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Atomic-Scale Study of Ge-Induced Incommensurate Phases on Si(111) *

WU Rui(吴蕊)[1], WANG Li-Li(王立莉)[1**], ZHANG Yi(张翼)[1], MA Xu-Cun(马旭村)[1], JIA Jin-Feng(贾金锋)[2], XUE Qi-Kun(薛其坤)[1,2]

[1] *Institute of Physics, Chinese Academy of Sciences, Beijing 100190*
[2] *Department of Physics, Tsinghua University, Beijing 100084*

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Two Ge-induced incommensurate phases, γ and β, on Si(111) are observed and studied by in situ scanning tunneling microscopy. The γ phase consists of aligned triangular domains whose stacking sequence is faulted with respect to the Si(111)-1×1 surface. The β phase consists of two kinds of triangular domains whose stacking sequences are faulted and unfaulted with respect to the Si(111)-1×1 surface, respectively. In the β phase, two types of domain walls, "zigzag" and "face-to-face", form to release the strain. The triangular domains all exhibit a quasi-1×1 hexagonal close-packed structure. By studying the structural evolution from magic clusters to incommensurate structures, the structure models for γ and β phases are proposed.

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Adsorption on semiconductor surfaces in the submonolayer coverage regime is often determined by adsorbate-substrate interaction and can result in ordered, periodic and commensurate reconstructions in many systems.[1] When coverage increases to one monolayer (ML), the adsorbate-adsorbate interaction becomes important and can accumulate strain in the system. One of the effective ways to release the strain is formation of an incommensurate structure composed of domains and domain walls. It occurs typically in metal on semiconductor surfaces,[2] such as Al/Si(111),[3,4] Al/Ge(111),[5] Ga/Si(111),[6−8] Ga/Ge(111),[9,10] In/Si(111),[11] In/Ge(111),[12] Cu/Si(111),[13] Cu/Ge(111).[14]

On the other hand, for semiconductor/semiconductor heteroepitaxial systems such as Ge/Si(111), where both the adsorbate-adsorbate and adsorbate-substrate interactions are strong and directional, the situation may become more complicated. Ge/Si(111) has been a model system for studying semiconductor heteroepitaxy, due to its simplicity and large lattice mismatch (4.2%) between Ge and Si.[15] Initial growth of Ge on the Si(111)-7×7 surface has been extensively investigated.[15−18] When Ge coverage is less than 0.10 ML, Ge atoms tend to substitute the corner Si adatoms in the faulted half unit cells of Si(111)-7×7.[16] At 1/3 ML, Ge atoms occupy the surface T_4 sites forming ($\sqrt{3}\times\sqrt{3}$) R30°-Ge reconstruction (called $\sqrt{3}\times\sqrt{3}$ hereafter).[17] At a coverage of 0.50 ML, Ge clusters form and are arranged in a regular hexagonal structure without covering the corner holes and dimer rows of Si(111)-7×7.[18] However, little work has been conducted for the situation close to ~ 1 ML, which is very important for understanding subsequent growth of Ge films. In this work, by using scanning tunneling microscopy (STM) we have carefully investigated the surface structure near this critical coverage. Two Ge-induced incommensurate phases (γ and β) are observed. By studying their atomic structures, the strain relaxation mechanism is discussed.

Our experiments were carried out in an ultra-high vacuum (better than 2×10^{-10} mbar) variable-temperature STM system combined with molecular beam epitaxy. Ge (99.999%) was evaporated onto Si(111)-7×7 surfaces at a typical flux rate of 0.05 ML/min (1 ML $= 7.8\times10^{-14}$ atoms/cm^2). The γ and β phases were prepared by depositing ~ 0.6 ML and ~ 0.9 ML Ge on Si at a substrate temperature of 550°C, respectively. In addition, to study their formation process, we first prepared the well-known $\sqrt{3}\times\sqrt{3}$ structure by depositing 1/3 ML Ge on Si at 550°C. Then, 1/6 ML Ge was deposited onto the $\sqrt{3}\times\sqrt{3}$ structure at room temperature. Lastly, annealing to 320°C or 420°C was conducted to track different structures. All the STM images were taken at room temperature with a sample bias of +2 V.

