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Deposition dynamics of droplet-free Si nanoparticles in Ar gas using laser ablation

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Abstract

Droplet-free deposition of Si nanoparticle films has been studied applying time-resolved imaging of Si nanoparticles formed by laser ablation of Si targets in Ar gas. We found that Si nanoparticles can be deposited not only on substrates facing to the targets but also on substrates placed beside the target. We further confirmed using a scanning tunneling microscope (STM), Si nanoparticles with sizes of 5–8 nm are deposited on substrates placed beside the target and using a scanning electron microscope (SEM) on the substrates, no droplets are observed.

Keywords: Pulsed laser deposition; Droplet-free; Si nanoparticle; Time-resolved imaging

1. Introduction

Since nanostructured Si systems can emit light efficiently, it is possibly applied to novel opto-electronic materials and to integrated opto-devices onto Si devices. Laser ablation can be used to fabricate such Si nanostructures because surface modification of the nanoparticles and impurity doping can be attained by using the pulsed feature of laser ablation. On the other hand, deposition of droplets is a common problem of laser ablation. Therefore, extensive investigations have been carried out to deposit droplet-free films and demonstrated that the number of droplets can be decreased by: (a) the eclipse method that utilizes a spatial filter between targets and substrates [1,2]; (b) the off-axis method [3]; (c) a method using a velocity filter [4]; (d) a method choosing carefully wavelength and fluence of ablation laser, and atmosphere gas [5,6] and (e) the porosity of a target [7]. In spite of these investigations, complete elimination of droplets has not been achieved. Compared to Si ablation in He gas and Ne gas whose atoms are lighter than Si atom, Si atoms and ions produced by laser ablation in Ar gas are confined in a limited region since Ar atoms are heavier than Si atoms. Consequently, Si nanoparticles are expected to deposit on substrates placed beside the targets without droplets. So we have investigated the growth dynamics of Si nanoparticles in Ar gas utilizing a second laser [8]. The second laser was irradiated to probe grown Si nanoparticles in inert gas.

In this work, we applied the second laser irradiation method in order to observe time-resolved photoluminescence (PL) from Si nanoparticles formed by laser ablation of Si targets in Ar gas. Based on the formation dynamics of Si nanoparticles, we have tried to obtain droplet-free films of Si nanoparticles.

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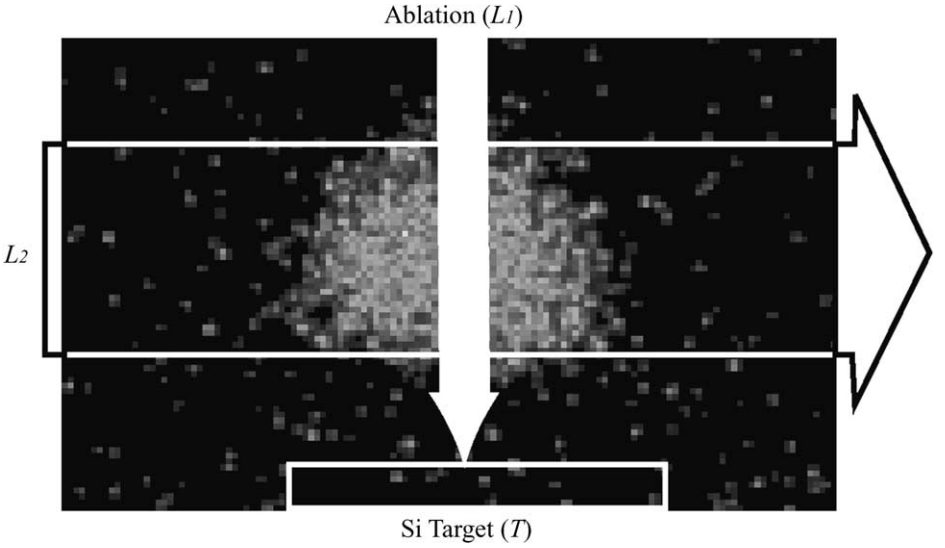

Fig. 1. A time-resolved 2-D spatial distribution of PL from Si nanoparticles at a delay time of 2.0 ms.

2. Experimental

Fig. 1 shows a typical two-dimentional (2-D) distribution of time-resolved PL from Si nanoparticles and illustrate the experimental configuration used. For observing time-resolved distributions of PL from Si nanoparticles, p-type CZ–Si (8–12 Ω cm) targets (T) were ablated by a Q-switched Nd:YAG laser light (532 nm, 5 J/cm^2, 7 ns) (L_1) in 5 Torr Ar gas filled in a vacuum chamber. Si nanoparticles were excited by irradiation with 355 nm pulsed light from another Nd:YAG laser (L_2) at 0.19 J/cm^2. Resulting PL distributions were observed using an gated ICCD camera (Princeton Instruments, ITE/CCD-576-G/RB-E). As shown in Fig. 1, 2-D spatial distributions of PL were observed in the track of the sheeted second laser beam.

