# Experimental Evidence for Spin-Split Bands in a One-Dimensional Chain Structure

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Gold atom chains on vicinal Si(111) surfaces exhibit an unusual doublet of half-filled bands, whose origin has remained uncertain. The splitting is identified by angle-resolved photoemission as a spin splitting induced by the spin-orbit interaction (Rashba effect), in agreement with a theoretical prediction by Sánchez-Portal, Riikonen, and Martin. This interaction leads to a characteristic pattern of avoided band crossings at a superlattice zone boundary. Two out of four crossings are avoided, with a minigap $E_G = 85$ meV and a $k$ offset of 0.05 Å$^{-1}$.

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One-dimensional electrons interact much more than higher-dimensional electrons, because they cannot avoid each other when moving along the same line in space. The Fermi liquid model of nearly free electrons loses its validity and gives way to highly correlated electrons with counterintuitive properties. The most notable prediction has been the separation of spin and charge in a one-dimensional electron gas [1]. In recent years, atomic chains at semiconductor surfaces have emerged as atomically precise one-dimensional systems [2–6]. The electrons in the chains are electronically decoupled from the substrate, as long as they remain in the gap of the semiconductor. In fact, one of these systems, the Si(557)-Au surface, exhibits a band splitting that has been taken as an example of spin-charge separation [4]. This interpretation was subsequently found to be incompatible with the persistence of the splitting at the Fermi level [5]. Nevertheless, the unusual pair of nearly half-filled bands remains an enigma. An interesting model has been derived from band calculations [6]. It involves a spin splitting, despite the fact that both silicon and gold are nonmagnetic. The spin splitting is caused by the spin-orbit interaction in the reduced symmetry of the surface, where inversion symmetry is broken (Rashba effect). Two-dimensional analogs exist for this phenomenon at the surfaces of high $Z$ metals [7–9]. It would be interesting to know experimentally whether such an effect occurs in one dimension. Many-body calculations [10] indicate that spin-orbit interaction becomes particularly influential in one-dimensional systems.

While the theoretical model of the spin splitting [6] has received a substantial amount of interest, experimental verification of the spin polarization has been difficult. The small splitting $\delta k_F = 0.05$ Å$^{-1}$ requires high $k$ resolution in angle-resolved photoemission. That is difficult to combine with spin detection, which typically costs 4 orders of magnitude in count rate. Nevertheless, such experiments have been accomplished for two-dimensional systems, such as the W(110) and Au(111) surfaces [8]. In a ferromagnetic system, the Rashba splitting can be observed by reversing the magnetization of the sample and detecting the resulting band shifts [9]. We use the existence or absence of avoided band crossings to determine whether or not the spin-orbit interaction has caused a spin splitting of a nonmagnetic material. This method makes it possible to draw conclusions about the spin without spin analysis of the photoelectrons. The pattern of band crossings is consistent with a Rashba-type spin splitting and incompatible with three other common scenarios, i.e., a splitting of unpolarized bands, a ferromagnetic exchange splitting, or an antiferromagnetic splitting. This finding agrees with the theoretical prediction [6] and is consistent with the one-dimensional plasmon dispersion observed by electron energy loss spectroscopy [11].

The method distinguishing between these types of splittings is explained in Fig. 1. Band dispersions are shown for the doublet of nearly half-filled bands, which is observed for Si(553)-Au, Si(557)-Au, and other Au chain structures on vicinal Si(111) [12]. An extra feature of the band structures in Fig. 1 is backfolding at a $2 \times 1$ superlattice (vertical dashed lines at $k_x = \pm 0.41$ Å$^{-1}$). Such a doubling of the unit cell is common to Au chain structures on vicinal Si(111) with half-filled bands, and characteristic of one-dimensional systems in general (because of the Peierls distortion). Backfolded bands interact with the original bands, if they have the same quantum numbers (including the same spin). That leads to avoided crossings and minigaps. These are offset horizontally (in $k$) for a spin-orbit induced spin splitting (top), and vertically (in $E$) for three other common types of splittings (bottom), i.e., for unpolarized, ferromagnetic, and antiferromagnetic bands. As a result, the one-dimensional density of states exhibits one dip in the first case and two dips in the other three (panels on the right side of Fig. 1). We have been able to resolve the avoided crossings at high quality Si(553)-Au surfaces and clearly find the situation depicted in the upper panel for a spin splitting induced by the spin-orbit interaction.