Figures 1(a)–1(b) show the empty-state STM images acquired on the sample after ~ 0.6 ML Ge deposition at a substrate temperature of 550°C. The surface is mostly covered by triangle-like domains (marked by the white solid triangles) with the same orientation. The triangular domains are alternately separated by triangular regions without Ge (marked by a white dashed triangle in Fig. 1(b)). Most of the domains have $n = 4$ atoms on their sides. The domains assume a nearly 1×1 structure with a lattice spacing of 0.40 ± 0.02 nm. The domains have the same stacking sequence with respect to the substrate, since the alignments of atoms in different domains have no

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**Email: llwang@aphy.iphy.ac.cn

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relative shift (see the black line in Fig. 1(b)). We call this structure the γ phase.

When ~ 0.9 ML Ge was deposited under the same condition, the surface becomes more ordered, as shown in Figs. 1(c)–1(d). In this case, the surface is really completely covered by triangular domains (marked by the white triangles in Fig. 1(d)) with alternating opposite orientations. The triangular domains retain the same 1×1 structure as those in the γ phase, but have larger sizes with $n = 7$ atoms on their sides while $n = 4$, 5 and 6 also appear. In addition, the triangular domains with opposite orientations have different stacking sequences with respect to the Si(111)-1×1, since the atomic alignments in adjacent domains have a relative shift (see the black line in Fig. 1(d)). Basically, there are two types of domain walls between the triangular domains, which is ascribed to release the strain.[2] The more common one is a "zigzag" domain wall (marked by the white zigzag lines in Fig. 1(d)), where Ge atoms are arranged in a zigzag manner and are separated by 0.76 nm. The other type is a "face-to-face" domain wall (marked by the white parallel lines in Fig. 1(d)), where Ge atoms face each other and are separated by 0.44 nm. Note that face-to-face domain walls occur only around smaller domains (typically $n = 4$ atoms on the side). We call this structure the β phase. Intuitively, the Ge coverage of the β phase should be twice that in the γ phase.

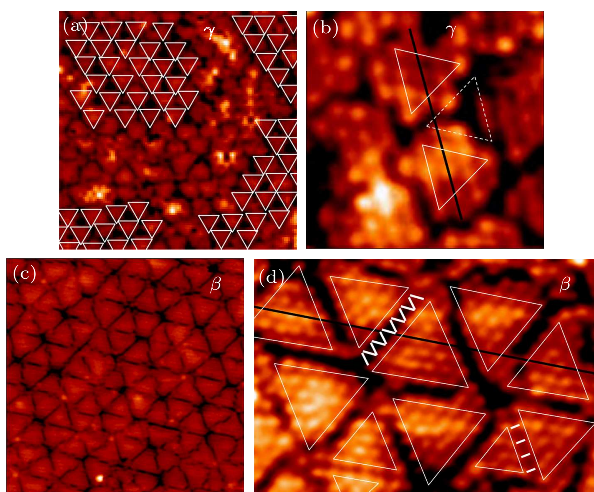

Fig. 1. Empty-state STM images of ~ 0.6 ML [(a) and (b)] and ~ 0.9 ML [(c) and (d)] Ge deposited on Si(111) at 550°C. Triangular domains are outlined by the white solid triangles. In (b), an empty triangular region without Ge is outlined by a white dashed triangle. In (d), the white lines between domains show two types of domain walls. The black lines in (b) and (d) indicate the relative positions of atomic alignments. The tunneling current setpoints are 0.02 nA in (a)–(c) and 0.2 nA in (d). Image sizes: (a) and (c) 30 nm × 30 nm, (b) 4 nm × 4 nm, and (d) 9.2 nm × 6.4 nm.

