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Self-lacing atom chains

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Abstract

The structural and electronic properties of self-lacing atomic chains on Pt modified Ge(001) surfaces have been studied using low-temperature scanning tunnelling microscopy and spectroscopy. The self-lacing chains have a cross section of only one atom, are perfectly straight, thousands of atoms long and virtually defect free. The atomic chains are composed of dimers that have their bonds aligned in a direction parallel to the chain direction. At low temperatures the atomic chains undergo a Peierls transition: the periodicity of the chains doubles from a $2\times$ to a $4\times$ periodicity and an energy gap opens up. Furthermore, at low temperatures ($T < 80$ K) novel quasi-one-dimensional electronic states are found. These quasi-one-dimensional electronic states originate from an electronic state of the underlying terrace that is confined between the atomic chains.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Nanowires play an important role in the continuing downscaling of electronic components. Besides the fact that nanowires can act as interconnects between nanoscale devices, they can also be active components themselves. The ultimate nanowire has a cross section of only one atom. However, the creation of these truly ‘atomic chains’ is far from trivial. One way to produce well defined atomic chains is by using a scanning tunnelling microscope and dragging or displacing the atoms one by one to the desired position [1–3]. Another approach to create well defined atomic chains is by using self-organization or self-assembly processes. For instance, the deposition of Pt or Au on Ge(001) leads, under the right conditions, to the formation of extremely well ordered nanowire arrays [4–10]. Such self-assembly processes are also reported on metal surfaces [11, 12].

One-dimensional systems are of particular fundamental interest since they exhibit a wealth of exotic phenomena, such as quantization of conductance and Luttinger liquid behaviour [13, 14]. In addition, atomic chains can undergo a so-called Peierls transition [15–21]. Peierls suggested that a linear monovalent chain is unstable with respect to a static lattice deformation of wavevector $2k_F$. The energy band is exactly half filled and the Fermi wavevector is located halfway through the Brillouin zone, i.e. at $k = \pm\pi/2a$. Under these conditions the atom chain is metallic. Doubling of the periodicity will lead to a modulation of the electronic density, often referred to as a charge density wave (CDW), and a folding

of the Brillouin zone in reciprocal space. The edges of the new Brillouin zone coincide with the Fermi wavevector. Since the presence of a periodic potential leads to the development of an energy gap at the edges of the Brillouin zone, the chain undergoes a metal-to-insulator transition. The opening of the energy gap at the edges of the Brillouin zone lowers the energy of the electrons below the energy gap. The deformation proceeds until the increase in elastic energy is balanced by the above mentioned gain in electronic energy.

Recent experiments have revealed that the growth of Pt on Ge(001) can lead to the formation of atomic chains that are almost ideal and can thus act as a playground to study the intriguing physical properties of one-dimensional systems [4–6, 20]. The chains are dimerized in the temperature window from 4 K up to at least 400 K. At low temperatures the chains undergo a doubling of the periodicity, which can be interpreted as a Peierls transition. The exact nature of the atomic chain remains, however, controversial.

In order to determine the chemical nature of the atomic chains we have dosed the Pt modified Ge(001) surface with CO [22, 23]. It is common knowledge that CO reacts with Pt, but is virtually inert to Ge. The fact that CO only adsorbs on the atomic chains provides strong evidence in favour of the assumption that the atomic chains are comprised of Pt dimers, rather than Ge dimers. Moreover, the observed adsorption geometry is consistent with first-principle density functional calculations by Sclauzero *et al* [24] of the interaction between an isolated monatomic Pt chain and a CO molecule. However, recent density functional theory (DFT) calculations [6, 25]

have revealed that an atomic chain consisting of Ge is actually lower in energy than a Pt chain. The atomistic models that have been forward for the Ge chain structure in [6] and [25], however, differ.

After a short review of the formation process of the self-lacing atomic chains we will focus our attention on the intriguing physical properties of these chains. High resolution scanning tunnelling microscopy (STM) images of the atomic chains, recorded over a wide bias range, show that at negative sample biases the chain dimers show up as two blobs located at the positions of the atoms, whereas at positive sample biases only an (empty) state located in between the two atoms that comprise the dimer is resolved. These observations are exactly the reverse of what one would naively expect for a regular adsorbed Ge dimer on a Ge(001) surface [26, 27]. Next we will discuss the Peierls transition that these chains undergo upon cooling down to 4.7 K. At temperatures below 80 K novel quasi-one-dimensional electronic states, which are located in the troughs between the atomic chains, are found. The spatial variation of these electronic states and their relative energy position can easily be understood within the framework of the simple ‘quantum mechanical particle in a box’ model.

