Continuous Transition from Two- to One-Dimensional States in Si(111)-(5×2)−Au

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A strong, gold-induced surface state is found on single-domain Si(111)-(5×2)−Au at low temperatures. Its band dispersion is one dimensional near the Fermi level E_F and gradually becomes two dimensional towards the bottom of the band, thus providing a model for a continuous transition in dimensionality. A Peierls-like gap is observed in the one-dimensional portion of the band near E_F.

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The investigation of one-dimensional electronic systems has frequently relied on three-dimensional crystals with embedded chains, such as rows of transition metals stabilized by an ionic lattice. Recently, progress has been made in fabricating one-dimensional chain structures of adsorbates on top of two-dimensional surfaces. The independent control of substrate and overlayer makes it possible to construct a wide variety of systems with tailored electronic properties. Semiconductor surfaces lend themselves towards such a program since their broken bonds naturally have a tendency towards rearranging into long-range reconstructions. A recent example is the observation of a one-dimensional, metallic band at a highly stepped Si(111) surface covered with 0.2 monolayers of gold [1]. Apparently, the rigid substrate prevents the inherent tendency of such a system to undergo a Peierls transition and become insulating. Instead, this structure shows indications of spin-charge separation [1], as predicted by Luttinger and others for one-dimensional metals. This surface exhibits a 5×1 diffraction pattern that suggests a relation to a 5×2 structure obtained on flat Si(111) at about 0.4 monolayers gold coverage. The latter exhibits a one-dimensional band also, which has been reported to be metallic in photoemission at room temperature [2]. Inverse photoemission results have suggested that the metallic states arise from Si adatoms, not from Au [3]. There exist several other one-dimensional structures of the type $m \times 1$ on Si, such as the metallic (4×1)-In structure [4] and 3×1 structures induced by Ag, Li, Na, K, and Mg [5].

We explore the electronic states of the Si(111)-(5×2)−Au surface at low temperature (≈16 K), where we find the spectral features to become much more pronounced than at room temperature. A gold-induced band is observed that is half-filled with respect to a 5×1 unit cell, but avoids being metallic by doubling the periodicity along the chain into 5×2 and forming a Peierls gap. Surprisingly, the Au-induced surface state band loses its one-dimensional character below the Fermi level and displays a gradually increasing dispersion perpendicular to the chains. Such a continuous transition from two-dimensional to one-dimensional electronic states is highly unusual. Canonical low-dimensional systems contain bands with well-defined dimensionality. Having a dimensional crossover within the same band makes the Si(111)-(5×2)−Au system an interesting model for low-dimensional electronic states and for the phase transitions associated with them.

A single-domain Si(111)-(5×2)−Au structure was created on a stepped Si(111) substrate oriented 1.1° towards the $[\bar{1}\bar{1}2]$ azimuth. Scanning tunneling microscopy (STM) images of this surface [6] showed well-separated, one-dimensional chains running parallel to the steps in the $[\bar{1}10]$ direction, in agreement with previous STM studies [7]. The Si(111)-(7×7) substrate (n-type, 10^{18} cm^{-3}) was prepared according to a method that has demonstrated single-domain 7×7 structures extending over >400 nm and straight steps with kink densities as low as one in 2×10^4 edge atoms [8]. Au was evaporated in a vacuum of $<10^{10}$ Torr with the substrate at 700 °C. The optimum coverage for the 5×2 structure was calibrated by LEED and by maximizing the intensity of the Au surface state (in steps of 0.1 monolayer). The Si(111)-(7×7) states were characterized under the same conditions [9], including the surface photovoltage effects at low temperature [10]. Photoelectron spectra were acquired with a hemispherical Scienta SES200 spectrometer equipped with angle and energy multidetection and coupled to an undulator beam line at the SRC. The energy resolution was 20 + 7 meV (photons + electrons), the angular steps were 1° along the chains and 0.3° perpendicular to them. All angles ϑ are referred to the [111] direction.

