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Controlled damaging and repair of self-organized nanostructures by atom manipulation at room temperature

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Abstract
The possibility of controlled local demolition and repair of the recently discovered self-organized Pt nanowires on Ge(001) surfaces has been explored. These nanowires are composed of Pt dimers, which are found to be rather weakly bound to the underlying substrate. Using this property, we demonstrate the possibility of carrying the constituting dimers of the Pt nanowires from point to point with atomic precision at room temperature. Pt dimers can be picked-up in two configurations: (i) a horizontal configuration at the tip apex, resulting in double tip images and (ii) a configuration where the Pt dimer is attached to the side of the tip apex, resulting in well-defined atomically resolved images.

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

Since Eigler and Schweizer demonstrated the possibility of manipulating the position of Xe atoms on a Ni(110) surface [1] there have been several attempts to identify other systems allowing the creation of nanostructures in an atom-by-atom fashion. A number of gas/metal and metal/metal systems have been discovered and used in the construction of structures like quantum corrals [2], molecular cascades [3], other nanostructures or just writing names or symbols on surfaces [4, 5]. Most of these systems are stable only at cryogenic temperatures. However, several molecular structures have also been produced on surfaces in manipulation experiments at room temperature (RT) along with low temperature experiments [6–8].

So far no reports have been published dealing with the manipulation of self-organized nanostructures: for instance, the controlled creation and repair of defects in such structures. Manipulation of such self-organized nanostructures is by no means self-evident. The self-organization is obviously driven by the minimization of the total free energy of the system. The taking away of building blocks may therefore lead to a (partial or local) collapse of the structure. Indeed we have obtained evidence for such behavior in the present study.

There are several methods through which atoms can be manipulated on surfaces at low temperatures using a scanning tunneling microscope (STM). First is the dragging of atoms on the surface using the STM tip [1]. Several more examples followed this method using other adsorbate/surface systems [4, 9]. Another method is picking up the atoms/molecules from the surface using the STM tip and placing them back onto the surface [10]. Prior to these demonstrations, local manipulation at the atomic scale was also reported on semiconductor surfaces [11–15]. One of the rare successful attempts to manipulate atoms with atomic precision on a metal surface at room temperature (RT) was reported by Fishlock *et al* [16] for the case of Br atoms on Cu(001).

Since molecules or atoms such as CO or Xe desorb from surfaces at elevated temperatures, cryogenic temperatures are a necessity for the precise positioning of the atoms/molecules

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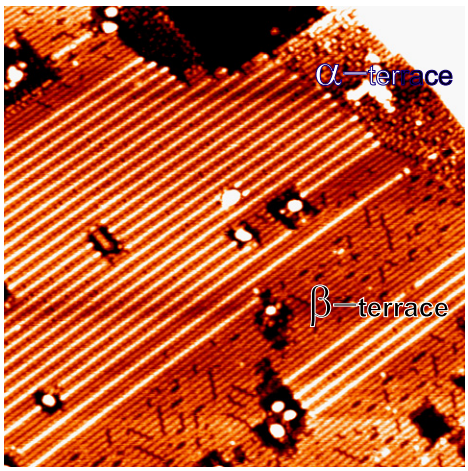

Figure 1. STM image of a patch of Pt nanowires (taken with −1.5 V sample bias and 0.4 nA tunneling current; size 50 × 50 nm^2). The nanowires are observed only on β terraces. In the upper part of the image an α terrace is visible. See [15] for a detailed description of the α and β terraces and their evolution.

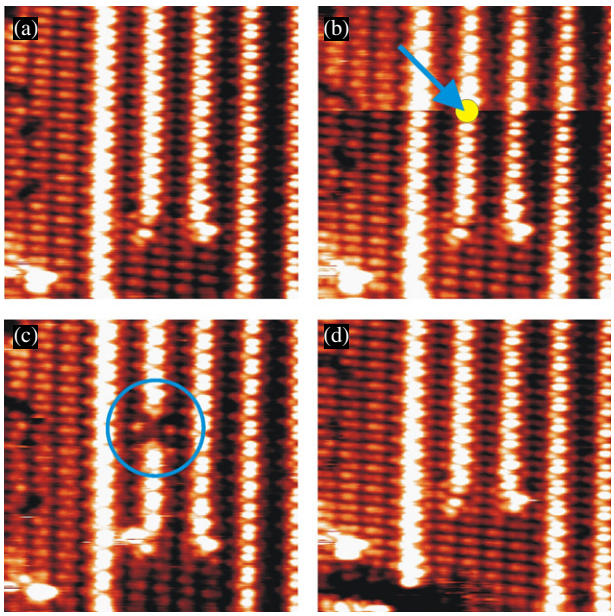

Figure 2. (a) Five Pt nanowires on an β terrace before manipulation (20 mV, 0.4 nA); (b) during scan, the pecking event is applied on the designated point; (c) rescan of the area after the pecking event; the missing nanowire piece is visible as a dent; (d) rescan of the same area after the application of a second pecking event on the dented point.

