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Surface electronic structures of the Eu- and Ca-induced so-called Si(111)-(5×1) reconstructions

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We have investigated the electronic structures of the so-called Eu- and Ca-induced Si(111)-(5×1) surfaces by using angle-resolved photoelectron spectroscopy (ARPES) and low-energy electron diffraction (LEED). The LEED patterns of these surfaces indicate that the periodicities of both surfaces are actually (5×4). In the ARPES study, seven surface states were observed on each (5×4) reconstruction. Of these surface states, the dispersions of five of them show good agreement with those of the Eu- and Ca-induced (3×2) honeycomb-chain-channel (HCC) surfaces and the dispersions of the two other states agree well with those of the Eu- and Ca-induced (2×1) Seiwatz surfaces along the $\bar{1}10$ direction—i.e., the direction parallel to the adsorbate chain. Taking the dispersion behavior of these surface states into account, we conclude that the interaction between the nearest-neighbor HCC chain and Seiwatz chain is quite small and that the electronic structure of one chain hardly affects the electronic structure of its neighboring chain. We also discuss the atomic structure of the Eu- and Ca-induced Si(111)-(5×1) reconstructions based on their electronic structures.

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I. INTRODUCTION

During the last decade, self-organized one-dimensional (1D) and quasi-1D superstructures, which are formed on semiconductor surfaces by the adsorption of metal atoms, have drawn much interest due to the possibility of observing various exotic physical phenomena such as formations of non-Fermi-liquid-like ground states, Peierls-like phase transitions, or order-disorder transitions.^{1–6} The observations of these exciting physical phenomena led to a profound interest in measuring the electronic structures of the 1D and quasi-1D reconstructions induced by the adsorption of rare-earth metals (REM's) and alkaline-earth metals (AEM's) on a Si(111) surface.

Eu has been reported to form a quasi-1D (3×2) reconstruction at a coverage of 1/6 ML on Si(111), and a series of 1D [$(n \times 1)$; $n=5, 7$, and 9] reconstructions at higher coverages culminating in a (2×1) phase at 0.5 ML.^{7–11} The Ca/Si(111) surface has been reported to form quasi-1D and 1D reconstructions that are quite similar to those formed by Eu.^{12–14} Of these reconstructions, the atomic structure of the lowest-coverage (3×2) phase has been proposed to be basically the same as that of the honeycomb-chain-channel (HCC) model^{15–17} for both the Eu- and Ca-induced phases [Fig. 1(a)].^{8–11,14,18–22} The surface electronic structures of the Eu- and Ca-induced (3×2) phases resemble closely not only each other, but also the surface electronic structure of a Li-induced Si(111)-(3×1) HCC surface,^{18–22} and support indirectly the proposed structural model. Regarding the highest-coverage phase, the basic atomic structures of both the Eu- and Ca-induced (2×1) phases have been proposed to follow the Seiwatz structure²³ illustrated by the model in Fig. 1(b).^{9,11–14,18,21,24} The observation of two surface states, which originate from Si atoms that form a Seiwatz chain, on both the Eu- and Ca-induced (2×1) phases^{18,21,24} supports the structural model.

In contrast to the two end phases, the electronic structures of the intermediate phases [the (5×1), (7×1), and (9×1)

phases] have not been studied so far, though these reconstructions may show interesting physical phenomena. Further, although the atomic structures of these intermediate phases are considered to be basically formed by combinations of the HCC and Seiwatz structures,^{9–14} different structural models have been proposed for the so-called (5×1) phase. Structural models with a periodicity of (5×2) and adsorbate coverages of 0.4 monolayer (ML) or 1/3 ML have been proposed for the Eu- and Ca-induced reconstructions,^{10,14} and a model with a (5×4) periodicity and an adsorbate coverage of 1/3 ML has been proposed for the Eu-induced reconstruction.¹¹ [Figure 1(c) shows the (5×4) periodic model proposed in Ref. 11.] However, no strong evidence has been reported regarding the atomic structure of this intermediate phase, and the determination of the surface electronic structure of this phase is an important input to the structure determination. Moreover, the determination of the electronic structure of the so-called (5×1) phase is essential to fully understanding the physical properties of divalent-atom-induced quasi-1D reconstructions.