The lower panel in Fig. 1 represents the three other cases, where the black and gray bands are shifted in $E$, not $k$. A pair of unpolarized bands is shown, which are distinguished by an extra quantum number, for example,

 

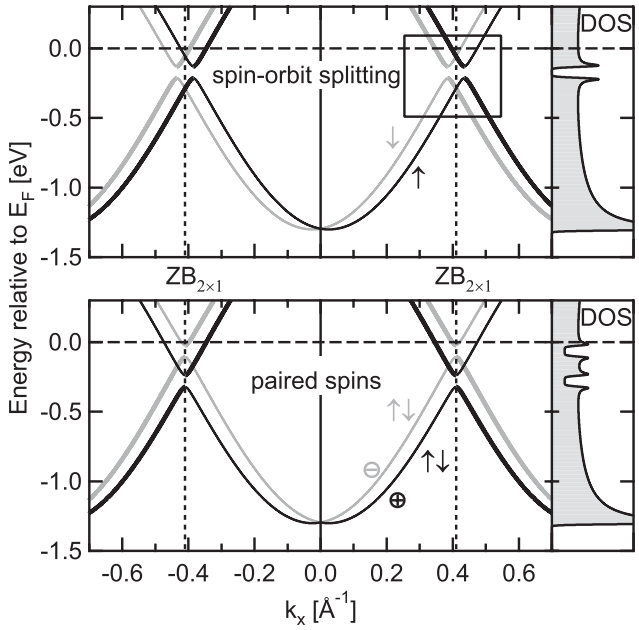


FIG. 1. Possible candidates for the split bands that are characteristic of atomic chain structures formed by Au on vicinal Si(111) surfaces. The dispersion is shown in the direction $k_x$ parallel to the chains. A spin splitting caused by spin-orbit interaction (top) can be distinguished from other splittings (bottom) via the pattern of avoided crossings at the $2 \times 1$ zone boundary $ZB_{2\times1}$. In the first case, the avoided crossings are offset horizontally (in $k_x$), in the second vertically (in $E$). Hence, the DOS exhibits either one gap (top right) or two (bottom right).

even/odd mirror symmetry (+ and −). At a Si surface, these could be $p_{x,y}$ states that become degenerate at $k = 0$. If the two unpolarized bands have the same quantum numbers, all four band crossings at each zone boundary are avoided. In the following, we discuss other situations, where the bands differ in details, but exhibit the same pair of avoided crossings shifted in $E$.

For a ferromagnet, the two bands in Fig. 1 (bottom) are distinguished by spin, with black bands corresponding to majority spin and gray bands to minority spin. Only bands with equal spin exhibit avoided crossings. Ferromagnetic bands would also exhibit a finite energy splitting at $k = 0$.

For an antiferromagnet, black bands correspond to up-spins localized to the spin-up sublattice or down-spins on the spin-down sublattice at the magnetic $2 \times 1$ zone boundary, while the gray bands are for up-spins localized to the spin-down sublattice and vice versa. Furthermore, the bands would be split only in the vicinity of the antiferromagnetic $2 \times 1$ zone boundary.

In all these cases, the observation of an avoided crossing tells us that the two participating bands have the same quantum numbers, particularly the same spin. In a sense, we use the bands themselves as internal reference for the spin quantum number instead of an external spin analysis.

The Si(553)-Au surface exhibits the same doublet of bands as Si(557)-Au, where the band calculation [6] has been performed, but it can be produced with higher perfection [12–14]. The undisturbed chain segments are longer and the resulting bands are sharper [12], which is essential for making small avoided crossing visible. The surface was prepared as described previously [5,12]. Particular attention was paid to keep the wafer strain-free during the 1250 °C flash, while holding it firmly enough to allow efficient cooling to the measurement temperature of 50 K. This temperature was chosen to be higher than the base temperature of 20 K, in order to prevent a photovoltage shift and broadening. This effect was further suppressed by choosing $p$-type samples, which exhibit a low surface Schottky barrier. The measurements were performed with synchrotron radiation at a photon energy $h\nu = 34$ eV, where the surface-to-bulk cross section ratio reaches a maximum. A Scienta 200U photoelectron spectrometer with energy and angle multidetection was used. The energy resolution was set to 15 meV (photons) and 20 meV (electrons), which was sufficient to reach the intrinsic broadening, which is dominated by inhomogeneities in the Fermi level pinning position. In contrast to previous work [5,12], the polarization of the light was not parallel to the chains, but nearly perpendicular.