and keeping them fixed on the surface. Metal-on-metal systems, including Co or Fe on Cu(111) also require low temperatures due to the high diffusivity of the adatoms on the surfaces. Studies so far clearly demonstrate the possibility of positioning the atoms/molecules at atomically precise positions on the surfaces at cryogenic temperatures. At low temperatures, the low diffusion barrier of atoms on surfaces would present a finite lifetime problem of the structures created atom by atom at elevated temperatures. Several semiconductor/semiconductor systems seem suitable candidates for atom manipulation at room temperature. However, even though Si or Ge dimers are quite stable on Si(001) or Ge(001) surfaces they still possess low diffusion barriers. This makes them hard to fix on certain positions [17, 18]. For this reason we choose to address STM-induced creation and repair of defects in self-organized Pt nanowires (NWs) on Ge(001).

We demonstrate that the individual Pt dimers of Pt nanowires grown on Ge(001) surfaces [19] can individually be manipulated on the surface using the STM tip at room temperature. Prior to the preparation of the Pt nanowires, the Ge(001) surfaces were cleaned by several cycles of annealing at 1100 K and argon ion sputtering [20]. Subsequently, we deposited about half a monolayer of Pt at room temperature. The Pt nanowires were formed after a 10 min anneal at 1050–1100 K.

Figure 1 shows an STM image of a Ge(001) surface with Pt nanowires. These wires are one atom wide and basically run from surface defect to surface defect. On the extended flat parts of the surface the Pt nanowires can be as long as several hundreds of nanometers. They typically occur in domains with 1.6 nm separation between neighboring wires. Note that figure 1 is deliberately chosen to demonstrate the effect of defects, primarily the steps on the surface. The preparation and general properties of single-atom thick, thousands of atoms long and defect-free Pt nanowires on Ge(001) surfaces are described in detail in [19].

During constant current STM imaging, at different sample biases, we have observed occasionally that pieces of nanowires can be kicked off the string of atoms while testing very low voltages. This indicates that the building blocks of the Pt chains are not so strongly bound to the substrate, since there were no visible defects on the underlying terraces after the kick-off events. Sometimes such an event led to a major distortion of the wires. It appeared that these linear structures collapsed over an extended length.

Consequently, we have tested a controlled way of manipulating these Pt nanowires. The procedure applied is as follows: first, a normal STM scan is taken on a patch of Pt NWs (figure 2(a)) using 0.4 nA tunneling current and −20 mV sample bias. During the second scan of the same area the following routine is applied to the STM tip: the STM tip is positioned on a designated point above a nanowire, the tunneling current is increased to 20 nA and after a very short delay (about 1 ms) the bias voltage is decreased to −2 mV and immediately set back again to −20 mV. Again after a short delay, the set point for the tunnel current is reduced back to 0.4 nA and the scan is continued. The effect of this quite short procedure on the STM image is visible in figure 2(b). The next scan of the same area with the same scan parameters reveals the absence of one Pt dimer of the Pt chain (figure 2(c)). Since the tip acts like a bird's beak we refer to this procedure as 'pecking'. During the pecking, the tunnel resistance of the tip–wire junction is reduced to only 100 kΩ, for which it is known that a jump-to-contact can occur, see, for example, [21–23]. After this event we tried to peck the same point with the same tip and the result is shown in the following scan after the re-pecking (figure 2(d)). This clearly indicates that the picked up piece of the Pt chain has been carried on the tip. In other words

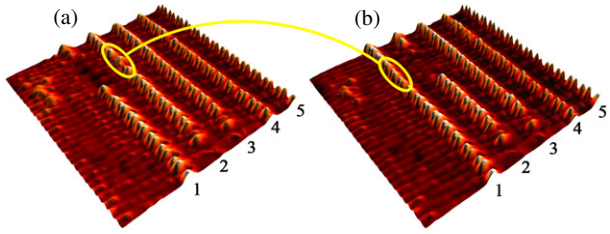

Figure 3. An array of 5 Pt nanowires on a Ge(001) surface. (a) Nanowire 1 has a defect which consists of four missing Pt dimers (upper left). (b) Four individual Pt dimers are picked up from nanowire 2 and deposited in the defect of nanowire 1. The dimers are picked up and deposited in a one-by-one fashion.

we have been able to generate a Pt (dimer) vacancy within the nanowires and repair it in the next pass by dropping it from the STM tip.

It should be pointed out here that the apex of our W tip is probably covered with Pt atoms due to the many pick-up–carry–drop processes. The latter probably also explains why it is relatively easy to pick up a Pt dimer from a Pt nanowire.

In figure 3 an array of five Pt nanowires is shown. In the upper left part of this image four Pt dimers are missing. Subsequently, we have picked up Pt dimers in a one-by-one fashion from nanowire 2 and placed these Pt dimers in the large defect site of nanowire 1. Between every carry-and-drop step the area of interest is scanned at least once to make sure that the structures formed are stable. The success rate of this pick-up–carry–drop process is almost 100%. The symmetry of the pick-up and drop events is most probably related to the fact that the only stable adsorption site for the Pt dimer is in the trough between the substrate dimer rows.