2. Experimental details

Experiments were performed with an ultrahigh-vacuum low-temperature scanning tunnelling microscope (Omicron) in the temperature window from 4.7 K to room temperature. Ge(001) samples are cut from nominally flat 3 in by 0.3 mm, single-side polished nearly intrinsic n-type wafers. Samples are mounted on Mo holders and contact of the samples to any other metal during preparation and experiment has been carefully avoided. In contrast to silicon, germanium crystals cannot be cleaned by flash annealing alone [28, 29]. Therefore, we cleaned the germanium crystal by applying cycles of argon ion bombardment followed by annealing at temperatures of 1100 (± 25) K. After several of these cleaning cycles the Ge(001) samples were atomically clean and exhibited a well ordered (2×1)/ $c(4 \times 2)$ domain pattern [29, 30]. Subsequently, an equivalent of 0.2–0.5 ML of platinum is deposited onto the surface at room temperature. Platinum is evaporated by resistively heating a W wire wrapped with high purity Pt (99.995%). After Pt deposition the sample is annealed at 1050 (± 25) K for 10 min. and then cooled down to room temperature before placing it into the STM for observation. We observed the formation of two types of terraces: the so-called α -terraces, which contain a high density of missing dimer defects, and the so-called β -terraces, which are proposed to consist of both Pt and Ge atoms. Only on the latter type of terraces did we observe the formation of one-dimensional, kink free and virtually defect free atomic chains.

3. Results and discussion

Pt is deposited on the Ge(001) surface at room temperature and the self-organization only takes place upon annealing at temperatures that exceed 1050 K. At room temperature the Pt atoms dive into the germanium substrate. This process

goes along with the formation of missing dimer defects in the outermost layer of the Ge(001) surface as well as with the ejection of Ge atoms onto the terrace. Upon mild annealing at temperatures around 400–500 K these ejected Ge atoms rearrange themselves into small epitaxial Ge islands that eventually annihilate at pre-existing steps at higher annealing temperatures. Annealing at temperatures that exceed 1050 K leads to a very intriguing surface structure. Besides α -terraces that resemble highly defected, but otherwise quite regular, Ge(001) terraces rather well, another type of terrace is also formed. These newly formed terraces, referred to as β -terraces, are also comprised of dimers and dimer rows. However, the dimer rows deviate from the regular Ge dimer rows in various ways. First, the structure seems to suggest that not all of the dimers within a dimer row are pure Ge dimers. Second, the missing dimer defects tend to organize themselves into missing dimer chains that are aligned along $\langle 310 \rangle$ directions. Third, on these Pt modified terraces one frequently finds chains of atoms that are located within the troughs of the substrate dimer rows. The atoms that form these chains dimerize, leading to a $2 \times$ periodicity along the chain direction. The chain dimers are initially oriented parallel to the substrate dimers. However, upon surpassing a critical local coverage and critical temperature the dimers rotate by 90° , resulting in a zipper-like formation of the Pt chains [31]. The self-organization of the dimers into well aligned and kink free chains is guided by the troughs between the substrate dimer rows. Interestingly, neighbouring troughs are always empty. We believe that this is due to the fact that a configuration of two filled adjacent troughs causes too much surface stress.

In figure 1(a) a scanning tunnelling microscopy (STM) image of the Pt modified Ge(001) surface is shown. In the upper part of the image a large β -terrace can be observed, whereas in the lower right two small α -terraces separated by a monatomic step are present. The dimerized atomic chains (see figure 1(b)) are exclusively found on the β -terraces. The majority of these chains are found in well ordered patches; however, many isolated atomic chains are observed as well (see figure 2). The α -terraces resemble the regular Ge(001) terraces very well. It should however be pointed out that the concentration of missing dimer defects is very high. Most of these missing dimer defects occur in so-called $2+1$ complexes, i.e. two missing dimer defects, followed by a regular dimer and another missing dimer defect. It is common knowledge that these $2+1$ missing dimer defects arise due to the presence of subsurface metal (in this case probably Pt) atoms [32–34]. On the α -terraces we occasionally found well oriented Ge pyramids (see figure 2 for an example).

