Atomic Scale Observation of the Initial Stage of Thin Film Formation by PLD Method

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Abstract

We have observed the initial stage of film formation process by pulsed laser deposition (PLD) method on the Si(111)7×7 surface to obtain the information of the interface between the substrate and the films. In the present work, we have concentrated to observe the initial stage for SiC and MnSi thin films fabricated by the PLD method.

We used an ultra-high-vacuum scanning tunneling microscope (UHV-STM) for the observation. HOPG, C_{60} and Mn targets were used for the PLD experiment. While the substrated used in the experiment is Si(111)7×7. The number of shots varied from 1-10 shots.

From the results of the experiment, we determined the characteristic of adsorption site as well as morphological characteristic for particles in each target material. We suggested that particles ablated from HOPG and Mn target materials which have dangling bonds preferred to adsorb onto adatoms of Si(111)7×7 which have dangling bonds as well, whereas particles ablated from C_{60} target material preferred to adsorb onto the mechanical structure of the Si(111)7×7 surface because of the neutrality of electricity of the particle. We also suggested that HOPG and Mn target particles preferred to adsorb more on faulted half unit cell rather than unfaulted half unit cell due to the small energy difference between the particles and the faulted half unit cell. While due to the charge neutrality, the difference between the faulted half unit cell and the unfaulted half unit cell was small for the C_{60} target particles. Also, we suggested that HOPG and Mn target particles preferred to cluster while C_{60} particles preferred to spread onto the surface.

These results are expected to be useful information for clarification of mechanism of the deposition process of PLD method.

Keywords: PLD, STM, Initial stage, Si(111), HOPG, C_{60}, Mn

1. INTRODUCTION

In recent years, the technique for fabrication of ultrathin film of good quality is in demand on order to develop the more miniaturezed and high performance semiconductor and optical devices. In such fabrication of ultra thin film, the quality of the film strongly influenced by the number of atom that is belongs to the interface is comparable to that of the film itself. From this point of view, we believe that it is very important to obtan the information of the interface between the substrate and the film. However, it is almost impossible to observe the interface after the film has been formed on the substrate. So, we tried to obtain the interface information by observing the initial stage in the fabrication of ultra thin films. In the initial stage, molecules and atoms composing the films are adsorbing onto the Si substarte surface in the scale at an atomic level.[1]

In this work, we concentrated on the SiC[1] and MnSi[2),3)] thin films which are fabricated by the Pulsed Laser Deposition (PLD) method[4]. These films of good quality have not been obtained, nor have the mechanisms of film formation not been clarified yet. So, we observed the adsorption of particles ablated from the target using the PLD method onto Si(111)7×7 surface intending to obtain the information of the interface that will be the key knowledge for fabricating good quality of ultra thin films.

2. EXPERIMENTAL SETUP

An ultra-high-vacuum scanning tunneling microscope (UHV-STM) was ised for the observation of the surface morphologies. Fig.1 shows the experimental apparatus used for this work. The apparatus consisted of three UHV chambers. We used Si(111) as sample substrate and HOPG, C_{60} and Mn as target. Sample and target substrates were introduced in Chamber III. A clean Si(111)7×7 surface was obtained in Chamber II by high temperature flashing method. A Q-switched 2 ω -Nd:YAG laser (wavelength of 532nm) with laser energy of 0.1mJ/pulse was used for the PLD process.

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The sample was obtained by ablating laser to target at a range of 20mm between the target and the Si substrate. The sample then was observed in Chamber I using STM measurement.

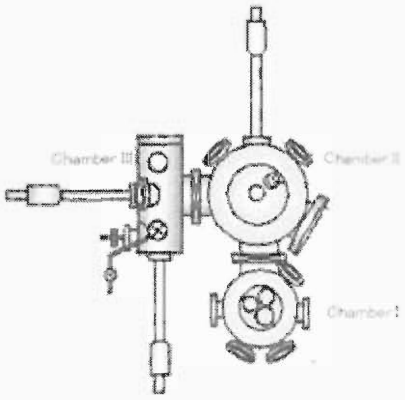


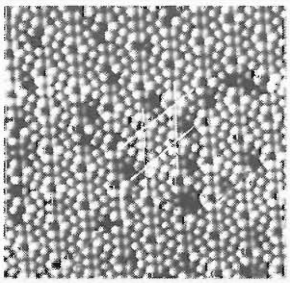
Fig. 1. Schematic diagram of experimental apparatus.

