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Ordered 1 x 1 SiH_2 Monolayer Adsorption on Si(001) from Cracked Si_2H_6 and Its Application to ALE Growth Operation

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1. Introduction

Atomic-layer epitaxy (ALE) is a layer-by-layer epitaxial film growth technique and expected to be used as a useful tool to develop nanometer-scaled hetero- and homo-structured devices [1]. The ALE technique uses one monolayer (ML) saturation adsorption reaction called a self-limited adsorption mechanism. As for Si ALE, hydride molecules, Si_nH_m, are promising precursors for their potential advantages of being free of contaminants, however, the saturation coverages are generally less than 1 ML at room temperature (RT).

We have previously proposed to apply thermally cracked Si_2H_6 as a source gas to Si ALE [2-7]. With cracked Si_2H_6, the Si saturation coverage increases and 1 ML adsorption controllability is obtained at RT, and we have also demonstrated Si ALE [4]. The Auger electron spectroscopy (AES), reflection high-energy electron diffraction (RHEED) and theoretical reaction path analyses suggest that :SiH_2 produced by cracking Si_2H_6 plays an important role on the saturation coverage increase [5-7].

In this work, we have further investigated the saturation adsorption mechanism and structure by hydrogen temperature-programmed desorption (TPD) and scanning tunneling microscope (STM) measurements. Upon cracked Si_2H_6 exposure on Si(001), we have observed for the first time a STM image of an self-ordered 1x1 :SiH_2 monolayer adsorption surface. The results demonstrate that the utilization of :SiH_2 from cracked Si_nH_m becoms a key nanotechnology for Si layer-by-layer growth operation.

2. Experimental

Thermally-cracked-Si_2H_6 adsorption experiments were carried out using a load-locked ultrahigh vacuum chamber with a base pressure of $< 5 \times 10^{-10}$ Torr. Substrates used were p-type Si(001). To thermally crack Si_2H_6, we used a alumina tube thermal-cracking cell with a tungsten spiral filament in it. The cracking temperature (T_c) was determined by the filament temperature. The absolute accuracy is within ± 50 °C. Research-grade Si_2H_6 (99.999%) were used.

To investigate the surface structure of thermally-cracked-Si_2H_6-exposed surface, hydrogen TPD experiments were carried out after exposing a clean Si(001) surface to atomic hydrogen or to thermally-cracked-Si_2H_6. The substrate was resistively heated at a rate of 2 °C/s for TPD. To determine the adsorption surface structure and its reaction mechanisms, we also measured the STM image of the thermally-cracked-Si_2H_6 exposed surface.

3. Results and Discussion

It has been established that a fully atomic H exposed Si(001) surface changes to 1x1 dihydride (:SiH_2) surface [8]. First, we have measured the β_2 (:SiH_2 dihydride state) / β_1 (-HSi-SiH- monohydride state) peak intensity ratios observed in the hydrogen TPD spectra. The results are shown in Fig. 1.

Upon exposure to atomic H, a 2 x 1 RHEED pattern of

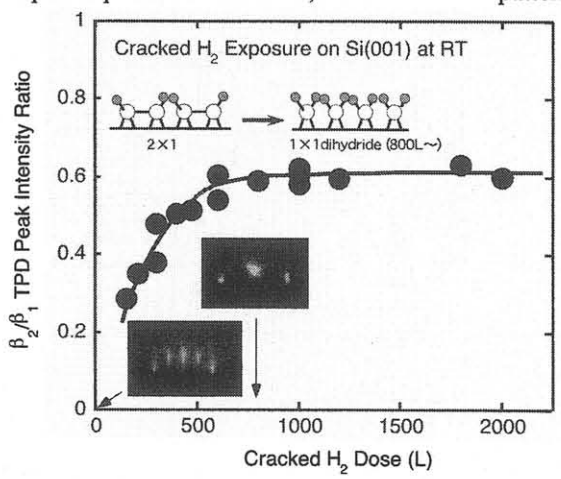

Fig. 1 β_2 / β_1 peak intensity ratios observed in the hydrogen TPD spectra obtained after exposing clean Si(001) surfaces at RT to cracked H_2. A 2 x 1 RHEED pattern of the clean surface changes to 1 x 1 after exposure to cracked H_2 doses of > ~800 L.

the clean surface changes to 1 x 1 and the β_2 / β_1 peak intensity ratio saturates when the cracked H_2 dose > ~800 L, indicating that the β_2 / β_1 peak intensity ratio of the dihydride surface is 0.61.

