

# Uniform Ag Thin Film Growth on an Sb-terminated Si(111) Surface

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## ABSTRACT

We report on the room-temperature-growth of highly uniform and ultrathin Ag films on Sb-terminated Si(111) surfaces, as evidenced from a scanning tunneling microscopy (STM) study in an UHV system. With predeposition of one monolayer (ML) of Sb, uniform growth of Ag islands was observed at room temperature. The Sb layer suppresses the surface diffusion of Ag atoms on Si surface and increases the Ag island density, and then the increased island density is believed to cause coalescence of Ag islands before the beginning of multilayer growth in higher coverages, resulting in the growth of atomically flat and uniform islands on the Sb surfactant layer.

## I. INTRODUCTION

The growth of metals on semiconductors has been a long-standing issue in current research on semiconductor devices. The ohmic and Schottky barrier contacts on silicon have been important applications of this branch of semiconductor technology. Ag growth on Si has been an especially interesting subject in the area of metal growth on semiconductors due to its abrupt interface and electronic potential [1]-[11]. However it has been impossible to achieve epitaxial layer-by-layer growth of Ag on Si(111) at room temperature. Ag films grown on Si(111) at room temperature have been shown to follow the Stranski-Krastanov mode of growth (layer plus island growth) [7], [8]. Up to the level of a monolayer, Ag atoms locate themselves on subunits of the  $7\times 7$  cell due to the strong interaction between Ag atoms and the Si dangling bonds on the  $7\times 7$  unit cell. Above a monolayer coverage, 3 dimensional (3D) Ag islands can be found on the intermediate Ag layer [8]. It is only at low temperatures that the growth mode of Ag on Si(111) restricts itself to 2D growth [11].

Since ultrathin metallic films with atomically flat surfaces are important model systems in the study of 2D phenomena, the observation of the 2D-layer like growth of Ag on Si(111) at low temperature was a remarkable discovery. Recently the epitaxial growth of hetero-interface systems such as Ge/Si has been demonstrated [12]-[18]. The smooth layer-by-layer growth of Ge on Si(100) has been

achieved through the use of As and Sb as surfactants, and the role of surfactants in heteroepitaxy has emerged as an issue of fundamental interest. The role of surfactants in layer-by-layer growth has also been studied in connection with the homoepitaxy of Ag [19]-[22], since in the absence of a surfactant multi-layer growth prevails at room temperature.

The ideal surfactant must segregate on the surface during growth leaving the substrate and growing film unperturbed and restricting islanding. A great deal of research into how this happens has been done [12]-[25]. Although the kinetic effect of surfactants in metal-on-metal growth systems is still controversial [19]-[23], three models have been suggested. The first is that surfactant suppresses surface diffusion of metal atoms on the metal substrate [23], which increases island density [20]. Due to the increased island density, islands coalesce into larger ones before multilayer growth occurs. The second model suggests that surfactant lowers interlayer diffusion barriers, which can affect effectively the downward mass transport since the upward interlayer diffusion barrier is larger than the downward one [22], so adatoms on islands can easily hop down the descending steps of the island. In this case, the 2 dimensional (2D) island size is enlarged, which is favorable for layer-by-layer growth. The third suggestion is that Sb lowers the mobility along step edges, inducing dendritic island shapes, which further enhances interlayer transport [22].

However, few application of surfactant to the layer-by-layer growth of metals on semiconductors have been reported [24], [25]. In one study, changes in the growth mode caused by the use of the surfactant Sb in Ag growth on Si(111) at elevated temperature (475 K) were measured using low energy electron microscopy (LEEM) [24]. Very dense nucleation of Ag islands on the Sb-terminated Si(111) surface at elevated temperature (475 K) was observed, while the large and widely spaced 3D Ag islands [24] were observed in the growth of Ag on clean Si(111) at this temperature. Although Sb does not completely suppress 3D island formation, almost completely close-packed Ag film was grown using Sb as a surfactant.

