Experimental and theoretical determination of sigma bands on (" $2\sqrt{3} \times 2\sqrt{3}$ ") silicene grown on Ag(111)

Weimin Wang, Weine Olovsson and Roger Uhrberg

Linköping University Post Print

N.B.: When citing this work, cite the original article.

Original Publication:

Weimin Wang, Weine Olovsson and Roger Uhrberg, Experimental and theoretical determination of sigma bands on (" $2\sqrt{3} \times 2\sqrt{3}$ ") silicene grown on Ag(111), 2015, Physical Review B. Condensed Matter and Materials Physics, (92), 20, 205427.

<http://dx.doi.org/10.1103/PhysRevB.92.205427>

Copyright: American Physical Society

<http://www.aps.org/>

Postprint available at: Linköping University Electronic Press

<http://urn.kb.se/resolve?urn=urn:nbn:se:liu:diva-123801>

Experimental and theoretical determination of σ bands on (" $2\sqrt{3} \times 2\sqrt{3}$ ") silicene grown on Ag(111)

W. Wang,^{*} W. Olovsson, and R. I. G. Uhrberg*Department of Physics, Chemistry, and Biology, Linköping University, S-581 83 Linköping, Sweden*

(Received 26 May 2015; revised manuscript received 9 September 2015; published 23 November 2015)

Silicene, the two-dimensional (2D) allotrope of silicon, has very recently attracted a lot of attention. It has a structure that is similar to graphene and it is theoretically predicted to show the same kind of electronic properties which have put graphene into the focus of large research and development projects worldwide. In particular, a 2D structure made from Si is of high interest because of the application potential in Si-based electronic devices. However, so far there is not much known about the silicene band structure from experimental studies. A comprehensive study is here presented of the atomic and electronic structure of the silicene phase on Ag(111) denoted as $(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ in the literature. Low energy electron diffraction (LEED) shows an unconventional rotated (" $2\sqrt{3} \times 2\sqrt{3}$ ") pattern with a complicated set of split diffraction spots. Scanning tunneling microscopy (STM) results reveal a Ag(111) surface that is homogeneously covered by the (" $2\sqrt{3} \times 2\sqrt{3}$ ") silicene which exhibits an additional quasiperiodic long-range ordered superstructure. The complex structure, revealed by STM, has been investigated in detail and we present a consistent picture of the silicene structure based on both STM and LEED. The homogeneous coverage by the (" $2\sqrt{3} \times 2\sqrt{3}$ ") silicene facilitated an angle-resolved photoelectron spectroscopy study which reveals a silicene band structure of unprecedented detail. Here we report four silicene bands which are compared to calculated dispersions based on a relaxed $(2\sqrt{3} \times 2\sqrt{3})$ model. We find good qualitative agreement between the experimentally observed bands and calculated silicene bands of σ character.

DOI: [10.1103/PhysRevB.92.205427](https://doi.org/10.1103/PhysRevB.92.205427)

PACS number(s): 68.37.Ef, 71.15.Mb, 73.20.-r, 79.60.-i

I. INTRODUCTION

The two-dimensional (2D) honeycomb structure of carbon (C) atoms, i.e., graphene, has attracted great attention because of its outstanding electronic properties due to sp^2 hybridization. The electronic structure shows a steep linear dispersion near the Fermi energy (E_F) forming a so called Dirac cone at specific points in k space. This part of the electronic structure resembles what is expected for massless fermions. Inspired by the exceptional properties of graphene, a lot of effort now goes into the search for other 2D sp^2 hybridized materials. As the nearest neighbor to C in group IV of the periodic table, Si is one of the most promising candidates. A 2D graphenelike structure made from Si is of high interest because of the application potential in Si-based electronic devices. Such a 2D honeycomb structure of Si has been theoretically studied since the 1980s [1] and was named silicene in 2007 by Guzmán-Verri and Yan Voom [2]. Unlike graphene, free-standing silicene is calculated to be stable only in a slightly buckled geometry. The reason is that the s and p valence orbitals of Si are prone to form an sp^3 hybridization which leads to a tetrahedral, three-dimensional, configuration rather than the planar sp^2 hybridization of a two-dimensional structure. For a small buckling, calculations do predict an electronic structure with Dirac cones [3] at the $\bar{K}$ points of the silicene Brillouin zone. However, there is no convincing experimental evidence of the presence of Dirac fermions in monolayer silicene. An electron band with a linear dispersion was found by Vogt *et al.* [4] near a $\bar{K}$ point of silicene grown on Ag(111) and it was discussed as emission from a modified Dirac cone with a gap between π and π^* bands. However, this idea has been questioned by several authors [5–8], and the linear band is instead suggested to orig-