Figure 2 focuses on the region of the four crossings between the split bands and their backfolded counterparts (indicated by a small box in Fig. 1, upper right). The raw data at the top of Fig. 2 show the strong primary bands, together with a hint of faint backfolded bands. The latter are brought out more clearly in the bottom panel using a high-pass filter in $E$ which eliminates smooth variations. It is clear that the strong bands are interrupted at two points by avoided gaps, and that these gaps are offset horizontally from each other. This pattern matches the top panel of Fig. 1, i.e., a spin splitting induced by the spin-orbit interaction. At the other two crossings there is no indication of a dip in the band intensity, which rules out the other three scenarios (unpolarized, ferromagnetic, and antiferromagnetic). A model calculation of the bands was performed using a two-band model with cosine electron dispersion plus the $2 \times 1$ Fourier component of the superlattice potential. This model fits the data well and gives a minigap $E_G = 85$ meV and a $k$ offset of 0.05 $\text{Å}^{-1}$ between the crossings.

The data can be analyzed more quantitatively by fits to individual spectra, as shown in Fig. 3. The top panel (a) gives an angle-resolved energy-distribution curve (EDC) with $k_x$ fixed to one of the avoided crossings (arrow in Fig. 2). The main peak is clearly split into a double-hump structure, which represents the two gray spin-down bands at the top and bottom of a minigap (see the box in Fig. 1 top right). The peak at $E_F$ is from the black up-spin band. Its counterpart lies near the bottom of the spectrum, but is too far from the avoided crossing to be visible. A fit to the EDC reveals the superlattice gap $E_G = 85$ meV [15], which is comparable to the values obtained for two-dimensional

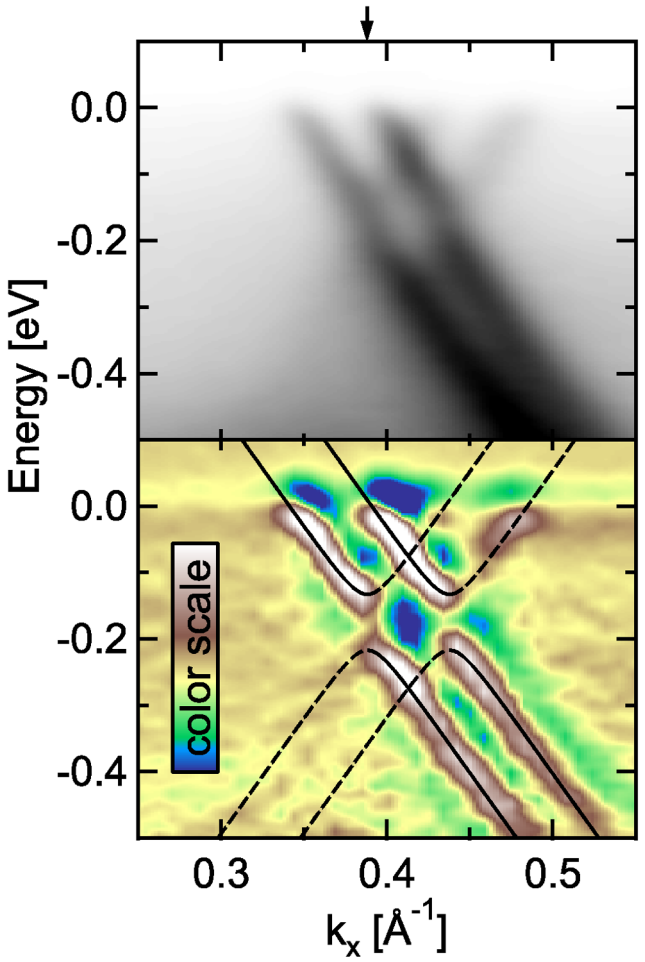


FIG. 2 (color online). Angle-resolved photoemission data of the band crossings near the $2 \times 1$ zone boundary (raw data on top and high-pass filtered at the bottom). The avoided band crossings are offset horizontally, which matches the spin-orbit scenario in Fig. 1, top. Unpolarized, ferromagnetic, and antiferromagnetic bands would exhibit a vertical offset between avoided crossings. The $E$, $k_x$ range corresponds to the small box in Fig. 1, upper right.

