High-resolution photoemission study of the discommensurate (5.55×5.55) Cu/Si(111) surface layer

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We have mapped the electronic bands of the “quasi-(5×5)" monolayer structure of Cu on Si(111) near the Fermi energy at high angular and energy resolution. Although this unusual system does not exhibit a true long-range periodicity, well-defined bands and sharp Fermi surface contours are observed. The fundamental Fermi surface inside the first surface Brillouin zone has the shape of a hexagon and possesses strong nesting features. However, no relation to the unusual translational symmetry could be established. Strong umklapp bands and Fermi surface contours are observed with umklapp vectors that correspond to the reciprocal lattice of the slightly expanded and rotated discommensurate CuSi layer, and to that of the quasi-(5×5) domain pattern formed by the regular dislocation network. We find thus a one-to-one correspondence between the electronic bands and the complex structure model that has been established by Zegenhagen et al. [Phys. Status Solidi B 204, 587 (1997)]. This suggests that the two-dimensional electron gas formed on this surface does not induce the formation of the discommensurate structure but rather takes a spectator role.

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I. INTRODUCTION

The growth of copper on Si(111) has attracted not only academic but also technological attention for the past two decades and is nowadays one of the most relevant metal-on-semiconductor interfaces for modern device technology. One of the interesting issues in this system is the well-defined and stable interface layer formed during elevated-temperature growth. Examination of this layer by low-energy electron diffraction (LEED) and He atom scattering revealed a (5.55×5.55) periodicity that is not commensurate with the Si(111) substrate. These findings have stirred a lot of interest in the surface science community 10 years ago. Structural investigations came up with three different models for the local bonding, which involve three different surface stoichiometries. From Auger electron diffraction measurements, Chambers, Anderson, and Weaver suggested a coplanar CuSi structure where Cu atoms occupy the H₃ sites in a vertically compressed Si(111) surface double layer. Chambers and Rhodin have measured the position and the dispersion of the Cu 3d band by angle-resolved ultraviolet photoelectron spectroscopy (ARUPS). From comparing their data to band-structure calculations, they favor a simple CuSi structure with Cu substituting for Si in the S₃ sites terminating the surface. A bit later, Zegenhagen et al. were able to identify two inequivalent Cu sites by means of an x-ray standing wave study, thus completing the CuSi model by adding one additional Cu atom into H₃ sites to form a corrugated Cu₄Si₂ layer. This model could recently be confirmed by Cu 2p x-ray photoelectron diffraction (XPD). While these probes were sensitive to the average local bonding geometry, they could not give further insight into the discommensurate character of the layer. This aspect also proved to be a challenge to scanning tunneling microscopy (STM). The pronounced bias dependence of the images and the variety of local structures that invariably appear on STM topographs, make determination of the surface unit cell and the film registry on the Si(111) substrate very difficult. The conclusions were that a dense network of misfit dislocations develops due to the large lattice mismatch, that at least three electronically and structurally distinct local phases arrange to form a quasi-aperiodically ordered structure, and that the observed quasi-(5×5) periodicity is the moiré periodicity arising from placing the Cu-Si layer on the (1×1) template of the Si(111) lattice plane. In the latter work, it is conjectured that the layer is vertically compressed to be nearly planar, and that this compression is accompanied by a lateral expansion of 9.7% and a ±3.3° rotation, whereby a moiré period of 5.55₃×₁ results. Here, a₃×₁ = 3.84 Å is the Si(111) surface lattice vector. A complete tiling of the surface can be achieved by having three different types of domains that are connected via 5.5₃₃×₁. Earlier transmission electron microscopy (TEM) results have shown large domains of typically 1 µm size that show either a plus or a minus 3.5° rotation with respect to the substrate directions, fully supporting this model. There is thus domain formation at two different length scales (ca. 20 Å, bounded by the dislocation network, and 1 µm for either clockwise or counterclockwise rotation). These results, comprehensively reviewed by Zegenhagen et al. and schematically depicted in Fig. 1, for the convenience of the reader, gave a full flavor of the complexity of this interface, and the general interest has since decayed.

