



# C<sub>70</sub> self-assembly on In- and Tl-adsorbed Si(111) $\sqrt{3} \times \sqrt{3}$ -Au surfaces



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## ABSTRACT

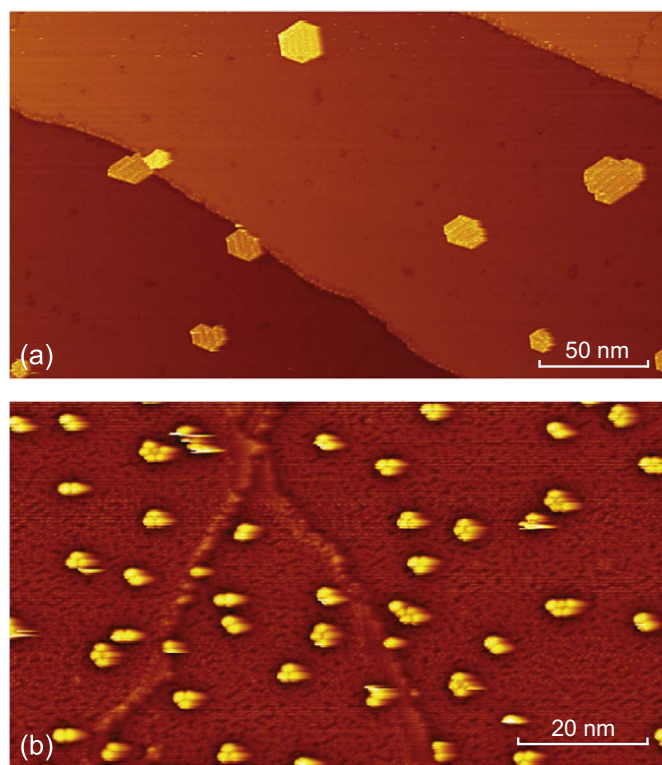
Behavior of C<sub>70</sub> fullerenes adsorbed onto the In- and Tl-modified Si(111) $\sqrt{3} \times \sqrt{3}$ -Au surfaces at room temperature (RT) and 112 K has been studied using scanning tunneling microscopy observations and compared with the known results for the C<sub>60</sub>s on the same surfaces. During island growth at 112 K, both C<sub>70</sub> and C<sub>60</sub> are characterized by the same critical island size  $i=1$ . The difference is in an island shape as C<sub>70</sub>s tend to form chain-like islands built of double or triple molecular rows along the  $[\bar{1}10]$  Si(111) substrate directions. At RT, C<sub>70</sub>s have a lower mobility as compared to C<sub>60</sub>s. In contrast to the C<sub>60</sub> magic islands, the C<sub>70</sub> islands do not demonstrate any strong preference for specific shapes or sizes. Extended C<sub>70</sub> arrays exhibit a stripe-like 3×1 reconstruction where a single row of bright fullerenes mediated by a double row of dim fullerenes. The reconstruction is due to the different orientations of C<sub>70</sub>s within the layer which does not relate to the structure of the underlying Au/Si(111) substrate. This is in contrast to the hexagonal reconstructions of C<sub>60</sub>s where the bright fullerenes occupy the specific adsorption site atop Au trimers. The difference in the behavior of C<sub>70</sub> and C<sub>60</sub> is plausibly affected by the non-spherical shape of C<sub>70</sub> molecule and a greater intermolecular C<sub>70</sub>-C<sub>70</sub> interaction.

## 1. Introduction

The adsorption of fullerenes onto metal and semiconductor surfaces has attracted considerable interest due to a variety of the fascinating phenomena underlying fullerene self-assembly. Among such phenomena one can mention developing modulations in the close-packed fullerene monolayers which shows up as appearance of molecules having two different contrast in scanning tunneling microscopy (STM) images: “bright” and “dim”. The bright-dim contrast was observed in C<sub>60</sub> monolayers on many noble-metal surfaces, including Ag(111) [1,2], Ag(100) [3–5], Au(111) [6–9], Cu(111) [10], Cu(100) [11]. The apparent height difference between bright and dim molecules can amount to  $\sim 1\text{--}2 \text{ \AA}$ . However, the origin of this contrast remains a debated subject as it can result from the electronic effects, the molecular orientations, or the geometric effects due to substrate reconstruction. As for the metal/silicon surface phases, i.e. surface reconstructions induced by adsorption of metal submonolayers onto Si crystalline substrates, the distinct dim-bright contrast was observed in the C<sub>60</sub> monolayers on the pristine Si(111) $\sqrt{3} \times \sqrt{3}$ -Ag surface [12,13] and Si(111) $\sqrt{3} \times \sqrt{3}$ -Au surface modified by In [14,15] or Tl [16] adsorption. In particular, the dim fullerenes in the C<sub>60</sub> monolayers on Si(111) $\sqrt{3} \times \sqrt{3}$ -Ag constitute  $\sim 10\%$  of all fullerenes and reside  $\sim 1.6 \text{ \AA}$  lower than other fullerenes. It was suggested that formation of the dim