Figure 1 compares angle-resolved photoelectron spectra of Si(111)-(5×2)−Au and Si(111)-(7×7). The clean 7×7 surface exhibits two surface states at 0.1 eV and 0.7–0.8 eV below E_F, which are assigned to adatoms and rest atoms, respectively [9]. The spectrum of the (5×2)−Au surface is dominated by a single, well-pronounced state. Its surface character is established by three criteria, i.e., the lack of dispersion with photon energy ($22 \leq h\nu \leq 48$ eV), the absence of underlying bulk features in the clean Si spectra (see Fig. 1), and the location of the Au band outside the projected bulk bands [10] (Fig. 2, bottom). The proximity of the surface state to the bulk band edge might look suspicious at first glance. However, it is fairly common for surface states to track the edge of the bulk bands that they are split off from. Remnants

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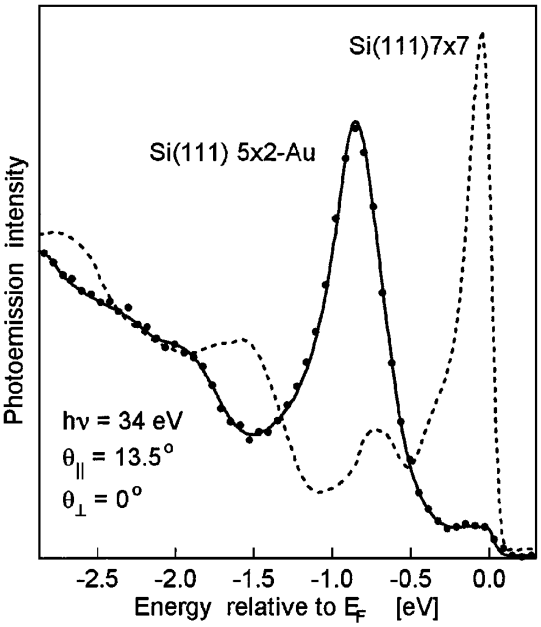

FIG. 1. Angle-resolved photoemission spectrum from the Si(111)-(5 × 2)−Au chain structure compared to the Si(111)-(7 × 7) substrate. The 7 × 7 adatom state at E_F is replaced by a gold-induced surface state below E_F.

of the Si(111)-(7 × 7) surface states are still present in the (5 × 2)−Au spectra, such as a weak Fermi edge and the rest atom state at 0.7–0.8 eV below E_F (open circles in Fig. 2, bottom). Such a slightly gold-deficient surface with Si(111)-(7 × 7) patches is selected in order to avoid the two-dimensional ($\sqrt{3} \times \sqrt{3}$)−Au phase [6].

The gold-induced state displays a strong band dispersion parallel to the chains (Fig. 2, top left and bottom). The bandwidth of 0.95 ± 0.03 eV indicates substantial coupling between electronic orbitals along the chain. The low effective mass $m^* = 0.5m_e$ at the bottom of the band suggests high electron mobility. In the direction perpendicular to the chains, we observe a smaller dispersion, i.e., 0.14 ± 0.03 eV at the band minimum and a negligible 0.03 ± 0.03 eV at the maximum (Fig. 2, top right). $\vartheta_\perp = 4°$ corresponds to the boundary π/d of the Brillouin zone perpendicular to the chains ($d = 16.6$ Å = chain spacing).

In view of the small perpendicular dispersion and the well-separated chain structure in STM, we use a one-dimensional approximation to the 5 × 2 Brillouin zone as a starting point (Fig. 3, bottom, horizontal lines). The one-dimensional zone boundary of a single chain is $\text{ZB}^{n\times1} = \pi/a$ ($a = 3.8$ Å = period along the chain). It becomes $\text{ZB}^{n\times2} = \frac{1}{2}\text{ZB}^{n\times1}$ after doubling the period a. The boundaries of the actual 5 × 1 and 5 × 2 Brillouin zones are zigzag lines straddling their one-dimensional approximations. The 5 × 1 reciprocal lattice, which dominates the LEED pattern, is given by open circles in Fig. 3.