More recently, metal-semiconductor interfaces have gained renewed interest from a different point of view. In the monolayer regime, some are found to represent low-dimensional materials that can exhibit exotic physical phenomena such as Peierls-like instabilities and surface charge-density wave (CDW) formation. For such scenarios the Fermi surface of the two-dimensional (2D) layer needs to have specific nesting features, i.e., parallel contours that can be connected by one particular so-called nesting vector q*, and that often render the electronic system quasi-one-dimensional. The CDW transition is driven by a singularity in the electronic susceptibility occurring at the wave vector q*, which in the truly 1D electron gas is equal
II. EXPERIMENTAL

The experiments were performed in a modified Vacuum Generators ESCALAB 220 spectrometer that is described elsewhere.\textsuperscript{24} The Si(111)-7×7 surface was prepared by cycles of Ar\textsuperscript{+} sputtering (500 V) at room temperature and consecutive flashing/annealing. The n-type crystal (0.07 Ω cm) of dimensions 1.0×0.4×0.03 cm\textsuperscript{2} was heated resistively. The quality and cleanliness of the surface were checked with LEED and x-ray photoelectron spectroscopy (XPS). From XPD measurements, we determined the crystal orientation to within better than 1°.

Copper was evaporated from a hot tungsten filament to give a deposition rate of about 0.1 monolayers (ML) per minute as calibrated by XPS measurements. The pressure was kept below 2×10\textsuperscript{-9} mbar during deposition. About 1–3 ML of Cu were deposited on the sample at room temperature. Subsequent heating of the crystal to temperatures of 500–600 °C reduced the Cu coverage to about 0.8 ML and led to the formation of the quasi-(5×5) phase as judged from the well-defined LEED pattern.

For the photoemission experiments He Iα (21.2 eV) and He Iβα (40.8 eV) radiation was used for excitation, produced in a microwave-driven high-flux He discharge lamp with a toroidal grating monochromator (Gammadata Burklin AB, Sweden). The energy resolution of the electron analyzer was set to about 40 meV, the angular resolution to better than 1° full width at half maximum. Data acquisition was made either by taking series of energy spectra while scanning one photoelectron emission angle (polar angle θ or azimuthal angle φ) to produce energy dispersion plots, or by recording photoelectron intensities at the Fermi level while scanning both emission angles across the full hemisphere to produce Fermi surface maps.\textsuperscript{24} The Fermi energy was determined by fitting the Fermi edge measured on an Ag polycrystal. All experiments were performed at room temperature, at a base pressure below 3×10\textsuperscript{-10} mbar. Typical measuring times for the individual data sets were about 5 h.

III. RESULTS AND DISCUSSION

In Fig. 2(a), the normal emission spectrum from a quasi-(5×5) Cu/Si(111) layer of 0.8 ML thickness is shown. It is dominated by the strong Cu 3d emission in the binding-energy region between 2 eV and 5 eV, in agreement with the earlier study by Chambliss and Rhodin.\textsuperscript{5} An additional peak appears at 1.37 eV that they missed, maybe due to their limited energy (0.2 eV) and angular resolution, or due to different light polarization. We can identify the origin of this peak by comparing the spectrum to the normal emission spectrum from the clean Si(111)-7×7 surface that is given as the lower curve in Fig. 2(a), and which shows two prominent features at 2.0 eV and 0.9 eV, labeled as B and S2, respectively. The feature B is associated with a direct transition from the uppermost Si(111) bulk band of Λ\textsubscript{3} symmetry.\textsuperscript{21} We have compared the dispersion of the 1.37 eV peak on the Cu/Si interface (see below) with the dispersion of feature B measured on the Si(111)-7×7 (not shown) and
found them to be very similar, which establishes the bulk origin of this extra spectral feature. Compared to the clean Si surface, the peak (B) appears here shifted by 0.63 eV to lower binding energy. This result indicates that band bending of the valence band towards the Fermi level has occurred on the surface. Since the binding energy of the valence band maximum (VBM) on the $7 \times 7$ surface is $\approx 0.63$ eV, the VBM for the quasi-$(5 \times 5)$ surface is estimated to be located very close to the Fermi level. This indicates that a $p$-type inversion layer is created by the formation of the interface.