inate from 2D surface or interface states that appear when the silicene layer has formed. As suggested by Cahangirov *et al.* [9], the linear dispersion found in Ref. [4] can arise from new states due to strong hybridization between Si and Ag orbitals.

Silicene is mostly synthesized on Ag(111) substrates because of a low Si-Ag intermixing and a “matching” 3 to 4 ratio between the lattice constant of silicene and that of the Ag(111) surface [4,5,10–15]. Several modifications of silicene with different mixes of sp^2 and sp^3 hybridization and different orientations with respect to the Ag surface lattice have been observed. Depending on the orientation, the silicene layer shows specific reconstructions involving buckling and thus a deviation from an “ideal” sp^2 hybridization. The most commonly observed periodicities are $(4 \times 4)\alpha = 0^\circ$, $(\sqrt{13} \times \sqrt{13})$ type I $\alpha = \pm 27^\circ$, $(\sqrt{13} \times \sqrt{13})$ type II $\alpha = \pm 5.2^\circ$, $(\sqrt{7} \times \sqrt{7})\alpha = \pm 19.1^\circ$, and $(2\sqrt{3} \times 2\sqrt{3})\alpha = \pm 10.9^\circ$, where α is the nominal angle between the silicene and the Ag(111) surface unit cells derived from atomic models [10]. The formation of the different reconstructions depends on substrate temperature, deposition rate, and coverage. They all form within a small region of parameter space and samples generally exhibit more than one silicene reconstruction [16–19]. Angle-resolved photoelectron spectroscopy (ARPES) experiments have focused on the (4×4) phase since it is aligned with the (1×1) unit cell of Ag(111) [4,5,7]. However, to the best of our knowledge, there is no clear evidence, neither from real space, nor from reciprocal space studies of a (4×4) silicene surface without the coexistence of other phases. The difficulty to grow large domains of a single silicene phase has so far hampered investigations of the electronic structure of individual silicene phases by ARPES.

In this paper we present a comprehensive study of a silicene layer that is known in the literature as $(2\sqrt{3} \times 2\sqrt{3})R30^\circ$ silicene due to its approximate periodicity with respect to the Ag(111) surface lattice, as observed by LEED [10,20].

^{*}Corresponding author: weiwa49@ifm.liu.se

This structure is formed by silicene layers that are rotated by either $+10.9^\circ$ or -10.9° with respect to Ag(111). The presence of both orientations of the silicene layer was verified experimentally in our study (see the Supplemental Material [21]). Structural information was obtained by STM and LEED, and the electronic structure was determined by ARPES. By comparing to the results of electronic band structure calculations, based on a $(2\sqrt{3} \times 2\sqrt{3})$ model of the silicene layer, we could unambiguously identify electronic bands of silicene (σ bands) in the ARPES data.

