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An ultrahigh vacuum scanning tunneling microscope for use at variable temperature from 10 to 400 K

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We report on the construction of an ultrahigh vacuum (UHV) scanning tunneling microscope (STM) specially designed for operation in the entire range of sample temperatures between 10 and 400 K. The sample is cooled by means of a liquid helium continuous-flow cryostat, while the supporting manipulator and the surrounding devices remain at room temperature. This allows rapid variation of the sample temperature. The standard instruments for surface preparation and analysis and the STM are contained in a single UHV chamber. By rotation of the manipulator the sample can be positioned in front of any of these instruments without changing the sample temperature. The performance of the microscope is demonstrated by two examples of images of xenon adsorbed on platinum(111) showing: (a) the evolution of the morphology of a submonolayer of xenon from adsorption at 17 K up to desorption at about 90 K and (b) atomically resolved images of the hexagonal incommensurate rotated phase for xenon at monolayer completion.

I. INTRODUCTION

The scanning tunneling microscope (STM) is established as a powerful tool for the investigation of solid surfaces and thin adsorbed layers. In many of these investigations one is interested in equilibrium structures and temperature dependent effects. This requires a variable and well controlled sample temperature. On the other hand, high resolution is necessary to study individual adsorbates or to achieve intramolecular contrast. This resolution can only be obtained at low temperature where the motion of the adsorbates is sufficiently reduced. In order to satisfy these two requirements, we need a temperature-variable and low-temperature STM, which allows accurate setting and control of the sample temperature as well as the possibility of rapid changes of the sample temperature.

The majority of STMs currently in use are fixed-temperature instruments. Those which operate at low temperature are mostly organized into two different sections: a room-temperature analytical part and a low-temperature STM part. The latter is connected to a cold stage or introduced into a dewar filled with liquid nitrogen or helium. The advantage of this procedure is the high stability (small thermal drift) of the STM. With this construction, however, these STMs usually suffer from one or more of the following drawbacks: (a) During the transfer of the sample to or from the analytical part of the chamber the sample temperature cannot be controlled; (b) the large mass, which has to be cooled, does not allow a rapid cycling of the sample temperature; and (c) increasing the sample temperature can cause a massive desorption from the cold walls and will result in a contamination of the sample surface. Moreover, these setups usually complicate the placement of other instruments close to the cold sample.

The investigation of clean and adsorbate covered metal surfaces requires the preparation and characterization of ordered surface structures. Besides the STM, traditional surface science techniques like Auger electron spectroscopy (AES) and low-energy electron diffraction (LEED) are needed to extract information on the metal surface chemical composition and long-range crystallographic order. These techniques and complementary methods as, for example, temperature programmed desorption (TPD), are also extremely useful for the characterization of the adsorbate systems. It is therefore desirable to operate the corresponding instruments, including the STM, in a single UHV chamber and under identical experimental conditions (sample temperature, pressure, coverage, etc.).

The above considerations led us to devise a special UHV STM, in which only the sample is cooled. For this purpose, a new manipulator was built that allows the sample to be transferred between the microscope and the other equipment without changing the sample temperature. For use with the analytical devices (AES, LEED, TPD, etc.) the sample temperature can be varied between 10 and 1200 K, while the STM can be operated in the temperature range from 10 up to 400 K.

II. INSTRUMENT DESIGN

A. General

The main difference between this instrumental setup and the low-temperature instruments mentioned before is the central manipulator, which provides the cooling as well as the positioning of the sample. At the same time, its stable construction allows STM images with atomic resolution to be recorded in a wide temperature range.

To simplify the handling of the sample, the cryostat with the heat exchanger and the sample holder are both integrated parts of a single manipulator. The actual design of this manipulator is governed by the conflicting requirements of a simultaneously strong thermal coupling and the weakest possible vibrational coupling between the sample holder and the heat exchanger.
There are two additional challenges:

(a) In order to perform rapid changes in sample temperature (some K/s), the total mass that has to be heated or cooled must be as small as possible. Hence, a miniaturized sample holder that is thermally decoupled from the rest of the manipulator was used.