3. Results and discussion

Fig. 2 shows the total intensity of the PL from Si nanoparticles as functions of a delay time of the second laser from the initial ablation laser. The curve (a) was obtained from the measurements after the target has been previously ablated for several hundreds of shots. More intense PL from Si nanoparticles is observed compared with the PL intensity without previous ablations shown by the curve (b). This suggests that Si nanoparticles deposit backward on the target in 5 Torr Ar gas and that the deposited nanoparticles are desorbed by the following ablation through processes such as thermal shock wave. Droplets are generally emitted radially from the ablated spot and fly straight. Therefore, in Ar gas, droplet-free films of Si nanoparticles are expected to be deposited on substrates not facing to the target but coplanar with the target plane.

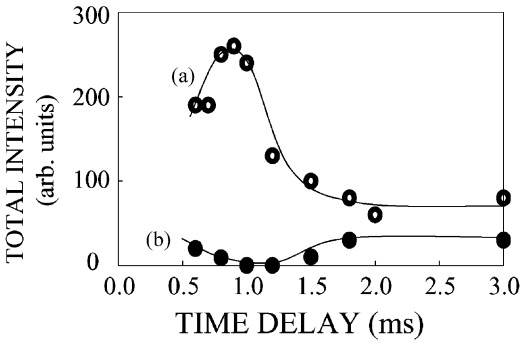

Fig. 2. Total PL intensity of Si nanoparticles formed by laser ablation in Ar gas for (a) a previously ablated Si target and (b) a fresh target.

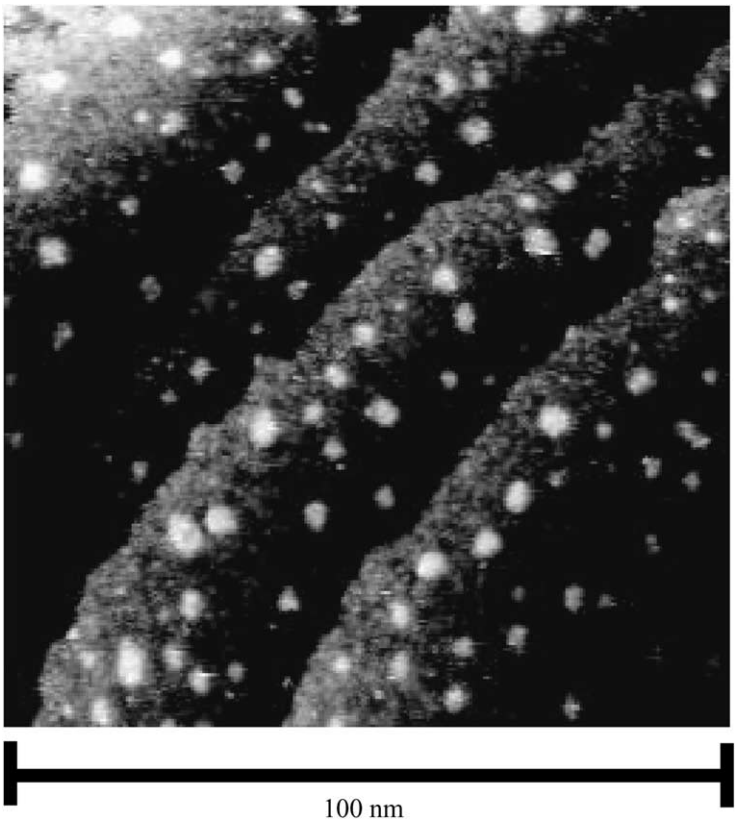

Fig. 3. A typical STM image of Si nanoparticles deposited on an organic single molecule film.

In order to confirm the backward deposition, self-assembled monolayer (SAM) films formed on Au films were placed 5 mm apart from the ablated spot on Si targets. Fig. 3 shows a typical scanning tunneling microscope (STM) image of Si nanoparticles on the SAM film after laser ablation of a Si target in 5 Torr Ar gas [9]. We observed Si nanoparticles with diameter in a range of 5–8 nm, which are seen as bright spots on Au terrace.

