Band Splitting for Si(557)-Au: Is It Spin-Charge Separation?

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(Received 24 October 2000)

It has been proposed that the Si(557)-Au surface exhibits spin-charge separation in a one-dimensional electron liquid. Two narrowly spaced bands are found which exhibit a well-defined splitting at the Fermi level. That is incompatible with the assignment to a spinon-holon pair in a Luttinger liquid. Instead, we propose that the two bands are associated with two nearly degenerate atomic chains, or a chain of step atoms with two broken bonds. Such an assignment explains why the surface is metallic despite an even number of electrons per unit cell.

DOI: 10.1103/PhysRevLett.86.4632 PACS numbers: 73.20.At, 71.10.Pm, 79.60.Jv

Low-dimensional structures induced by metals at the Si(111) surface exhibit a variety of interesting electronic states. There have been reports of correlation effects destroying the metallicity of surfaces with an odd electron count per unit cell [1,2], of anomalous surface corrugation either by charge density waves [3], or by large atomic displacements [4], of metallic nanowires [5], of a surface state with mixed dimensionality [6], and of spin-charge separation in a Luttinger liquid [7,8]. The phenomenon of spin-charge separation in a one-dimensional metal has been sought as one of the truly novel phases of electrons [7,9]. The predicted spectral function [9] exhibits two peaks, one containing predominantly the spin degree of freedom (spinon), the other the charge (holon). These two quasiparticle peaks exhibit different dispersions with different group velocities, but they have to join at the Fermi level E_F and recombine into an electron.

One of the best candidates for spin-charge separation is the Si(557)-Au surface, a highly stepped Si(111) surface that contains one Au chain and one step per unit cell (Fig. 1A). A one-dimensional, metallic state was found on that surface [7], suggesting that the threat of a Peierls transition to a semiconductor can be avoided by rigidly anchoring a metallic chain to a step. A splitting of the surface state near E_F was observed and taken as an indication for spin-charge separation [7]. We find that this is a well-ordered, metallic surface, indeed, but demonstrate that the band splitting does not vanish at the Fermi level. Therefore, it cannot be attributed to the spinon-holon splitting in a Luttinger liquid. Our data are best explained by two nearly identical surface states. Such an assignment is quite unusual as well, indicating that there are two nearly identical chains within the unit cell that create a pair of bonding/antibonding surface states. In fact, we observe such a doublet of chains by scanning tunneling microscopy (STM). Another possibility is a pair of degenerate orbitals on the same chain, such as the two broken bonds of the Si atoms at the step edge.

The interaction between electron liquids in chain structures has been an active field of theoretical research [10]. Electron liquids become more exotic in one dimension and many of the models cannot be solved in higher dimensions. Chain models have been taken as analogs of two-dimensional systems, such as high temperature superconductors with multiple CuO_2 layers in the unit cell. The critical temperature T_c depends on the number of CuO_2 layers, suggesting that hopping between adjacent layers is a key to superconductivity [10]. It has been disturbing that photoemission experiments have not resolved the interlayer splitting expected from band calculations, but a spin-charge separated Luttinger liquid might offer an explanation. Calculations show [10] that the non-Fermi-liquid nature of the states competes with interlayer hopping and might be winning in that case. Our results for

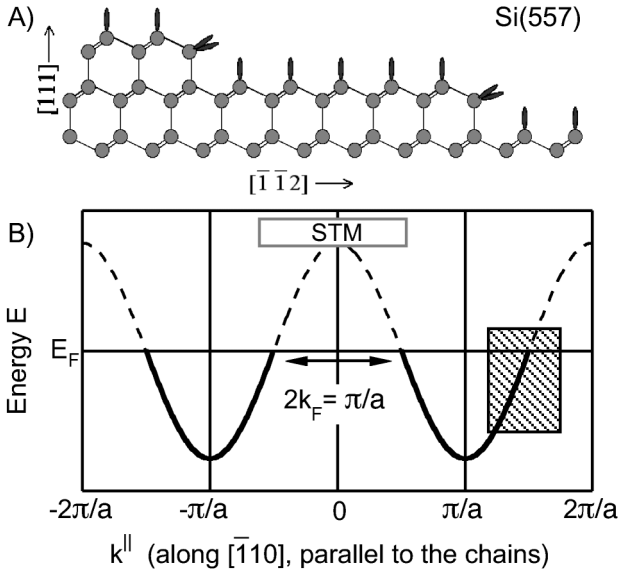

FIG. 1. (A) Side view of the unreconstructed Si(557) surface showing seven unpaired electrons per unit cell in broken bond orbitals. Adding one unpaired electron from a single Au atom gives an even number of eight electrons. Extra Si atoms contribute four electrons each and do not alter the parity of the electron count. (B) Schematic of the band topology for Si(557)-Au (excluding the observed band splitting). Data for the hatched (E, k) area are shown in Fig. 2.