(b) Small masses, however, are more sensitive to vibrations than large ones. Good cooling characteristics require good thermal, i.e., good mechanical, contact of the sample holder to the heat exchanger. Even after minimization of the vibrational coupling between these two parts an intolerable transmission of vibration might remain. Additional care has been taken to reduce the amplitude of these residual vibrations.

There are two types of perturbations from which the STM must be insulated: externally generated vibrations, which penetrate the vacuum system, and internally generated vibrations.

External perturbations (like floor vibrations, etc.) mostly consist of low-frequency but high-amplitude noise. They can be reduced efficiently by metal-spring damping. Therefore, the entire UHV chamber is suspended from the ceiling by four metal springs of about 2 m in length. They are coated with a layer of Armaflex for additional damping. To obtain a low resonance frequency in this damping stage the mass of the vacuum chamber was increased by adding blocks of lead, resulting in a total mass of about 600 kg for the suspended system.

The design of the manipulator could then be focused on the problem of possible high frequency noise, as it can emerge from the coolant flow, the STM itself, or enter the system as acoustic noise. The effects of this high frequency noise can be reduced by viscous damping, e.g., using organic materials like Viton, and by scaling down the amplitude with the help of an enlarged mass stiffly connected to the sample holder. Here we use a combination of both methods and a very rigidly constructed STM (a modified Besocke type), which, due to its high resonance frequencies, does not require multiple stages of vibrational damping. In this way, the noise level could be reduced far enough as to obtain STM images with an atomic resolution down to very low temperatures, i.e., at full power of the cooling stage.

B. UHV system

The basic idea of the UHV setup is that of a central manipulator which comprises both cryostat and sample holder and allows positioning of the sample towards the different preparational and analytical devices. There are two straightforward ways to put this into practice: (a) to have all devices aligned along the main axis of the chamber and to move the sample in front of them using a linear motion feedthrough, or (b) to locate the sample in the central point of the chamber with all devices arranged in a surrounding circle and aiming their focus at this central point. In this case all sample positioning is accomplished by a rotary motion feedthrough.

The second possibility was chosen because it can be realized with a comparatively smaller UHV ensemble. To facilitate future extensions, a modular UHV chamber was built consisting up to now of two independent modules that are connected via a conflat flange 300 mm in diameter. Figure 1 gives a schematic drawing of the main (upper) module, which houses the sample and the surface analytical components. The lettered flanges hold the following instruments: (a) ion sputter gun, (b) LEED, (c) AES, (d) QMA, (e) xyz translator that holds the STM, and (f) central manipulator. Top: side view, indicating instrument positions. The bottom flange (g) connects the upper to the lower module carrying the vacuum pumps. Bottom: top view (equatorial cross section). The STM housing (e) is facing outward.
Variable temperature STM

The inner end of the pipe, which also has been left out of Fig. 2(A), is shown from a side view in a smaller scale in Fig. 2(B); at the very end of the pipe (a), i.e., in the center of the chamber, a massive copper block (d) is mounted and supports the sample holder. It actually does not touch the pipe directly but via Viton rings (f) so that the vibrations transmitted by the pipe are damped before propagating into the copper block. The liquid helium cryostat (g) is centered inside the pipe and does not make direct contact to the copper block, either.

The design of the cryostat is adapted from the cryostats that have been successfully used in various helium scattering machines in our institute. It has been slightly enlarged to enhance the cooling capacity.

The best way to avoid vibrations originating from the fluid/gas bubble mixture is to flow cold helium gas instead of helium liquid through the heat exchanger. In order to reach low temperatures (\(\approx 10\) K), however, it is necessary to use liquid helium as the coolant. The He flow introduces a non-negligible acoustic noise. A common method to bypass this problem is to operate the cryostat according to a “ballistic” cooling scheme, in which the flow is periodically turned off for the time it takes to record several STM images. This, however, is not feasible in our case as we require a well defined and stable sample temperature for studying the kinetics of adsorbate systems.

The solution to this problem (the vibrational decoupling of the sample holder and the heat exchanger) is to use a soft copper braid (h) as a connection between the heat exchanger and sample holder. This braid provides good heat transfer but greatly reduces the transmission of vibrations stemming from the liquid helium flow.