To further understand the atomic structure and bonding configuration, we conducted a two-step deposition and post-annealing treatment. Figures 2(a)–2(c) show the typical high-resolution empty-state STM images with 1/6 ML Ge deposited on $\sqrt{3}\times\sqrt{3}$ surface at room temperature and annealed to 320°C. Figure 2(a) indicates that many triangular clusters are formed on the $\sqrt{3}\times\sqrt{3}$ surface and all of them have the same orientation, whose edges point exclusively to $\langle 11\bar{2}\rangle$. The clusters with $n = 2$, 3, 4, 5, 6 atoms on their sides (marked by the white circles) would be referred to as magic clusters for their enhanced abundance and stability.[19] As shown in Fig. 2(b), clusters with $n = 4$ atoms are the most abundant. Furthermore, the fact that annealing to 420°C results in aggregation of all the clusters and the formation of γ and β phases (not shown) reveals that triangular domains of γ and β phases are made of the triangular magic clusters. Thus, the structure model of the γ and β phases could be proposed based on that of the magic clusters.

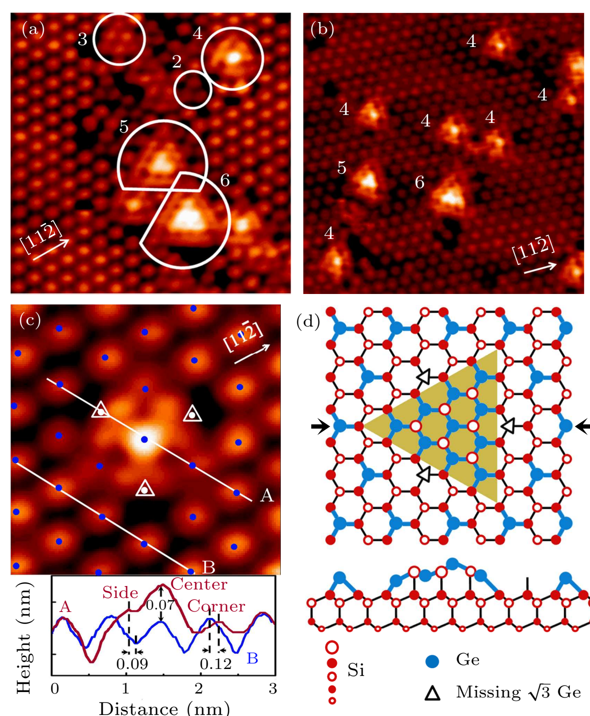

Fig. 2. (a)–(c) Empty-state STM images of 1/6 ML Ge deposited on the Si(111)-($\sqrt{3}\times\sqrt{3}$) R30°-Ge surface at room temperature and annealed to 320°C. In (a), the magic triangular clusters of $n = 2$, 3, 4, 5, 6 are outlined by the white circles. In (c), three white small triangles indicate three characteristic vacancies on the surrounding $\sqrt{3}\times\sqrt{3}$ lattice, and the line profiles (A and B) corresponding to the white lines (A and B) are shown at the bottom. The tunneling current setpoint is 0.1 nA. Image sizes: (a) 10 nm × 10 nm, (b) 14 nm × 14 nm, and (c) 3.5 nm × 3.5 nm. (d) Proposed model for the magic triangular cluster of $n = 4$ shown in (c).