FIG. 1. (Color online) (a) HCC structure of the Si(111)-(3×2) surface with an adsorbate coverage of 1/6 ML, (b) Seiwatz structure of the Si(111)-(2×1) surface with an adsorbate coverage of 0.5 ML, and (c) the structural model of the so-called Si(111)-(5×1) surface proposed in Ref. 11. Solid circles are metal atoms and open circles are Si atoms. The dashed lines indicate the unit cell of each surface.

In this paper, we present detailed angle-resolved photoelectron spectroscopy (ARPES) measurements and low-energy electron diffraction (LEED) observations of the so-called Eu- and Ca-induced (5×1) reconstructions. In the LEED study, we found that the periodicities of both the so-called Eu- and Ca-induced (5×1) surfaces are actually (5×4) . The ARPES measurements were performed along the $[\bar{1}10]$ and $[11\bar{2}]$ directions of the Eu/Si(111)- (5×4) and Ca/Si(111)- (5×4) —i.e., directions parallel and perpendicular to the adsorbate chains (Fig. 1). Seven surface states were observed on each (5×4) reconstruction. Among these surface states, the dispersions of five of them show good agreement with the dispersions of the surface states of Eu- and Ca-induced (3×2) HCC surfaces in the $[\bar{1}10]$ direction. This result indicates that these surface states originate from the orbitals of Si atoms that form a HCC structure. The dispersions of the two other surface states agree well with those of the Eu- and Ca-induced (2×1) Seiwatz surfaces in the $[\bar{1}10]$ direction, and thus we conclude that they originate from orbitals of the Si atoms that form a Seiwatz structure. In the $[11\bar{2}]$ direction, although one surface state follows the $5 \times$ periodicity, we cannot discuss the dispersion features of the other surface states since they are only observed in small $k_{\parallel}$ regions. Based on the electronic structures and the LEED patterns, we also discuss the atomic structures of the so-called Eu- and Ca-induced (5×1) reconstructions.

II. EXPERIMENTAL DETAILS

The ARPES measurements and the LEED observations were performed at beamline 33 at the MAX-I synchrotron radiation facility in Lund, Sweden. Photoemission spectra were obtained using an angle-resolved photoelectron spectrometer and linearly polarized synchrotron radiation light using three different photon energies ($h\nu=17$, 21.2, and 35 eV). The total experimental energy resolutions were ~ 70 meV at $h\nu=78$ eV, ~ 50 meV at $h\nu=21.2$ eV, and ~ 118 meV at $h\nu=35$ eV, and the angular resolution was $\pm 2^\circ$. A Si(111) sample (*n*-type) with a 1.1° miscut towards the $[\bar{1}1\bar{2}]$ direction was cleaned by direct resistive heating following the procedure described in Refs. 25 and 26. After the annealing, a sharp (7×7) LEED pattern was observed, and neither the valence-band spectra nor the Si $2p$ core-level spectra showed any indication of contamination. The Eu-induced reconstruction was prepared by depositing Eu onto a clean Si(111)- (7×7) surface at a substrate temperature of ~ 1100 K, and the Ca-induced reconstructed surface was prepared by depositing Ca onto a clean surface at a substrate temperature of ~ 1000 K. The base pressure was below 8×10^{-11} Torr during the measurements and below 5×10^{-10} Torr during the Eu and Ca evaporations.