3. EXPERIMENTAL RESULTS AND DISCUSSION

3.1. STM image results

To obtain a good PLD experiment, the Si substrate used in the experiment has to be without any impurities so that there only would be particles from the target that were deposited on the surface of the substrate. To achieve this, we prepared a clean Si(111)7×7 surface and observed it with STM measurement.

Fig.2 shows an experiment result of a STM image of Si(111)7×7 surface structure. The measurement range is 20nm×20nm. In the image, a clean Si(111) surface with a 7×7 reconstruction structure is clearly visible such as shown in the rhombus shape in the image. Defect on the surface can be seen such as shown with the "arrow D" in the image. Comparing this image to the DAS model as shown in Fig.3, we can say that a clean Si(111)7×7 surface structure was obtained.



Measurement area: 20nm×20nm

Fig. 2. STM image of Si(111)7×7 structure surface.

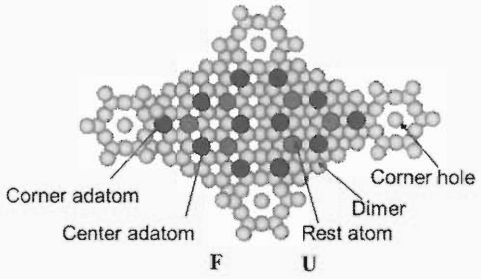
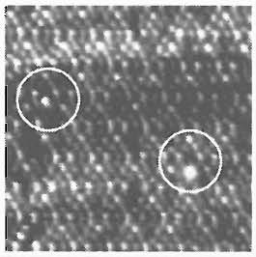
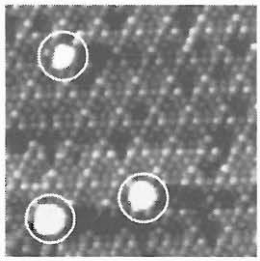


Fig. 3. DAS Model of Si(111)7×7.

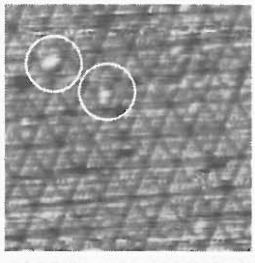
Fig. 4 indicated the STM images of particles from (a) HOPG target, (b) C_{60} target and (c) Mn target each adsorbed on a clean Si(111)7×7 surface in the PLD experiment. The measurement area for each image is 20nm×20nm. In all the STM images, particles brighter than Si adatom, which were surrounded by circles, can be observed clearly adsorped onto the Si(111)7×7 surface. Also we can observed that the Si(111)7×7 surface is not damage after the PLD process.



(a) HOPG target at 3 shots



(b) C_{60} target at 3 shots



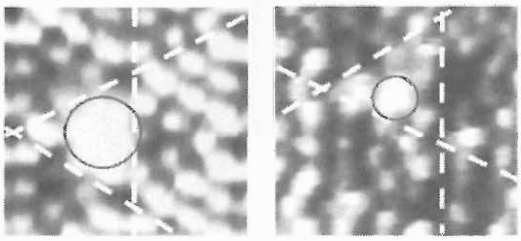
(c) Mn target at 3 shots

Measurement area:20nm×20nm

Fig. 4. Particles adsorbed onto the Si(111)7×7 surface after the PLD process.