In order to grow a uniform and smooth Ag film on Si at room temperature, we chose Sb as a surfactant in this study. At first we obtained the structures of Sb overlayer on Si(111). Ag films were grown upon this Sb-terminated surface. Small sized Ag islands were formed at the initial stage of Ag growth without the wetting layer of Ag. As the Ag coverage increased, flat Ag islands formed uniformly across the Sb-terminated surface and coalesced into large dendritic islands. The kinetic origin of the flat and uniform growth of Ag film on Sb-terminated Si(111) is believed to be the suppression of the diffusion of Ag atoms owing to the Sb surfactant, which will be discussed in session IV. Even though Sb does not completely suppress islanding effects in the initial stage of Ag growth in the

Ag/Sb/Si(111) system, this work clearly showed that the use of an Sb surfactant encourages uniform growth of Ag films on an Si substrate. This observation is technologically very important, since it means that atomically smooth and uniform ultrathin metal films can be grown on semiconductors at room temperature.

## II. EXPERIMENT

We obtained a clean and defect free Si(111)- $7\times 7$  surface by repeated cycles of flashing. A Si sample was kept in a vacuum at 700 °C for several hours in order to remove the thick silicon oxide layer, and it was then heated up to 1230 °C by resistive heating, followed by subsequent annealing. As shown in Fig. 1, the  $7\times 7$  structure of Si(111) was clearly observed. Since the faulted halves of  $7\times 7$  unit cells have larger charge densities in their filled states, adatoms in the faulted halves are seen to protrude more than those in the unfaulted halves. An optical pyrometer was used to measure the temperature of the Si substrate. Sb was evaporated from a water cooled Knudsen cell. In order to obtain an Sb monolayer on Si(111), 3 ML of it was deposited on a Si(111)- $7\times 7$  substrate at a substrate temperature of 600 °C. This is below the desorption temperature for the first monolayer of Sb, but above the desorption temperature for the second layer of Sb. The Sb overlayer structures were monitored with UHV-LEED and STM.

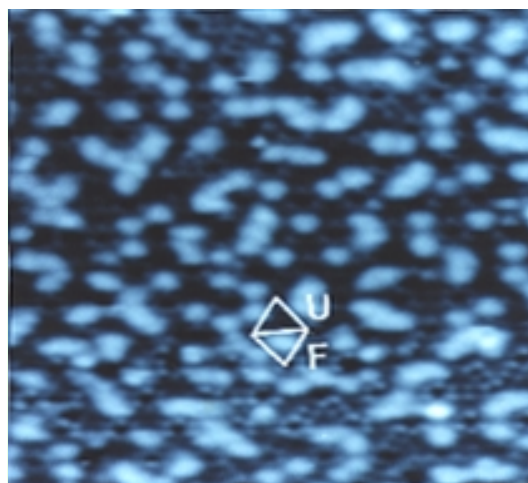


**Fig. 1.**  $7\times 7$  structure of clean Si(111) surface. Scan area  $270 \text{ \AA} \times 280 \text{ \AA}$ ,  $V_S = -2.5 \text{ V}$ ,  $I_t = 0.5 \text{ nA}$ .

Ag was evaporated from a hot tungsten filament under epitaxial growth conditions ( $0.1 \sim 0.2 \text{ ML/min}$ ). The thicknesses of Sb and Ag films were measured using the quartz crystal oscillator. A monolayer (ML) of Ag was defined as one atom per  $1 \times 1$  surface unit cell of the bulk Ag(111) substrate lattice ( $1.4 \times 10^{15}$  atoms per  $\text{cm}^2$ ), and for the growth on the clean Si(111) the definition of monolayer on Si(111) was also considered ( $7.83 \times 10^{14}$  atoms per  $\text{cm}^2$ ).

