally gradient Si-Ge thin films prepared by the IBE method [3]. However, compositionally gradient area was 12 mm in width. For phase identification of the compositionally gradient thin films, X-ray diffractometer was used. In the measurements, irradiated width of the samples is 3 mm at sample-source distance of 200 mm, divergence angle of 0.5 ° and diffraction angle of 30 °. The irradiated width determined the compositional resolution for the phase identification in the compositionally gradient thin films. Therefore, to obtain phases of ten different compositions in a compositionally gradient thin film, it is required to have compositionally gradient area of more than 30 mm in width.

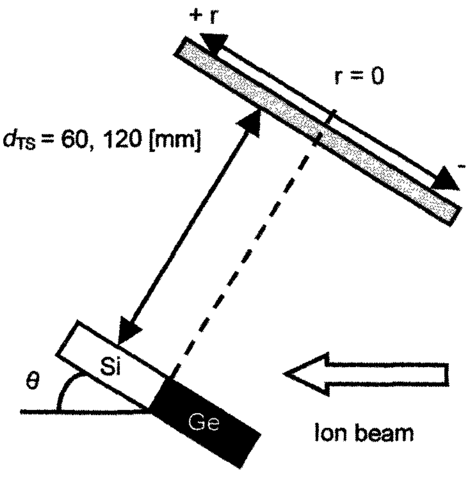
The purpose of this study is to prepare Si-Ge compositionally gradient thin films having the compositionally gradient area of more than 30 mm in width. For this purpose, we carried out (1) the optimization in preparation condition of compositionally gradient thin films and (2) the enlargement of compositionally gradient area in the Si-Ge thin film. Using the results above, the enlargement of compositionally gradient area in the Si-Ge thin film was accomplished by the ion beam irradiation on a segmented Si-Ge target.

2. EXPERIMENTAL APPARATUS AND METHOD

Figure 1 (a) shows schematic illustration of the experimental setup on the IBE system. An intense pulsed ion beam was extracted from a magnetically-insulated diode, which was connected to a pulse power generator "ETIGO-II". A polyethylene flashboard was attached to an anode as an ion source. The high voltage of 1 MV (peak) was applied between the anode and cathode with the pulse width of approximately 50 ns. To prevent a current of electrons between the anode and cathode, the transverse magnetic field of approximately 1 T was generated by the cathode as a theta-pinch coil. The ions, which were produced by flashover of polyethylene flashboard, were mainly composed of protons (more than 85 %) and some carbon ions. The ion beam passed through the cathode with vane structure, and was geometrically focused on a target. Accordingly, the energy density of the ion beam on the target was 60 J/cm^2, when the target was set at the distance (d_{AT}) of 230 mm from the anode.



(a) The schematic illustration of the experimental setup on the IBE system.



(b) The arrangement of target and substrate for the preparation of compositionally gradient Si-Ge thin film.

Fig. 1 Experimental setup.

For the preparation of thin films, single crystal Si and/or Ge plates were used as targets. This investigation was carried out for (1) the optimization in preparation condition of compositionally gradient thin films, and (2) the enlargement of compositionally gradient area in Si-Ge thin film. In the former, using the Si plate as a target, Si thin films were prepared on single crystal Si substrates by changing the incident angle (θ) of the ion beam or the distance (d_{TS}) between the target and substrate. In the latter, as can be seen in Fig. 1 (b), the compositionally gradient Si-Ge thin film was prepared on a stainless steel (SUS304) substrate by irradiating the ion beam on the target, which consisted of the Si and Ge plates. Ablation plasma induced by the ion beam irradiation was deposited on each substrate that was kept at RT in a vacuum of 1×10^{-4} Torr. The Si and Si-Ge thin films were prepared by ion beam irradiation of fifteenth and once, respectively. Experimental conditions are listed in Table I.

The thickness of the Si thin films was measured with a surface profiler. The composition of the compositionally gradient Si-Ge thin films were determined by an energy dispersive X-ray spectroscope (EDS). The EDS was equipped with a scanning electron microscope (SEM), which was operated at the acceleration voltage of 10 kV. By this voltage, the incident depth of electrons corresponded to the thickness (approximately 300 nm) of the compositionally gradient Si-Ge thin film. Thus, the intensity of X-ray from the substrate was almost same as that of the background of EDS spectra. Moreover, the composition of the thin films was determined by $\Phi(\rho, z)$ method [5] using a Cliff-Lorimer factor, which was obtained from the spectrum of a standard Si-50 at. % Ge bulk.