II. EXPERIMENTAL AND THEORETICAL DETAILS

Samples were prepared *in situ* in ultrahigh vacuum (UHV) systems with base pressures in the 10^{-11} Torr range. The Ag(111) crystal was cleaned by repeated cycles of sputtering by Ar^+ ions (1 keV) and annealing at approximately 400°C until a sharp (1×1) LEED pattern was obtained. Atomically resolved STM images, as well as the presence of a sharp Shockley surface state in ARPES, confirmed a high quality of the Ag(111) surface. About 1 monolayer (ML) of Si was deposited from a heated Si wafer piece at a rate of about 0.03 ML/min. The Ag(111) substrate was kept between 280 and 300°C during deposition. STM images were recorded at room temperature using an Omicron variable temperature STM at Linköping University. LEED and ARPES data were obtained at the MAX-lab synchrotron radiation facility using the beamline I4 end station. Data were acquired at room temperature by a Phoibos 100 analyzer from Specs with a two-dimensional detector. The energy and angular resolutions were 50 meV and 0.3° , respectively. Angle integrated Si $2p$ core-level spectra were measured at room temperature. DFT calculations were performed using the WIEN2k package [22] employing the generalized gradient approximation (GGA) for exchange and correlation. The periodic slab model includes nine layers of Ag atoms, one layer of silicene, and a vacuum layer of 15 Å. The positions of the atoms were fully relaxed using the projector augmented wave method (PAW) Vienna *ab initio* simulation package (VASP) code [23] except for the bottom six Ag layers. The energy cutoff of the plane-wave basis set was 250 eV, and the k -point mesh was $4 \times 4 \times 1$.

III. RESULTS AND DISCUSSION

Figure 1(a) is a large-scale $\sim 90 \times 90 \text{ nm}^2$ topographic filled-state image showing an overview of the $(2\sqrt{3} \times 2\sqrt{3})$ silicene layer. STM images obtained at different places on the surface verified the homogeneous coverage of the Ag(111) surface by $(2\sqrt{3} \times 2\sqrt{3})$ silicene. It should be noted that there are two differently oriented domains, differing by a rotation angle of $\sim 8^\circ$. The 8° difference implies that there are two variants of the $(2\sqrt{3} \times 2\sqrt{3})$ reconstruction, one rotated by 26° and the other by 34° with respect to Ag(111). From here on we use the short notation “ $2\sqrt{3}$ ”, where the quotation marks indicate that the real silicene layer deviates from an ideal $(2\sqrt{3} \times 2\sqrt{3})$ periodicity.

A close-up image of a $42 \times 42 \text{ nm}^2$ area [Fig. 1(b)] shows a clear quasiperiodic long-range order in the form of brighter parts arranged in a hexagonal fashion. The orientation of this hexagonal structure is rotated by $\sim 9^\circ$ clockwise relative to

the Ag(111) surface, as illustrated by the white and green dashed lines. This phase has been reported by Feng *et al.* [24], which they described as a $(2\sqrt{3} \times 2\sqrt{3})$ reconstruction with an obvious moiré pattern, due to the quasiperiodic brighter parts observed by STM, see Fig. 1(b). This long-range pattern is actually not a consequence of moiré interference but a consequence of a mismatch of about 2% in the lattice constants of silicene and Ag(111) [10,25], resulting in ideal and distorted honeycomb rings in bright and dark parts, respectively [26]. Figure 1(c) is a detailed STM image ($3.8 \times 3.8 \text{ nm}^2$) of Fig. 1(b). A fast Fourier transform (FFT) of the STM image in Fig. 1(a) results in two sets of Fourier components, each set forming a hexagonal pattern, see Fig. 1(d). The split into two hexagonal sets of components is just a consequence of the presence of two “ $2\sqrt{3}$ ” domains differing by $\sim 8^\circ$. This FFT map can be directly compared to the LEED pattern in Fig. 1(e) to gain important information about this complex surface structure. The strong Fourier components within the green and blue circles of Fig. 1(d) correspond to the two brightest spots within the circles of the LEED pattern. Thus, these LEED spots originate from two $\pm 4^\circ$ rotated “ $2\sqrt{3}$ ” domains explaining the major deviation from an ideal $(2\sqrt{3} \times 2\sqrt{3})$ diffraction pattern. Figure 1(e) also shows two weaker spots within the circles which originate from the moiré-like hexagonal structure. One set of spots is rotated by $+25^\circ$ with respect to the -4° domain of the “ $2\sqrt{3}$ ” phase and the other set of spots is rotated by -25° with respect to the $+4^\circ$ domain. The periodicity of the moiré-like pattern is approximately 3.7 times larger than that of the “ $2\sqrt{3}$ ” structure. These assignments have been derived from a detailed FFT analysis of single domain STM images and experimental LEED patterns, see the Supplemental Material [21].