To find out the atomic structure of the magic clusters, we examined the most abundant magic cluster of $n = 4$ in detail (see Fig. 2(b)). The STM observation shows that three atoms (marked by the triangles) around this cluster are missing from the surrounding

$\sqrt{3}\times\sqrt{3}$ lattice. The comparison of profile A on the cluster and profile B on the surrounding $\sqrt{3}\times\sqrt{3}$ lattice reveals the registration of this cluster. The center atom is right above the original $\sqrt{3}$ adatom site (T_4 site), but moves upwards by $\sim$ 0.07 nm. The three corner atoms are almost on the original $\sqrt{3}$ adatom sites (T_4 sites), but move outwards by $\sim$ 0.12 nm. On the other hand, the six side atoms are approximately above the midpoint sites (degenerate T_4 sites) of two neighboring $\sqrt{3}$ adatoms, but move outwards by $\sim$ 0.09 nm and upwards by $\sim$ 0.02 nm. As a result, this cluster has a nearly 1×1 structure with a lattice spacing of 0.40 ± 0.02 nm, and its stacking sequence is faulted with respect to the Si(111)-1×1 surface. We also examined the magic cluster of $n=5$ (see Fig. 2(a)). Likewise, the characteristic vacancies around the cluster are found in the surrounding $\sqrt{3}\times\sqrt{3}$ lattice. Also, its three interior atoms are right above the original $\sqrt{3}$ adatom sites (T_4 sites), but significantly deviate upwards, while its side atoms are approximately on the original $\sqrt{3}$ adatom sites (T_4 sites).

In view of these characteristics of magic clusters, a model (see Fig. 2(d)) for the magic cluster of $n=4$ is proposed, which essentially consists of a Ge-Si bilayer embedded in the $\sqrt{3}\times\sqrt{3}$ lattice.[7] The cluster is composed of one Ge atom at the center, six additional Si atoms (which are presumably invisible in the empty-state STM image) in the interior, and nine Ge atoms on the side. The center Ge atom is bound to three of six additional Si atoms and belongs to the outer half of the Ge-Si bilayer, which is consistent with the observed higher contrast. The six additional Si atoms in the interior are located on the atop sites of Si(111)-1×1 to saturate the dangling bonds of ten Ge atoms as much as possible, which enhances the stability of this cluster. Meanwhile, their existence could better explain the increased lattice spacing ($\sim$ 0.40 nm) in the magic clusters. The side Ge atoms are bound not only to the inside additional Si atoms, but also directly to the outside Si surface, similar to the $\sqrt{3}$ adatoms. Such an unusual bonding configuration for the side atoms is qualitatively consistent with the STM observation that they are only slightly higher than the surrounding $\sqrt{3}$ adatoms. The stacking sequence of ten Ge atoms is faulted with respect to the Si(111)-1×1 surface and is consistent with the STM observation of their locations on T_4 sites. Naturally, there are three characteristic vacancies left in the surrounding $\sqrt{3}\times\sqrt{3}$ lattice, which avoids a conflict of bonding between an additional Ge atom on the vacancy site (marked by triangles) and the side atoms. In fact, the γ phase consists of aligned magic clusters of $n=4$. Therefore, this model is also applied to that of the triangular domains of the γ phase.

The model for clusters of $n=4$ could be generalized to magic clusters with different sizes, $Ge_{n(n+1)/2}Si_{n(n-1)/2}$, where n is the number of side Ge atoms. For $n<4$, the model includes the case of a single $\sqrt{3}$ Ge adatom, which can be treated as a special "cluster" of $n=1$. For the cluster of $n=2$ (see Fig. 2(a)), there are three Ge atoms on the sides and one Si atom at the center. For the cluster of $n=3$ (see Fig. 2(a)), there are six Ge atoms on the sides and three Si atoms in the interior. Due to the lack of second-layer atoms, clusters of $n=2$ and 3 should be considered as transitional species from one $\sqrt{3}$ adatom to bilayer nanostructures. For $n\geq4$, all the clusters described by this $Ge_{n(n+1)/2}Si_{n(n-1)/2}$ model have bilayer structure. The absence of clusters of $n\geq8$ implies that $n=7$ should be the upper limit for the size of such bilayer clusters. This coincides with Si(111)-7×7 and implies that the γ phase composed of magic clusters of $n=7$ can form thermodynamically. However, kinetically Ge atoms congregate to form clusters of $n=4$. Then, only a small amount of Ge atoms congregate to these clusters and most Ge atoms congregate to form new clusters on the spare triangular half, as the beginning of the β phase. Actually, Figs. 1(a) and 1(b) show the evidence. Hence, this $Ge_{n(n+1)/2}Si_{n(n-1)/2}$ model for larger clusters is also suitable for the triangular domains of the β phase. The only difference is that the stacking sequence changes alternately between faulted and unfaulted with respect to the Si(111)-1×1 surface. To release the strain domain walls occur in the β phase.