### III. Ag GROWTH ON CLEAN Si(111)

On the clean Si(111)- $7 \times 7$  surface we deposited Ag epitaxially at room temperature. At the submonolayer level of coverages we observed small islands which preserved the  $7 \times 7$  unit cell pattern as shown

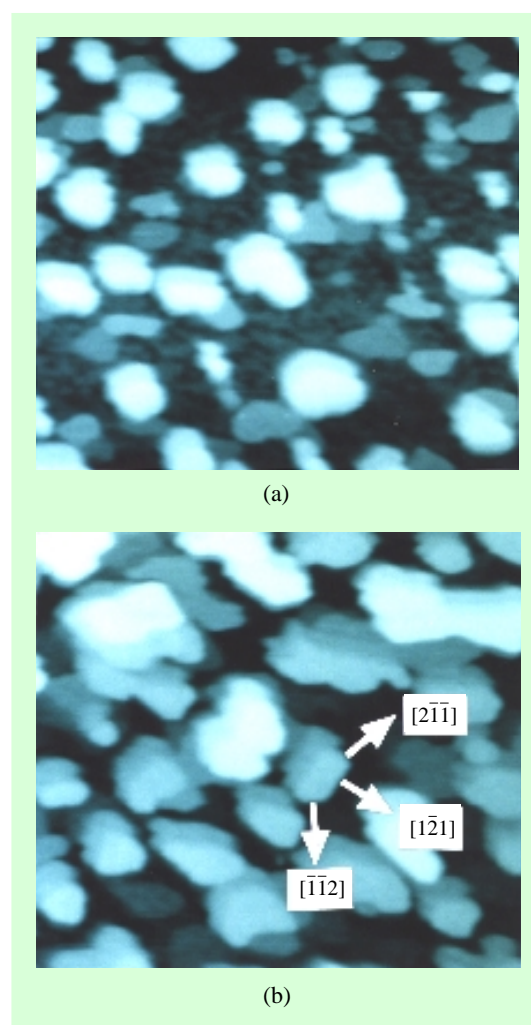


**Fig. 2.** Partly packed triangular islands and their coalescence in  $0.23 \text{ ML}$  Ag films grown on Si(111)- $7 \times 7$  surface at room temperature. Scan area  $260 \text{ \AA} \times 320 \text{ \AA}$ ,  $V_S = -2.5 \text{ V}$ ,  $I_t = 1.0 \text{ nA}$ .

in Fig. 2. This result is similar to that obtained in Ag deposition at substrate temperatures of  $90 \sim 130 \text{ }^\circ\text{C}$  [8]. The dangling bonds of rest atoms and adatoms on the  $7 \times 7$  unit cell are the most favorable sites for an adsorbed Ag atom in the initial stage of Ag deposition, and among them, the dangling bonds in the faulted halves of  $7 \times 7$  units were more reactive to the Ag atoms. The bonding probability on the faulted halves of the whole Ag islands was estimated to be 61% at  $0.23 \text{ ML}$ , which is estimated as  $0.4$  monolayer referenced to Si(111) surface. As Ag islands grow, the size and shape of islands change from circular types to partially packed triangular ones, which is attributable to the characteristics of the subunits of  $7 \times 7$  structure. The height of the Ag layer adsorbed on the Si adatoms was estimated to range from  $0.5$  to  $1 \text{ \AA}$ .

Abrupt changes in growth pattern above the dangling-bond saturation coverage can be clearly observed in Fig. 3. Flat islands with relatively large areas of  $300\sim 13000 \text{ \AA}^2$  and with various heights from 1.6 to  $7 \text{ \AA}$  appear upon the first intermediate Ag layer with deposition of a 0.9 ML (1.6 monolayer referenced to Si(111) surface) Ag film (Fig. 3(a)). At 1.5 ML (2.4 monolayer referenced to Si(111) surface) of Ag film, the height of flat Ag islands increased ranging from 1.6 to  $10 \text{ \AA}$ , and then the Ag islands coalesced. The island heights were estimated from the z-directional PZT bias voltages in the constant current mode and their histogram. These flat islands have an interesting structural behavior which is very different from that of the intermediate Ag layer. The height of the first Ag layer ranged from 0.5 to  $1 \text{ \AA}$  but the step heights of the second and subsequent Ag layer ranged from 1.5 to  $2.5 \text{ \AA}$ . These step heights of the flat islands are comparable to the  $2.36 \text{ \AA}$  step height of an Ag(111) surface. This suggests that the surface normal facets of Ag islands be in the direction of (111).