Next we present the effect of many single dimer pick-up events on the STM tip and its resolution. Here we have applied a slightly different manipulation method. The method we applied for the second part of this paper is described in detail by Lyo and Avouris in [11]. On a designated point above a nanowire the feedback loop is opened, the tip is approached to the surface and subsequently a voltage pulse (−3.0 V) is given, the tip is retracted and the feedback loop is closed again. The main difference with the earlier described method is the opening of the feedback loop during the pick-up experiment.

We believe that the Pt dimer can be picked up in two different configurations at the apex of the tip, labeled configurations I and II.

Upon picking up a Pt dimer we frequently observe double tip images. We believe that in these cases the Pt dimer is attached to the apex of the tip in a horizontal position (configuration I, see figure 4(a)). In figure 4(b) we placed a Pt dimer from the tip apex back to the surface. The Pt dimer is attached horizontally at the tip apex (configuration I), resulting in a double tip image in the lower part of figure 4(b). After dropping the Pt dimer, the resolution changes back to an atomically sharp one (upper half of figure 4(b)).

Many other single Pt dimer pick-up events do not lead to double tip effects and therefore we believe that in these cases the Pt dimer is attached to the side of the tip apex (configuration II). Imaging with such a Pt dimer modified tip results in atomically sharp images. In figure 4(c) we have picked up

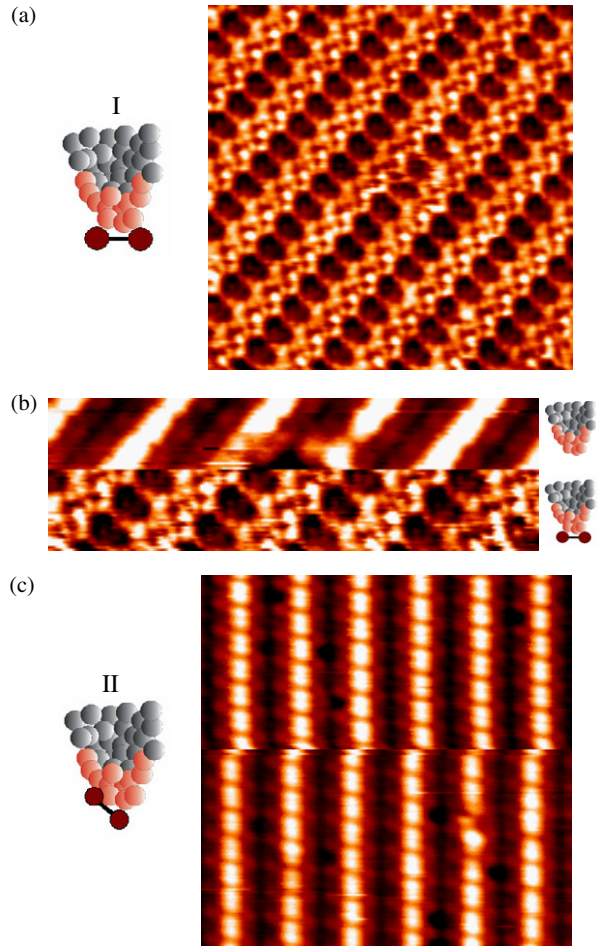

Figure 4. (a) STM image (10 × 10 nm) recorded on an array of Pt nanowires just after a single dimer pick-up event, resulting in a double tip image (configuration I). (b) STM image (10 × 3 nm) in which a Pt dimer is dropped from the STM tip during imaging. The lower half of the image is scanned with a double tip (Pt dimer at the tip apex in configuration I), while the upper half of the image, after the dropping event, is again atomically sharp. (c) STM image (10 × 10 nm) recorded on an array of Pt nanowires. In the middle of the image a single dimer is picked up from one of the Pt chains. The resolution of the STM image remains essentially unaltered (configuration II).

a single Pt dimer in the middle of the image (scanning from bottom to top). As is shown in figure 4(c) the resolution remains essentially unaltered after the pick-up event.

Combining all knowledge from the Pt nanowire manipulation experiments we can elaborate on many possibilities for scientific innovations. We may just mention the possibility of the controlled creation of caves of empty spaces within ordered domains of Pt nanowires. This could lead to unexplored electron confinement features [24, 25].

This manipulation method is also applicable to other systems provided that: (i) the binding of the elementary building block to the underlying substrate is weak enough, (ii) upon removing an elementary building block the self-organized nanostructure remains stable.

Summarizing, we have shown that it is possible to manipulate self-organized Pt nanowire arrays on Ge(001) by means of STM. Pt dimers can be picked up and released at a

predefined position with atomic precision at room temperature. The latter allows creating or repairing defects in Pt nanowires and thus opening the possibility to study in a controllable way the effects of defects in a one-dimensional electronic system [17, 18]. A Pt dimer that has been picked up by the tip can be in two stable configurations: a horizontal one at the tip apex and a more diagonal one at the side of the tip apex.

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