In figure 3 an STM image of a chain free β -terrace is shown. The dimer rows consist of two types of dimers. Gurlu *et al* [4] proposed that one of the two types of dimers is a regular Ge–Ge dimer, whereas the other type is a mixed Pt–Ge dimer. The two types of dimers alternate within a dimer row and the ordering registry of an adjacent dimer row is always out of phase. The mixed dimer has a rather small buckling angle ($< 10^\circ$) and theoretical calculations suggest that the Pt atom buckles inwards and the Ge atom outwards [25, 35]. The regular Ge–Ge dimer has a buckling angle of about 15° – 20° .

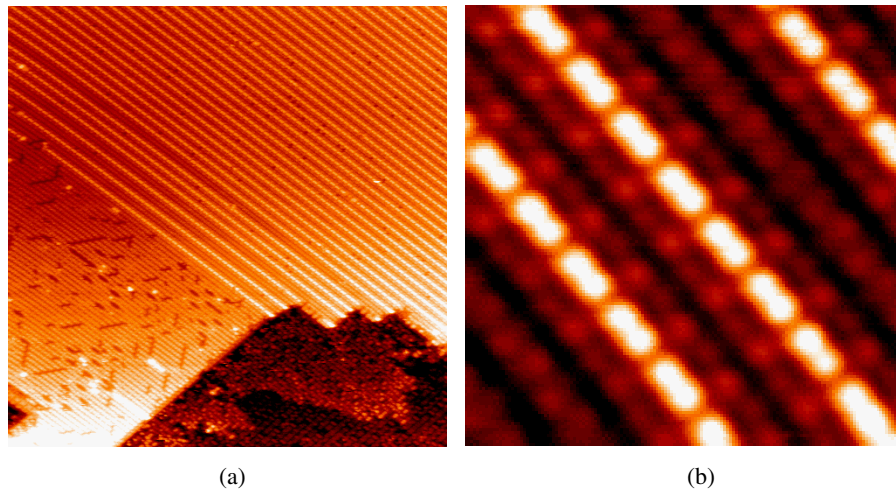


Figure 1. (a) STM image ($80 \text{ nm} \times 80 \text{ nm}$) of a Pt modified Ge(001) surface. Two types of terraces, the so-called α -terraces (lower right) and β -terraces (upper part). The atomic chains are exclusively found on the β -terraces. $V = -1.5 \text{ V}$. (b) STM image ($8 \text{ nm} \times 8 \text{ nm}$) of a β -terrace containing a few dimerized atomic chains. $V = -1.5 \text{ V}$.

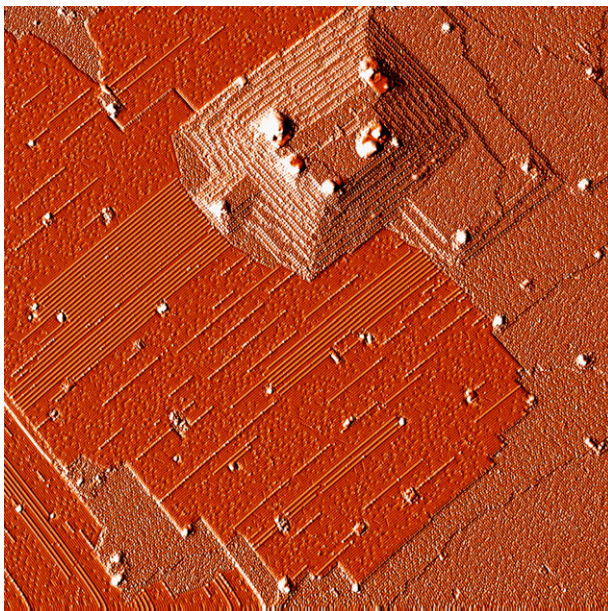


Figure 2. Large scale STM image ($250 \text{ nm} \times 250 \text{ nm}$) showing the two types of terraces and a well ordered Ge pyramid.

