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Single atomic manipulation and writing with scanning tunnelling microscopy at low temperatures

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In the work reported in this paper, we have used a low-temperature scanning tunnelling microscope (LT-STM) system to manipulate accurately single atoms. We show how we can use a LT-STM to image and modify a bulk Ag(111) surface and manipulate Ag atoms from substrate and evaporated adsorbates on Ag(111) substrates. We present a synergistic combination of STM-induced modification and ordered arrays of nanometre-scale structures. In particular, we demonstrate the ability to modify Ag atomic nanometre structures on the Ag(111) substrate, and some English letters and a Chinese character can be written by single Ag atoms coming from the substrate and evaporated adsorbates on Ag(111). In this way, we supply an effective basis to explore the fundamental physical properties of a nanometre structure and to develop nanotechnology with a 'bottom-up' approach.

Keywords: low-temperature scanning tunnelling microscope, single atomic manipulation, artificial nanometre structure

PACC: 6116P, 6820

1. Introduction

At the turn of 21st century, nanotechnology has become the driving force in all the other technologies. As a promising technology possessing tremendous market potential, it has gained much attention all over the world. Especially in the electronics area, electronic components such as transistors on computer chips are becoming increasingly smaller, while their performance capabilities are growing. The concept of microelectronics is being replaced by 'nanoelectronics'.

To realize the nanotechnology, two approaches can be used: the 'bottom-up' and 'top-down' approaches. The 'top-down' approach means using mechanical and etching technologies to manufacture nanostructures; while the 'bottom-up' approach means creating organic and inorganic structures by arranging atoms or molecules, which need to artificially control atoms and molecules in structure, composition, shape, size and position, etc. The 'top-down' or 'bottom-up' techniques can judge the developing level of nanotechnology in this area. However, considering the expenses and accurate equipment required by the 'top-down' approach and the limited technology in the current state, the 'bottom-up' approach could become a more useful method in the future development of nanotechnology.

Using the 'bottom-up' method to fabricate nanometre-scale structures on a surface proves a tremendous challenge. This challenge has been approached from various directions. Studies using the scanning tunnelling microscope (STM) invented in 1982 by Binning and Rohrer[1] have demonstrated its ability not only in observing surface structures but also in assembling nanostructures by manipulating individual atoms and molecules at both cryogenic and room temperature.[2−6]

The STM can image the surface of conducting materials in atomic-scale detail. Like other microscopes, we can use the STM to extend our vision to a realm where our eyes cannot see. In a tunnelling microscope we conventionally record an image that is a map of the trajectory of a probe tip over a surface while the height of the probe tip is constantly adjusted to maintain a constant tunnelling current between the tip and the surface. Such images reflect both the topography and electronic structure of the surface. The lateral movement of individual adatoms and molecules in a controlled manner on solid surfaces and the construction of structures on a nanoscale were first demonstrated by Eigler and collaborators at 4K in 1990.[7]

Noble metal surface properties such as Ag(111) have been the focus of experimental interest for

two decades, from the initial observation via angle-resolved photoemission spectroscopy (ARPES) by Gartland *et al*,[8] through to recent studies of the defect scattering and lateral confinement on surface nanostructures via a STM by Crommie *et al*[9] in the USA and Braun and Rieder in Germany.[10] They have characterized the effective masses, Fermi wave vectors, surface-state and surface electron lifetime, and most of these results agree well with theoretical predictions. For the studies of surface properties, a nanometre-scaled artificial atomic structure, which cannot be found in nature, generally needs to be constructed by atomic and molecular manipulation using the STM at low temperatures.

In this study, we have used a home-made LT-STM system to accurately manipulate single atoms. The flexibility of the manipulation technology allows us to control accurately the position of atoms and, forming an artificial nanostructure, to explore their basic physical properties. We demonstrate how we can use an LT-STM to image and modify a bulk Ag(111) surface and to manipulate Ag atoms from the substrate and the evaporated adsorbates on Ag(111) substrates. We also demonstrate a synergistic combination of STM-induced modification and an ordered array of nanometre-scale structures. In particular, we demonstrate the ability to modify Ag atomic nanometre structures on the Ag(111) substrate, and some letters and a Chinese character have been written for the first time by single Ag atoms coming from the substrate and evaporated adsorbates on the Ag(111) substrate.

2. Experimental details

The home-made ultra-high vacuum (UHV) LT-STM is specially designed for working at an atomic scale at low temperatures with high stability and small drift. This system is equipped with a STM and a cold finger stage, and the base pressure is less than 5×10^{-13} Pa. Its temperature can be varied between 6–300 K. Due to the design of the He cryostat, a very low helium consumption is achieved and it needs to be refilled with only 4 litres of liquid helium every 48 h when the STM is at its lowest temperature. The sample transfer can also be done at low temperatures. A helium-cooled manipulator allows us to prepare, analyse and transfer the samples at temperatures down to 20 K.

