UHV-STM manipulation of single flat gold nano-islands for constructing interconnection nanopads on MoS$_2$

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Abstract. We demonstrate manipulation of metallic islands containing nearly a million atoms with a precision of one lattice spacing on a MoS$_2$ surface, one at a time. Optimizing the growth conditions yields triangular shape metallic nano-islands 40 nm in lateral size and 12 nm in height on the MoS$_2$ surface. The manipulation of these nano-islands is done one at a time using the scanning tunneling microscope, and a fully planar 4 pad nanostructure is demonstrated, where one apex of each triangular nano-island is pointing towards a central working MoS$_2$ area of 12 nm x 24 nm in which atomic cleanliness is preserved. The feedback loop conditions to achieve this manipulation are discussed. This fully planar 4 pads nano-structure is ready to be interconnected by a multi-tip system.

1. Introduction

The measurement of a current-voltage characteristics on a single atomic wire [1] (a single molecule [2]) demands an atomic precision of the interconnection between the atomic wire (the molecule) and the end surface atoms of the contacting pads [3,4]. This is not achievable with modern e-beam nanolithography [5] nor with the nanostencil technique [6], which are both limited to a lateral precision of about a nanometer. With a scanning tunneling microscope (STM) working in a manipulation mode, the precise manipulation of ultra-flat nanometer size metallic clusters on a semiconductor surface opens the way of fabricating an artificial nanostructure for interconnecting an atomic wire (a single molecule) with more than 2 metallic pads in a fully planar configuration. To reduce the lateral extension of the contact area between the nano-pads and the supporting surface, each pad must have a triangle shape with atomically facetted terminations.

To explore the fabrication of such a multi-pad metallic nano-structure, we start from the self-assembly of metallic gold nano-islands on an MoS$_2$ surface. After a careful control of the growth conditions to obtain flat triangular Au nano-islands, we demonstrate experimentally how a nano-island can be manipulated in a pushing mode on the MoS$_2$ surface by the tip of the STM and with a precision better than 0.1 nm. The experimental tunneling current and feedback loop conditions for this manipulation are detailed. As an application of this manipulation process, the assembly of a 4 metallic pads nano-structure is presented. The structure consists of 4 triangular nano-islands pointing towards a 12 nm x 24 nm atomically clean and flat MoS$_2$ working area. This nanostructure is ready to be connected by a multi-STM system equipped with 4 ultra sharp metallic tips [7].
The STM images and the nano-island manipulation were carried out at room temperature using an Omicron UHV-STM with a base pressure of $3 \times 10^{-8}$ Pa. The STM tip was made by chemical etching a 0.25 mm diameter tungsten wire in 2 M NaOH. Freshly etched tips were transferred into the vacuum chamber and cleaned by electron bombardment. The typical radius of curvature of the tip apex is 10 nm, as measured using a transmission electron microscope.

2. Sample preparation and nano-island fabrication

The 1 cm$^2$ MoS$_2$ wafer was fabricated from bulk molybdenite mined in Australia. The MoS$_2$ substrate was freshly cleaved from this wafer. Prior to the nano-island growth, the surface was degassed at 400 °C for 3 hours to remove surface impurities. The gold nano-islands were prepared by thermal evaporation at 400 °C with a deposition rate of 0.02 nm/s. The total thickness of deposited gold was 1 nm, as measured by a quartz microbalance. After the gold deposition, the sample was kept at 400 °C for another 1 hour in order to facilitate the self-assembly of crystalline Au nano-island on the MoS$_2$ surface. The nano-island growth conditions were optimized to achieve a majority of almost equilateral triangular nano-islands which measure 30 nm on a side. A small number of 22 nm triangles (<3%) were still found on the MoS$_2$ surface by STM. The full process was optimized by imaging each batch using a 10 keV SEM FEG electron microscope from FEI. At optimized conditions, each of the triangular nano-islands have a sharp apex with radii of curvature measuring 4.5 nm and 3.3 nm for the 30 nm and 22 nm lateral size nano-islands respectively (see Fig. 1).