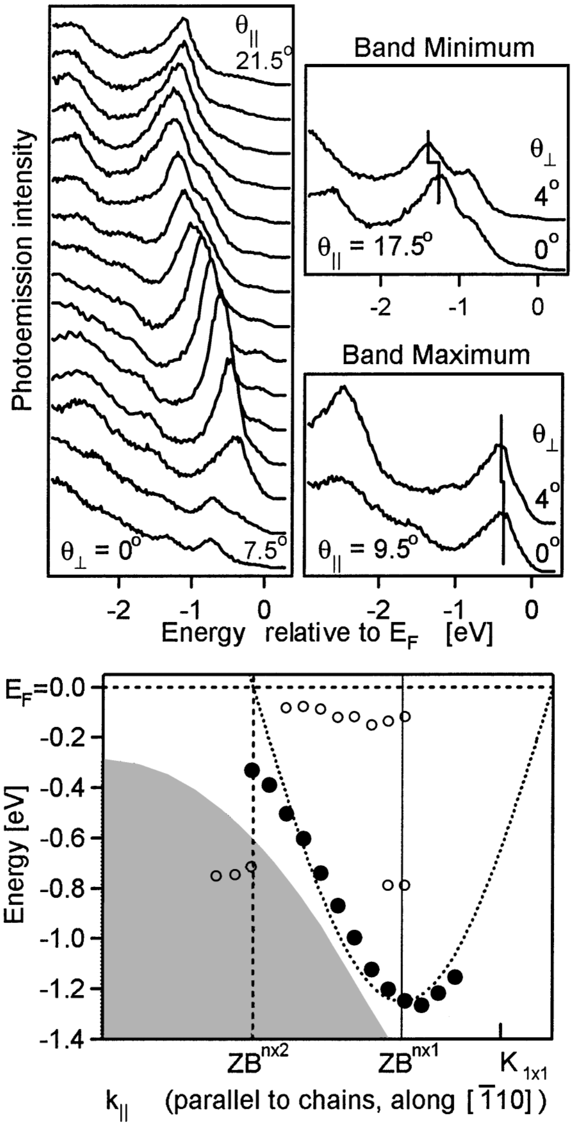

FIG. 2. Band dispersion of the gold-induced state on Si(111)-(5 × 2)−Au. The top panels provide raw data in the directions parallel and perpendicular to the chains ($\vartheta_\parallel$ along $[\bar{1}10]$ and $\vartheta_\perp$ along $[11\bar{2}]$). The surface state (solid circles) can be approximated by a half-filled band (dotted curve). However, it circumvents the Fermi level via doubling the periodicity along the chain from 5 × 1 to 5 × 2 and opening a Peierls gap at the new zone boundary $\text{ZB}^{n\times2}$.

In a one-dimensional Brillouin zone with period a, the surface state resembles a half-filled, metallic band, as indicated by the dotted cosine function in Fig. 2. The top of such a band would be located at $\mathbf{k} = 0$, and the Fermi level crossing would occur at $\text{ZB}^{n\times2} = \pi/2a = 0.41$ Å^{-1}. That lies close to the crossing of a metallic band observed on the highly stepped Si(111)-(5 × 1)−Au structure [1]. In the 5 × 2 structure, the band never reaches the Fermi level [solid circles in Fig. 2 (bottom)]. Instead, it bends over and leaves a gap below E_F. The location of the gap at $\text{ZB}^{n\times2}$ is suggestive of a Peierls gap associated with the doubling of the 5 × 1 periodicity to 5 × 2.