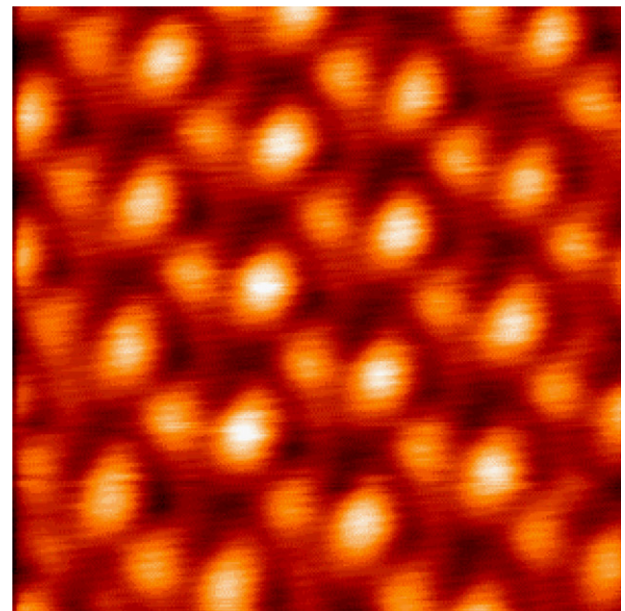


Figure 3. STM image of a β -terrace taken at $T = 80 \text{ K}$. The dimer rows consist of two types of dimers. Adjacent dimer rows are always out of phase. Scan size $4 \text{ nm} \times 4 \text{ nm}$, sample bias is -0.3 V and tunnel current is 0.5 nA .

Within the framework of Gurlu's model it is easy to understand that missing dimers prefer to align along (310) directions. Moreover this model would also explain why atomic chains that are separated 1.6 nm apart are always in phase and atomic chains that are separated 2.4 nm apart are always out of phase. The latter is in agreement with available STM data. In figure 4 an STM image of a β -terrace covered with four atomic chains recorded at room temperature is depicted. The chains that are spaced 2.4 nm apart are out of phase and the two atomic chains that are spaced 1.6 nm apart are in phase. The buckling angle of the substrate dimers at the left side of the atomic chains is slightly larger than the buckling angle of the dimers at the right side of the atomic chain. This asymmetry is due to the ordering

of the dimers of the underlying terrace. On one side of the atomic chain (in between the dimers of the chains) one has a mixed Pt-Ge dimer that is only slightly buckled, whereas on the other side one has a more strongly buckled, regular, Ge-Ge dimer.

In figure 5 a series of STM images of a β -terrace covered with two atomic chains recorded at 4.7 K at various sample biases in the range from -1.5 to 1.5 V is shown. The atomic chains run from the bottom to the top of the images. The atomic chains are dimerized and visible as two separate protrusions (red/yellow) in the voltage range from -1.5 to $+0.1 \text{ V}$. For voltages in the range from $+0.3$ to $+1.5 \text{ V}$ the

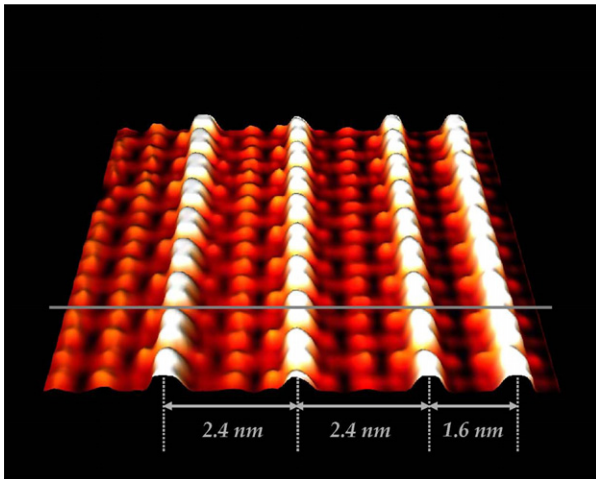


Figure 4. STM image (size 8 nm \times 7 nm) of several atomic chains on a Pt modified Ge(001) terrace recorded at room temperature. The atoms within the chains are dimerized. Without exception the dimers are always in phase with the dimers in the neighbouring chains if the spacing is 1.6 nm and they are always out of phase with their neighbours if the spacing is 2.4 nm. Sample bias -1.35 V and tunnelling current is 0.54 A.

dimers appear as single elongated protrusions (yellow). The observed bias dependence is just the opposite of what one would intuitively expect for a regular adsorbed Ge dimer on

a Ge(001) surface, namely one single elongated protrusion located in between the atoms at a negative sample bias and two separate protrusions located on top of the atoms at positive sample bias [26, 27]. It should be pointed out that the left chain exhibits a $2\times$ periodicity since it is located at the edge of an array of chains. The right chain (negative sample biases) exhibits a $4\times$ periodicity which is only visible for negative sample biases in the range from -1.5 to -0.7 V.