 0031-9007/01/86(20)/4632(4)$15.00

one-dimensional chains on Si represent the opposite limit where chain hopping dominates. Any remaining spectral features of Luttinger liquid behavior must be at a smaller energy scale (<0.05 eV).

Our two-band assignment also solves the puzzle of why the Si(557)-Au surface is metallic, despite an even number of electrons per unit cell. Usually, an even count produces a semiconductor because the opening of a gap lowers the energy of the occupied states. Two half-filled bands are a viable alternative, however, and provide a natural explanation of the metallicity of Si(557)-Au. This surface and the Si surfaces with correlation gaps [1,2] both defy conventional wisdom that an even electron count produces a semiconductor and an odd count produces a metal.

Gold adsorbed on Si(111) surfaces forms two distinct, one-dimensional structures, i.e., the highly stepped Si(557)-Au surface discussed here [7,11] and the flat Si(111)-(5 × 2)-Au surface [6,12–14]. Both exhibit chains of atoms along the $[\bar{1}10]$ direction when imaged by STM, and in both cases the unit cell contains five atomic rows of the unreconstructed surface. The Si(557)-Au surface contains an additional double step per unit cell (see Fig. 1A). Although the two Si-Au structures have common features, some of the basic electronic characteristics are quite different, such as the Au content and the electron count. Si(557)-Au contains 0.2 ML of Au, which is equivalent to a single Au row per unit cell [7,11]. Si(111)-(5 × 2)-Au contains twice as much Au, i.e., 0.4 ML or the equivalent of two Au rows per unit cell [14]. For determining the number of unpaired electrons per unit cell let us begin with the unreconstructed Si(557) and Si(111)-(5 × 1) surfaces and count the number of broken bonds (bold in Fig. 1A). Both structures contain an odd number of electrons in broken bonds, 7 for Si(557) and 5 for Si(111)-(5 × 1). Next, we add Au atoms with an unpaired s electron each. According to the reported coverage we have one Au electron for Si(557)-Au and two Au electrons for Si(111)-(5 × 1)-Au, resulting in a total of 8 electrons for Si(557)-Au and 7 for Si(111)-(5 × 1)-Au. Finally, silicon atoms may be added to the unit cell during reconstruction. They contain four electrons each and do not affect the parity of the electron count. After doubling the unit cell along the chains the count becomes even in both cases.

The Si(557)-Au surface was prepared from n-doped, 1 Ωcm wafers with a miscut of 9.5° from [111] towards $[\bar{1}\,\bar{1}2]$. The surface quality was optimized by varying the Au coverage and the annealing conditions and observing the resulting structures with STM. After preparing a clean substrate by heat cleaning at 1250 °C [15] we deposited gold at 650 °C with a subsequent anneal at 950 °C. We confirmed the ratio of 1:2 of the Au coverages for Si(557)-Au and Si(111)-(5 × 2)-Au, using LEED and STM. Below the optimum coverage we observe 7 × 7 patches in both cases, together with triple steps and triple terraces for Si(557). Above the optimum coverage the steps bunched on Si(557), and Si(111)-(5 × 2)-Au terraces appeared in between. The flat Si(111)-(5 × 2)-Au exhibited patches of the $(\sqrt{3} \times \sqrt{3})$-Au phase. Details of the preparation will be reported elsewhere [16].