Figure 1: (a) Constant current STM image of the MoS$_2$ surface showing a majority of 30 nm lateral size Au nano-islands recorded at $I = 20$ pA and $V = 1.0$ V for $G = 24$ %. To avoid multiple tip effect, the tip was cleaned by pulsing the bias voltage up to 3 V before imaging. (b) Constant current line scan on a 22 nm lateral size nano-island and (c) on a 30 nm nano-island with the same scanning conditions as in (a).
On the MoS$_2$ surface, the self-assembled Au nano-islands can be imaged by STM at a very low tunneling currents, typically $I < 20$ pA, as presented in Fig. 1a. When the tip apex is clean enough to preserve the atomic resolution of the MoS$_2$ surface and when it does not pick up any of the small nano-islands (lateral size typically ~5 nm) while scanning, the STM images are stable over night and are identical to the SEM images of the same surface. On the MoS$_2$ surface, the atomic resolution is preserved around the nano-island, showing the absence of wetting layer around those nano-islands. On top of the Au nano-islands, atomic resolution STM images were also obtained showing a nice hexagonal structure typical of (111) surface of Au. This confirms the crystallinity of those nano-islands with a preferred Au(111) orientation on the MoS$_2$ surface. Figure 1b and 1c present the STM images of two nano-islands with a lateral dimension of 30 nm and 22 nm with an apparent height of 12 nm and 9 nm respectively.

3. STM Manipulation of a single nano-island

As compared to the imaging conditions given above, increasing $I$ by 2 orders of magnitude caused brushing and subsequent removal of the Au nano-islands from the MoS$_2$ surface in the area scanned by STM. This phenomenon has been reported by several groups for different cluster and surface materials [8]. However, between the stable imaging and brushing conditions, there is a large range of possible STM feedback loop conditions to fine tune both the magnitude and the force acting on a single cluster during the brushing. To our knowledge, this fine tuning has not been explored for nano-clusters on semi-conductor surfaces with the prospect to manipulate only one cluster at a time with a positioning precision better than 0.1 nm.

![STM image](image)

Figure 2: (a) Constant current STM image ($I=20$ pA, $V = 1.0$V and $G = 24\%$) of a 30 nm lateral size nano-island used to study the manipulation threshold current and feedback loop conditions. The $N = 256$ STM lines scans presented in (b) were all recorded at the same location. (b) Representation of the time succession of the 256 lines scans recorded from A to B in 180 s for $I = 80$ pA, $V = 1.0$ V and $G = 50\%$. The nano-island imaged in (a) starts to move after $N = 38$ line scans.

To determine the threshold current and feedback loop gain ($G$) to manipulation a single nano-island on MoS$_2$, the following experimental sequence was followed. First, well isolated triangular nano-islands, 30 nm in lateral size were identified and imaged at a very low $I$ with large STM feedback loop gain $G$. All the manipulation experiments were performed on 30 nm nano-islands having the same lateral size and apparent height. Second, the STM was switched from the imaging mode to a
single scan mode with a scan generated always at the same pre-determined location and uni-
directional. During a single scan, the STM traverses over the nano-island at a point in the middle of a
nano-island facet, as presented in Fig. 2a. We call this the “impact point”. Third, up to a maximum of
700 identical scans were performed exactly at this impact point, while carefully noting any resultant
displacement of the targeted nano-island. To minimize possible deformation of the nano-island facet at
the impact location, all the scans were recorded in a constant current mode. Below \( I = 20 \text{ pA} \), as much
as 700 scans performed during a time interval of 490 s do not affect the position of the nano-island
even at low G.

To observe the first occurrence of a nano-island manipulation by the STM tip, I and G were
systematically varied in small steps starting from \( I = 20 \text{ pA} \) and a large G. The experimental procedure
is illustrated in Fig. 2b for \( I = 80 \text{ pA} \) and \( G = 50\% \). The nano-island starts to move after \( N = 38 \) scans.
After \( N = 256 \) scans, the nano-island had been moved 2.5 nm laterally, having been displaced from its
initial position by the STM tip. It should be noted that this observation is not to be confused with the
lateral drift of the scans due to some instabilities of our instrument. The lateral drift of our instrument
was measured to be one order of magnitude lower i.e. 2.7 nm/hour. The 256 scans were recorded in
180 s. During this time period involving manipulation of this particular nano-island, the apparent drift
was only 0.12 nm, recorded at \( I = 20 \text{ pA} \).

![Figure 3: Statistics of the nano-island manipulation on MoS₂.](image)

(a) Average number \(<N>\) of scans required to move one nano-island as a function of the tunneling current I, for 2 values of the feedback loop gain (circles: \( G = 50\% \) and triangle \( G = 24\% \)). (b) Probability of manipulating a nano-island after \( N \) scans as a function of the tunnel current \( I \) for \( G = 50\% \) (square: \( N < 10 \), triangle: \( N < 100 \), lozenge: \( N < 200 \)). No Manipulation was observed below \( I = 20 \text{ pA} \), \( V = 1.0 \text{V} \) in this constant current mode.