III. RESULTS AND DISCUSSION

Figure 2(a) shows the LEED pattern of a single domain so-called Eu/Si(111)- (5×1) surface obtained at 100 K with a primary electron energy of 85 eV. The observation of $5 \times$

FIG. 2. LEED patterns of (a) the so-called Eu/Si(111)- (5×1) surface and (b) the so-called Ca/Si(111)- (5×1) surface obtained at 100 K. Primary electron energies are (a) 85 eV and (b) 86 eV. Sharp $5 \times$ spots are observed in the $[11\bar{2}]$ direction together with dim streaks in (a). In (b), strong $5 \times$ spots are observed in the $[11\bar{2}]$ direction and weak $5 \times$ spots are observed in the $[1\bar{2}1]$ and $[2\bar{1}1]$ directions together with weak dim streaks.

spots in the $[11\bar{2}]$ direction and the absence of $5 \times$ spots in the $[1\bar{2}1]$ and $[2\bar{1}1]$ directions indicate that a very-high-quality single-domain surface was obtained. Together with these sharp $5 \times$ spots, dim streaks are observed in Fig. 2(a). The positions of these dim streaks indicate that this surface has actually a (5×4) periodicity.

The LEED pattern of the so-called Ca/Si(111)- (5×1) surface was obtained at 100 K with a primary electron energy of 86 eV [Fig. 2(b)]. In this pattern, clear $5 \times$ spots are observed in the $[11\bar{2}]$ direction, while only faint $5 \times$ spots are observed in the $[1\bar{2}1]$ and $[2\bar{1}1]$ directions. This result indicates that a predominantly single-domain Ca-induced surface was obtained. Dim streaks that originate from the $4 \times$ periodicity of this Ca-induced reconstruction are observed in Fig. 2(b) though their intensities are weaker than the intensities of the $4 \times$ streaks observed on the Eu-induced reconstruction. The observation of (5×4) periodicity on both the Eu- and Ca-induced reconstructions suggests that the other intermediate phases are not formed by a simple combination of (3×2) and (2×1) phases and may have $4 \times$ periodicities along the chains as well. Further, these LEED results show that single-domain surfaces with a quite high quality were obtained, and thus the ARPES spectra of both the Eu- and Ca-induced surfaces can be analyzed without the ambiguity that would have otherwise been caused by contributions from the two other domains.

The ARPES spectra of the Eu/Si(111)- (5×4) surface are shown in Fig. 3, together with the surface Brillouin zones (SBZ's) of the Si(111)- (1×1) , (2×1) , (3×1) , (3×2) , and (5×4) surfaces [Fig. 3(f)]. Figures 3(a)–3(c) are the spectra measured along the $[\bar{1}10]$ direction—i.e., the direction parallel to the Eu chains—using $h\nu=17$ eV, 21.2 eV, and 35 eV, respectively. The spectra in (d) and (e) are measured along the $[11\bar{2}]$ direction—i.e., the direction perpendicular to the Eu chains—using $h\nu=17$ eV and 21.2 eV. All spectra are normalized to the background intensities at the Fermi-level position (E_F , which is indicated by a dashed line in each figure), which was determined by measuring the metallic Fermi edge of a Ta foil fixed on the sample holder. The spectra in the $[11\bar{2}]$ direction were measured using the in-plane polarization geometry (the electric field of photons is

FIG. 3. (Color online) ARPES spectra of the Eu/Si(111)-(5 $\times$ 4) surface measured along the $[\bar{1}10]$ direction using (a) $h\nu=17$ eV, (b) 21.2 eV, and (c) 35 eV and along the $[11\bar{2}]$ direction using (d) $h\nu=17$ eV and (e) 21.2 eV. The spectra in the $[\bar{1}10]$ direction were obtained using the out-of-plane polarization geometry, and the spectra in the $[11\bar{2}]$ direction were measured using the in-plane polarization geometry. The states labeled S_1 – S_5 , Σ_1 , and Σ_2 are surface states. (f) SBZ's of the Si(111)-(1 $\times$ 1), (2 $\times$ 1), (3 $\times$ 1), (3 $\times$ 2), and (5 $\times$ 4) surfaces. $\bar{K}$ and $\bar{M}$ are the symmetry points of the (1 $\times$ 1) SBZ, $\bar{J}$ and $\bar{J}'$ are those of the (2 $\times$ 1) SBZ, and $\bar{A}$ and $\bar{C}$ are those of the (3 $\times$ 1) SBZ.