In the remainder of this paper, we want to focus on the electronic bands in the vicinity of the Fermi energy $\epsilon_F$. Figures 2(b,c) show series of ARUPS spectra taken with a photon energy of 21.22 eV (He I$\alpha$ radiation) along the $\bar{\Gamma}M$ and $\bar{\Gamma}K$ directions of the Si(111) $(1 \times 1)$ surface Brillouin zone (SBZ). All the spectra show a Fermi edge and a steep background beginning immediately below $\epsilon_F$ and rising monotonously towards the onset of the Cu 3$d$ band. Superimposed on this background is a fast-dispersing feature that can be seen along both directions at binding energies below 1 eV. The particular polar angle range from $\theta = 10^\circ$ to $\theta = 20^\circ$ has been selected in order to illustrate the Fermi level crossings of this feature. It is obvious that the quasi-$(5 \times 5)$ surface is metallic. In the previous ARUPS measurements on this surface by Chambliss and Rhodin, they also have observed the same electronic states near the Fermi level. However, in their spectra these features are much broader than those of the present study due to their poorer energy resolution, and they could not conclude whether the observed surface state crosses the Fermi level or not. The metallicity that we find is consistent with the data of an early inverse photoemission study of this interface where a structure of relatively high intensity was observed at the Fermi level. From our very detailed measurements of the metallic states it emerges that there are actually two features that disperse along $\bar{\Gamma}K$ and that cross the Fermi level at a few degrees apart (see below).

In order to trace all the Fermi-level crossings as a function of $k$, we have compiled the photoemission intensities at $\epsilon_F$ for all emission angles and plotted them in the $k_{||}$ plane. At a given azimuth, $k_{||}$ is calculated as $k_{||} = 1/h\sqrt{2m(h\nu-\Phi)}\sin \theta$ where $\theta$ is the polar emission angle measured from the surface normal, $h\nu$ the photon energy, $m$ the free-electron mass and $\Phi$ the work function that is taken to be 5.3 eV. Figure 3(a) shows the resulting Fermi surface contours obtained with He I$\alpha$ excitation. Centered inside the first Si(111) $(1 \times 1)$ SBZ a regular hexagon appears with its sides parallel to the zone boundaries. Stronger replica of these straight hexagon sides appear in all six neighboring SBZ's, showing a strictly sixfold rotational symmetry. The intensities of the contours, on the other hand, are modulated to give only a threefold symmetry. The photoemission matrix elements responsible for the measured intensities are thus sensitive to the threefold symmetry of the local bonding configuration within the Cu$_2$Si film.

The observed Fermi-level crossings are extremely sharp in $k$ space. This is clearly seen in momentum distribution curves (MDC's), i.e., in intensity line scans across the Fermi surface map. The maxima near 0.5 and 1.5 Å$^{-1}$ in Fig. 3(b) are very sharp and reproduce well in the two inequivalent $\bar{\Gamma}M$ directions. The curves have been sampled with 1° steps in polar angle, meaning that the dominant peak in the second SBZ has a full width at half maximum of less than 3°. The precise Fermi wave vector $k_{F(1)}$ measured from $\bar{\Gamma}$ is found to be $0.48(2)$ Å$^{-1}$, which equals $0.51(2)$ times the distance $\bar{\Gamma}M = 0.945$ Å$^{-1}$ on the Si(111) $(1 \times 1)$ surface. The two curves along the $\bar{\Gamma}M$ and $\bar{\Gamma}M'$ directions illustrate the marked inequivalence with respect to photoemission intensities. In Fig. 3(c), we show the $k_{||}$ dispersion of the contours in the 2nd SBZ along the dashed curve (A-B) indicated in Fig. 3(a). Four pairs of metallic states are clearly identified that are separated in azimuth angle by 6.5° within each pair. Since this azimuthal scan samples the second SBZ's not far from equivalents of the $\bar{\Gamma}M$ line, and since there is only one strong Fermi-level crossing along the $\bar{\Gamma}M$ direction, these data indicate that the photoemission measurement picks up Fermi surface contours simultaneously from the two micrometer-sized domains that exhibit either a clockwise or
counterclockwise rotation of about 3° with respect to the substrate low-index directions. From such data we may now venture to measure the exact size and shape of the Fermi surface. In order to confirm the visual impression of a hexagonal shape, we inspect the line scan along the $G\overline{1}$ direction, which is also shown in Fig. 3. For a regular hexagon with the measured value for $k_W$ (1) of 0.48(2) Å$^{-1}$ along the $G\overline{M}$ direction, we expect a corner distance of $k_W$ (2) = $k_W$ (1) $\times \sqrt{3}/2$, which is 0.55(2) Å$^{-1}$. Although the width of the peak in Fig. 3 indicates the presence of two bands crossing the Fermi level, the one further out in $k_i$ is very nearly at this position. The conclusion from this preliminary analysis is thus that the quasi-(5$\times$5) Cu/Si$_{111}$ layer has to first order a Fermi surface with the shape of a hexagon that spans very nearly half the linear dimensions of the (1$\times$1) SBZ. However, complications arise due to the two rotational domains and due to additional features related with the quasi-(5$\times$5) superstructure, as will be discussed further below.