This feature of island formation differed strikingly from epitaxial growth. Up to the dangling-bond saturation coverage, uniform layer growth was established due to the strong interaction between adsorbed Ag atoms and substrate Si atoms. However, above the dangling-bond saturation coverage the interaction between adsorbed Ag atoms comes to play an important role in film growth due to saturation of the Si



**Fig. 3.** Large size flat 3D islands on the intermediate Ag layer and their coalescence. (a) 0.9 ML film grown on Si(111)-7 $\times$ 7 surface at room temperature. Scan area  $650 \text{ \AA} \times 790 \text{ \AA}$ ,  $V_s=2.0 \text{ V}$ ,  $I_t=1.0 \text{ nA}$ . (b) 1.6 ML film,  $650 \text{ \AA} \times 790 \text{ \AA}$ ,  $V_s=2.0 \text{ V}$ ,  $I_t=1.0 \text{ nA}$ .

dangling bonds. Since the activation energy for surface diffusion of Ag adatoms on the intermediate layer seems to be less than or equal to the thermal energy, Ag adatoms can easily diffuse resulting in the formation of large metallic islands at room tempera-

ture. Hence the average size of island became larger above the dangling-bond saturation coverage. The features of multilayer growth shown in Fig. 3(b) are attributable to this kinetic mechanism. The orientation of thick Ag islands seems to match closely with Si(111) substrate, since the step edges of thick Ag islands are composed of the straight lines and the normal direction of step edges indicated by arrows in Fig. 3(b) is one of six directions  $[\bar{1}\bar{1}2]$ ,  $[11\bar{2}]$ ,  $[\bar{1}2\bar{1}]$ ,  $[1\bar{2}1]$ ,  $[2\bar{1}\bar{1}]$ , and  $[\bar{2}11]$  in Si(111) crystal orientation.

In case of low temperature (90K) Ag growth on Si(111)- $7\times 7$ , interlayer exchange increased due to the small island size and the roughness of island edges [11]. However in the present system island sizes are larger and the edges are smoother than those observed in low temperature growth.

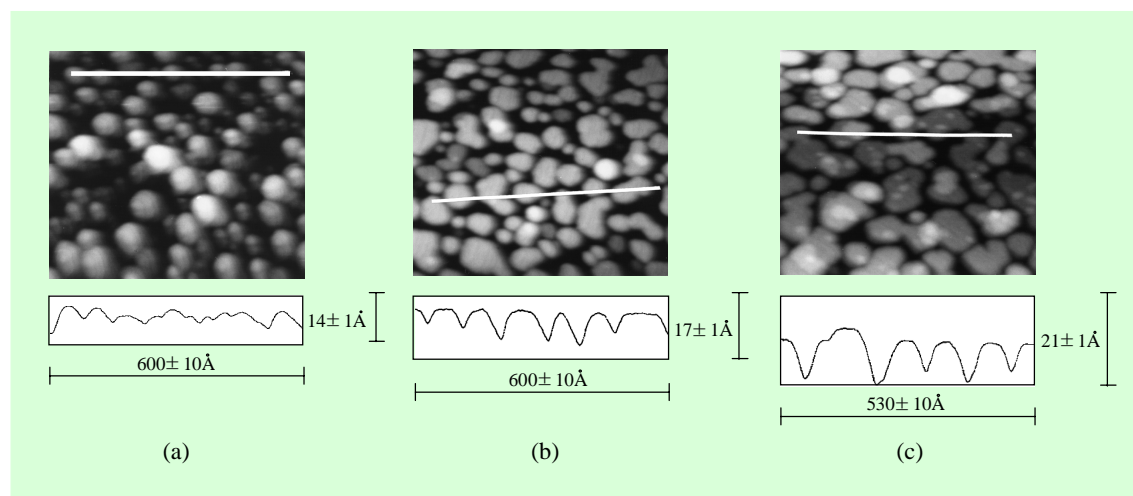
#### IV. Ag GROWTH ON Sb-TERMINATED Si(111)

