Applied Physics Express 4 (2011) 085002
DOI: 10.1143/APEX.4.085002

Assembling Indium Atoms into Nanostructures on a Cleaved InAs(110) Surface

Kyoichi Suzuki*, Stefan Fölsch[1], and Kiyoshi Kanisawa

NTT Basic Research Laboratories, NTT Corporation, Atsugi, Kanagawa 243-0198, Japan
[1]*Paul-Drude-Institut für Festkörperelektronik, 10117 Berlin, Germany*

Received June 8, 2011; accepted June 30, 2011; published online July 20, 2011

Single-atom manipulation of indium adatoms was achieved on a cleaved InAs(110) surface in a scanning tunneling microscope (STM) at low temperature. In addition to vertical manipulation by reversible STM tip–sample transfer of atoms, lateral manipulation by means of tip-induced atom hopping was achieved with an anisotropic tendency preferencially along the [001] in-plane direction by sweeping the sample bias voltage on the target adatom. Scanning tunneling spectroscopy of the adatoms assembled in a line was also achieved with an optimized tip–sample distance. The results show the molecular-like behavior of interacting adatoms.

As semiconductor devices become more advanced and more highly integrated, further miniaturization and further reduction of power consumption are desired. Atomic-scale devices are the ultimate ones for meeting these demands. In addition to the scale merit, new functions are expected from quantum mechanical effects as well. A promising way to construct these devices is to use a scanning probe microscope system. Up to now, several groups have succeeded in manipulating a single atom at a time in/on semiconductor surfaces.[1–5] Regarding atomic-scale structures "on" a semiconductor surface, vertical manipulation and repositioning of adatoms by means of reversible tip–surface transfer have been achieved on an InAs(111)A epitaxial surface with a low-temperature scanning tunneling microscope (LT-STM).[1] To make precise structures, the ability to perform not only vertical manipulation, but also lateral manipulation would provide more flexibility. However, lateral manipulation has not been achieved on the InAs(111)A surface because the adatom is significantly bound to the potential pocket originating from the surface reconstruction.

Here, we investigate the availability of adatom manipulation, including lateral manipulation, on an InAs(110) surface using LT-STM. Compared with other surfaces that have been successfully used for atom manipulation, a clean InAs(110) surface has certain features, such as a moderate surface reconstruction, due to unpolarized characteristics composed of atomic rows of both cation (In) and anion (As), and no surface Fermi level pinning. The biggest advantage is that a wide and flat area of over ten micrometers square without any atomic steps can be easily obtained simply by cleaving in an ultrahigh vacuum (UHV).[6] In other words, complex preparation of the surface, such as by epitaxy, evaporation, or deposition, is not necessary before the atom manipulation.

A piece of a 400-μm-thick commercially available undoped InAs(001) substrate was loaded into a UHV chamber at 10^{-10} Torr and cleaved with tweezers to obtain a (110) surface. Then, the sample was transferred to the LT-STM setup that had been cooled to 4.8 K in UHV on the order of 10^{-11} Torr without breaking the UHV condition. The details of the experimental setup and preparation are same as those in ref. 6. Electrochemically etched tungsten wire was used as the STM tip. During the *in-situ* tip preparation by applying a voltage pulse or by bringing the tip into contact with the sample surface far enough away from the measurement place, the tip is terminated with In atoms. Therefore, we can start to transfer In atoms one-by-one from the tip to the surface despite the fact that no native In adatoms are present at the cleaved surface.

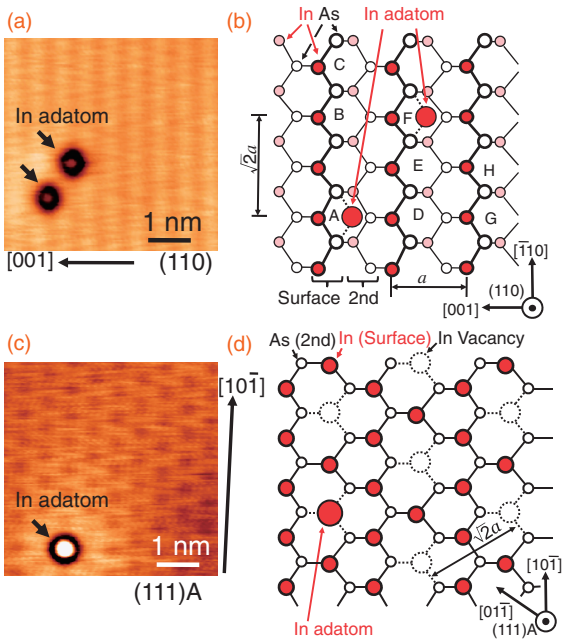

Fig. 1. STM images of In adatoms on (a) InAs(110) surface (sample bias $V = -1.0$ V, feed back tunneling current $I = 200$ pA) and (c) InAs(111)A surface (+0.1 V, 100 pA). The different apparent sizes of the In adatoms are caused by the different V. (b) and (d) Corresponding stick-and-ball models. a is the lattice constant of InAs. Dotted lines indicate dangling bonds and the existence of ionic-bonds with nearest neighbor atoms.