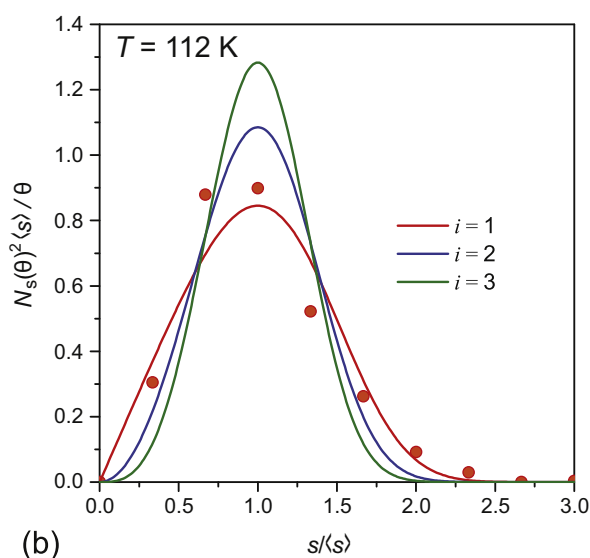
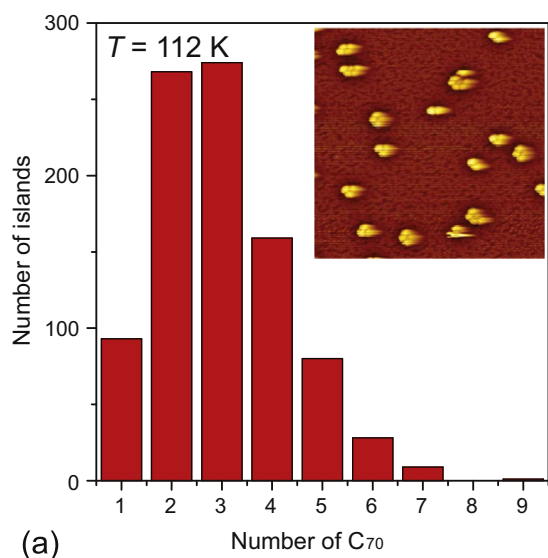
C<sub>60</sub> is associated with a disintegration of Ag trimer of Si(111) $\sqrt{3} \times \sqrt{3}$ -Ag surface beneath a given fullerene [13]. In the C<sub>60</sub> monolayers on the In- or Tl-adsorbed Si(111) $\sqrt{3} \times \sqrt{3}$ -Au surfaces, the bright fullerenes are arranged in the well-defined two-dimensional (2D) lattices [14,16]. Note that adsorption of  $\sim 0.1\text{--}0.2 \text{ ML}$  of In or Tl onto Si(111) $\sqrt{3} \times \sqrt{3}$ -Au was used to remove domain walls, characteristic of the pristine surface [17], and to produce almost defect-free highly-ordered and homogeneous Si(111) $\sqrt{3} \times \sqrt{3}$ -Au surfaces [18–20]. These surfaces preserve atomic arrangement of the pristine surface [21] but contains also a 2D gas of In or Tl adatoms. The bright C<sub>60</sub> on these surfaces were recognized to reside directly atop the Au trimers, the most energetically unfavorable adsorption sites [14,15]. This fact has a remarkable sequence for the C<sub>60</sub> island growth leading to size selection and magic C<sub>60</sub> island formation. For example, more than 80 % of C<sub>60</sub> islands grown on In-adsorbed Si(111) $\sqrt{3} \times \sqrt{3}$ -Au surface can have identical shape and size, namely each contains 37 fullerenes arranged in a regular hexagon [15]. Bearing in mind this advantageous property of the particular fullerene-substrate pair, it seemed of interest to explore how changing of the spherical fullerenes C<sub>60</sub> for the elongated fullerenes C<sub>70</sub> would affect their growth mode. It is worth noting, however, that in many adsorption phenomena, C<sub>70</sub> differ from C<sub>60</sub> not only by the size and shape but rather by the different charge transfer [22], kinetic characteristics [23] and intermolecular interaction [24].