The homogeneous coverage of the Ag(111) surface by the “ $2\sqrt{3}$ ” silicene layer opens up for an unambiguous ARPES study of the electronic structure. Initially, the clean Ag(111) substrate was studied in order to establish its contribution to the ARPES data. Figure 2(a) (left) shows bulk emission (B) and some emission intensity with a symmetric dispersion around the $\bar{M}$ point of the Ag (1×1) surface Brillouin zone (SBZ) using a photon energy of 19 eV. After the formation of the “ $2\sqrt{3}$ ” silicene, there is no change in the bulk emission, while there is a clear change of the emission symmetric around the $\bar{M}$ point which now exhibits an energy maximum well below E_F at 0.4 eV (middle). A comparison of spectra obtained at two different photon energies shows that the bulk emission B crosses E_F at different k values [$1.11 \text{ \AA}^{-1}$ when the photon energy is 19 eV (middle) and $1.16 \text{ \AA}^{-1}$ in the 26 eV data (right)]. The band S_{Ag} does not change the shape and the energy maximum remains at the same position, which confirms its 2D character. In a recent paper [8], a calculation was presented indicating that the emission here labeled S_{Ag} is actually due to hybridization between Si and Ag orbitals. A similar band is also observed by ARPES for the Ag_2Sn surface alloy on Ag(111) [27], which supports the interpretation that S_{Ag} is not specifically related to the silicene layer, but instead stems from a modification of the electronic structure of the outermost Ag layer.

The experimental 2D band structure of “ $2\sqrt{3}$ ” silicene is presented along the $\bar{\Gamma}\bar{M}$ and $\bar{\Gamma}\bar{K}$ lines of the Ag (1×1) SBZ

FIG. 1. (Color online) (a) Large scale filled-state STM image, $\sim 90 \times 90 \text{ nm}^2$, of “ $2\sqrt{3}$ ” silicene on two terraces of the Ag(111) substrate. The blue lines indicate the orientations of the two domains which differ by $\sim 8^\circ$. The image has been compensated for thermal drift. (b) Atomically resolved filled state STM image of a $42 \times 42 \text{ nm}^2$ area showing honeycomb rings and a long-range, quasiperiodic, hexagonal pattern. The white dashed line is a guide to the eye of the orientation of this quasiperiodic pattern. The inset is a $1.8 \times 1.8 \text{ nm}^2$ filled state STM image of the clean Ag(111) surface. The green dashed line indicates the orientation of the Ag(111) surface unit cell. The angle between the green and white dashed lines is about 9° . (c) Atomically resolved STM image of a $3.8 \times 3.8 \text{ nm}^2$ area showing the well-ordered honeycomb rings within the brighter parts of the STM image in (b). The green diamond depicts a “ $2\sqrt{3}$ ” unit cell. All STM images were recorded at room temperature in constant current mode with a tunneling current of 200 pA and a sample bias of -1.2 V in (a), -1.5 V in (b) and (c), and -0.1 V for the clean Ag(111) [inset in (b)]. (d) Fast Fourier transform (FFT) of the STM image in (a) showing two sets of Fourier components forming two hexagons with a rotational difference of $\sim 8^\circ$ [$\pm 4^\circ$ relative to the Ag(111) lattice] as indicated by the blue lines. (e) LEED pattern obtained from “ $2\sqrt{3}$ ” silicene at an electron energy of 40 eV. The two bright spots inside each circle are due to diffraction from the two “ $2\sqrt{3}$ ” domains. These diffraction spots match the split Fourier components encircled in (d). The two weaker LEED spots inside the circles originate from the long-range, quasiperiodic, structure. A detailed analysis of STM, FFT, and LEED results is available in the Supplemental Material [21].