The photoemission data in Fig. 2 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 interval was 1/4° with multidetection along the chain direction. The data were taken at a temperature of about 100 K with frequent recleaning at 850 °C. All measurements were performed with p-polarized light in the emission plane normal to the surface that contains the steps and chains. Several photon energies were used in order to demonstrate that there is no coupling to bulk states, which would manifest itself by a change of band energies with $h\nu$ at fixed parallel momentum $k^{\parallel}$ (compare Fig. 2, left and right). As with other Si(111) surfaces, we found an optimum in the cross section of surface states near $h\nu = 34$ eV. At $h\nu = 27$ eV the surface signal is only half as large (compare the signal-to-background ratio in Fig. 2, top). For the He line at $h\nu = 21.2$ eV used in Ref. [7] the Fermi edge becomes so weak that the splitting of the bands at E_F is difficult to determine. That explains why our conclusion is

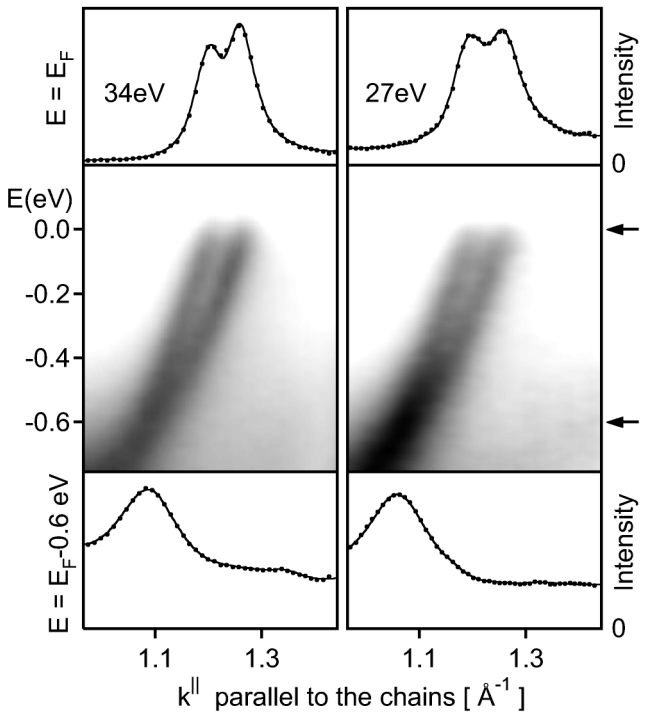

FIG. 2. Band dispersion of the metallic surface state on Si(557)-Au near the Fermi level E_F, measured by E, k multidetection (center). High photoemission intensity is shown dark. Two nearly degenerate bands are observed with a splitting that increases towards E_F. The momentum distributions at E_F (top panels) clearly show a splitting, while spinon and holon bands in a Luttinger liquid would have to converge at E_F.

different from [7], even though our data are quite similar at $h\nu = 21.2$ eV.

The experimental band dispersion in Fig. 2 (center) exhibits two closely spaced bands (in dark) crossing the Fermi level E_F. Right at E_F there is a clear splitting, as demonstrated by the momentum distribution in Fig. 2 (top). Farther down in energy the splitting decreases and leads to a single, unresolved peak in Fig. 2 (bottom). The band crosses E_F about half way to the boundary of the Brillouin zone, which is located at $\pi/a = 0.82$ Å^{-1} (where $a = a_{\text{cubic}}/\sqrt{2} = 3.84$ Å is the atomic spacing along the chains). The band topology is given schematically in Fig. 1B (without the band splitting). All four crossings in Fig. 1B are observed. Figure 2 focuses onto one of them in the hatched region of Fig. 1. A Lorentzian fit to the curves in Fig. 2 top yields a pair of peaks at $k^{\parallel} = 1.20$ Å^{-1}, 1.26 Å^{-1}. After reducing these values to the 1st Brillouin zone one obtains a pair of Fermi wave vectors $k_F = 0.44$ Å^{-1}, 0.38 Å^{-1}. They are perfectly symmetric about half filling, i.e., $k_F = 0.54\pi/a$, $0.46\pi/a$. Together the two bands carry two electrons and provide an even electron count. The slope of the bands in Fig. 2 center gives a Fermi velocity $v_F = \hbar^{-1}\partial E/\partial k = -1.0 \times 10^6$ m/s. For comparison, consider the values for bulk Au ($k_F = 1.21$ Å^{-1}, $v_F = +1.4 \times 10^6$ m/s) and for the Au(111) surface state ($k_F = 0.153$ Å^{-1}, 0.176 Å^{-1}, $v_F = +0.8 \times 10^6$ m/s [17]). The band topology is quite different, i.e., holelike for chains compared to electronlike for bulk and surface. At best, one could compare the chain bands in the 2nd zone with bulk Au.