Nevertheless, this reduction is not sufficient. In order to scale down the amplitude of the remaining vibrations the sample holder is tautly connected to the massive copper block (d). As the copper block is about a factor of 1000 heavier than the sample holder, the amplitude of the vibrations is strongly reduced. On the other hand, the thermal contact between the sample holder and the copper block should be as small as possible. This can be achieved by using “pushing and pulling screws” (i). The result is a strong mechanical contact with low heat conductivity across these thin and long screws. This combination of a massive block and the two special connections (h, i) was already employed by Michely.

The sample holder itself is comprised of a copper ring (j), which supports the sample (e) and the “race-way” (k) of the STM (cf. Sec. II D), which fixes the sample to the copper ring. The sample holder is connected to the heat exchanger via the soft copper braid, which is electron-beam welded to the copper ring (j). The sample temperature is measured with a Ni-CrNi thermocouple spot welded directly to the sample. The sample holder and the sample are grounded, as required for AES and LEED.

To operate the STM, it is imposed onto the race-way using the xyz translator. After contact, the sample temperature should be kept below 400 K in order to prevent damage to the piezos if they are heated above their Curie temperature as well as to prevent degradation of the glues used to affix it to the race-way.
the piezos. With the STM lifted up from the race-way the sample can be electron-beam heated to 1200 K by a filament located about 0.3 mm below the sample, inside a bore in the copper block. It is affixed to the pipe without making contact with the copper block or the sample holder.

To summarize, only the sample holder, i.e., a very small mass, has to be cooled or heated. Mechanical coupling between the cryostat and sample holder is minimized by using a copper braid. The remaining vibrations, including those that enter via the pipe, are damped by Viton spacers and by scaling down the amplitude using a large second mass. This mass is in good mechanical but poor thermal contact with the sample holder.

**D. STM**

The body of the STM is derived from the Besocke-type piezo walker. Due to its symmetric design, this “beetle”-type STM is very stable with regard to thermal drift and its compact shape leads to high resonance frequencies. Contrary to the original design, the STM has been turned upside down. This allows one to leave the sample on the manipulator and move and remove the STM instead. The technical details of a similar STM and a description of the race-way, a special sample ring used for the tip-sample coarse approach, can be found in Ref. 6.

As scan actuators in the STM four piezoelectric PZT tubes are used. The piezos are glued to an aluminum base plate using low out-gas conducting epoxy. The three “feet” of the STM are made from stainless steel balls glued to the outer three piezos also with conducting epoxy. When placed on the race-way, the STM thus operates at the same potential as the sample. The outer electrodes of the tubes are sectioned into four equal 90° quadrants along the length axis in order to allow xy motion. For the wiring of the outer electrodes Kapton-coated 0.04-mm-diam copper wire is used. The race-way is made from molybdenum. To avoid the steel balls sticking to the race-way at UHV conditions, the ring’s surface has been carbidized.

The possibilities of the STM have been extended to allow for two different types of scan: (a) the classical mode of operation in which the outer piezos move the whole STM body in the xy direction while the center piezo provides z motion and height control of the tip; and (b) only the center piezo is used, which then provides xy and z motion. This is achieved by cutting the outer electrodes of the center piezo around the circumference halfway down the length of the tube thus giving eight instead of four independent electrodes. The lower four electrodes (near the tip) control the tip height while the upper four are used for xy scanning. By using this distribution of signals the maximum scan width compared to the width in the classical mode is reduced by a factor of 1.7. The advantage of this second mode is that the microscope body does not move during the scan, thus allowing faster scanning.

The STM itself is not very efficiently cooled via the race-way and remains at an elevated temperature (close to RT), even when placed on a sample at low temperature. This results in a nonvanishing drift (mostly in z direction) when tunneling on a cold sample. After some 10 min of cooling, however, the lateral drift is sufficiently small to take high quality images and stay approximately at the same place. The advantage is that the scan range does not change significantly when scanning at different sample temperatures. Even after several hours of operation at 10 K the scan width has been found to be only 20% smaller than at RT. This has to be compared to a reduction by a factor of 5.5 when cooling the entire piezo to 4.2 K.