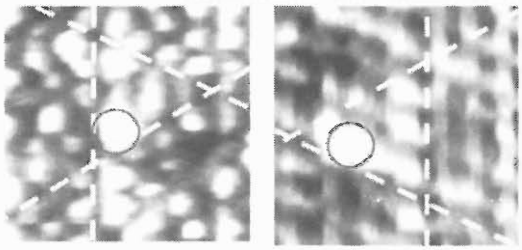
3.2. Adsorption sites

From the STM images, we classified the adsorption sites of particles as indicated in Fig. 5(a)~(g) and compared them with the DAS model as shown in Fig. 6. Site *A* is adsorption site for particle on the center of a half unit cell. Site *B* is on the center adatom, Site *C* is on the corner adatom, Site *D* is the site between the center adatom and corner adatom, Site *E* is on the rest atom, *Dim* is the site on the dimer and *CH* is the site on the corner hole.



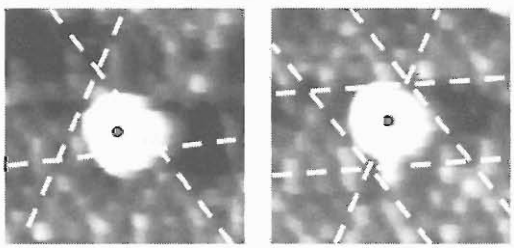
(a) Site *A* (b) Site *B*

Measurement area: 5nm×5nm



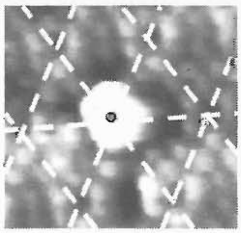
(c) Site *C* (d) Site *D*

Measurement area: 5nm×5nm



(c) Site *E* (d) *Dim*

Measurement area: 10nm×10nm



(e) *CH*

Measurement area: 10nm×10nm

Fig. 5. Classification of adsorption sites on STM images.

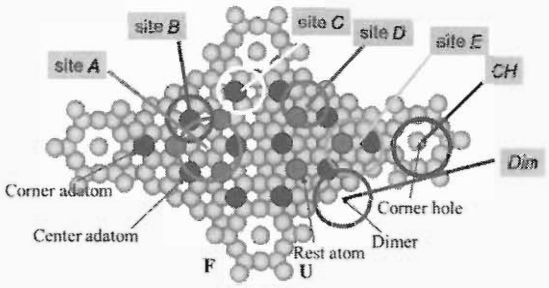


Fig. 6. Classification of adsorption sites on DAS model.

To further investigate the adsorption sites, we calculated the probability of adsorption sites for each target materials and concluded it in a table as shown in Tab. 1.

Tab. 1 Probability of adsorption site

Target materials	HOPG		C_{60}		Mn	
Adsorption sites	F [%]	U[%]	F [%]	U[%]	F [%]	U[%]
site *A*	17	3	7	7	10	3
site *B*	26	7	5	6	17	10
site *C*	24	10	6	7	28	26
site *D*	10	3	-	-	4	2
site *E*	-	-	20	16	-	-
Dim	-		16		-	
CH	-		10		-	

From the table, particles from HOPG and Mn targets prefer to adsorb on site *B* and site *C* than other sites. Both of these sites are on the adatoms. While for C_{60} target, the particles prefer to adsorb onto site *E*, *Dim* and *CH*. All of these sites are on the hollow part of the Si(111)7×7 surface. This is because, for the HOPG and Mn targets the particles that adsorbed onto the Si(111)7×7 surface are atoms which has dangling bonds as well as the adatom of Si(111)7×7 surface. While for the C_{60} target, the particles that adsorbed onto the Si(111)7×7 surface are molecules which do not have dangling bond which is not bond well with adatom of Si(111)7×7 surface and prefer to adsorbed onto the hollow part which is the mechanical structure of the surface.

Also from the table, we can see the particles from HOPG and Mn targets prefer to adsorb more on the faulted half unit cell than on unfaulted half unit cell due to the small difference of energy level between the particles and the faulted half unit cell. While C_{60} particles randomly adsorb onto the Si(111)7

× 7 surface due to the electrically neutral characteristic of C_{60} particles.