parallel to the photoelectron emission plane), and the spectra measured along the $[\bar{1}10]$ direction were obtained using the out-of-plane polarization geometry (the photoelectron emission plane is perpendicular to that of the in-plane polarization geometry). The symbols $\bar{M}$ and $\bar{K}$ are the symmetry points of the (1 $\times$ 1) SBZ, the symbols $\bar{J}$ and $\bar{J}'$ are those of the (2 $\times$ 1) SBZ, and the symbols $\bar{A}$ and $\bar{C}$ are the symmetry points of the (3 $\times$ 1) SBZ. In the $[\bar{1}10]$ direction, the angle-resolved photoelectron spectra were recorded at every 1° from emission angles (θ_e) of 0° to 70° using $h\nu=17$ eV and 21.2 eV and from $\theta_e=0^\circ$ to 40° using $h\nu=35$ eV. In the $[11\bar{2}]$ direction, the spectra were recorded at every 1° from $\theta_e=0^\circ$ to 20° . The states labeled S_1 – S_5 , Σ_1 , and Σ_2 are in the

band gap; i.e., they are surface states. Further, compared to the spectra observed using other photon energies, the cross section of the S_1 and S_4 states are larger using $h\nu=17$ eV in both directions. These state do not cross the Fermi level, and no other state is observed at lower binding energies. We therefore conclude that the electronic structure of the Eu/Si(111)-(5 $\times$ 4) surface is semiconducting.

The band dispersions of the seven surface states S_1 – S_5 , Σ_1 , and Σ_2 are compared with the surface bands obtained experimentally for the Eu-induced Si(111)-(3 $\times$ 2) and (2 $\times$ 1) surfaces¹⁸ in Fig. 4 to discuss the surface electronic structure of the Eu/Si(111)-(5 $\times$ 4) surface. The open circles in Fig. 4 represent the peak and shoulder positions of the ARPES spectra obtained using $h\nu=17$ eV, the solid ones are those obtained using $h\nu=21.2$ eV, and the open triangles

FIG. 4. (Color online) Surface state dispersions of the Eu- and Ca-induced Si(111)-(5 $\times$ 4) surfaces along the $[11\bar{2}]$ and $[\bar{1}10]$ directions. The open circles represent the peak and shoulder positions of the ARPES spectra obtained using $h\nu=17$ eV, the solid ones are those obtained using $h\nu=21.2$ eV, and the open triangles are those obtained using $h\nu=35$ eV. Solid lines are the surface states of the Eu- and Ca-induced Si(111)-(3 $\times$ 2) surfaces derived from Refs. 18 and 22. Dashed lines are the surface states of the Eu- and Ca-induced Si(111)-(2 $\times$ 1) surfaces derived from Refs. 18 and 24. Gray vertical lines are the symmetry points of the (5 $\times$ 4) unit cell in the $[11\bar{2}]$ direction. The gray arrows indicate the $\bar{\Gamma}$ point of the second (5 $\times$ 4) SBZ.

represent the peak and shoulder positions obtained using $h\nu=35$ eV. The solid lines, labeled $S_{1\text{Eu}(3\times 2)}-S_{3\text{Eu}(3\times 2)}$, $\Sigma_{1\text{Eu}(3\times 2)}$, and $\Sigma_{2\text{Eu}(3\times 2)}$, are the surface states of the Eu-induced Si(111)-(3 $\times$ 2) surface and the dashed lines, labeled $SS_{1\text{Eu}(2\times 1)}$ and $SS_{2\text{Eu}(2\times 1)}$, are the surface states of the Eu-induced Si(111)-(2 $\times$ 1) surface derived from Ref. 18. Of the gray vertical lines, which indicate the symmetry points of the (5 $\times$ 4) unit cell in the $[11\bar{2}]$ direction, the line indicated by a gray arrow corresponds to the $\bar{\Gamma}$ point of the second (5 $\times$ 4) SBZ.