in Figs. 2(b) and 2(c), respectively. Two downward dispersing bands, S_1 and S_2 , are clearly observed in the $\bar{\Gamma}\bar{M}$ direction, while a third band, S_3 , with an upward dispersion starting from $\bar{\Gamma}$ can be better seen along $\bar{\Gamma}\bar{K}$, where the upward dispersion is evident changing to a downward dispersion further out in the SBZ. Along $\bar{\Gamma}\bar{K}$, S_2 cannot be detected while S_1 appears with an intensity and dispersion similar to the results along $\bar{\Gamma}\bar{M}$. The dispersive feature BU in Fig. 2(b) is due to umklapp scattered Ag bulk emission appearing in the outer parts of the Ag (1×1) SBZ, see feature B in Fig. 2(a). In addition to these three dispersive bands, there is electron emission S_4 close to E_F at normal emission. On samples which are only partly

covered by silicene, we observed the Shockley surface state of clean Ag(111) near normal emission. However, it is clearly separated from the S_4 emission and we therefore also assign S_4 to the silicene layer. Constant energy contours are shown in Fig. 2(d) at binding energies of 0.5 and 1.0 eV. The contour of S_1 is a hexagon with the same orientation as the hexagon formed by the Ag(111) (1×1) SBZ, while the S_2 and S_3 bands give rise to a sixfold pattern of lobes, pointing along $\bar{\Gamma}\bar{M}$ and $\bar{\Gamma}\bar{K}$, respectively.

In order to further analyze the S_1 – S_4 bands, we performed first-principles density functional theory (DFT) calculations using the full-potential linearized augmented plane wave

FIG. 2. (Color online) (a) Valence band dispersions of (i) clean Ag(111) using a photon energy of 19 eV (left) and (ii) “2√3” silicene on Ag(111) using 19 eV (middle) and 26 eV photons (right). The data were obtained around $\bar{M}$ in the $\bar{\Gamma}\bar{M}$ direction of the 1×1 SBZ of Ag(111). The S_{Ag} band appears after the formation of silicene. The dispersion does not change with photon energy, compare middle and right panels, but the bulk related emission B does. This verifies the 2D character of S_{Ag} . (b) and (c) Valence band dispersions of “2√3” silicene on Ag(111) obtained using a photon energy of 19 eV. The bands were mapped along the $\bar{\Gamma}\bar{M}$ and $\bar{\Gamma}\bar{K}$ lines of the Ag (1×1) SBZ, respectively. Three dispersive silicene bands, S_1 , S_2 , and S_3 , are observed around $\bar{\Gamma}$. A dispersive feature (BU) due to umklapp scattering of the bulk Ag sp emission (B) is also observed and the position fits with scattering by a $2\sqrt{3}$ reciprocal lattice vector. A fourth silicene related feature S_4 is observed as an almost flat band at E_F close to $\bar{\Gamma}$. (d) Constant energy contours obtained at 0.5 and 1.0 eV below E_F . The S_1 , S_2 , and S_3 bands seem to be degenerate at $\bar{\Gamma}$ at 0.5 eV leading to a spot at the center of the 0.5 eV contour. The six lobes stretching out from $\bar{\Gamma}$ originate from the S_3 band. In the 1.0 eV contour plot, the S_1 band gives rise to the inner hexagon, six lobes are generated by emission from S_2 , while the S_3 emission results in the outer weak hexagon. The contour due to the BU emission is also indicated.

(FPLAPW) WIEN2k simulation package [22]. We model the experimental “2√3” surface by the structure shown in Fig. 3(a). This model should quite accurately represent the structure of the brighter parts in the STM images (see Fig. 1).