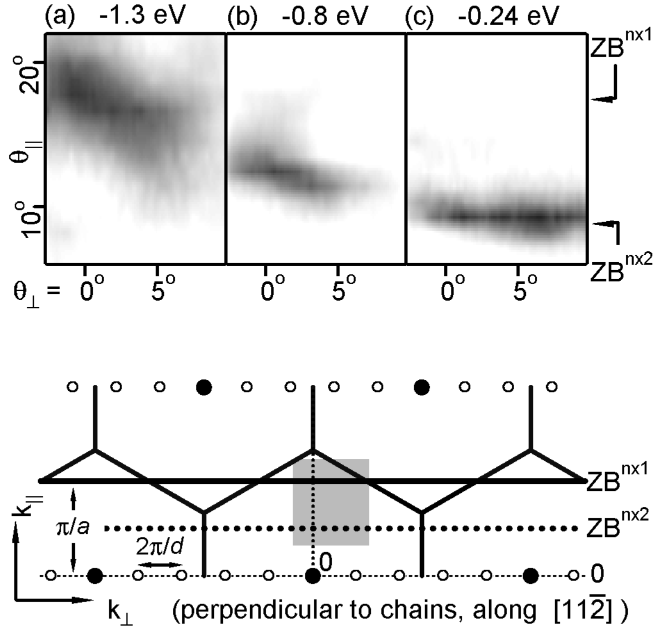

FIG. 3. Top: Constant energy contours in **k** space for the surface state on Si(111)-(5 × 2)−Au, obtained from angular photoemission distributions at $h\nu = 34$ eV (high intensity shown dark). The emission at the bottom of the band in (a) spreads diagonally in both **k** directions, indicating two-dimensional behavior. The horizontal contour at the top of the band in (c) is one dimensional, because it does not depend on $k_\perp$. It coincides with the period-doubled zone boundary ($\mathrm{ZB}^{n\times2}$), suggesting that it has triggered a Peierls gap. Bottom: The one-dimensional Brillouin zone of a single chain, compared to the bulk Brillouin zone (hexagonal lines, solid circles) and a 5 × 1 diffraction pattern (open circles). The zone boundaries are $\mathrm{ZB}^{n\times1}$ and $\mathrm{ZB}^{n\times2}$ before and after doubling the period a along the chain. The gray rectangle represents the **k** range of panels (a)−(c).

This picture of a Peierls gap is corroborated further when considering the band dispersion perpendicular to the chains. The states closest to E_F are one dimensional and thus prone to a Peierls instability. Their dimensionality can be assessed most reliably by compiling an extensive set of data over two-dimensional **k** space, such as in Fig. 3 (top). Dark features represent high photoemission intensity. They correspond to constant energy contours of the band structure, the analog to energy surfaces in three dimensions. For the top of the band, one finds a nearly horizontal contour [Fig. 3(c)], i.e., the energy does not depend on $k_\perp$ (k perpendicular to the chains). That characterizes a one-dimensional band. Furthermore, the contour lies very close to the Brillouin zone boundary $\mathrm{ZB}^{n\times2}$, where one would expect the opening of a Peierls gap. These two observations suggest that the states in Fig. 3(c) trigger the formation of the doubled 5 × 2 structure. There is an intriguing connection with the exotic, metallic (5 × 1)−Au structure on highly stepped Si(111) [1], which has a Fermi level crossing near the contour in Fig. 3(c). Apparently, the underlying steps suppress the Peierls distortion and preserve a half-filled band similar to the dotted curve in Fig. 2.

While a Peierls gap remains an interesting aspect to explore further, we focus our attention onto a surprising change in dimensionality that occurs when going from the top of the band in Fig. 3(c) towards the bottom in Fig. 3(a). At the bottom, one finds a diagonal energy contour which indicates a two-dimensional band dispersion (both parallel and perpendicular to the chains). The tilt of the energy contour vanishes gradually towards the top of the band in Fig. 3(c), where we have a horizontal, one-dimensional contour. The wave function corresponding to such a band must be quite unusual. At the top of the band, it appears to be localized to a chain; at the bottom, it extends out to neighboring chains. That would be contrary to the expectation that states at higher energies are more delocalized. In order to demonstrate that such a band topology is not at odds with basic band theory, we provide a model band structure in Fig. 4. It reproduces the observed band topology after folding the band in Fig. 2 back into the first $n \times 2$ zone, i.e., folding the band minimum back to $\mathbf{k} = 0$ (Γ). At the Γ point in Fig. 4, the chains are coupled by a string of intermediate orbitals, which we choose to be antibonding to match the experiment (Γ has higher energy than Y). The chains decouple at the band maximum XZ because the intermediary orbitals lie on nodal planes and have zero amplitude. A more realistic band structure will have to take the complex arrangement of Si and Au atoms at the 5 × 2 surface into account, which is not yet agreed upon.