Our experiments were performed in the UHV LT-STM, operating down to a temperature of 6 K and using etched W tips under a pressure of about 1×10^{-12} Pa. Clean Ag(111) surfaces were prepared and characterized before transportation with a cold finger. The Ag(111) surfaces were cleaned using cycles of Ne-ion bombardment followed by annealing to 700–800 K in the UHV chamber until the STM identified a surface region free from defects. This system was also equipped with low-energy electron diffraction (LEED) to characterize the sample surface.

Ag was evaporated from a tantalum crucible heated by a feedback controlled electron beam and was deposited at room temperature. A quartz crystal thickness monitor determined the deposition rate. Ag thickness was measured in monolayers (ML) where 1ML=1.4×10^{15} atoms/cm^2, the number of atoms per unit area of a Ag(111) surface. In this experiment, 0.04 ML Ag (5.6×10^{13} atoms/cm^2) was deposited according to the records of the quartz monitor.

After evaporation of Ag on to the surface, the sample was cooled to 6 K for imaging. In order to take STM images, tunnelling currents between 100 pA and 5 nA were used. Bias voltages refer to the sample voltage with respect to the tip.

3. Results

3.1. Manipulation of evaporated adsorbates

A variety of different atomic manipulation processes have been demonstrated with the STM. We may divide these processes into two classes: lateral processes and perpendicular processes. In this experiment, lateral processes were carried out to purposefully rearrange matter on the atomic scale.

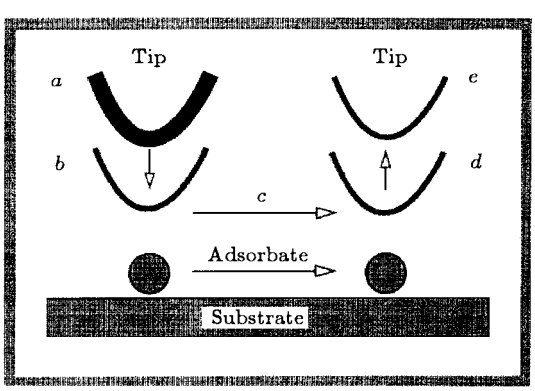


Fig.1. The procedure of lateral manipulation, involves first positioning the tip on top of the adsorbate at normal imaging distance (a). Then the tip is moved closer to the surface, thereby increasing the force between the tip and adsorbate (b). If the force is high enough, the adsorbate will follow the tip during movement (c) to the final position (d). There the tip is retracted (e).

In lateral processes an adsorbed atom or molecule is caused to move along the surface. We may view the act of rearrangement as a series of steps, which results in the selective making and breaking of chemical bonds between atoms or, equivalently, as a procedure that causes a configuration of atoms to evolve along some time-dependent potential energy hypersurface from an initial to a final configuration. Both points of view should prove useful in understanding the physical mechanism by which atoms may be manipulated with the STM.

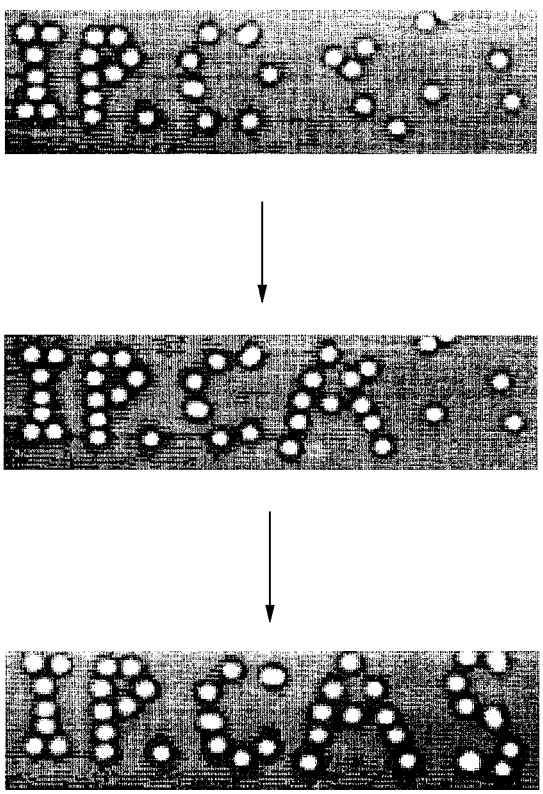


Fig.2. The construction of five letters (IP.CAS means Institute of Physics, Chinese Academy of Sciences). These four images show the process of constructing the five letters with 41 Ag atoms from evaporated adsorbates on Ag(111). Each Ag atom is individually positioned with the tip of the STM, and the average nearest-neighbour distance of Ag atoms is 1.833 nm. The experimental imaging parameters are 33.4 nm×18 nm, V=280 mV and I=1.5 nA.