Scanning electron microscope (SEM) observation was performed in order to investigate the deposition of droplets on substrate (S) placed at different positions as illustrated in Fig. 4. When Si nanoparticles were deposited at a conventional position (a) facing to the target, droplets are deposited on the Si substrates as shown in Fig. 4a. When Si nanoparticles were deposited on substrates placed just beside a target (b), droplets are observed to have several trails in directions emanating from the ablated spot. The trails are tracks of ejected droplets that have shaved the films of Si nanoparticles deposited by previous ablations. In order to prevent the droplets, a Si wall with a height of 1 mm was placed between a substrate and a target as illustrated in Fig. 4c. We found that deposition of droplets is completely suppressed by the wall. Since the wall is low enough compared with the dimension of ablation plume, Si nanoparticles formed in the gas phase can fly back to the substrate and deposit on it. When a film of Si nanoparticles was deposited on a substrate placed at a position (d) which is 2 mm lower than the target, web-like films of Si nanoparticles were observed. This may be due to the lower annealing temperature of deposited films of Si nanoparticles exposed to ablation plasma because the substrate at position (d) is placed further from the plasma.

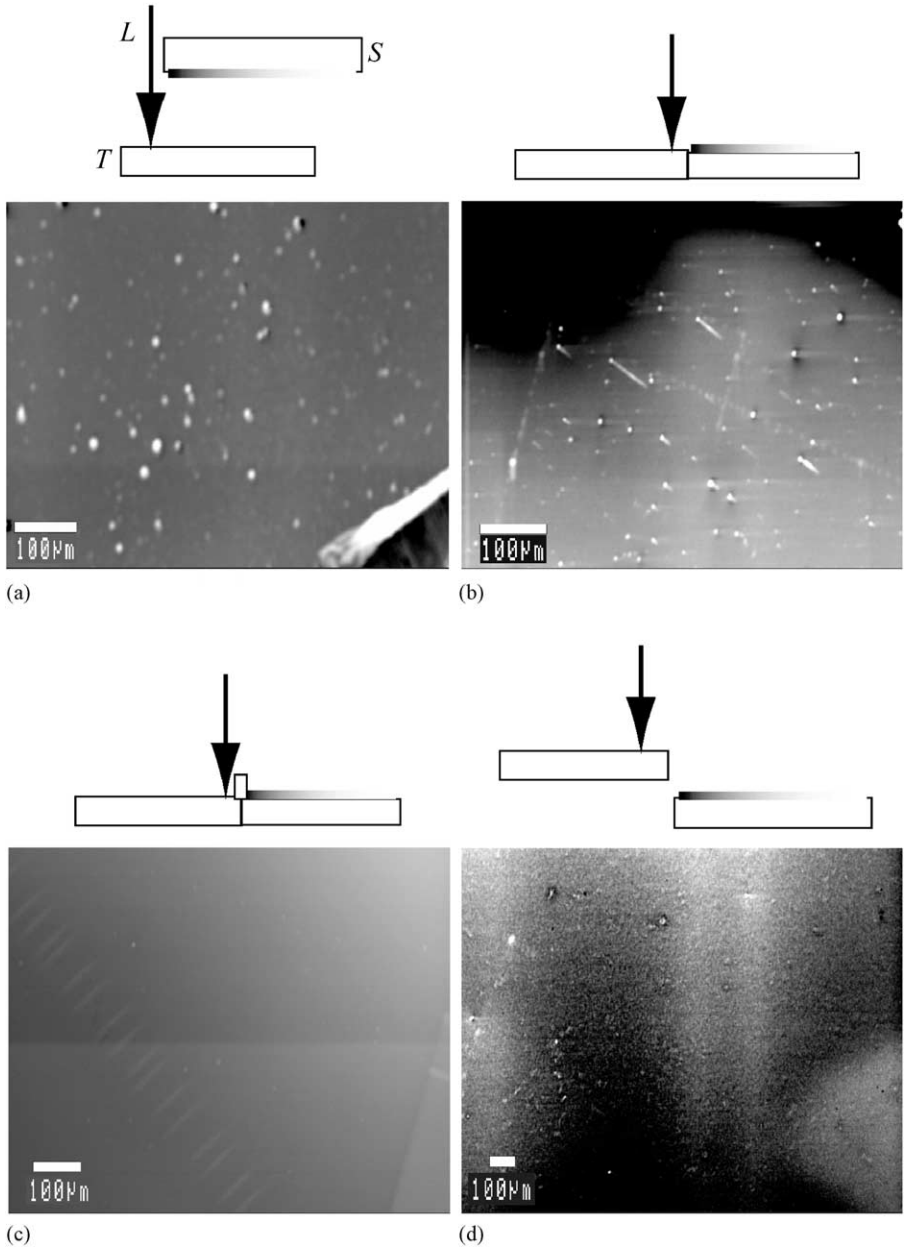

Fig. 4. SEM images of droplets on films of Si nanoparticles deposited at positions (a) opposite to a target, (b) beside a target, (c) beside a target with a wall and (d) below a target.

4. Conclusion

In conclusion, we have investigated droplet-free deposition of Si nanoparticles based on time-resolved imaging of photoluminescence from Si nanoparticles. We found that droplet-free films of Si nanoparticles can be backward deposited on substrates placed beside the Si targets in Ar gas.

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