In summary, we find a continuous transition from two-dimensional to one-dimensional behavior for the dominant surface band of the Si(111)-(5 × 2)−Au chain structure. States at the top of the band (close to the Fermi level) are purely one-dimensional, explaining the one-dimensional characteristics of that surface. However, the dispersion perpendicular to the chains increases gradually towards the bottom of the band. This observation is quite different from canonical low-dimensional systems, where each band has a well-defined dimensionality, such as a surface state versus a bulk band [11]. A suggestion is made for the type of wave functions that can generate a band of continuously

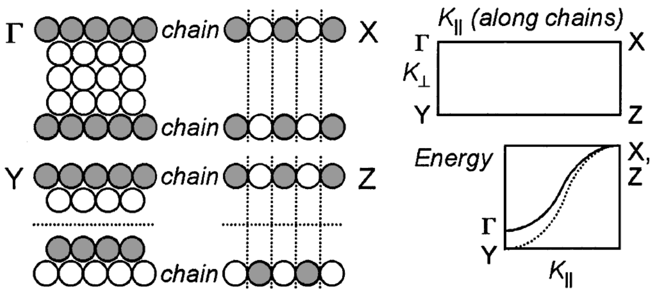

FIG. 4. Model orbitals for a band that changes its dimensionality. Two chains of s, p_z orbitals are coupled by intermediate orbitals at Γ ($\mathbf{k} = 0$). They become decoupled at the zone boundary XZ (= $\mathrm{ZB}^{n\times2}$), because the intermediate orbitals fall onto nodal planes (dotted straight lines). Orbitals with plus (minus) signs are shown as hatched (open) circles.

variable dimensionality. Such an unusual system may exhibit other instabilities beyond the Peierls transition, such as those discussed for Group IV adsorbates on Si(111) and Ge(111) [12].

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[1] P. Segovia, D. Purdie, M. Hengsberger, and Y. Baer, Nature (London) **402**, 504 (1999).

[2] I. R. Collins, J. T. Moran, P. T. Andrews, R. Cosso, J. D. O'Mahony, J. F. McGilp, and G. Margaritondo, Surf. Sci. **325**, 45 (1995). In this paper and in Ref. [3], the miscut is opposite to ours and much larger (4° versus 1.1°). Therefore, the uphill and downhill directions are interchanged.

[3] I. G. Hill and A. B. McLean, Phys. Rev. B **55**, 15 664 (1997).

[4] I. G. Hill and A. B. McLean, Phys. Rev. B **56**, 15 725 (1997); Phys. Rev. B **59**, 9791 (1999); Phys. Rev. Lett. **82**, 2155 (1999); H. W. Yeom, S. Takeda, E. Rotenberg, I. Matsuda, K. Horikoshi, J. Schaefer, C. M. Lee, S. D. Kevan, T. Ohta, T. Nagao, and S. Hasegawa, Phys. Rev. Lett. **82**, 4898 (1999).

[5] S. C. Erwin and H. H. Weitering, Phys. Rev. Lett. **81**, 2296 (1998).

[6] STM results by D. Y. Petrovykh *et al.* (to be published).

[7] J. D. O'Mahony, J. F. McGilp, C. F. J. Flipse, P. Weightman, and F. M. Leibsle, Phys. Rev. B **49**, 2527 (1994); M. Shibata, I. Sumita, and M. Nakajima, Phys. Rev. B **57**, 1626 (1998).

[8] J. Viernow, J.-L. Lin, D. Y. Petrovykh, F. M. Leibsle, F. K. Men, and F. J. Himpsel, Appl. Phys. Lett. **72**, 948 (1998); J.-L. Lin, D. Y. Petrovykh, J. Viernow, F. K. Men, D. J. Seo, and F. J. Himpsel, J. Appl. Phys. **84**, 255 (1998).

[9] R. Losio, K. N. Altmann, and F. J. Himpsel, Phys. Rev. B **61**, 10 845 (2000).

[10] P. Pervan, K. Markert, and K. Wandelt, Appl. Surf. Sci. **108**, 307 (1997). Based on the band bending reported for 0.3 monolayers of Au on p- and n-type Si(111), we have placed the top of the valence band at -0.3 eV in Fig. 2. The photovoltage causes some uncertainty in the exact value of the observed gap of 0.3 eV.

[11] The closest alternative to our finding is the formation of a surface resonance when a surface state enters a region of bulk bands. That is a discontinuous transition, however. We do not observe any two- or three-dimensional band that could give rise to such a resonance.

[12] G. Santoro, S. Scandolo, and E. Tosatti, Phys. Rev. B **59**, 1891 (1999).