After this fine determination of the threshold current and feedback to move a nano-island, we have
explored the full range of the feedback parameters variation with the objective to speed up the
manipulation process and to enlarge the distance a nano-island can be manipulated, while retaining the
manipulation precision as demonstrated in Fig.2. On the same impact point and for the same tip apex,
Fig. 3a indicates the average number \(<N>\) of identical STM scan lines required to move a nano-island
as a function of I for a fixed G. For a 100 % chance of success during a single STM line scan, the
threshold current must be larger than 0.1 nA. It should be mentioned here that this threshold is slightly dependent on the STM tip, and varies slightly from one tip to another. A better statistical analysis of the manipulation phenomenon is presented in Fig. 3b where the probability to manipulate a nano-island after a certain number of scans repeated on the same impact point is given for as a function of I. For I > 200 pA a nano-island can be manipulated with a 100 % success, irrespective of the feedback loop gain G. The magnitude and sign of the bias voltage had no direct influence on the determined threshold for manipulation of the nano-islands.

4. Construction of nano-pads interconnects.

To explore further the capabilities of constructing a nanostructure by using this manipulation process, simple investigations were performed such as approaching 2 nano-island together to attempt coalescence, rotating a nano-island by positioning the tip on an apex of the triangular nano-island, searching for MoS\textsubscript{2} surface defects at the initial position of a nano-island, and manipulating a few of the 22 nm lateral size nano-islands. The conclusion of these explorations is that a nano-island is a solid

Figure 4: A sequence of STM images taken during the construction of a 4 pads nano-structure. The nano-islands A, B, C and D are manipulated on a MoS\textsubscript{2} surface. (a) the surface after the Au nano-island self-assembly process. (b) to (d) various stages of the construction where 15 other nano-islands have been manipulated out of the central area one by one. At the final stage, one apex of each triangle is pointing towards the center of the nano-structure where an atomically clean 12 nm x 24 nm MoS\textsubscript{2} surface can be recovered. Imaging conditions as described in Fig. 1 and manipulation conditions as described in Fig. 2. Scale bar: 60 nm.
object which is very stable, since no coalescence was observed. The 22 nm nano-island can also be manipulated but at lower I. Although there are surface atomic defects on natural MoS$_2$ samples, they are not correlated to the position of the nano-islands. This suggests that the dominant force between the tip apex and the nano-island is a repulsive force. Increasing I and lowering the gain increases the strength of the repulsion. This repulsive interaction should exceed the adhesion of a single nano-island, for the kicking process to be successful. After a certain number of tip apex kicks, the nano-island is destabilized and can be manipulated on the surface. This tentative explanation of the observed manipulation requires further investigation.

In Fig. 4, a sequence of images is presented demonstrating how 4 nano-islands can be manipulated to construct a fully planar metallic 4 nano-pads structure. The end apex of each pad is pointing towards a central MoS$_2$ surface area where atomic corrugation is recovered. First, we have identified 4 nano-islands A, B, C and D having the same apparent lateral size and height and a suitable orientation to form the 4 pads. Then, we have identified 15 nano-islands which are to be cleared out one by one from the target area. Around the 4 pads, this cleaning creates a free area of 150 nm x 150 nm dimension, easily recognized by a moderate resolution SEM [7]. The closest distance between neighboring nano-islands in the 4 nano-pad structure are 3 nm, 6 nm and 10 nm respectively. The diagonals of the 4 nano-pad structure are 12 nm and 24 nm respectively. Several other nanostructures can be constructed along similar lines. For instance, we have nanostructured nano-pads contacts with an inter pad distance less than 2 nm, or a 4 pointed star using 30 nm nano-islands whose apices are pointing outwards.

5. Conclusion

We have demonstrated how single gold nano-islands 30 nm in size and 12 nm in height can be manipulated at room temperature with a UHV-STM, on a semi-conductor MoS$_2$, with an appropriate choice of STM feedback parameters. We foresee that the precise manipulation of single ultra-flat metallic nano-islands on a semi-conductor surface will open a new way of fabricating planar metallic contact pads to interconnect an atomic wire or a molecular to macroscopic probes, preserving the atomic cleanliness of the surface. Commercially available UHV compatible multi-STM systems where each tip apex is positioned independently on a surface using top SEM imaging [7], provide direct electrical access to the 4 pads presented in Fig. 4 without the need of any nano-lithography step. Consequently, there is no physical contact between the macroscopic contacting electrodes with the semi-conductor surface supporting the nano-island pads.

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