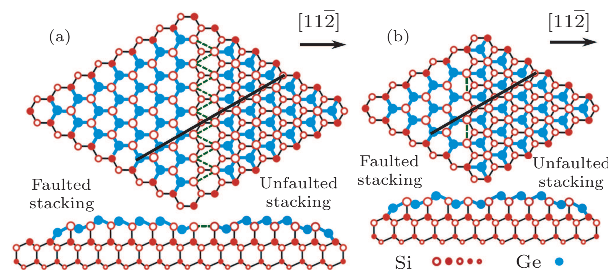

Fig. 3. Proposed models for two types of domain walls in β phase: zigzag (a) and face-to-face (b). The green dashed lines indicate the covalent bonds between Si atoms in the domain walls, and the black lines indicate the relative shifts of atomic alignments in the adjacent domains.

Figure 3 shows the proposed models for two types of domain walls in the β phase based on the generalized Ge-Si bilayer model. Figure 3(a) corresponds to the zigzag domain wall formed between the domain of $n=7$ and the domain of $n=6$, and Fig. 3(b) the face-to-face domain wall formed between the domain of $n=4$ and the domain of $n=6$, indicated by white lines in Fig. 1(d). Both are composed of two triangular domains, one is faulted (with $\langle 11\bar{2}\rangle$ edges) and the other is unfaulted (with $\langle\bar{1}\bar{1}2\rangle$ edges) with respect to the Si(111)-1×1 surface, which causes a relative shift

of atomic alignments. The black lines in Figs. 3 and 1(d) indicate the relative shift and the coincidence between the model and the experimental results. For the zigzag domain wall, the distance between the two rows of Si atoms is shortened with the lattice expansion of the Ge-Si bilayer so that they could be covalently bound in a zigzag chain (marked by the green dashed lines). For the face-to-face domain wall, two Si atoms are covalently bound to saturate each other and form a Si dimer chain (marked by the green dashed lines). The covalent bonds in the Si dimer chain are in a higher-energy state because the distance between Si atoms is increased. This is the reason why face-to-face domain walls seldom occur and appear only around small triangular domains of $n = 4$. Although the real widths of these two domain walls (0.76 nm and 0.44 nm) are 50% and 43% smaller than the values calculated based on the models (about $4a$ and $2a$, where a is the lattice parameter of Si(111)), which is reasonable since the 1×1 structure in domains is more than 10% expanded, the experimental and calculated ratios between the widths of these two kinds of domain walls are approximate, and the larger the domain is, the more compressed the domain wall is.

In summary, two Ge-induced phases on Si(111) near 1 ML coverage are studied using STM. The $\mathrm{Ge}_{n(n+1)/2}\mathrm{Si}_{n(n-1)/2}$ bilayer models are proposed for γ and β phases. The triangular domains of both phases basically exhibit the 1×1 structure. In the γ phase, the domains with $\langle 11\bar{2}\rangle$ edges are faulted with respect to the Si(111)-1×1 surface, while in the β phase the domains with $\langle 11\bar{2}\rangle$ and $\langle \bar{1}\bar{1}2\rangle$ edges are faulted and unfaulted with respect to the Si(111)-1×1 surface, respectively. To reduce the strain energy at increased Ge coverage, the zigzag and face-to-face domain walls occur in the β phase.

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