In order to verify the 2D nature of the interface-related bands, we have measured a corresponding data set by using He II ($h\nu = 40.8$ eV). The Fermi surface map for this photon energy is shown in Fig. 4(a), again with the (1 $\times$ 1) SBZ indicated in order to give the relevant $k$ space

![Image](https://example.com/image.png)
The higher photon energy of He II leads to a marked shift of the main contour to higher values in the first SBZ, where the Fermi surface is still well centered around the zone centers. In the second SBZ, however, the shift is no longer true, and the Fermi surface at six more places. Where we have overlap with the He I data, the contours look quite similar, with a rather straight section parallel to the zone boundaries and the split contours that appear to bend around the zone centers. Rather than completing the picture, this additional data set at higher photon energy raises more questions.

We suggest that the answer to this puzzling and energy-dependent deviation from periodicity in k space lies in the quasiperiodic structure of the Cu/Si overlayer leading to umklapp scattering with discommensurate surface lattice vectors. Such umklapp processes can lead to strong additional photoemission features as a function of angle and energy. In order to visualize umklapp bands, Fig. 5 displays photoemission dispersion plots along the \( \Gamma M \) and \( \Gamma K \) directions. The white dashed lines represent the band edges of the bulk bands projected onto the \( \text{Si}(111) \) (1×1) SBZ. The metallic states are positioned well within the bulk band gap, and the states are thus clearly identified as surface states. The two strong Fermi surface crossings near 0.5 and 1.5 Å\(^{-1} \) along the \( \Gamma M \) direction, which were discussed above, are associated with two dominant bands [Fig. 5(a)] that are, at first sight, related to each other by the symmetry of the \( \text{Si}(111) \)-(1×1) Brillouin zone (see below). A similar but broader band appears along the \( \Gamma K \) direction [Fig. 5(b)]. These data clearly identify the hexagonal Fermi surface centered at \( \Gamma \) to be of the hole type. In addition to these dominant bands, there are a number of weaker bands that also cross the Fermi level. These crossings, some of which are marked by white lines, can also be seen as extra peaks in the MDC’s of Fig. 3(b), clearly visible on either side of the main Fermi surface peak in the first SBZ. Along the \( \Gamma M \) line, these features appear to be roughly periodic, with umklapp vectors of the order of 1/11 of a \( \text{Si}(111)-(1×1) \) reciprocal lattice vector \( q_{1×1} \) [indicated by arrows in Fig. 5(a)]. This value corresponds to half the periodicity \( q_{5×5} \) of the quasi-(5×5) lattice. In order to understand this reduced spatial frequency, we have to consider the 2D character of the k-space geometry. The situation is sketched in Fig. 6(a). Due to the sixfold arrangement of quasi-(5×5) umklapp vectors, and due to the hexagon shape of the Fermi surface, the contours from two umklapp vectors that lie 120° apart combine to form extra straight lines constituted by two parallel hexagon sides. These extra lines are...
and the contour of the first umklapp vector along the halfway features rather than that along the Fermi surface centered at \( \Gamma \) reciprocal lattice point situated just halfway between the Fermi surface centered at \( \Gamma \) and the contour of the first umklapp vector along the \( \Gamma M \) line. They are thus responsible for the apparent reduction of spatial frequency along this direction. The Fermi surface near the hexagon corners (\( \Gamma K \) direction) is sampled in these halfway features rather than that along the \( \Gamma M \) direction. This explains why the Fermi-level crossings marked in Fig. 5(a) are not precisely separated by 1/2 \( q_{3}\times 5 \). In fact, these data imply a slight deviation from a perfectly hexagonal shape of the Fermi surface, with the hexagon corners pushed slightly outwards. This impression may well arise due to the presence of two bands crossing the Fermi level at very close positions along the \( \Gamma K \) direction [see Fig. 3(b)].