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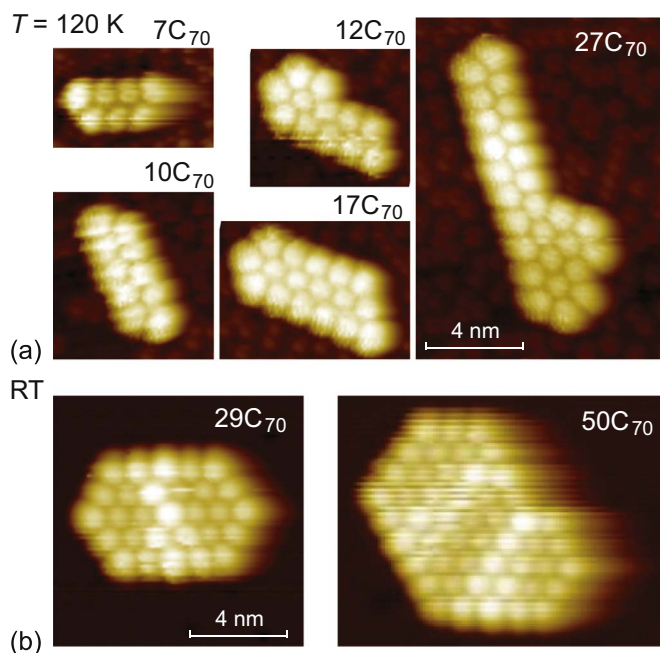


**Fig. 1.** STM images illustrating early stage of  $C_{70}$  island growth on In-adsorbed  $\text{Si}(111)\sqrt{3} \times \sqrt{3}$ -Au held at (a) RT and (b) 112 K. Scale: (a)  $300 \times 165 \text{ nm}^2$ , (b)  $100 \times 55 \text{ nm}^2$ .

In the present paper, we report on the results of the STM study of  $C_{70}$  adsorption and self-assembly on In- and Tl-adsorbed  $\text{Si}(111)\sqrt{3} \times \sqrt{3}$ -Au surface which results are compared with those reported for  $C_{60}$  on the same surfaces. It has been found that non-spherical shape of  $C_{70}$  alters significantly the growth mode. Starting from the early stages, the islands tend to adopt elongated shapes. In the extended arrays, the molecules are arranged in the linear superstructure with alternating sequence of bright and dim  $C_{70}$  rows, namely one bright row is typically mediated by two dim rows. The superstructure does not demonstrate a clear resemblance with the structure



**Fig. 2.** (a) Experimental  $C_{70}$  island size distribution at 112 K. Inset shows a fragment of the surface with  $C_{70}$  island array under consideration. (b) Experimental scaled island size distribution (red circles) superposed on the simulated scaling functions  $f_i$  for critical island size  $i=1, 2$ , and 3 (red, blue and green lines, respectively). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)



**Fig. 3.** Selection of  $C_{70}$  islands grown at (a) 112 K and (b) RT.

of the underlying Au/Si(111) substrate and is plausibly a result of the interactions between molecule in various orientations.

## 2. Experiment

Our experiments were performed with an Omicron VT-STM operating in an ultrahigh vacuum ( $\sim 2.0 \times 10^{-10}$  Torr). Atomically-clean  $\text{Si}(111)7 \times 7$  surfaces were prepared *in situ* by flashing to  $1280^\circ \text{C}$  after the samples were first outgassed at  $600^\circ \text{C}$  for several hours. Gold was deposited from an Au-wrapped tungsten filament, indium and thallium from the Ta crucibles and fullerenes,  $C_{70}$  (BuckyUSA, purity 99.5%), from a resistively heated Ta boat. For STM observations, electrochemically etched tungsten tips cleaned by *in situ* heating were employed. To prepare the In- or Tl-modified  $\text{Si}(111)\sqrt{3} \times \sqrt{3}$ -Au surfaces, the  $\text{Si}(111)\sqrt{3} \times \sqrt{3}$ -Au surface was first formed by Au deposition onto  $\text{Si}(111)7 \times 7$  surface held at  $600^\circ \text{C}$  and then  $\sim 0.5 \text{ ML}$  of In or Tl was