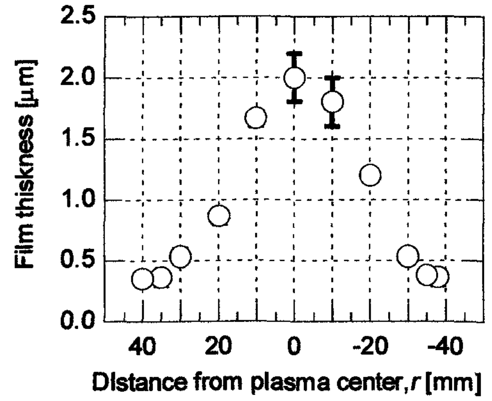
Table I Experimental conditions

Main component of ion beam	H^+
Energy density	60 J/cm^2
Distance from anode to target (d_{AT})	230 mm
Targets	Si, Ge
Substrates	Si, SUS304
Incident angle of ion beam (θ)	30, 40 °
Distance from target to substrate (d_{TS})	60, 120 mm
Substrate temperature	RT
Pressure	1×10^{-4} Torr
Number of shots	1, 15 shots

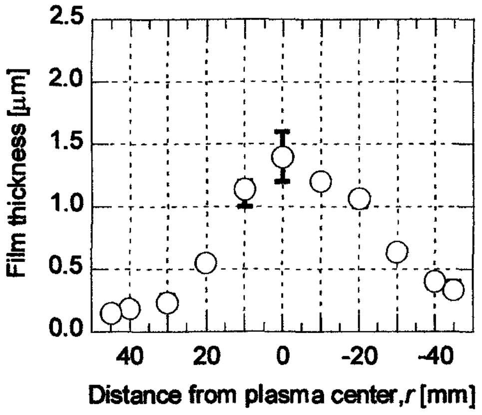
3. RESULTS AND DISCUSSION

3.1 The optimization in preparation condition of compositionally gradient thin films

Figure 2 show thickness distribution of Si thin films prepared at (a) θ = 40 and (b) 30 °. Such thin films were prepared under the condition of d_{TS} = 60 mm. In Fig. 2 (a), at the distance (r) from plasma center being r = 0 mm, the thickness of the Si thin film was the largest in the whole r range. Additionally, the thickness distribution of the Si thin film was found to be almost symmetrical to the axis of r = 0 mm. On the other hand, in Fig. 2 (b), although the thickness of the Si thin film was maximum at r = 0 mm, the distribution was unsymmetrical being inclined toward the negative r side. Therefore, the thickness distribution of the Si thin films was clarified to become unsymmetrical with decreasing θ from 40 to 30 °.



(a) $\theta = 40$ °



(b) $\theta = 30$ °

Fig. 2 The thickness distribution of Si thin films prepared at (a) $\theta = 40$ and (b) 30 °. Such thin films were prepared under the condition of $d_{TS} = 60$ mm.

Figure 3 shows schematic illustration of preparation mechanism in Si thin film with unsymmetrical thickness distribution. In Fig. 3, the Si target is assumed to present at the position of $d_{AT} = 230$ mm. The ion beam, which is extracted from the anode, is geometrically focused at the position of $d_{AT} = 140$ mm. Subsequently, the ion beam diverges before the interaction with the Si target. At the condition of $\theta = 30$ °, the distance from the anode differs 17 mm in the plane of the Si target. In our previous work, the energy density of the ion beam has been known to decrease 20 % for the difference of 17 mm [6]. Thus, a larger amount of ablation plasma seems to occur from the position in the Si target, where is nearer to the anode. As a result, the thickness distribution of the Si thin film, which was shown in Fig. 2 (b), was considered to have unsymmetrical. Moreover, to enlarge the compositionally gradient area in Si-Ge thin films, the change of the thickness distribution is required to be less. The finding leads to that the negative r region having a gentle slope in Fig. 2 (b) can be prepared Si-Ge thin film with a larger compositionally gradient area.

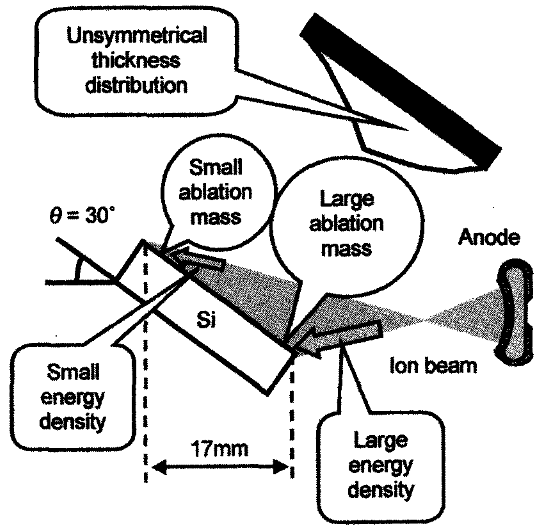
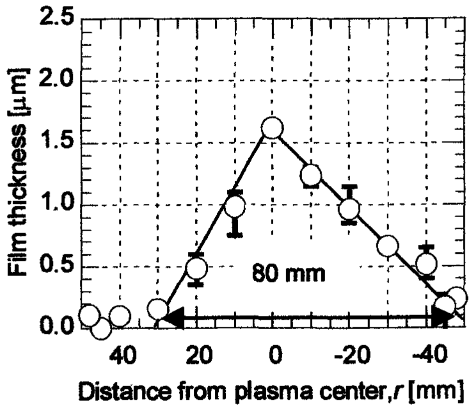
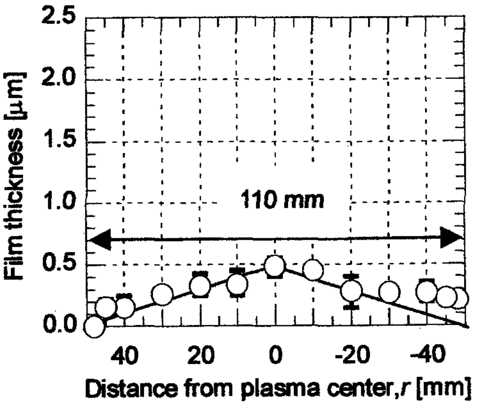