The lateral process consists of the steps depicted in Fig.1. The adsorbate to be moved is first located with the STM in its imaging mode and then the tip is placed near the adsorbate, step (a). The tip–adsorbate interaction is then increased by lowering the tip toward the adsorbate, step (b). This is achieved by changing the required tunnel current to a high value and letting the feedback loop move the tip to a height which yields the higher current demanded. The tip is then moved laterally across the surface under the constant current condition (c) to the desired destination (d), pulling the adsorbate along with it. This process is terminated by reverting to the imaging mode (e), which leaves the adsorbate bound to the surface at the desired destination. Figure 2 shows the process of constructing the letters of IP.CAS (an abbreviation of 'Institute of Physics, Chinese Academy of Sciences') with the lateral manipulation method.

3.2. 'Digging out' atoms for atomically resolved imaging and manipulation of substrate atoms

Atoms from the substrate can also be manipulated via the processes of 'digging out' atoms under suitable parameters to make the native substrate atoms and cluster be dug out to the surface as adsorbates. In Fig.3, a hole and some clusters, including various numbers of Ag atoms around the hole, can be clearly seen. The manipulation of atoms dug out from the surface is similar to the process of evaporated adsorbate atoms. The difference is that the atoms or clusters obtained by the 'digging out' technique are generally composed of various numbers of Ag atoms. Sometimes one needs to pull out a single atom from the clusters step-by-step to construct a nanometre structure.

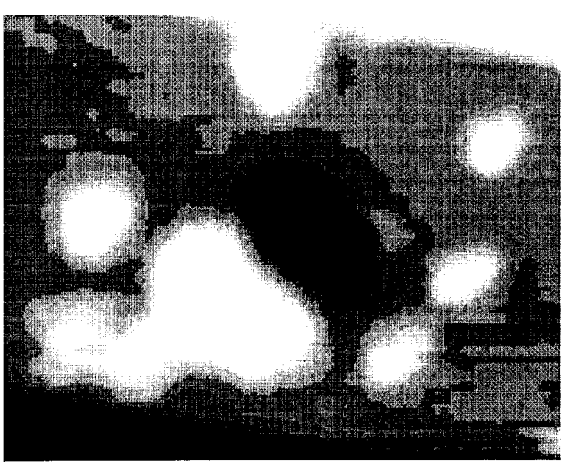


Fig.3. A hole and some atoms and clusters around the hole formed after the 'digging out' step (14 nm×10.6 nm, 1.2 nA, 19 mV).

The 'digging out' step can also be used to improve the tip state (shape and stability). A general limitation encountered in the STM is the lack of precise knowledge of the morphology and chemical composition of the tip. Despite this uncertainty, the STM has become a versatile tool for probing surface phenomena

with unprecedented resolution and control. Nevertheless, a well-defined tip may enable new applications and expand the capabilities of the STM. In this work, we have used the 'digging out' method to prepare *in situ* a tip by transferring a single atom or a few atoms from the substrate surface to the tip apex (or sharpening the tip). A single Ag atom or cluster was reversibly transferred between the surface and the tip through controlled manipulation by changing the parameters such as tunnelling current, voltage and distance. With this atom-terminated tip, the atomically resolved surface corrugation of the Ag(111) sample was obtained, which was not easy to achieve with a bare tip (Fig.4). The nearest-neighbour atomic distance of the Ag(111) surface is 0.289 nm.

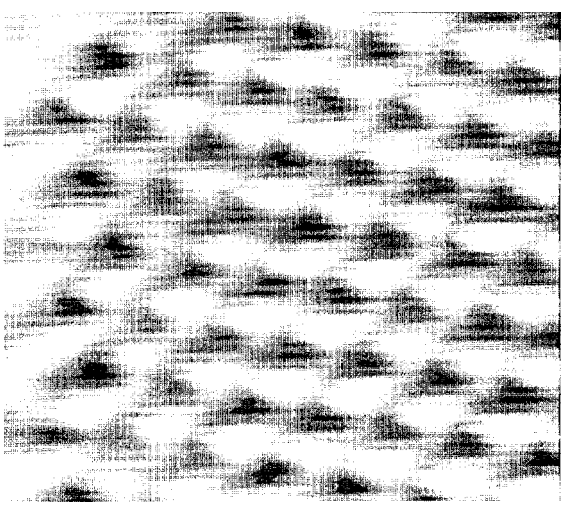


Fig.4. An atomically resolved STM image of the Ag(111) surface (2.2 nm ×2.2 nm, 1.3 nA, 45 mV).