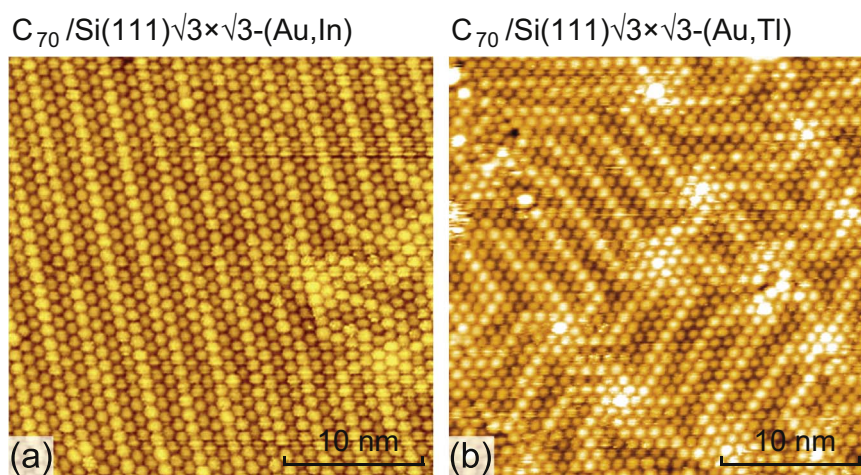


Fig. 4.  $30 \times 30 \text{ nm}^2$  STM images of  $C_{70}$  arrays grown on (a) In-adsorbed and (b) Tl-adsorbed  $\text{Si}(111)\sqrt{3} \times \sqrt{3}$ -Au surfaces at RT.

deposited onto this surface held at room temperature (RT) followed by brief ( $\sim 15$  s) annealing at  $\sim 600^\circ \text{C}$  for In and  $\sim 350^\circ \text{C}$  for Tl.

### 3. Results and discussion

First of all we would like to remark that the main regularities of  $C_{70}$  adsorption and layer growth were the same for both In- and Tl-adsorbed  $\text{Si}(111)\sqrt{3} \times \sqrt{3}$ -Au surfaces. Fig. 1 illustrates the early stage of  $C_{70}$  layer growth at RT. Formation of the scarce islands indicates that  $C_{70}$  molecules demonstrate a relatively high mobility. However, nucleation of islands not only at the step edges but also at the terraces yields a rough estimation of diffusion length on the order  $\sim 50$ – $100$  nm. This is lower than the corresponding value for RT migration of  $C_{60}$  on the same surfaces, where islands nucleate exclusively at the step edges even when the terraces are more than  $100$  nm in width. Note that a slower migration of  $C_{70}$  as compared to  $C_{60}$  can be thought as a general trend as a sequence of the difference in the molecule shape. In particular, the same rule was found to hold for the migration of  $C_{60}$  and  $C_{70}$  on  $\text{Si}(111)\sqrt{3} \times \sqrt{3}$ -Ag surface [23]. The lower mobility of  $C_{70}$  molecules can be attributed to their non-spherical shape bearing in mind that rolling is rather typical for fullerene motion on surfaces.

