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Improving Crystalline Quality of Pulsed-Laser-Microcrystallized Si Thin Films by the Two-Step Irradiation Method

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Introduction: In the previous meeting, we have reported that PLA method can produce a better microcrystallized Si film quality on crystallization-induction (CI) layer of YSZ $[(ZrO_2)_{1-x}(Y_2O_3)_x]$, compared with that on glass substrate[1]. However, the quality of Si film degrades at the high laser energy density E (the upper of Fig. 1). Annealing at the low E can produce a good quality of Si film, but it takes a long time for the complete crystallization of the entire a-Si film. In order to shorten annealing time for full crystallization with good quality, we have proposed a new irradiation method or two-step method, in which an a-Si film will be irradiated using two kinds of E (the lower of Fig. 1). First, in the initial state, it is irradiated at the low E (E_i) for a short time to occur nucleation at the interface between Si and underlayer. Next, in the second step of growth state, the Si film is irradiated at the high E (E_g) to speed up the growth from nuclei, and thus accelerate the crystallization of the Si film.

In this Meeting, we present the detail investigation results of the two-step method, comparing with the conventional irradiation method.

Experimental: A YSZ CI layer is deposited on a quartz substrate at a substrate temperature of 50 °C by sputtering. Then, an a-Si film is deposited on a YSZ/glass substrate by e-beam evaporation method at 300 °C. Next, crystallization of a-Si film is carried out in N_2 ambient by a pulse Nd:YAG laser (λ = 532 nm) with a repetition frequency of 10 Hz and a pulse duration of 6 ~ 7 nsec. In the two-step method, the total pulse number N is kept constant of 100 while pulse numbers of initial state N_i and growth state N_g are changed from 10 to 90 and 90 to 10, respectively, so as to $N = N_i + N_g$. The crystallization degree of Si film is estimated by Raman spectroscopy.

Results: Figure 2 shows Raman spectra of Si/YSZ/glass for conventional and two-step methods, whose irradiation conditions are shown in the inset. It can be seen that a-Si phase of the two-step method is reduced while its c-Si peak is higher and sharper than those of the conventional one. It seems that the crystalline quality of Si film is improved by using the two-step method.

We investigated the dependences of FWHM of c-Si peak and crystalline fraction X_c on the E_i for Si/YSZ/glass, whose results are shown in Fig. 3. E_g, N_i, and N_g are 106 - 109 mJ/cm^2, 10, and 90, respectively. At E_i = 18.5 ~ 22 mJ/cm^2, X_c is maximum while FWHM is minimum. It is considered that this value of E_i is nearly optimum for enhancement of Si film crystalline fraction and crystallinity.

In Fig. 4, we compare the relationship between FWHM and X_c for the two-step (△) and conventional (●) methods. In the conventional method, the rightmost datum is the highest X_c since increasing E over this datum condition induces obviously surface roughness due to partial melting. It can be seen from this figure that the crystallized Si films by the two-step method has more 4 times larger X_c with the smaller FWHM of ~ 6 cm^{-1} than those by the conventional one. This indicates that crystalline quality of Si films is improved with higher X_c by the two-step method. It can be considered that nucleation sites are controlled at the interface between Si film and YSZ layer in the initial state of the low E_i.

Summary: In the presentation, we will discuss more results in detail. In order to further improve the crystalline quality of Si film, we will try to use 3 kinds of energy density E: low E (initial state) – middle E (middle state) – high E (growth state) for crystallization.

Reference: [1] M. T. K. Lien et al., Abstract JSAP 74th Autumn Meeting, 2013, 19p-B4-4.

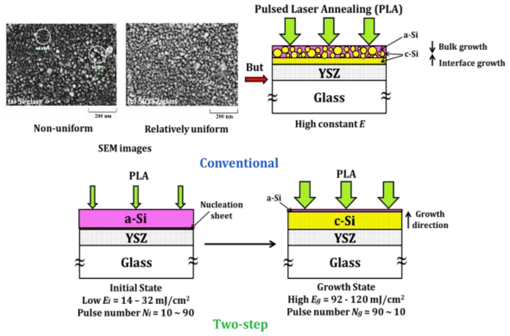

Fig. 1 Comparison between the conventional (upper) and the new or two-step (bottom) methods.

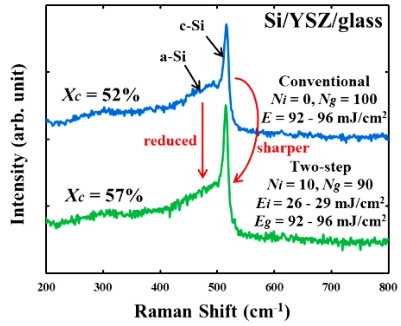

Fig. 2 Raman spectra of Si/YSZ/glass for conventional and two-step methods.

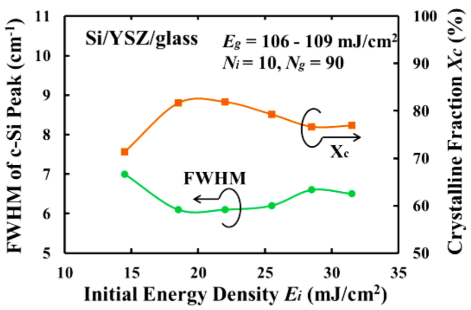

Fig. 3 Dependences of FWHM of c-Si peak and crystalline fraction X_c on the initial energy density E_i for Si/YSZ/glass in the two-step method.

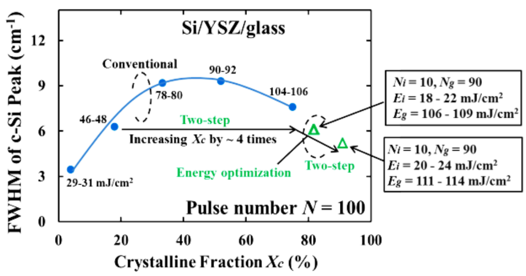

Fig. 4 Comparison of dependences of c-Si peak FWHM on crystalline fraction Xc between the two-step (△) and conventional (●) methods for Si/YSZ/glass.