Quantum corrals provide a new method for spectroscopically probing this quantity, and open new possibilities for studying the interaction of a two-dimensional electron gas with microscopic scattering objects. The atoms are precisely positioned using the tip of the STM, and this enables one to image the quantum mechanical interference patterns. The spectroscopic capabilities of the STM allow the observation of the local density of states (LDOS) inside quantum corrals at different energies.

Figures 5(a) and 5(b) show the formation of a simple quantum corral and a Chinese character by lateral manipulation of substrate Ag atoms dug out from the Ag(111) surface. Figure 5(a) is similar to the work first created by Crommie *et al.*[11] In Fig.5(a), the Ag atoms have also been positioned in a circular geometry with a diameter of 8 nm and a circular centre; the average distance between two atoms is 4 nm. The circular pattern shown in Fig.5(a) can be attributed to the effects of the surface electron state of Ag atoms.

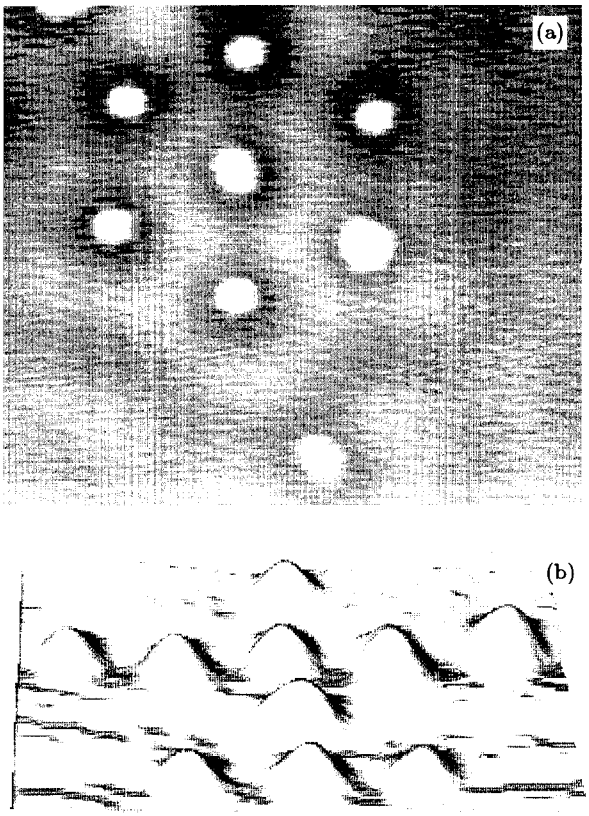


Fig.5. (a) Construction of a simple quantum corral with a ring of 6 Ag atoms, with a diameter of 8 nm and a central Ag atom. Each Ag atom comes from the native substrate and is individually positioned with the tip of the STM. The surrounding quantum interference patterns are visible. The experimental imaging parameters are 17 nm×14.5 nm, V=63 mV and I=1.1 nA. (b) The construction of a Chinese character [士 (SHI) means a soldier or person trained in an intelligentsia, shown here as a three-dimensional STM image] with 10 Ag atoms on Ag(111). Each Ag atom that comes from the native substrate is individually positioned with the tip of the STM. The average nearest-neighbour distance of the Ag atoms is 0.1875 nm. The experimental imaging parameters are 1.6 nm×9.6 nm, V=28 mV and I=1.7 nA.

4. Conclusions

In summary, we have shown that it is possible to manipulate a single native substrate and evaporated adsorbate atoms on Ag(111) in a controlled manner to accurately construct or write artificial nanometre-scale structures, including quantum corrals, some letters and a Chinese character, by using a lateral STM manipulation method at low temperatures. This provides an efficient basis from which to explore the basic physical properties of nanometre structures and to develop nanotechnology using a 'bottom up' approach.

References

[1] Binning G, Rohrer H, Gerber Ch and Weibel E 1982 *Phys. Rev. Lett.* **49** 52

[2] Jung T A, Schlittler R R and Gimzewski J K 1996 *Science* **271** 181

[3] Meyer G and Rieder K H 1998 *MRS Bulletin* **1** 28

[4] Silva S L and Leibsle F M 1999 *Surf. Sci.* **441** L904

[5] Getzla M 2001 *Chin. Phys.* **10** 186

[6] Eende O 2001 *Chin. Phys.* **10** 100

[7] Stroscio J A and Eigler D M 1991 *Science* **254** 1319

[8] Gartland P O and Slagvold B J 1975 *Phys. Rev.* B **12** 4047

[9] Crommie M F, Lutz C P and Eigler D M 1993 *Nature* **363** 524

[10] Braun K F and Rieder K H 2002 *Phys. Rev. Lett.* **88** 096801

[11] Crommie M F, Lutz C P and Eilger D M 1993 *Science* **262** 218