Fig. 2 shows β_2 / β_1 peak intensity ratios observed in the hydrogen TPD spectra obtained after exposing clean surfaces at RT to a cracked Si_2H_6 doses of 150000 L. The β_2 / β_1 ratios are almost constant irrelevant to the dose, indicating that the surface structures were determined in the initial adsorption stage. The ratio changes form 0.43 for a noncracked Si_2H_6 exposed surface covered with :SiH_2 + H [9] to ~0.61 when T_c > ~550 °C. The RHEED pattern also changes from weak 2x1 to 1x1 when T_c > ~550 °C. The results suggest that upon cracked Si_2H_6 exposure, fully dihydride surface is formed when T_c > ~550 °C.

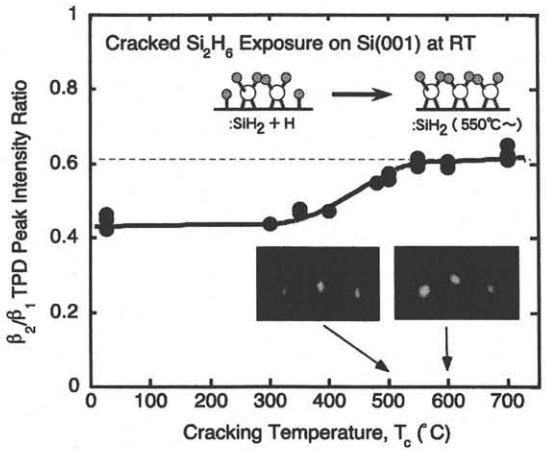

Fig. 2 β_2 / β_1 peak intensity ratios observed in the hydrogen TPD spectra obtained after exposing clean Si(001) surfaces at RT to a cracked Si_2H_6 doses of 150000 L with the T_c being varied from RT to 700 °C. A 1 x 1 RHEED pattern is obtained when T_c > ~550 °C.

Fig. 3 shows STM images obtained from (a) a clean Si(001) surface, and after exposure at RT to a cracked Si_2H_6 dose of (b) 3000 L and (c) 6000 L with T_c = 600 °C. The images are obtained with negative sample biases to observe the occupied states. The dimer row image observed from the clean surface ((a)) is clearly splitted into two rows ((b), (c)). The first gas phase Si_2H_6 cracking path is Si_2H_6 -> :SiH_2 + SiH_4 with :SiH_2 much more reactive than SiH_4 [7] and together with the results of the hydrogen TPD experiments, the STM images indicate that two :SiH_2 molecules adsorb on the underneath dimerized two Si atoms. Although the RHEED pattern is 1x1, very weak 2x1 periodicity observed in the STM images is ascribed to the fact that the underneath dimerization is not fully dissolved.

4. Conclusions

We have demonstrated ordered 1x1 :SiH_2 monolayer adsorption on Si(001) at RT from thermally cracked Si_2H_6 and that :SiH_2 is effectively produced and utilized as a ALE self-limited precursor by cracking Si_2H_6. It is expected that our proposed cracking method to produce :SiH_2 becomes a key nanotechnology for Si layer-by-layer growth operation.

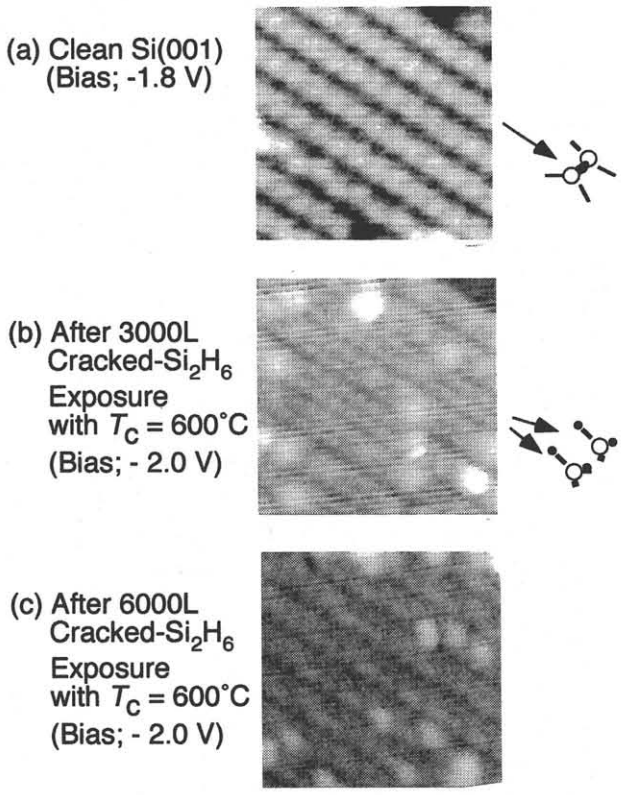

Fig. 3 STM images obtained from (a) a clean Si(001) surface, and after exposure at RT to a cracked Si_2H_6 dose of (b) 3000 L and (c) 6000 L. Upon cracked exposure, ordered 1x1 dihydride rows were observed.

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