The dependence of Sb overlayer structures on the Sb coverage and temperature of the Si(111)- $7\times 7$  substrate has been studied extensively [26]-[29]. These studies show that the Sb-terminated Si(111) surface involves complex reconstructions. In order to obtain an Sb monolayer on Si(111), Sb was deposited on a Si(111)- $7\times 7$  substrate at a substrate temperature of 600 °C. At a substrate temperature of 600 °C, a monolayer of Sb is reported to have three orientational domains of  $2\times 1$

structures, and a disordered  $7\times 7$  structure [26]-[28]. The interaction of Ag atoms with Si atoms is depressed due to the passivation of Si dangling bonds with Sb in the growth on Sb-terminated surface, and therefore, the agglomeration of Ag atoms occurs at the submonolayer coverage without the wetting layer of Ag.

In the STM images taken from 1.1 ML Ag film grown on a  $2\times 1$  Sb layer, closely packed round islands were observed as shown in Fig. 4(a). The average diameter of islands is small, and the surface is compactly covered. The height of these round islands is estimated to range from 1 to 3 ML height of Ag(111), where the monolayer step height is 2.36 Å. Since Ag islands, thinner than 3 ML, are unstable on Sb-terminated Si surface, small islands of irregular heights were formed initially. The vertical scale of the STM image was calibrated based on the bilayer step height of Si(111). The density of small flat islands increases with increasing film thickness. At 2.1 ML the surface is nearly filled with flat islands as shown in Fig. 4(b). The formation of islands with flat surfaces continues up to 3.2 ML as shown in Fig. 4(c). From the analysis of island heights it is suggested that the small round shaped islands observed in the Ag film of 1.1 ML tend to coalesce into flat Ag islands at higher coverages. In fact at 3.2 ML these flat islands coalesced resulting in the formation of dendritic islands.

The heights of such islands are much larger than the step height of Ag(111).



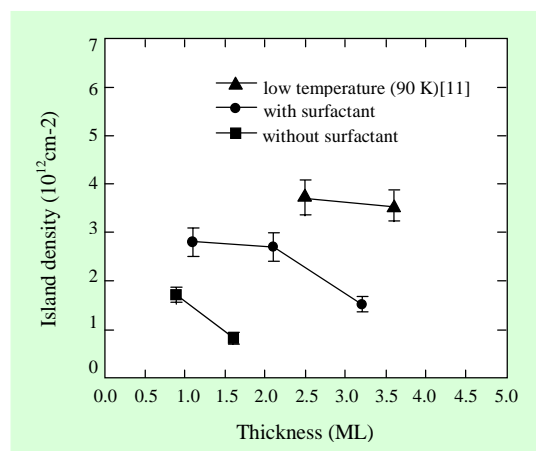
**Fig. 4.** STM images and line profiles along the white line. (a) 1.1 ML Ag film grown on an Sb-terminated Si(111) surface at room temperature, scan area  $650 \text{ \AA} \times 790 \text{ \AA}$ ,  $V_S = -2.9 \text{ V}$ ,  $I_t = 0.5 \text{ nA}$ . (b) 2.1 ML Ag film, scan area  $650 \text{ \AA} \times 790 \text{ \AA}$ ,  $V_S = -2.9$ ,  $I_t = 0.5 \text{ nA}$ . (c) 3.2 ML Ag film, scan area  $650 \text{ \AA} \times 790 \text{ \AA}$ ,  $V_S = -2.0 \text{ V}$ ,  $I_t = 0.5 \text{ nA}$ .

At 2.1 ML, islands with heights between 7 and 9  $\text{\AA}$ , corresponding to 3 or 4 MLs of Ag(111), are dominant, although a few shorter islands can also be found. In addition, the nucleation of small islands on the flat islands is occasionally observed as shown in Fig. 4(b). Fig. 4(c) shows that for a Ag film of 3.2 ML, flat islands coalesced into large islands. Furthermore at 3.2 ML a large number of flat Ag nuclei are formed on the coalesced flat islands as shown in Fig. 4(c). The step height of small islands nucleated on flat islands is also estimated to be a monolayer step height of Ag(111).