As shown in Fig. 4, the dispersion features and relative binding energies of S_1-S_3 agree well with those of $S_{3\text{Eu}(3\times 2)}$ and $S_{3\text{Eu}(3\times 2)}$ in the $[\bar{1}10]$ direction. The dispersion features and relative binding energies of Σ_1 and Σ_2 show

good agreement with those of the $\Sigma_{1\text{Eu}(3\times 2)}$ and $\Sigma_{2\text{Eu}(3\times 2)}$ states as well. Further, the photoemission cross sections of S_1-S_3 , Σ_1 , and Σ_2 coincide with those of $S_{1\text{Eu}(3\times 2)}-S_{3\text{Eu}(3\times 2)}$, $\Sigma_{1\text{Eu}(3\times 2)}$, and $\Sigma_{2\text{Eu}(3\times 2)}$; i.e., S_1 and $S_{1\text{Eu}(3\times 2)}$ have quite small photoemission cross sections in the measurement using the in-plane geometry along the $[\bar{1}10]$ direction, while the cross sections of the other states do not have such dependence. Based on the good agreements in the dispersion features, the relative binding energies, and the photoemission cross section, we conclude that the origins of S_1-S_3 , Σ_1 , and Σ_2 are orbitals of Si atoms that form a HCC structure likewise the surface states of the Eu/Si(111)-(3 $\times$ 2) surface.

The dispersion features of S_4 and S_5 are similar to those of the $SS_{1\text{Eu}(2\times 1)}$ and $SS_{2\text{Eu}(2\times 1)}$ along the $[\bar{1}10]$ direction. That is, both the S_4 state and $SS_{1\text{Eu}(2\times 1)}$ state disperse first upward and then downward, and S_5 and $SS_{2\text{Eu}(2\times 1)}$ hardly disperse along this direction. This similarity indicates that the origins of the S_4 and S_5 states are orbitals of Si atoms that form a Seiwatz structure likewise the $SS_{1\text{Eu}(2\times 1)}$ and $SS_{2\text{Eu}(2\times 1)}$ states of the Eu/Si(111)-(2 $\times$ 1) surface. Here we note that a difference in magnitude of buckling might cause different dispersion widths and the difference in dispersion width of the S_4 state and that of the $SS_{1\text{Eu}(2\times 1)}$ state might indicate that the bucklings for Si atoms forming Seiwatz chains are different in these two reconstructions. The good agreement between S_1-S_3 , Σ_1 , and Σ_2 and the surface states of Eu/Si(111)-(3 $\times$ 2) and the similarity between S_4 and S_5 and the surface states of Eu/Si(111)-(2 $\times$ 1) show that the atomic structure of the Eu/Si(111)-(5 $\times$ 4) surface consists of both the HCC and the Seiwatz structures.

In contrast to the $[\bar{1}10]$ direction, the SBZ's are small and some surface states are overlapped in the $[11\bar{2}]$ direction. However, although these two cases make it difficult to discuss the dispersion of the surface states in this direction, it is clearly shown in Fig. 4 that the dispersion widths of the surface states are small and that the S_4 state follows a 5 $\times$ periodicity and not the 3 $\times$ periodicity of the (3 $\times$ 1) unit cell or the 2 $\times$ periodicity of the (2 $\times$ 1) unit cell [S_4 disperses upward from the $\bar{\Gamma}$ point to the symmetry point of the (5 $\times$ 4) unit cell, which is indicated by a gray vertical line, and then downward to the $\bar{\Gamma}$ point of the second (5 $\times$ 4) SBZ]. Based on the observation of surface states that follow the (3 $\times$ 1) periodicity and those which follow the (2 $\times$ 1) periodicity in the $[\bar{1}10]$ direction and the observation of a surface state that follows a 4 $\times$ periodicity in the $[11\bar{2}]$ direction but with a small dispersion, we conclude that the interaction between the outermost Si atoms of a HCC chain and those of the nearest-neighboring Seiwatz chain is quite small and that the electronic structure of one chain hardly affects the electronic structure of its neighboring chain.