Scanning tunnelling spectroscopy experiments show that these atomic chains are metallic at room temperature [4], but become less metallic at low temperatures. In addition, upon cooling down to temperatures of 4.7 K the periodicity of the atomic chains doubles from a $2\times$ periodicity to a $4\times$ periodicity [20]. This $4\times$ to $2\times$ transition is only observed for atomic chains that are located in a patch of atomic chains. Isolated atomic chains or chains that are located at the edge of a patch maintain their $2\times$ periodicity; see figure 6. This discrepancy can, however, easily be understood if we interpret the transition in terms of a Peierls distortion. An ideal one-dimensional atomic chain has a Peierls transition temperature of 0 K. An additional coupling with the substrate or with neighbouring chains will increase the Peierls transition temperature. Therefore, we anticipate that the isolated chains or chains located at the edge of the patch have a Peierls transition temperature lower than 4.7 K, whereas the atomic chains within a patch have a Peierls transition temperature

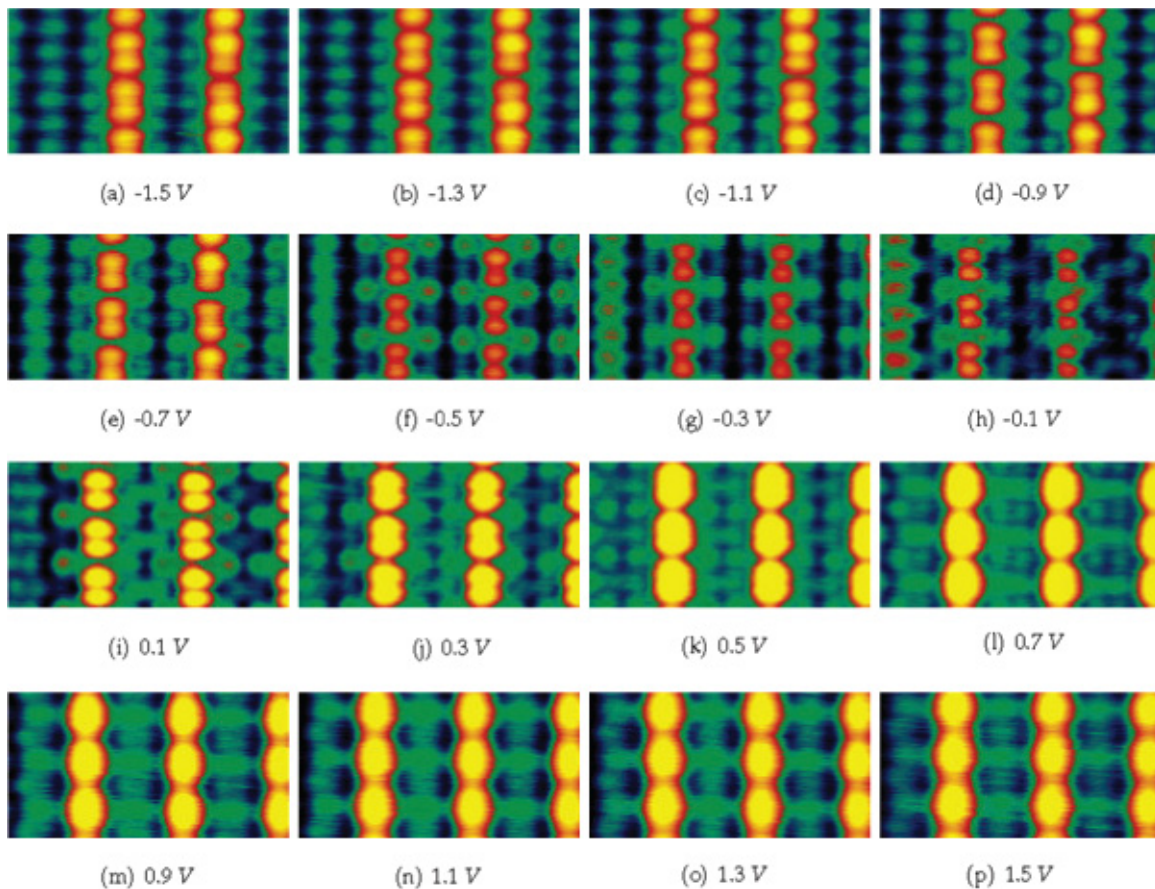


Figure 5. Series of STM images recorded at voltages ranging from (a) -1.5 V to (p) 1.5 V from the same area (size 2.4 nm \times 4.5 nm). The tunnelling current is 0.5 nA and the images are recorded at 4.7 K. The colour scale is the same for all images (0 nm is black, 0.12 nm is yellow). The yellow/red dimerized chains are the atomic chains.