The so called $(2\sqrt{3} \times 2\sqrt{3})$ type II model in Ref. [18] and $(\sqrt{7} \times \sqrt{7})$ silicene on $(2\sqrt{3} \times 2\sqrt{3})$ Ag(111) model in Ref. [25] presented similar atomic structures as our model by showing the two topmost Si atoms in the $2\sqrt{3}$ unit cell

FIG. 3. (Color online) (a) Top and side views of a unit cell of the relaxed model of $(2\sqrt{3} \times 2\sqrt{3})$ silicene on Ag(111). Only the Si atoms (blue) and the first layer Ag atoms (gray) are included. The Si atoms on top of the silver atoms give rise to the honeycomb pattern observed by STM. The slab used in the band structure calculations is composed of one buckled Si layer, three relaxed, slightly buckled, Ag layers, and six bulk Ag layers. (b) Calculated band structure for $(2\sqrt{3} \times 2\sqrt{3})$ silicene on Ag(111) where only contributions from the Si atoms are plotted. The red and green circles represent the $p_x p_y$ and p_z states of the Si atoms, respectively. The bigger the circles are, the larger the contribution is from the Si atoms to the wave functions at the various k points. The band structure is plotted in the $2\sqrt{3} \times 2\sqrt{3}$ SBZ. Note that the $\bar{\Gamma}\bar{K}_{2/3}$ and $\bar{\Gamma}\bar{M}_{2/3}$ lines correspond to $\bar{\Gamma}\bar{M}_{\text{Ag}}$ and $\bar{\Gamma}\bar{K}_{\text{Ag}}$ in Figs. 2(b) and 2(c), respectively. Several of the calculated bands, labeled Σ_1 – Σ_4 , show qualitative agreement with the experimental bands S_1 – S_4 in Figs. 2(b) and 2(c). The calculated bands below a binding energy of about 2 eV were not observed experimentally.

sitting on top of Ag atoms. The height difference between highest and lowest Si atoms is 1.0 [18] and 1.19 Å [25], respectively, which agree nicely with our value of 1.19 Å. The silicon atoms that are located on top of silver atoms form a honeycomb pattern. Between the bright parts, the honeycombs show some distortions which are not included in the model.

Calculated bands along the $\bar{\Gamma}\bar{K}_{2/3}$ and $\bar{\Gamma}\bar{M}_{2/3}$ symmetry lines of the $2\sqrt{3} \times 2\sqrt{3}$ SBZ are shown in Fig. 3(b). Red and green circles represent $p_x p_y(\sigma)$ and $p_z(\pi)$ bands of the silicene layer, respectively. The bigger the circles are, the larger the contribution is from the Si atoms to the wave functions at the various k points. Comparing the experimental and calculated bands, one finds striking qualitative similarities in the binding energy range from 0 to ~ 1.5 eV. (i) The steep Σ_1 band in the calculation reproduces the dispersion of the S_1 band. The Σ_1 energy at $\bar{\Gamma}$ is 0.9 eV, while the experimental value is 0.5 eV. (ii) The upward dispersive “bands” labeled $\Sigma_{2,3}$ and Σ_3 close to $\bar{\Gamma}$ agree qualitatively with the experimental S_3 band. It should be noted that the particular shape shown by $\Sigma_{2,3}$ and Σ_3 has not been reported in any published band structure calculation of silicene on Ag(111). Thus, the initial upward dispersion observed experimentally, which is reproduced theoretically, appears to be a consequence of the particular buckling of the silicene layer in this case. Between Σ_1 and Σ_3 there are some dispersive bands, indicated by Σ_2 , which fit qualitatively with the experimental S_2 band. Confined to a small region around $\bar{\Gamma}$, there are silicene states Σ_4 , at 0.5 eV, i.e., 0.4 eV higher than the Σ_1 maximum. These theoretical findings fit nicely with the characteristics of S_4 which we therefore assign to Si $p_x p_y$ states. To summarize, we find that the model calculation reproduces the shapes of the S_1 , S_3 , and S_4 bands quite well and, to a somewhat lesser extent, also the S_2 band. Quantitatively, the calculation locates the bands ~ 0.4 eV lower