The dispersion plot for the electronic bands along the \( \Gamma K \) direction, which is shown in Fig. 5(b), shows the two-band character of the main feature much more clearly. The broad peak at the Fermi level separates into two distinct bands at binding energies higher than 0.5 eV. Along this direction, the 2D umklapp vectors lead to features with a periodicity of 1/\( \sqrt{3}\times q_{5}\times 5 \) according to the model of Fig. 6(a). In this \( k \)-space section, we realize the high degree of complexity that arises due to this strong umklapp scattering also at higher binding energies. A one-to-one identification of individual bands is not possible here without the help of a band-structure calculation for the Cu/Si interface. Fortunately, the situation at the Fermi level appears to be simpler due to the presence of one dominating band or maybe two bands that are nearly degenerate except along the \( \Gamma K \) direction.

We now address the problem of the inconsistent positions of Fermi surface contours in the second SBZ, as observed with He I and He II excitation [cf. Figs. 3(b) and 4(b)]. In doing so, we need to consider all the subtleties of the structural model as described by Zegenhagen et al.,\textsuperscript{15} i.e., that the nearly coplanar Cu\textsubscript{3}Si layer is laterally expanded by 9.7% and rotated by \( +3^\circ \) relative to the underlying Si(111) lattice within the individual domains formed by the quasi-(5\texttimes 5) dislocation network. Clarifying the origin of the surface state requires a band structure calculation for this surface compound, which is not available to the present study. Nevertheless we can speculate on the character of the electronic states that cross the Fermi energy. In this energy region one expects Cu 4\( s \), Cu 3\( d \) and Si 3\( sp \) valence electrons. Previous ARUPS and x-ray absorption studies\textsuperscript{33,34} have shown that the hybridization of Cu 3d with Si valence states lies in-plane of the quasi-(5\texttimes 5) layer and that of Cu 4s lies perpendicular to the layer. This indicates that the Cu-Si bonding in the quasi-(5\texttimes 5) layer is likely to follow the typical transition-metal-silicide bonding scheme, e.g., in a bulk Cu silicide (Cu\textsubscript{3}Si),\textsuperscript{35} but in two dimensions. Thus, one would expect that the metallic state that is observed in the present study, is a partially filled antibonding state resulting from an in-plane hybridization of Cu 3d and Si 3\( sp \) states. The measured band dispersion and the holelike character of the Fermi surface are fully consistent with this antibonding picture.

An important consequence of this assignment is that these states are weakly coupled to the underlying substrate,\textsuperscript{33} and that the electrons thus probe essentially the potential of the expanded and slightly rotated lattice within the Cu\textsubscript{3}Si layer. The relevant periodicity is thus not described by the Si(111)-(1\texttimes 1) reciprocal lattice but rather by one that is shrunk by roughly 10% and rotated by \( +3^\circ \). Figure 6(a) illustrates the resulting geometries that arise from surface umklapp scattering off these periodicities. The Fermi surface related with the antibonding band is sketched as a bold hexagon centered at the origin \( \Gamma_{00} \) of the two-dimensional momentum space. A

FIG. 6. (a) Schematic drawing of the various Fermi surface contours obtained in the first and second surface Brillouin zone by quasi-(5\texttimes 5) umklapp scattering of the hexagonal Fermi surface. In the second SBZ, the umklapp vectors \( q_{(5\times 5)} \) are centered at the reciprocal lattice point \( \Gamma^\text{CuSi}_{10} \) of the slightly expanded and rotated Cu\textsubscript{3}Si layer and not at \( \Gamma_{11}^{(1\times 1)} \) of the underlying Si(111)-(1\texttimes 1) lattice. (b) Complex umklapp pattern of Fermi surface contours obtained by superimposing two domains rotated by \( \pm 3^\circ \). (c) Overlay of this umklapp pattern of Fermi surface contours on the real data. The He I and He II excited Fermi surfaces have been brought to matching \( k \)-vector scales.
replica of this Fermi surface is plotted, again in bold lines, in one of the neighboring SBZ’s. Note that this contour is not centered around the reciprocal lattice point $\Gamma_{10}$ of the Si(111)-(1×1) surface, but around the slightly shifted $\Gamma_{CuSi}^{10}$ point associated with the expanded and rotated Cu$_2$Si layer. Within each rotational domain, the periodic network of dislocations leads to the (5.55×5.55) or quasi-(5×5) periodicity relative to the Si(111)-(1×1) lattice. Umklapp scattering off the dislocation network potential thus produces Fermi surface replica centered at positions $\Gamma_{00} + \tilde{q}_{(5×5)}$ and $\Gamma_{10} + \tilde{q}_{(5×5)}$, where $\tilde{q}_{(5×5)}$ is one of the six symmetry-equivalent quasi-(5×5) reciprocal lattice vectors. Figure 6(a) shows the intricate line pattern that is formed by the entity of all these umklapp Fermi surfaces, with the apparent reduction of spatial frequencies along the $\Gamma M$ direction discussed earlier. The presence of roughly equal amounts of domains rotated by $+3^\circ$ or $-3^\circ$ should lead to the rather messy picture plotted in Fig. 6(b). In Fig. 6(c), the complete umklapp pattern is overlayed upon the experimental He I and He II Fermi surface maps, all of which are given in the same momentum scale. The direct comparison yields some interesting observations:

(i) The experimental data show some but not all of the umklapp Fermi surface contours.

(ii) Where they are observed, they appear at positions that are predicted by our simple model.

(iii) The measured intensities of the contours vary strongly within the individual SBZ and from one to the other.

(iv) The relative intensities of different umklapp contours are very different for He I and He II excitation.

(v) The dominant contours measured in the second SBZ correspond to quasi-(5×5) umklapp bands, and not to the Fermi surface centered around $\Gamma_{10}$. Different umklapp vectors $\tilde{q}_{(5×5)}$ dominate for He I and for He II.

Overall, this comparison gives convincing evidence that the momentum distribution of the measured surface state reflects all aspects of the complete structural model as described by Zegenhagen et al.\cite{15}

The striking dependence of the umklapp scattering intensities on the excitation energy can be understood on the basis of the one-step model of photoemission.\cite{36} The final state is here taken to be a time-reversed LEED state. During the characterization of the quasi-(5×5) phase by LEED, we noticed a pronounced change of relative intensities within the quasi-(5×5) spot sets as a function of electron energy. This is shown in Fig. 7, where LEED patterns at three different energies are given. At 100 eV the quasi-5×5 spots around each fundamental spot have very similar weights, while at the lower energies, the spots closer to the (0,0) beam are significantly stronger. At 44 eV and 100 eV there is also an increasing threefold component in the spot-intensity distribution due to the growing contribution of substrate scattering. Although we have not studied these effects systematically, the LEED patterns indicate that the energy dependence of the umklapp scattering intensities is a final-state effect. It is worth noting that our LEED patterns do not show the $\pm 3^\circ$ rotation of spots expected from the presence of rotational domains. At the energies that we used, which lie between 36 eV and 100 eV, electron diffraction from the underlying Si(111) substrate appears to be strong enough to lock the quasi-(5×5) spots at positions that are centered around the (1×1) reciprocal lattice vectors. There are reports that LEED patterns taken at very low energies show the presence of the rotated domains by a $\pm 3^\circ$ azimuthal splitting of the quasi-(5×5) spots centered around the (0,0) spot.\cite{37} The diffraction intensities must be coming from the first monolayer alone, like in our photoemission study of the metallic band.

The strong momentum dependence of the umklapp intensities is more interesting and surprising. In the following, we discuss two mechanisms that are known to distribute intensities nonuniformly over momentum space.

Coherent photoemission from inequivalent atoms within the Cu$_2$Si surface unit cell can interfere to modulate the measured intensity in momentum space. This is conveniently described by the so-called photoemission structure factor.\cite{38,39}
which can distribute band intensities in a characteristic fashion from zone to zone, but also within each SBZ. We have performed a calculation of the photoemission structure factor by using a simple tight-binding model for the Cu$_2$Si layer with spherically symmetric Cu 3$d$ and Si 3$p$ orbitals placed on the atomic sites. Preliminary results predict strong photoemission of the fundamental band (umklapp scattering is not contained in this model) inside the first SBZ and weak emission in all six neighboring zones, with little variation inside each zone except for enhanced emission along the zone boundaries. Although the calculation may be over simplified, these results might rationalize why the fundamental band is suppressed relative to the umklapp bands in the second SBZ’s, as is observed in Figs. 3–6. However, a more sophisticated calculation including the detailed electronic structure and surface umklapp scattering is required in order to fully clarify this effect.

A different mechanism that can redistribute photoemission intensities in momentum space has recently been discussed by Voit et al. They calculate the single-particle spectral function for solids with competing periodic potentials. The solutions to their simple model show two things: main bands and bands folded by the periodic potentials repel each other wherever they are close in energy and momentum and form energy gaps where they cross. Moreover, there is a shift of spectral weight from main bands to folded bands near these crossing points. This is due to the coherent interaction of the quasiparticles with the periodic potentials, and as such this is an initial-state effect and not to be confused with umklapp scattering in the final state, although the momentum shifts of the folded bands are the same. The quasi-(5×5) Cu/Si(111) surface is obviously a system with strongly competing potentials, and the Voit model should thus be relevant.