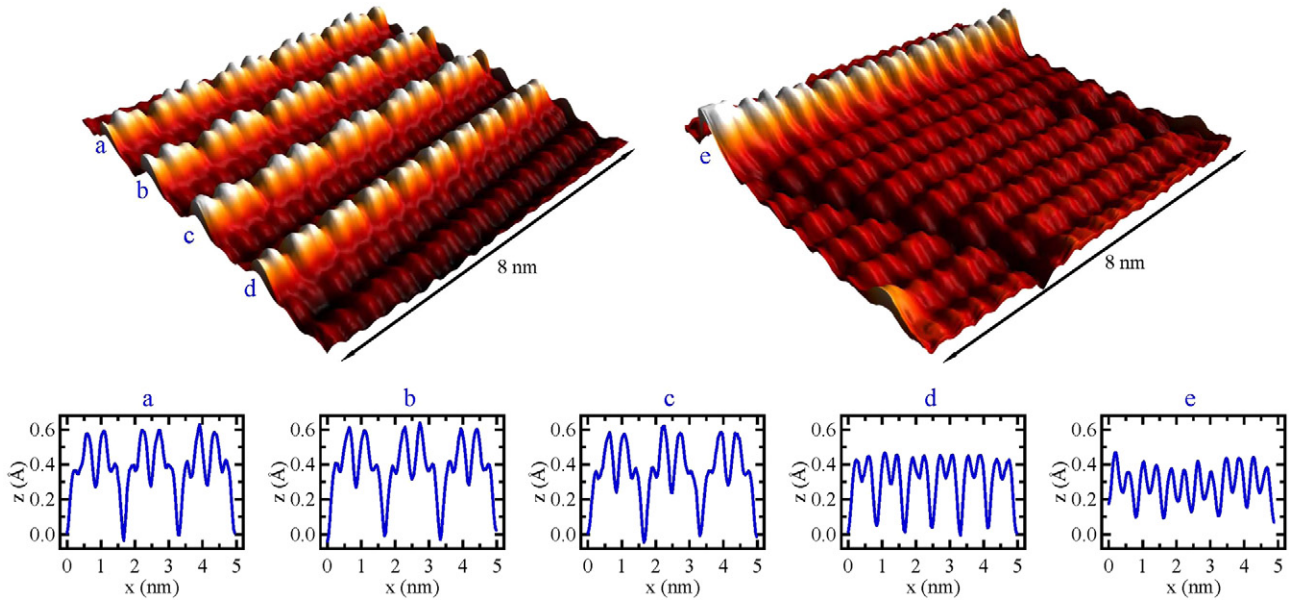


Figure 6. STM images recorded at 4.7 K. The atomic chains in the middle of a patch exhibit a 4× periodicity, whereas chains at the edge of a patch (left image) or isolated chains (right image) maintain their 2× periodicity.

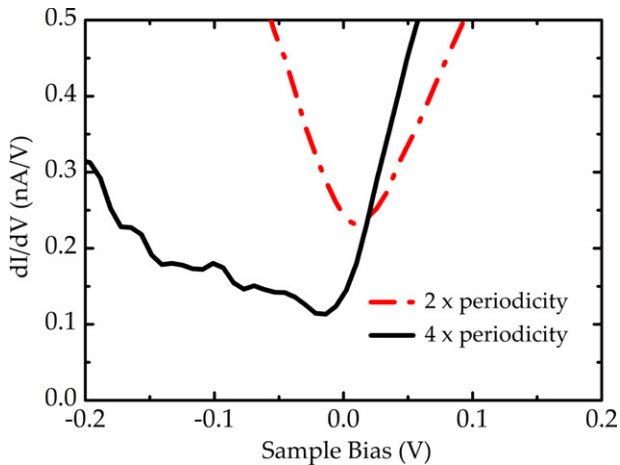


Figure 7. dI/dV spectra recorded on top of 2× and 4× atomic chains. The scanning tunnelling spectroscopy data are recorded with the same STM tip at a temperature of 4.7 K. Set points: sample bias −0.7 V and current 0.5 nA.