FIG. 4. (Color online) Si 2*p* core-level spectrum obtained at a photon energy of 135 eV in normal emission. The dots are the experimental data and the fitting curve is the sum of two spin-orbit split components (C1 and C2) and a Shirley background. The spectrum is dominated by C1. The C2 component, which has $\sim 15\%$ of the total intensity, is shifted from C1 by -0.46 eV. Fitting parameters: Spin-orbit split: 0.61 eV; Branching ratio: 0.46; Gaussian width: ~ 220 meV (C1) and ~ 260 meV (C2); Lorentzian width: 80 meV (C1 and C2). The asymmetry parameter of the Doniach-Šunjić line profile is 0.055.

in energy. A reason for the difference can be that in the present calculations self-energy effects are not included to obtain the quasiparticle band structure [28]. Even though the $(2\sqrt{3} \times 2\sqrt{3})$ model used for calculation does not exactly represent the real “ $2\sqrt{3}$ ” silicene structure, we conclude that the experimental bands observed by ARPES originate from the silicene layer. The comparison with theory identifies them as bands of mainly σ character. This is a report of electron bands that can be unambiguously assigned to silicene.

Angle-integrated Si $2p$ core-level spectra were also measured in order to gain further information about the atomic and electronic structures. Figure 4 shows a Si $2p$ spectrum measured from “ $2\sqrt{3}$ ” silicene using a photon energy of 135 eV. The spectrum consists of two components, C1 and C2. The intensity of C2 corresponds to $\sim 15\%$ of the total intensity. It is interesting to compare this result to the expectations based on the $(2\sqrt{3} \times 2\sqrt{3})$ model in Fig. 3(a). The two Si atoms, located on top of Ag atoms, constitute 14% of the 14 Si atoms in the unit cell. These atoms are the ones observed by STM both in the bright and dark parts, indicating that they are markedly different from the other silicon atoms. It is therefore natural to assign the C2 component of the Si $2p$ spectrum to these atoms. Although, the real “ $2\sqrt{3}$ ” silicene layer shows deviations from the atomic structure of the $(2\sqrt{3} \times 2\sqrt{3})$ model, we conclude that the Si $2p$ core-level data are consistent with the model used in this study.

IV. SUMMARY

By analyzing LEED and STM results, we have obtained a detailed structure determination of “ $2\sqrt{3}$ ” silicene on Ag(111). Due to strain, the silicene layer exhibits a quasiperiodic long-range modulation resulting in a hexagonal-like superstructure with a lattice constant ~ 3.7 times larger than that of a $2\sqrt{3}$ unit cell. The homogeneous coverage of the Ag(111) surface by this type of silicene made it meaningful to perform an ARPES study. As the main result, we have reported four silicene bands, S_1 – S_4 . By comparing to a band structure calculation on a relaxed $(2\sqrt{3} \times 2\sqrt{3})$ model we could unambiguously assign S_1 – S_3 to σ bands of the silicene sheet.

ACKNOWLEDGMENTS

Technical support from Dr. Johan Adell, Dr. Craig Polley, and Dr. T. Balasubramanian at MAX-lab is gratefully acknowledged. Financial support was provided by the Swedish Research Council (Contracts No. 621-2010-3746, No. 621-2014-4764, and No. 621-2011-4426) and by the Linköping Linnaeus Initiative for Novel Functional Materials supported by the Swedish Research Council (Contract No. 2008-6582). The calculations were carried out at the National Supercomputer Centre (NSC), supported by the Swedish National Infrastructure for Computing (SNIC).