First, we describe the observed positions of In adatoms on the (110) and (111)A surfaces. Figure 1 shows STM images of (110) and (111)A surfaces with In adatoms [(a) and (c)] and corresponding stick-and-ball models [(b) and (d)], respectively. On the (110) surface, rows composed of In and As atoms are regularly aligned without any vacancy sites. In Fig. 1(a), the rows of As atoms appear as bright rows at the sample bias voltage $V = -1.0$ V and feedback current $I = 200$ pA. The adatoms, which were positioned and made ionic-bonds to the adjacent As atoms, appear in constant-current imaging as a depression with a central protrusion. On the (111)A surface whose surface monolayer is composed of In, showing the polar characteristics of the surface, the adatom resides on the native In-vacancy site caused by the (2×2) surface reconstruction[7] and is

*E-mail address: suzuki.kyoichi@lab.ntt.co.jp

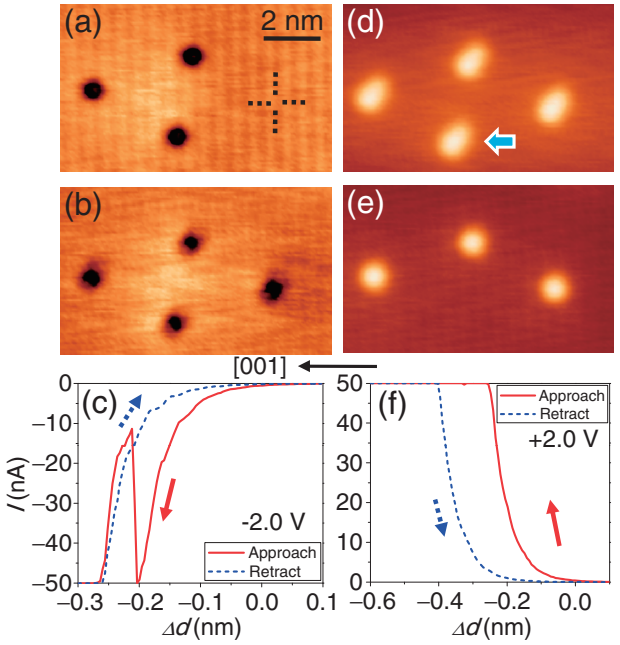

Fig. 2. STM images at the same position corresponding to the adatom putting-down process [(a) and (b)] (−1.0 V, 400 pA) and the picking-up process [(d) and (e)] (+2.0 V, 400 pA). I as a function of the change in the tip–sample distance (Δd) from the initial feedback distance for the putting-down process (c) and picking-up process (f).

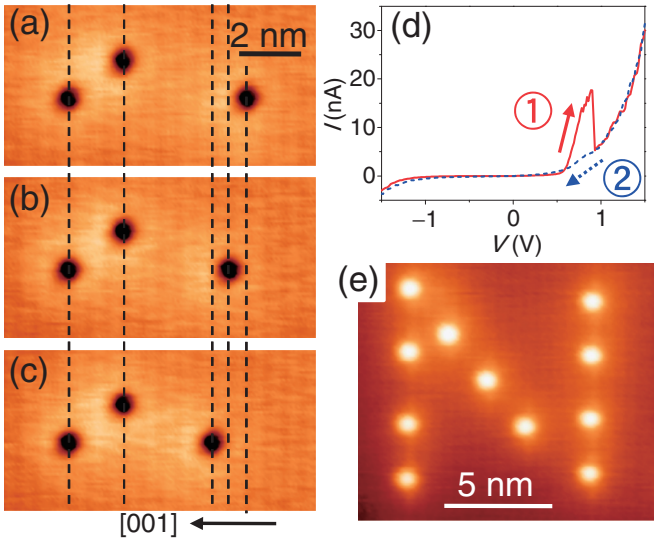

Fig. 3. (a)–(c) STM images corresponding to lateral manipulation (−1.0 V, 400 pA). Between the scanning images, V was swept above the target adatom on the right-hand side. (d) I as a function of V above the target adatom. (e) STM image (+1.0 V, 100 pA) of "N" made by eleven assembled adatoms.

stabilized by Coulomb interaction with the three As atoms at the vacancy [Figs. 1(c) and 1(d)]. For both the InAs(110)[8] and InAs(111)A[9] surfaces, the In adatoms are found to be positively charged.