higher than 4.7 K. It should be noted here that the 4× buckling registry of the dimers in neighbouring 4× chains is always nicely in phase. The latter is indicative of a chain–chain coupling that is mediated by the substrate. The coexistence of atomic chains with 2× and 4× periodicities has a great advantage: namely, at the same temperature and with the same scanning tunnelling microscope tip the electronic properties of both types of chains can be measured simultaneously. IV spectra recorded at 4.7 K reveal that the 2× chains are more metallic than the 4× chains (see figure 7). The non-zero value of dI/dV at the Fermi edge of the 4× chains is most likely due to the underlying β -terrace, which is slightly metallic too. The reduced metallicity of the 4× chains versus the 2× chains is confirmed by measurements of the inverse decay length of

the atomic chains [20]. The inverse decay length at the Fermi edge of a 4× chain is significantly larger than the inverse decay length at the Fermi edge of a 2× chain [20]. The higher inverse decay length of the 4× chains is indicative of the fact that the tunnelling electrons are collected from the nearby troughs rather than from the chains. For more details the interested reader is referred to ref. [20]. Finally, as will be discussed below, the increased value of dI/dV of the 4× chains just above the Fermi level is due to the presence of an empty one-dimensional electronic state in the troughs between the 4× chains [5, 36].

Another intriguing observation is the development of quasi-one-dimensional electronic states at low temperatures that are located in the troughs between the chains [5]. In figure 8 the local densities of states of arrays of atomic chains with a nearest neighbour spacing of 1.6 and 2.4 nm, respectively, are depicted. The $n = 1$ peak for a nearest neighbour chain spacing of 1.6 nm has a strong asymmetric appearance, characteristic of a one-dimensional electronic state (van Hove singularity) and lies about 90 meV above the Fermi level. For a nearest neighbour chain spacing of 2.4 nm two empty electronic states, lying about 40 and 160 mV above the Fermi level, are found. We suggest that these novel quasi-one-dimensional electronic states arise due to the confinement of a 2D electronic state of the underlying β -terrace that is located just below the Fermi level (see the lower curve of figure 8). As can be observed in figure 8, this 2D electronic state slightly shifts towards the Fermi level in chain decorated regions of the β -terrace. The relative energy position of the three confined states with respect to this 2D electronic state agrees very well with the additional confinement energy (E_c), which is given by

$$E_c = \frac{\hbar^2 \pi^2 n^2}{2m_e L^2} \quad (1)$$

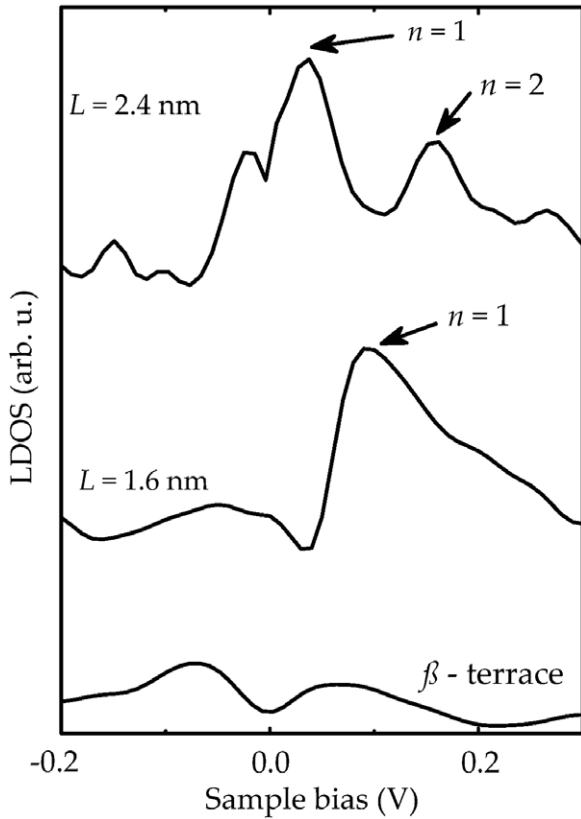


Figure 8. Local density of states (LDOS) measured in the troughs between the atomic chains at 77 K. Lower curve: LDOS of the underlying β -terrace. Middle curve: LDOS recorded in a trough with a nearest neighbour chain spacing of 1.6 nm. Upper curve: LDOS recorded in a trough with a nearest neighbour chain spacing of 2.4 nm.