The observed band splitting at E_F immediately rules out the spinon-holon splitting predicted for the Luttinger liquid. Spinon and holon bands have to converge at E_F [9]. The abrupt Fermi cutoff observed in Fig. 2 would also be difficult to explain by the predicted power-law decay of the spectral function towards zero at E_F [9]. Furthermore, having just a single band cross the Fermi level would not be compatible with the even electron count. The same argument rules out other splitting mechanisms for a single band, such as a spin splitting [17].

As our best interpretation of the data we propose a closely spaced doublet of ordinary bands crossing E_F. This is quite unusual, too, because semiconductor surfaces with an even electron count have a strong driving force to pair up electrons into completely filled bands. That leads to surface bands with a gap, instead of the observed metallic bands. In fact, we are not aware of any other semiconductor surface with such a band topology. Two nearly degenerate energy levels suggest two nearly identical orbitals within the unit cell that generate bonding/antibonding combinations, corresponding to even/odd superpositions of the two wave functions. For example, the two orbitals could originate from two chains in the unit cell or from two orbitals located on a single chain (such as the two broken bonds at the step edge in Fig. 1 or a set of in-plane, $p_{x,y}$ orbitals connecting atoms in a zigzag chain).

Actually, we find that the surface contains two chains per unit cell as shown in Fig. 3 (see the small dots in the inset). These STM images clearly demonstrate the one-dimensional, chainlike reconstruction of the Si(557)-Au surface. The repeat period of 1.9 nm consists of 5-2/3 silicon rows plus a step (Fig. 1A). The spacing between the two chains inside the unit cell is 2.2 ± 0.2 silicon rows. The spacing of the atoms along these chains is twice the lattice spacing ($2a = 7.68$ Å instead of $a = 3.84$ Å). Every other atom in each chain is missing. Adjacent chains seem to lack phase correlation since about half of the chains are arranged in a zigzag pattern, and the other half in a ladder structure, as seen on the right and left side of the inset in Fig. 3, respectively. The resulting diffraction pattern in LEED has the half-order spots suppressed and elongated perpendicular to the chains. A Fourier transform of the STM image gives analogous half-order streaks.

STM cannot distinguish whether these chains consist of Au or Si atoms. If they were Au atoms, the coverage of two half-occupied rows would be consistent with the

FIG. 3. STM images of the Si(557)-Au surface revealing a one-dimensional structure with two nearly equivalent chains of atoms in the unit cell (inset). The overview shows the derivative of the topography, the inset the topography itself. The $[\bar{1}\,\bar{1}2]$ direction (downhill) is to the right.

observed 0.2 ML (about one row per unit cell). If they were Si atoms, their pattern would match possible Si adatom arrangements, i.e., the zigzag rows would correspond to a 2×2 lattice and ladders to a $c(4 \times 2)$. Also, the chains atoms are resolved best when tunneling into empty states at positive sample bias (+2.0 V in Fig. 3, +1.6 V for the inset), which is typical for the adatoms on Si(111)-(7×7). A variety of possible chain configurations and their implications on the band structure will be discussed elsewhere [16], including a model where the step atoms with two broken bands give rise to the split bands.

In addition to the chains there are extra protrusions occupying random lattice sites in Fig. 3 (see the two large dots in the inset). Such defects comprise only 0.01 ML and can be expected to contribute little to the photoemission spectrum, even though they appear prominently in the STM topography. Similar protrusions have been observed on Si(111)-(5×2)-Au [12,13].

In summary, the combination of the real space information from STM and reciprocal space data from photoemission supports a Si(557)-Au surface consisting of one-dimensional chains and exhibiting a pair of nearly degenerate bands. As a result we have a metallic surface despite an even electron count. A spinon-holon splitting proposed previously can be ruled out by the observation of two separate Fermi level crossings. With its sharp Fermi level crossings and well-defined chain structure the Si(557)-Au structure is a nearly ideal, one-dimensional metal. It promises to become a fertile ground for producing derivatives with customized features, such as the electron count and the internal ladder structure. The periodicity of 1.9 nm between metallic chains and a nonmetallic substrate makes the Si(557)-Au structure comparable to the stripes that have been discussed in high temperature superconductors.

We are grateful to N. Shannon and R. Joynt for insights into spin- charge separation in a Luttinger liquid and to J. N. Crain for help with the experiment. This work was supported by the NSF under Awards No. DMR-9815416 and No. DMR-0079983. It was conducted at the SRC, which is supported by the NSF under Award No. DMR-0084402.

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