3.2 Particle distribution and clustering

To determite the distribution of particles on the Si(111)7 × 7 surface, we summarized the average number of particles adsorbed onto the Si(111)7×7 surface according to the number of shots as shown in Fig. 7. Fig. 8 shows particles adsorbed onto the Si(111)7×7 surface according to (a) HOPG, (b) C_{60} and (c) Mn targets. The upper STM images are after 2 shots and the lower are after 8 shots. The measurement area for each image is 20nm×20nm.

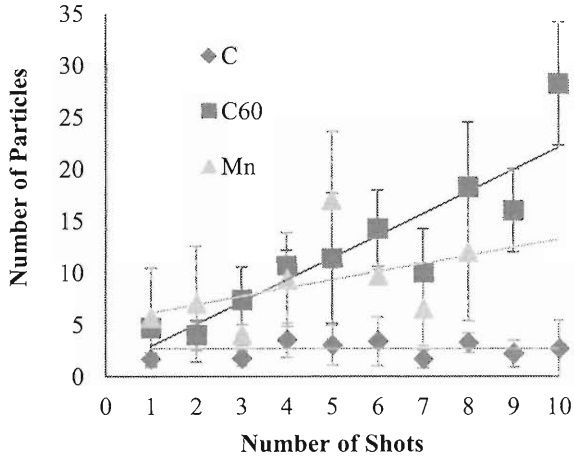
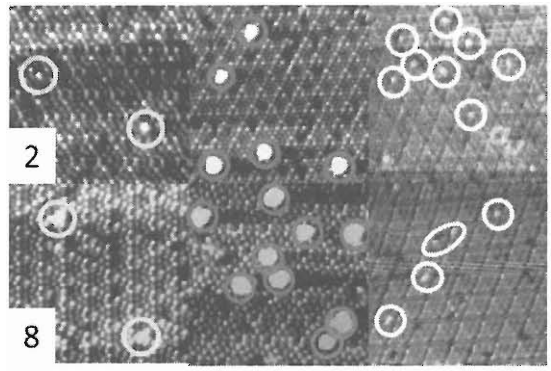


Fig. 7. Average number of particles according to the number of shots.



(a) HOPG (b) C_{60} (c) Mn

Measurement area: 20nm×20nm

Fig. 8. Particles adsorbed onto the Si(111)7×7 surface with different number of shots according to target.

From the graph, we can see that for C_{60} target, the number of particles increased propotional to the number of shots increased. For HOPG and Mn targets, eventhough the number of shots increased, the number of particles did not increased much.

From the STM images in Fig. 8, we can see clearly the different pattern of particle adsorption for each target in both number of shots. For HOPG target, the C particle size getting bigger, for C_{60} particles, there are no increase in the size but the particles are clearly spreaded on the Si(111)7×7 surface and for the Mn particles, is in the between the C particles and C_{60} particles. These indicated that no clustering to occur for C_{60} particles. While for C particles and Mn particles prefer to cluster onto Si(111)7×7 surface. The tendency to cluster can be summarize in Fig. 9.

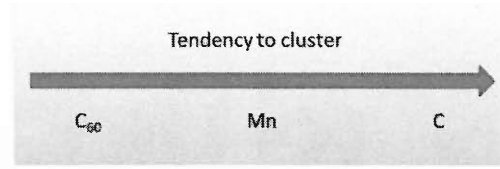


Fig. 9. Tendency to cluster for each target particle.

4. CONCLUSION

We have observed the adsorption of particles ablated from targets, especially the HOPG, C_{60} and Mn respectively onto a Si(111)7×7 surface intending to obtain the information of the interface of thin films by PLD method. Our results suggested that particles ablated from HOPG and Mn targets prefer to adsorb onto adatom as both the target particles and the adatom have dangling bonds whereas C_{60} particles which are electrically neutral preffered to adsorb onto the mechanical structure of the surface. We also suggested that the difference energy level plays important roll in determine the preferable adsorption site in half unit cell for HOPG and Mn target particles while for the C_{60} particles, due to the charge neutrality, no significant preferable adsorption site in half unit cell can be seen. Apart from that, C and Mn particles prefer to cluster while C_{60} particle prefer to spread onto the Si(111)7×7 surface.

These results are expected to be useful information for fabrication of thin films of good quality.

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