The presence of a HCC and a Seiwatz structure and the number of surface states suggest that there is one Eu atom per (3 $\times$ 2) unit and one Eu atom per (2 $\times$ 1) unit of the Eu/Si(111)-(5 $\times$ 4) surface. Since a (5 $\times$ 4) unit cell is basically formed by two (3 $\times$ 2) units and four (2 $\times$ 1) units,

these numbers correspond to the adsorption of six Eu atoms per (5×4) unit cell and thus to a 0.3-ML coverage for a Eu/Si(111)- (5×4) surface. This coverage supports the model shown in Fig. 1(c) as the atomic structure of the Eu/Si(111)- (5×4) surface. Moreover, the LEED pattern shown in Fig. 2(b) and the agreement between the surface states of the Ca/Si(111)- (5×4) surface and those of the Ca induced (3×2) and (2×1) surfaces (Fig. 4) suggest that the atomic structure of the Ca/Si(111)- (5×4) surface is the same as that of Eu/Si(111)- (5×4) . We therefore propose that the atomic structure of a Si(111)- (5×4) reconstruction, which is formed by a combination of divalent-atom-induced (3×2) and (2×1) reconstructions, is universal.

IV. CONCLUSION

In conclusion, we have studied the electronic structures of the so-called Eu- and Ca-induced Si(111)- (5×1) surfaces along the $[\bar{1}10]$ and $[11\bar{2}]$ directions using ARPES and LEED. The LEED patterns of both the Eu- and Ca-induced reconstructions show $\text{dim } \times 4$ streaks together with sharp $5 \times$ spots. These LEED patterns indicate that the periodicities of both the so-called Eu- and Ca-induced (5×1) surfaces are actually (5×4) . In the ARPES measurements performed along the $[\bar{1}10]$ and $[11\bar{2}]$ directions, seven surface states were observed on each (5×4) reconstruction. Among these surface states, the dispersions of five of them show good agreement with those of the Eu- and Ca-induced (3×2) HCC surfaces along the $[\bar{1}10]$ direction. This result indicates that these five surface states observed in the band gap originate from the orbitals of Si atoms that form a HCC structure.

The dispersions of the two other surface states agree well with those of the Eu- and Ca-induced (2×1) Seiwatz surfaces in the $[\bar{1}10]$ direction. Based on the observation of surface states that follow a (3×1) periodicity and those which follow a (2×1) periodicity in the $[\bar{1}10]$ direction, we conclude that the interaction between the outermost Si atoms of a HCC chain and those of the nearest-neighboring Seiwatz chain of the (5×4) reconstruction is quite small. This conclusion also indicates that the electronic structure of one chain hardly affects the electronic structure of its neighboring chain and is supported by the small dispersions of the surface states along the $[11\bar{2}]$ direction. Further, the presence of a HCC and a Seiwatz structure and the number of surface states suggest that the adsorbate coverage of a (5×4) reconstruction is 0.3 ML. This coverage supports the model shown in Fig. 1(c) as the atomic structure of the Eu/Si(111)- (5×4) surface. Moreover, taking the good agreement of the surface states of Ca/Si(111)- (5×4) with that of the Ca/Si(111)- (3×2) and (2×1) surfaces into account, we propose that the atomic structure of the Si(111)- (5×4) surface, which is formed by a combination of divalent-atom-induced (3×2) and (2×1) reconstructions, is universal.

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