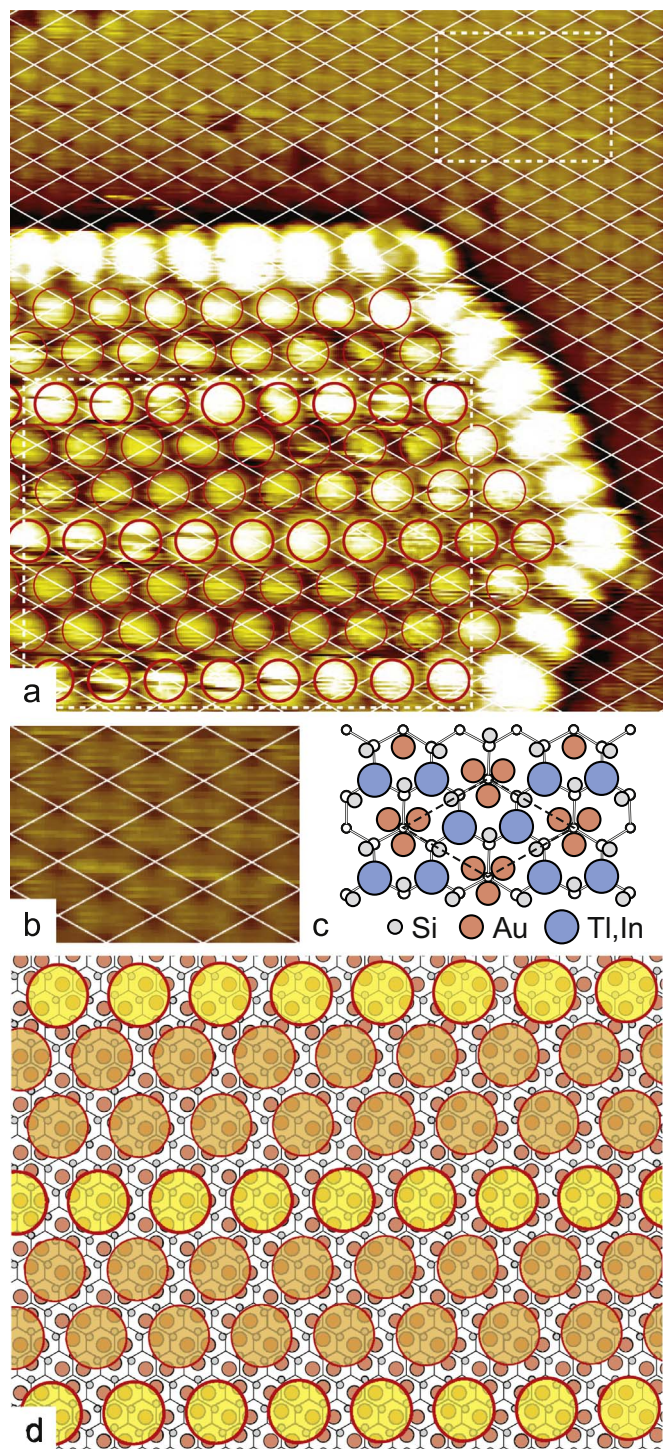
When adsorption of  $C_{70}$  takes place at low temperature of  $112$  K, fullerenes agglomerate into molecular islands at terraces with island density of  $\sim (0.5 - 1.0) \times 10^{-2} \text{ nm}^{-2}$ , hence the mean distance between islands is on the order of  $\sim 10$  nm. Fig. 1b shows STM image of such an array and Fig. 2a presents its island size distribution. These data can be processed using the scaling relation [25,26],  $N_s = \Theta \langle s \rangle^{-2} f_i(s/\langle s \rangle)$ , where  $N_s$  is the density of islands of size  $s$  (the number of molecules in the island),  $\Theta$  is the coverage of adsorbate,  $\langle s \rangle$  is the average island size, and  $f_i$  is the scaling function for the island size distribution for a critical island size  $i$ . Plotting  $N(s/\langle s \rangle) \langle s \rangle^2 \Theta^{-1}$  versus  $s/\langle s \rangle$  (that essentially yields the experimental scaling function  $f_i$ ) and comparing it with the simulated scaling functions, one can determine the size of critical island  $i$ . Such an evaluation is illustrated in Fig. 2b. One can see that the experimental data fit the simulated  $f_i$  plot for  $i=1$ . Remind that critical island size  $i$  is defined as the minimal size when the addition of just one more molecule makes the island stable. Hence,  $C_{70}$  dimer on the In- or Tl-adsorbed  $\text{Si}(111)\sqrt{3} \times \sqrt{3}$ -Au surfaces is already a stable island which is the same as reported for the  $C_{60}$  molecules [27,28]. It is worth noting that this is rather typical situation for fullerenes adsorbed onto the relatively inert surfaces [29,30], in which case the intermolecular interaction is greater than that of molecules with a substrate.

Peculiar feature of the  $C_{70}$  island arrays formed at  $112$  K is that a noticeable portion of the islands has a chain-like shape built of the double or triple molecular rows aligned along the main crystallographic directions of  $\text{Si}(111)$ ,  $[\bar{1}10]$ . Selected examples of such islands are

presented in Fig. 3a. When the islands are grown by  $C_{70}$  deposition at RT or by heating island arrays formed at  $112$  K to RT, the islands adopt generally a more compact shape (Fig. 3b). No noticeable preference for any specific island sizes and shapes are observed that is in contrast to the  $C_{60}$  fullerenes which under the same conditions demonstrate a strong tendency towards the growth of islands of magic sizes and shapes [15,16]. Remind that in the latter case the island size selection is controlled by the hexagonal superlattices ( $\sqrt{19} \times \sqrt{19} - R23.4^\circ$  for (Au, In)/ $\text{Si}(111)$  and  $\sqrt{7} \times \sqrt{7} - R19.1^\circ$  for (Au, Tl)/ $\text{Si}(111)$ ) formed by the bright  $C_{60}$  fullerenes which occupy the most energetically unfavorable adsorption sites atop the Au trimers.