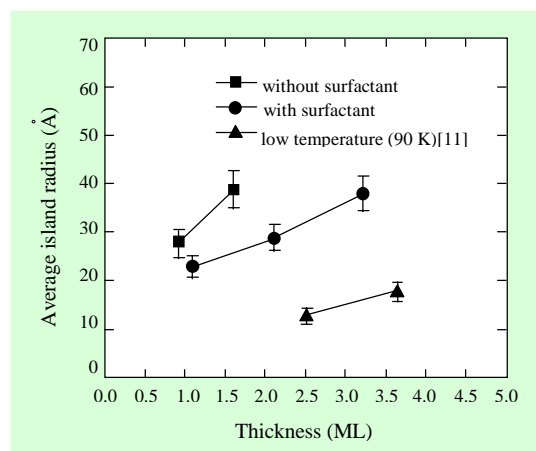
The height fluctuation of Ag islands is estimated to be less than 2 ML at 3.2 ML Ag films. Although the Sb-terminated Si(111) surface is not completely covered with Ag islands and the crystal orientations of Ag islands also do not match well

with Si(111) substrate even at 3.2 ML [30], the trend toward the growth of atomically flat and uniform Ag films when the surfactant Sb is present is clearly shown in this work.

The island densities of each film with and without Sb at room temperature as well as those at low temperature [11] are shown in Fig. 5. These indicate that in our study the island density increased when surfactant was used, compared to the sample without surfactant. This result suggests that the surface diffusion barrier of Ag atoms on an Sb buffer layer be higher than that on the intermediate Ag monolayer on clean Si(111), since island density depends on the surface diffusion barrier of the substrate. The increased island density causes the height of Ag islands to be limited to 3 or 4 ML before coalescence of



**Fig. 5.** The island densities of Ag films on a Si(111) surface with and without Sb surfactant at room temperature and at low temperature (90 K) [11].



**Fig. 6.** The average island sizes of Ag films on a Si(111) surface with and without Sb surfactant at room temperature and those of Ag film grown on Si(111) at low temperature (90 K) without surfactant [11].

the Ag islands. The small Ag islands eventually coalesce into larger ones before high 3D islands begin to grow.

The average island radius obtained from

Ag grown on Si(111) without surfactant and in the low temperature growth of Ag/Si(111) [11] are shown in Fig. 6. The multilayer growth of Ag film in Ag/clean Si(111) system is probably because of the low island density. In the LEEM study of Ag growth on Sb/Si(111) by van der Gon *et al.* [24], a similar dense nucleation effect of Ag islands was observed when Sb was used as a surfactant. However, since Ag was grown at elevated temperature in their work, the island size was much larger than in our work.

In the growth of Ge on Sb-terminated Si, precovered Sb atoms diffuse to the surface via an exchange mechanism. This process plays an important role in the layer-by-layer epitaxial growth [14], [17], [18]. However no clear evidence of Sb segregation and its role in Ag film growth are found in this work. The nucleation of new monolayer height Ag islands on the already formed flat islands in the 3.2 ML high Ag film may be considered as an indirect evidence of a small amount of Sb segregation.

## V. SUMMARY

We have observed the atomically flat and uniform growth of Ag on a Si(111) substrate treated with an Sb surfactant. STM images showed that a rough layer with round islands was formed having a 1~3 monolayer height in the 1.1 ML film, and afterwards atomically flat Ag islands grew uniformly on the Sb-terminated surface. The STM analysis of island density



and size in Ag growths on clean Si(111) and on Sb-terminated Si(111) surface suggests that the increment in island density be caused by an increase of surface diffusion barrier for Ag atoms on the Sb-terminated Si(111) surface. This kinetic effect of Sb seems to play a crucial role in the growth of flat and uniform Ag film on Si(111) on an atomic level at room temperature.

## VI. ACKNOWLEDGMENTS

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