-
- [1] M. T. Yin and M. L. Cohen, *Phys. Rev. B* **29**, 6996 (1984).
 - [2] G. G. Guzmán-Verri and L. C. Lew Yan Voon, *Phys. Rev. B* **76**, 075131 (2007).
 - [3] S. Cahangirov, M. Topsakal, E. Aktürk, H. Şahin, and S. Ciraci, *Phys. Rev. Lett.* **102**, 236804 (2009).
 - [4] P. Vogt, P. De Padova, C. Quaresima, J. Avila, E. Frantzeskakis, M. C. Asensio, A. Resta, B. Ealet, and G. Le Lay, *Phys. Rev. Lett.* **108**, 155501 (2012).
 - [5] D. Tsoutsou, E. Xenogiannopoulou, E. Golias, P. Tsipas, and A. Dimoulas, *Appl. Phys. Lett.* **103**, 231604 (2013).
 - [6] C.-L. Lin, R. Arafune, K. Kawahara, M. Kanno, N. Tsukahara, E. Minamitani, Y. Kim, M. Kawai, and N. Takagi, *Phys. Rev. Lett.* **110**, 076801 (2013).
 - [7] S. K. Mahatha, P. Moras, V. Bellini, P. M. Sheverdyaeva, C. Struzzi, L. Petaccia, and C. Carbone, *Phys. Rev. B* **89**, 201416(R) (2014).
 - [8] Y.-P. Wang and H.-P. Cheng, *Phys. Rev. B* **87**, 245430 (2013).
 - [9] S. Cahangirov, M. Audiffred, P. Tang, A. Iacomino, W. Duan, G. Merino, and A. Rubio, *Phys. Rev. B* **88**, 035432 (2013).
 - [10] H. Jamgotchian *et al.*, *J. Phys.: Condens. Matter* **24**, 172001 (2012).
 - [11] Z.-X. Gao, S. Furuya, J.-i. Iwata, and A. Oshiyama, *J. Phys. Soc. Jpn.* **82**, 063714 (2013).
 - [12] L. Chen, H. Li, B. Feng, Z. Ding, J. Qiu, P. Cheng, K. Wu, and S. Meng, *Phys. Rev. Lett.* **110**, 085504 (2013).
 - [13] P. Vogt *et al.*, *Appl. Phys. Lett.* **104**, 021602 (2014).
 - [14] J. Avila, P. De Padova, S. Cho, I. Colambo, S. Lorcey, C. Quaresima, P. Vogt, A. Resta, G. Le Lay, and M. C. Asensio, *J. Phys.: Condens. Matter* **25**, 262001 (2013).
 - [15] S. Cahangirov, V. O. Özçelik, L. Xian, J. Avila, S. Cho, M. C. Asensio, S. Ciraci, and A. Rubio, *Phys. Rev. B* **90**, 035448 (2014).
 - [16] P. Moras, T. O. Montes, P. M. Sheverdyaeva, A. Locatelli, and C. Carbone, *J. Phys.: Condens. Matter* **26**, 185001 (2014).
 - [17] C. -L. Lin *et al.*, *Appl. Phys. Express* **5**, 045802 (2012).
 - [18] H. Enriques *et al.*, *J. Phys. Condens. Matter* **24**, 314211 (2012).
 - [19] M. R. Tchalala *et al.*, *Appl. Surf. Sci.* **303**, 61 (2014).
 - [20] A. Acun, B. Poelsema, H. J. W. Zandvliet, and R. van Gastel, *Appl. Phys. Lett.* **103**, 263119 (2013).
 - [21] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevB.92.205427> for detailed analysis of LEED patterns and FFTs of STM images.
 - [22] P. Blaha, K. Schwarz, G. K. H. Madsen, D. Kvasnicka, and J. Luitz, *WIEN2k: An Augmented Plane Wave+Local Orbitals Program for Calculating Crystal Properties* (Karlheinz Schwarz, Technische Universität, Wien, Austria, 2001).
 - [23] G. Kresse and D. Joubert, *Phys. Rev. B* **59**, 1758 (1999).
 - [24] B. Feng *et al.*, *Nano Lett.* **12**, 3507 (2012).
 - [25] J. Gao and J. Zhao, *Sci. Rep.* **2**, 861 (2012).
 - [26] H. Jamgotchian *et al.*, *J. Phys.: Condens. Matter* **27**, 395002 (2015).
 - [27] J. R. Osiecki and R. I. G. Uhrberg, *Phys. Rev. B* **87**, 075441 (2013).
 - [28] F. Aryasetiawan and O. Gunnarsson, *Rep. Prog. Phys.* **61**, 237 (1998).