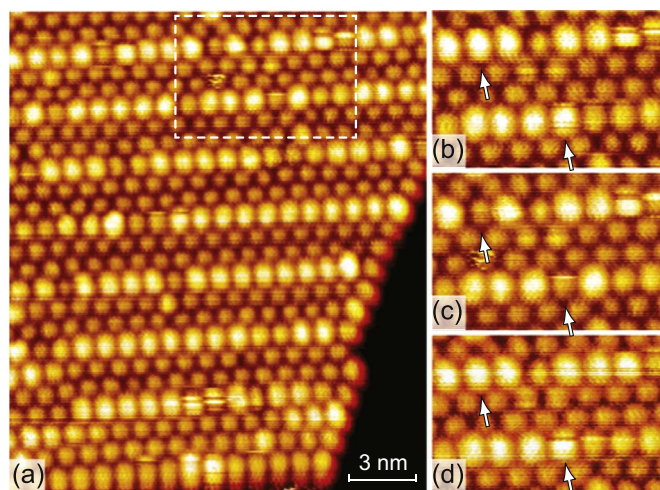
In case of the  $C_{70}$  fullerenes, another type of the dim-bright patterns develops. Its motifs appear in the islands starting from the size of about  $30$   $C_{70}$  fullerenes (Fig. 3b) and the pattern becomes well-defined in the extended  $C_{70}$  molecular arrays. Figs. 4a and b show such arrays formed on the In- or Tl-adsorbed  $\text{Si}(111)\sqrt{3} \times \sqrt{3}$ -Au surfaces, respectively. The  $C_{70}$  layer displays a stripe-like  $3 \times 1$  superstructure where single rows of the bright  $C_{70}$ s are mediated by double rows of dim  $C_{70}$ s. Following the notation proposed for the superstructured  $C_{60}$  monolayers on  $\text{Cu}(100)$  [11], the  $C_{70}$  superstructure can be defined as 1B2D since it has a one-bright-and-two-dim ordering sequence of molecular rows. The  $C_{60}$  monolayers on  $\text{Cu}(100)$  have a similar appearance in STM, albeit with the other ordering sequences, namely there are two phases, 2B1D and 1B1D [11]. However, in spite of an apparent similarity in STM appearance the driving force for developing striped superstructures in the two cases,  $C_{60}$  on  $\text{Cu}(100)$  and  $C_{70}$  on In- or Tl-adsorbed Au/ $\text{Si}(111)$  surfaces, is quite different. For  $C_{60}$  on  $\text{Cu}(100)$ , it develops due to superposing of the close-packed  $C_{60}$  film on a substrate of a different symmetry. In the present case, both the Au/ $\text{Si}(111)$  substrate and the closed-packed  $C_{70}$  layer possess basic hexagonal symmetry and the striped superstructure develops in the molecular layer due to the non-spherical shape of  $C_{70}$  molecules.

Let us consider now the structural properties of the 1B2D  $C_{70}$  monolayer in a greater detail. The  $C_{70}$  rows are aligned along the  $[\bar{1}10]$  directions of the  $\text{Si}(111)$  substrate. Hence, following the substrate symmetry there are three equivalent  $120^\circ$ -rotated domains. The 1B2D contrast between fullerene rows in STM images is the same at both, positive and negative, bias polarities. An apparent height difference between dim and bright fullerenes is  $0.3 \pm 0.1 \text{ \AA}$  in the case of  $C_{70}$  on In-adsorbed  $\text{Si}(111)\sqrt{3} \times \sqrt{3}$ -Au surface. In the case of Tl-adsorbed surface, the contrast is somewhat higher,  $\sim 0.5 \text{ \AA}$ , though all other features of the superstructures are the same. In the regions where three rotational domains intersect, the fullerenes have the greatest apparent height. Thus, they can be called “superbright” fullerenes by analogy with the superbright  $C_{60}$  molecules on  $\text{Ag}(111)$  [2]. Scanning tunneling spectroscopy measurements conducted on dim, bright and superbright