For the InAs(110) surface, vertical manipulation by reversible tip–surface transfer requires procedures similar to those for the (111)A surface.[1] Figure 2 shows the vertical manipulation processes on the (110) surface. The STM image at −1.0 V and 200 pA is shown in Fig. 2(a). Three In adatoms on the surface appear as dark spots. At the marked position, the STM tip approached by 0.3 nm without feedback control from the initial tip–sample distance tuned by the feedback. As a result, one atom was transferred to the surface from the tip [Fig. 2(b)]. The typical tunneling current profile as a function of the change in the tip–sample distance (Δd) during this putting-down process is plotted in Fig. 2(c). With decreasing distance from the initial position, the absolute tunneling current increased (solid line). At $\Delta d = -0.2$ nm, the current suddenly drops, corresponding to the release of an atom from the tip to the surface. As the distance was subsequently increased from the minimum, the current monotonically decreased (dashed line).

Figure 2(d) shows an STM image (+2.0 V, 400 pA) at the same place after the putting-down process. In this sample bias condition, adatoms appear as bright spots. Above the adatom indicated by the arrow, the STM tip is approached by 0.6 nm from the initial distance. As a result, the adatom disappears, as evident from Fig. 2(e). The typical tunneling current profile as a function of Δd during the picking-up process is plotted in Fig. 2(f). Although the current overflowed due to the limitation of the amplifier, we believe that the tunneling current suddenly decreases when an adatom is removed from the surface. The decreased tunneling current indicates a larger tip–sample distance, meaning that the adatom adhered somewhere to the tip other than the top.

In addition to the vertical manipulation, we found that lateral manipulation is available on the (110) surface by sweeping V. Three adatoms are positioned, as shown in Fig. 3. The tip was fixed above the adatom shown on the right-hand side of image (a) and V was swept from −1.5 to +1.5 V and *vice versa* at the fixed initial distance at −1.0 V and 200 pA. As a result, the target atom hopped laterally by one row in the [001] direction, as shown in image (b). I as a function of V during the lateral manipulation process is plotted in Fig. 3(d). During the sweep to the positive maximum voltage, I increased and suddenly dropped at about +0.9 V, corresponding to the lateral motion of the target adatom. Mostly, the drop point in the voltage corresponding to the adatom hopping is independent of the tip–sample distance. This suggests that the hopping is caused by the inelastic tunneling excitation rather than the field effect.

Repeating this, the target adatom was moved step by step as shown in Figs. 3(a)–3(c). Such movement in the [00$\bar{1}$] direction has hardly been observed, attributed to inversion asymmetry on the surface. In some cases, this lateral movement was accompanied by some movement in the [1$\bar{1}$0] or [$\bar{1}$10] direction. Similar lateral movement was observed when the picking-up process failed at positive V. Using the vertical and lateral manipulation techniques, we assembled eleven In adatoms into an "N" on the InAs(110) surface within 1 h [Fig. 3(e)].

Although it seems to be possible to position adatoms at any appropriate site at first glance, there are some restrictions. If the first adatom is positioned at site A in Fig. 1(b) for example, the nearest and second nearest sites for the next adatom are expected to be B and D, respectively. However, the adhesivity of the adatom to the surface is weaker than that to the (111)A surface because of the lower

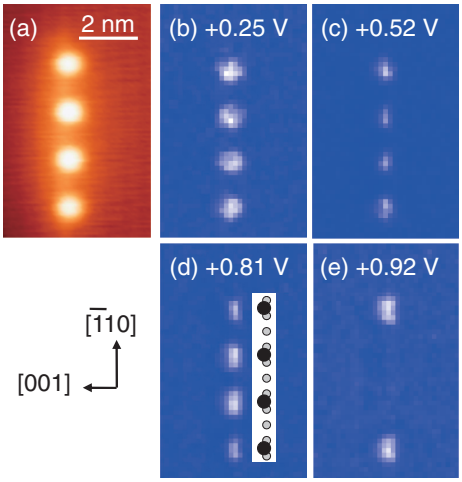

Fig. 4. (a) STM image of four assembled adatoms along [$\bar{1}$10] with an equal distance of $(3/\sqrt{2})a$ (+1.0 V, 200 pA). (b)–(e) $(dI/dV)/(I/V)$ maps at $V = +0.25$, +0.52, +0.81, and +0.92 V, respectively. Bright regions indicate high electron local density of states. The inset in (d) shows the corresponding alignment of assembled adatoms (black circles) and In atoms in the second atomic layer (gray circles).

ionic-bond strength and nonpolar characteristics of the (110) surface. The effect of repulsive Coulomb force between adatoms becomes relatively large. As a result, placing an adatom at site B or D is almost impossible. Even when the placement is successful, these adatoms, including the first one, are often unstable and disappear after several scans. On the basis of our experience so far, the nearest stable site is F and the distance between the adatoms is $\sqrt{3}a$, where a is the lattice constant of cubic InAs, about 0.605 nm. This is very different from the case of the (111)A surface, on which the second and more adatoms can be positioned at any vacancy site, including the adjacent site at the distance of $\sqrt{2}a$.[1)]