A complete analysis of the observed umklapp band intensities needs to combine all these discussed initial-state and final-state processes and is thus a formidable task and beyond the scope of this paper. Preliminary results show that applying the Voit model to a single tight-binding antibonding band, parametrized to match the fundamental Fermi surface in the first SBZ, can reproduce almost quantitatively the many bands shown in the dispersion plots of Fig. 5 as far as band positions are concerned (not shown). Some significant spectral weight shift to folded bands occurs in the second SBZ, qualitatively in accordance with the experimental data, but for a correct description of umklapp band intensities, umklapp scattering in the final state needs to be considered as well (see above). The periodicity of the underlying Si(111) substrate was found to play no significant role.

Let us now discuss possible scenarios for the formation of the quasi-(5×5) structure of the Cu$_2$Si layer. As described in the introduction, a metallic system in reduced dimensions becomes unstable against a lattice distortion due to the interaction of the electrons near the Fermi level with phonons at 2$k_F$. The strength of this interaction scales with the degree of Fermi surface nesting, and if it is strong enough, it manifests itself as a periodic lattice distortion and the formation of a charge-density wave. The hexagon-shaped Fermi surface of the quasi-(5×5) Cu/Si(111) surface does show some nesting, and we can identify a nesting vector of $2k_F = 0.96(2)$ Å$^{-1}$ oriented $\pm 3^\circ$ away from the $\Gamma M$ direction. However, this value is in no simple relationship to any of the periodicities present on this surface: $\vec{q}_{10}$ of the Si(111)-(1×1) is at 1.89 Å$^{-1}$, $\vec{q}_{CuSi}$ at 1.72 Å$^{-1}$, and $d_{(5\times5)}$ is 0.34 Å$^{-1}$. The closest match would be achieved with a (2×2) reconstruction with respect to the underlying Si(111) lattice, but there is no indication for such a reconstruction within the film, at least not at room temperature. Moreover, the data of Fig. 5 show clear Fermi-level crossings for all observed bands with no gap formation at $E_F$. We may, therefore, reasonably conclude that the formation of the quasi-(5×5) structure is due to the near balance between the adsorbate-substrate interaction and the local bonding within the Cu$_2$Si layer, and that none of the more exotic scenarios apply.

IV. CONCLUSIONS

We have performed Fermi surface and band mapping measurements by high-resolution photoemission spectroscopy to study the electronic structure of the quasi-(5×5) Cu/Si(111) surface near the Fermi level. It is clearly established that the surface is metallic. The Fermi surface is of the hole type; it has a hexagonal shape and spans half the linear dimensions of the Si(111)-(1×1) SBZ. In addition, several umklapp bands and Fermi surface contours are observed that relate directly to the complete structural model of the system, since the metallic band is associated with electrons that hybridize strongly within the Cu$_2$Si layer and weakly perpendicular to the film, the basic periodicity is that of the expanded film and not that of the underlying substrate. In the second SBZ’s the bands from the two rotational domains are well resolved, and quasi-(5×5) umklapp bands dominate over the fundamental bands, with a strong dependence of spectral weight on momentum and on excitation energy. Although some nesting appears to be present due to the hexagonal shape of the Fermi surface, there seems to be no conceivable connection to the formation of the quasi-(5×5) structure. The present study demonstrates that high-resolution Fermi surface and band mapping experiments with photoemission spectroscopy is a very powerful tool to study such complex systems as the commensurate quasi-(5×5) Cu/Si(111) surface.

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The ordering of an adlayer on a substrate is dictated by two competing forces, the adsorbate-adsorbate interaction and the adsorbate-substrate interaction. If the adsorbate-adsorbate interaction is dominant, the adsorbate layer may be incommensurate. If the adsorbate-substrate interaction is dominant, the adlayer is usually commensurate. If the two forces are competitive, the resulting structure can be discommensurate: weakly incommensurate domains form in which the adsorbate layer is highly strained. These domains are separated by a regular network of dislocations, where the strain is released (see, e.g., Ref. 15).

The data have been analyzed by the XPDPLOT package written by R. Fasel and R. A. Agostino, University of Fribourg.

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