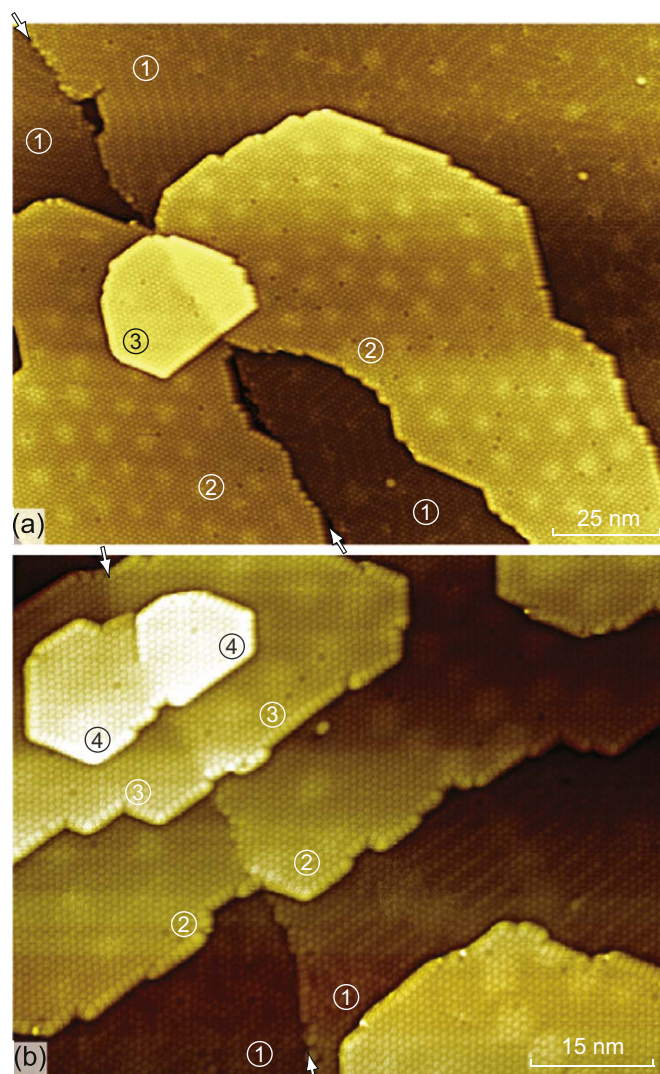


**Fig. 5.** Identification of the  $C_{70}$  adsorption sites within the molecular array. (a) STM image with coexisting  $C_{70}$  array and bare  $\text{Si}(111)\sqrt{3} \times \sqrt{3}$ -Au, In surface with the grid associated with the  $\sqrt{3} \times \sqrt{3}$  periodicity. (b) Enlarged region of the  $\text{Si}(111)\sqrt{3} \times \sqrt{3}$ -Au, In surface outlined by the dashed frame in (a). (c) Schematics of the  $\text{Si}(111)\sqrt{3} \times \sqrt{3}$ -Au surface. Au trimers are shown by red circles, Si atoms by gray and white circles, blue circles indicate the adsorption sites of In or Tl atoms. The  $\sqrt{3} \times \sqrt{3}$  unit cell is outlined to illustrate that depressions in (b) correspond to location of Au trimers [18,19]. (d) Schematics of the  $C_{70}$  array region outlined in (a) superposed on the structure of the underlying  $\text{Si}(111)\sqrt{3} \times \sqrt{3}$ -Au surface. Bright  $C_{70}$ s are shown by yellow circles, dim  $C_{70}$ s by orange circles. One can see that fullerene adsorption sites are random without any apparent difference between those for bright or dim fullerenes.

$C_{70}$  molecules have revealed that their spectra are very similar. This imply that STM contrast is primarily due to topography. However, modest bias-dependent variation in apparent height difference is an



**Fig. 6.** (a)  $19 \times 22 \text{ nm}^2$  high-resolution STM image of the  $C_{70}$  array acquired at 112 K. (b), (c), and (d) Successive snapshots of the same fragment of the array where  $C_{70}$  molecules changing their appearance are indicated by arrows.



**Fig. 7.**  $150 \times 120 \text{ nm}^2$  STM image illustrating the onset of the third-layer island growth. (b)  $85 \times 66 \text{ nm}^2$  STM image showing the onset of the fourth layer formation. The numbers in circles indicate the layer number in the multi-layer systems. Arrows indicate the location of the atomic step at the Au/Si(111) substrate surface.

indication that certain electronic effects cannot be completely excluded.