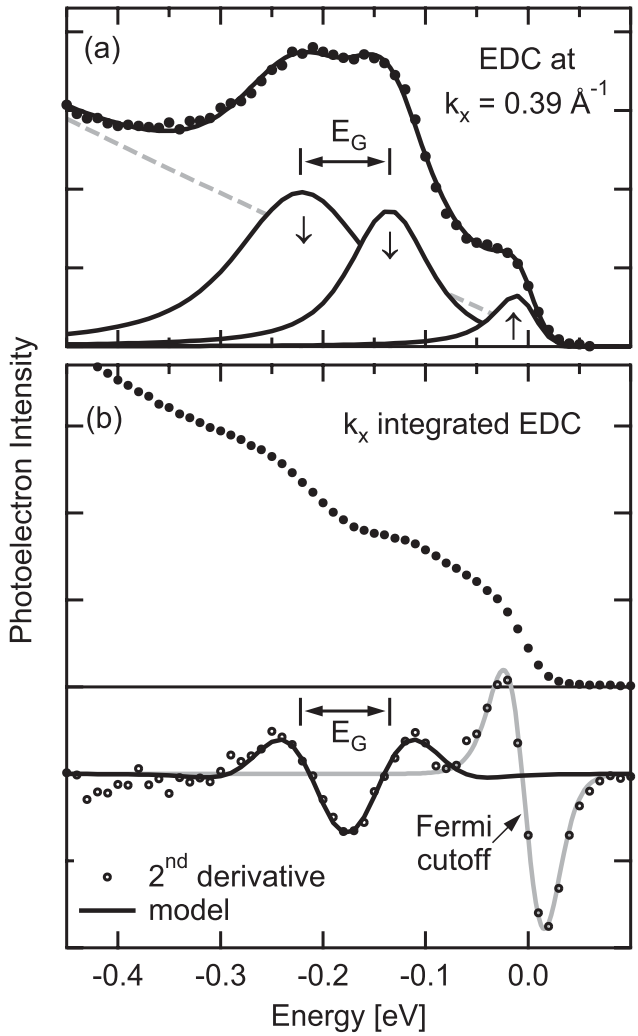


FIG. 3. (a) Angle-resolved EDC (dots) at the $k_x$ value of an avoided crossing (arrow in Fig. 2). The double-hump structure indicates a minigap $E_G$. The solid curves represent the fit result and the decomposed peaks [15]. (b) Angle-integrated EDC (top) and its 2nd derivative (bottom), integrated over the $k_x$ region shown in Fig. 2. The minigap causes a single dip in the DOS, which is characteristic of the spin-orbit scenario. The other three scenarios would have two dips in the DOS (see the panels on the right side of Fig. 1).

systems [16]. The corresponding $2 \times 1$ Fourier component of the superlattice potential is $V_{2\times1} = \frac{1}{2}E_G = 43$ meV.

The angle-integrated EDC in Fig. 3(b) can be compared to the one-dimensional density of states (DOS) on the right side of Fig. 1 as calculated from the band model. Clearly, there is only one dip in the spectrum, plus the Fermi cutoff. This observation again selects the scenario in Fig. 1 top, instead of the double dip in Fig. 1, bottom. Forming the second derivative for both the data and the DOS sharpens the edges of the gap and removes the slope of the background. The model fits the data well using the superlattice potential derived from the angle-resolved EDC [17].

In summary, we provide experimental evidence for a spin splitting in a one-dimensional chain structure. A method is used that does not require spin detection. Instead, the pattern of avoided band crossings identifies the spin direction. The finding solves a long-standing puzzle about the origin of an unusual doublet of half-filled bands observed for one-dimensional Au chains on vicinal Si(111) surfaces. It is consistent with a prediction from local density theory, and it extends the observation of Rashba-type spin splittings from two-dimensional to one-dimensional systems. It is likely that the split bands observed in several other Au chain structures have the same origin.

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