where m_e is the rest mass of an electron, L the spacing between neighbouring atomic chains and \hbar Planck's constant. Since the

potential well is not infinitely deep, the energy values should be renormalized. In [5] it is shown that the experimental data can be explained well for an effective well depth of about 210 meV and an effective mass of $0.6m_e$. Spatial maps of these electronic states show that the $n = 1$ states exhibit a maximum in the middle of the trough, whereas the $n = 2$ state exhibits a node in the middle of the trough (see figure 9). These observations are consistent with the simple 'quantum mechanical particle in a box' model that has been put forward in order to interpret the data [36]. The fact that the confined electronic states are only found between the atomic chains that exhibit a $4\times$ periodicity suggests that the Peierls transition and the development of the one-dimensional electronic states are in some way related to each other. In addition, the electron density displays an oscillatory behaviour with a wavelength of 0.8 nm along the chain direction. This oscillatory behaviour is a direct consequence of the Bloch nature of the electronic states in the trough.

4. Conclusions

The physical properties of self-lacing atomic chains on the Pt modified Ge(001) surface have been studied with low-temperature scanning tunnelling microscopy and spectroscopy. At room temperature the atomic chains are metallic and exhibit a $2\times$ periodicity. Upon cooling down the atomic chains undergo a doubling of the periodicity. Simultaneously with the doubling of the periodicity the metallicity of the atomic chains reduces drastically. Based on these observations we suggest that the atomic chains undergo a Peierls transition. Along with this transition we found the development of one-dimensional electronic states that are located in the troughs between the atomic chains. These novel electronic states arise due to the confinement of a 2D electronic state of the underlying β -terrace that is located just below the Fermi level.

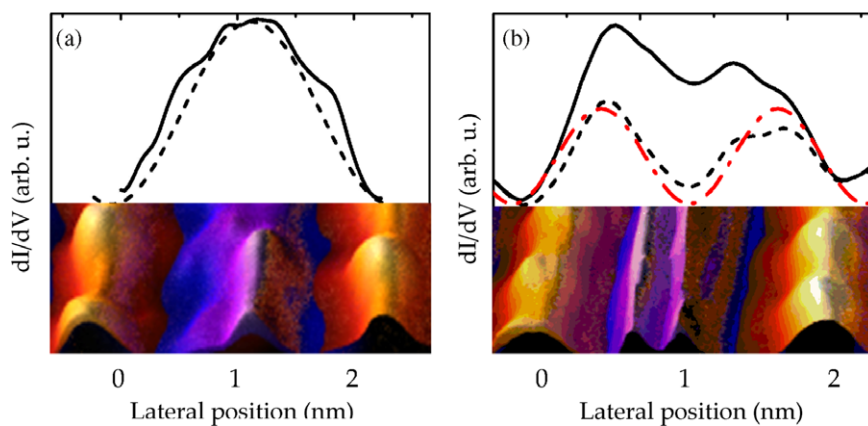


Figure 9. (a) Spatially averaged cross section of the differential conductivity (dI/dV) between the atomic chains with an interwire distance of 2.4 nm recorded with a sample bias of 40 mV, which corresponds to the $n = 1$ state (solid line). The dashed line shows a representation of the expected squared wavefunction $|\psi_1(x)|^2$. In the STM image, the topography (orange) and spatial map of the differential conductivity (dI/dV) (purple) of the same area taken at a sample bias of 40 mV are shown. (b) Spatially averaged cross section of the differential conductivity between the atomic chains with an interwire distance of 2.4 nm recorded with a sample bias of 160 meV, which corresponds to the $n = 2$ state (solid line). The dashed line shows a corrected distribution, in which the contribution of the tail of the $n = 1$ state is subtracted from the measured distribution. The red dashed-dotted line shows a representation of the expected squared wavefunction $|\psi_2(x)|^2$. In the STM image, the topography (orange) and spatial map of the differential conductivity (dI/dV) (purple) of the same area taken at a sample bias of 160 mV are shown.

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