It is worth noting that the 1B2D superstructure of the  $C_{70}$  layers has no resemblance with the structure of the underlying  $\text{Si}(111)\sqrt{3} \times \sqrt{3}$ -Au surface. This is illustrated in Fig. 5 which shows that fullerenes occupy random sites without any apparent difference between those for bright or dim  $C_{70}$ s. This is contrasted with the  $C_{60}$  monolayers where the bright fullerenes are always those residing atop the Au trimers, while dim molecules are those occupying all the other adsorption sites. Thus, one can conclude that the role of the intermolecular interactions in the superstructure formation is prevailing for the  $C_{70}$  layers, plausibly due to a greater  $C_{70}$ - $C_{70}$  binding as compared to the  $C_{60}$ - $C_{60}$  one.

Fig. 6a shows a high-resolution STM image with a fragment of the 1B2D  $C_{70}$  array. One can see that dim  $C_{70}$ s have a round shape, while bright  $C_{70}$  have an oval shape with a long size oriented perpendicular to the direction of molecular row. This is in apparent contradiction with the expectation that bright fullerenes stand upright, while dim fullerenes lie on the surface. In turn, this implies that arrangement of  $C_{70}$  in the molecular layer is controlled by the more complicated mechanisms. For example,  $C_{70}$  molecules were suggested to tend to arrange in a configuration in which electron-poor regions on one molecule face electron-rich regions on the adjacent molecule [24]. It should be noted also that fullerenes in the  $C_{70}$  layer are in continuous rotation as evidenced by lacking of the intermolecular features characteristic of molecules in a fixed orientation. Moreover, the layer changes dynamically even at 112 K and selected bright  $C_{70}$ s transfer to dim  $C_{70}$ s and vice versa, as illustrated in Fig. 6b, c, and d.

With prolonged  $C_{70}$  deposition, almost the whole surface becomes occupied by the  $C_{70}$  monomolecular layer. Formation of the second-layer islands starts when more than 80% of the surface is already covered by the first layer. Thus, at this stage the growth seems to follow the 2D layer-by-layer mode. However, when occupation of the second layer exceeds 20%, nucleation of the third-layer islands takes place (Fig. 7a), rapidly followed then by nucleation of the fourth-layer islands (Fig. 7b). Thus, the growth mode changes to the 3D multi-layer one. Each time, the next-layer islands nucleate preferentially where the atomic steps of the underlying Au/Si(111) substrate are situated. One can see that the step is inherited in the  $C_{70}$  molecular layers with a progressive smearing with each next layer. The third-layer islands appear as single molecular sheets that are spread over the step and cover both terraces. The fourth-layer islands typically contain screw dislocations. As for the superstructure of the  $C_{70}$  layers, one can see that the traces of the 1B2D superstructure of the first layer can be noticed in the next layers, though it concerns mainly the motifs of the superbright fullerenes. Note that a similar inheritance of the first-layer superstructure in the next layers were observed with the  $C_{60}$  fullerenes [14], though for a different origin of the superstructure. In the present case, the superstructure in the first layer is associated with a different orientation of elongated  $C_{70}$  molecules, hence a similar STM contrast of all molecules in the third and fourth layers implies that they are plausibly in the same orientation, namely oriented upright.

#### 4. Conclusions

In conclusion, self-assembly of  $C_{70}$  fullerenes adsorbed onto the In- and Tl-modified  $\text{Si}(111)\sqrt{3} \times \sqrt{3}$ -Au surfaces at RT and 112 K has been studied. As compared to the  $C_{60}$  fullerenes on the same surfaces, the difference in the behavior of  $C_{70}$ s stems from its non-spherical shape and a greater  $C_{70}$ - $C_{70}$  bonding. At 112 K,  $C_{70}$ s tend to form chain-like islands built of double or triple molecular rows along the  $[\bar{1}10]$  Si(111) substrate directions, while the  $C_{70}$  arrays formed at RT exhibit a stripe-like  $3 \times 1$  reconstruction where a single row of bright fullerenes mediated by double row of dim fullerenes. The reconstruction does not relate to the structure of the underlying Au/Si(111) substrate and originates from the different orientations of  $C_{70}$ s within the layer. As a result, the reconstruction does not cause any island size selection that

is in contrast to the hexagonal reconstructions of  $C_{60}$ s where the bright fullerenes occupying the specific adsorption sites control formation of the magic-size islands.

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