One of the features of STM is scanning tunneling spectroscopy (STS), with which we can investigate the spatial distribution of the electron local density of states (LDOS).[10)] The LDOS is obtained as the normalized differential conductance $[(dI/dV)/(I/V)]$ by sweeping V at fixed tip height and recording the dI/dV signal using the lock-in technique with the small voltage modulation (10 mV_{pp} was used here). Collecting these data for each pixel of a scanned surface area allows us to generate LDOS maps at a given energy eV relative to the Fermi level. A set point of +1.0 V and 200 pA, for example, is required to prevent unintentional lateral hopping of the adatoms when V is swept.

We demonstrate the results of STS on four adatoms assembled in a line along [$\bar{1}$10] with an equal distance of $(3/\sqrt{2})a$, shown in the STM image, Fig. 4(a). The LDOS maps at $V = +0.25$, +0.52, +0.81, and +0.92 V are shown in Figs. 4(b)–4(e), respectively. The bright regions indicate a high LDOS, corresponding to the high probability of the existence of electrons. The two LDOS maps in the lower energy region indicate the ground and second resonance energy states for the artificial molecule composed of four In adatoms. In Fig. 4(b), the LDOS is distributed to each adatom almost equally, but slightly concentrated at two adatoms in the middle. In contrast, in Fig. 4(c), the LDOS is mainly concentrated at the adatoms on both the upper and lower edges. This behavior is similar to that observed in the molecularly coupled adatom chain fabricated on the (111)A surface arising from atomic orbital states associated with the In adatom.[1)]

The two maps in the higher energy region [Figs. 4(d) and 4(e)] are the states with a different origin. We observe an anisotropic shape of each bright spot as compared to the uniform LDOS maxima in Figs. 4(b) and 4(c). We interpret this as an indication that the LDOS is concentrated on two In atoms in the second atomic layer [depicted as gray circles in the inset of Fig. 4(d)] below the assembled In adatoms (black circles). No enhanced LDOS appears on the second atomic layer In atoms without adatoms nearby. This indicates that, by the positively charged adatoms, delocalized surface states composed of the surface atoms are locally modified and the LDOS is confined in the adatom potential wells. In addition, these states below each adatom couple with each other, resulting in discrete quantization. The ground state is shown in Fig. 4(d). The two spots in the middle are brighter than the others. In contrast, the second state is shown in Fig. 4(e). Only the two outermost spots appear bright.

In conclusion, we investigated In adatom manipulation on a cleaved InAs(110) surface. We achieved not only vertical manipulation but also controlled lateral manipulation by means of tip-induced adatom hopping. By optimizing the sample bias voltage and feedback current, the STS is also available for adatoms without lateral movement. STS results show that the (110) surface is a good template for investigating quantum states in artificially assembled atomic-scale structures.

Acknowledgment This work was partly supported by a subsidy from the Strategic Japanese–German International Cooperative Program on Nanoelectronics supported by the Japan Science and Technology Agency (JST) and the German Research Foundation (DFG).

1) S. Fölsch, J. Yang, Ch. Nacci, and K. Kanisawa: Phys. Rev. Lett. **103** (2009) 096104.
2) Y. Sugimoto, P. Pou, O. Custance, P. Jelinek, M. Abe, R. Perez, and S. Morita: Science **322** (2008) 413.
3) I.-W. Lyo and Ph. Avouris: Science **253** (1991) 173.
4) G. Dujardin, A. Mayne, O. Robert, F. Rose, C. Joachim, and H. Tang: Phys. Rev. Lett. **80** (1998) 3085.
5) Y. J. Song, S. C. Erwin, G. M. Rutter, P. N. First, N. B. Zhitenev, and J. A. Stroscio: Nano Lett. **9** (2009) 4333.
6) K. Suzuki, K. Kanisawa, S. Perraud, M. Ueki, K. Takashina, and Y. Hirayama: J. Appl. Phys. **101** (2007) 081705.
7) A. Taguchi and K. Kanisawa: Appl. Surf. Sci. **252** (2006) 5263.
8) For example, in Fig. 3(e), the slightly brighter regions around the adatoms at positive V show the background LDOS on the (110) surface, indicating the adatoms charged positively.
9) J. Yang, S. C. Erwin, K. Kanisawa, Ch. Nacci, and S. Fölsch: Nano Lett. **11** (2011) 2486.
10) K. Suzuki, K. Kanisawa, C. Janer, S. Perraud, K. Takashina, T. Fujisawa, and Y. Hirayama: Phys. Rev. Lett. **98** (2007) 136802.