Fig. 3 Schematic illustration of preparation mechanism in Si thin film with unsymmetrical thickness distribution.



(a) $d_{TS} = 60$ mm



(b) $d_{TS} = 120$ mm

Fig. 4 The thickness distribution of Si thin films prepared at (a) $d_{TS} = 60$ and 120 mm. Such thin films were prepared under the condition of $\theta = 30$ °.

Figure 4 shows thickness distribution of Si thin films prepared at (a) d_{TS} = 60 and 120 mm. Such thin films were prepared under the condition of θ = 30 °. In Fig. 4, straight lines fitted to the measured data. The length between the points, where the lines cross the transverse axis, is defined as the spread length (l_s) of the Si thin films. As a result, the Si thin films prepared at d_{TS_1} = 60 and d_{TS_2} = 120 mm were found to have l_{s_1} = 80 and l_{s_2} = 110 mm, respectively. Accordingly, the divergence angle (Φ) of ablation plasma can be calculated as

$$\tan\Phi = \frac{(l_{s2} - l_{s1})/2}{d_{TS2} - d_{TS1}}$$

As a result, the ablation plasma of Si was clarified to diverge at Φ = 14 ° with going away from the target. In Fig. 4, with increasing in d_{TS}, the unsymmetrical thickness distribution was not clearly observed, whereas l_s was found to increase from 80 to 110 mm. In addition, the slope of thickness distribution became gentle with increasing l_S. Since the compositionally gradient area was conjectured to enlarge for the thickness distribution having a gentler slope, we attempted to prepare the compositionally gradient Si-Ge thin film at the condition of θ = 30 ° and d_{TS} = 120 mm.

3.2 Enlargement of compositionally gradient area in Si-Ge thin film

Figure 5 shows composition distribution of compositionally gradient Si-Ge thin film. The thin film was prepared by irradiating an ion beam on the target, which consisted of Si and Ge plates, furthermore, the deposition conditions were θ = 30 ° and d_{TS} = 120 mm.

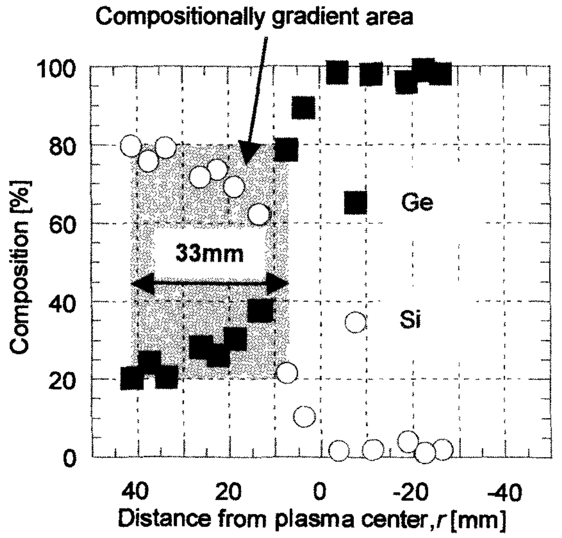


Fig. 5 The composition distribution of compositionally gradient Si-Ge thin film.

From the result of Fig. 5, the compositionally gradient area in the Si-Ge thin film, which was approximately 33 mm, was approximately 20 mm larger than that of our previous work [3]. One of the reasons to explain a larger compositionally gradient area was given that the thickness distribution having a gentler slope is achieved to control the deposition conditions as θ and d_{TS}.

4. CONCLUSIONS

From these experimental results, we have obtained the following conclusions:

1. Using the IBE method, the thickness distribution of the Si thin film prepared under the condition of θ = 40 ° is almost symmetrical to the axis of r = 0 mm.
2. When an ion beam is irradiated at θ = 30 ° to the Si target, the thickness distribution of the Si thin film is unsymmetrical being inclined toward the negative r side.
3. The spread length of the Si thin films increases from l_s = 80 to 110 mm with increasing in d_{TS} from 60 to 120 mm.
4. The compositionally gradient are in Si-Ge thin film is approximately 33 mm to control the deposition conditions as θ and d_{TS}.

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