The tip is electrochemically etched from a 0.2 mm tungsten wire. It is held by spring tension in a stainless-steel syringe tube epoxied onto an insulating Macor block, a machinable ceramic that has a thermal expansion coefficient close to that of our PZT-piezoelectric material. This block is epoxied onto the central piezo. For both epoxy connections insulating epoxy is used.

Special attention has been devoted to shielding the wire carrying the tunneling signal from the syringe tube to the feedthrough against the scan signals. Near the tip the grounded inner electrode of the center piezo shields the wire from the other piezos. Further up this wire is led separately through a stainless steel tube and is thus protected from crosstalk to the scan signals.

**E. Operation**

The sample preparation is performed under ultrahigh vacuum conditions (base pressure below $1 \times 10^{-10}$ mbar) and consists of repeated cycles of ion sputtering (1 keV Ar$^+$), heating to 750 K in a $1 \times 10^{-6}$ mbar oxygen atmosphere for 10 min, and annealing at $\approx 1200$ K. Afterwards, the Pt(111) crystal typically exhibits terraces $\approx 1000$ Å wide separated by single atomic steps. At a few places pinning centers can be found, which are due to the small residual contamination of the surface. As the STM is much more sensitive than LEED or AES even to minute impurity concentrations, the STM itself was used for the ultimate control of the surface smoothness and cleanliness.

After preparation the sample is cooled to low temperature. Cooling from RT to 10 K is performed within $\approx 6$ min using liquid helium as a coolant. In subsequent flash/cooling cycles, however, it takes less than a minute to cool the sample from 300 to 10 K.

In the case of rare gas adsorption, which is extremely sensitive to CO contamination, the sample has to be cleaned about every 2 h. An effective removal of the low temperature contamination can be achieved by flashing the sample to $\approx 650$ K. During the flash, the STM is lifted up from the race-way to avoid damage of the instrument. Once the sample has reached the desired low temperature, the experiment (e.g., the adsorption) starts. In the present experiments the adsorption temperature was chosen between $\approx 10$ K (full helium flux, no heating) and 100 K (reduced helium flux, controlled heating). For higher temperatures liquid nitrogen could be used as a coolant instead of liquid helium.

Adsorption is usually performed with the STM still lifted up from the race-way. The STM is then lowered onto the race-way and is allowed to adapt itself to the low temperature. This takes about 10 min. By using controlled heating of the sample, the STM can be operated at any temperature between 10 and 400 K. Temperature sequences can be re-
FIG. 3. Morphology of about 0.75 ML of xenon physisorbed on Pt(111). Each image was taken at the same location on the sample, at the same sample temperature of 17 K and using the same scanning parameters (scan width 6000×6000 Å², tip bias -400 mV, I = 2 nA). The images are represented as if illuminated from the left. (a) Right after adsorption at 17 K: many small xenon islands have nucleated on the platinum surface. The following images were taken after short annealing at increasing temperatures: 32 K (b), 38 K (c), 55 K (d), 80 K (e), and 93 K (f). The images were taken directly after quenching the sample to 17 K again. In (f) all the xenon is desorbed.

corded at a fixed location on the sample over a range of up to 100 K, limited only by the range of adjustment of the tip-sample distance using the central piezo (a few 100 Å); a change of more than 100 K would result either in a tip-sample crash or the loss of tunneling contact as the range of z correction is exceeded due to thermal contraction or expansion of the piezos and the sample holder. Thus one can, for instance, adsorb a film at 10 K and follow its morphology at the selected location up to 100 K before the tip-sample distance has to be enlarged mechanically. This is achieved by performing an appropriate rotation with the STM on the race-way, thus moving the tip away from the sample. In most cases, however, this causes the particular scan area to be lost.

An absolute sample temperature calibration was obtained by temperature programmed desorption recording the multilayer desorption peak of argon and of nitrogen. The exact procedure and experimental parameters were taken from Ref. 12. Using these reference points the sample temperature controller\(^{15}\) was calibrated with an estimated error of about ±1 K at 25 K (increasing to ±3 K at 10 K).
FIG. 4. Atomically resolved images of a full monolayer of xenon on Pt(111) taken at 13 K at two different locations in the vicinity of the step edges of vacancy islands of the underlying platinum surface. Due to the representation (apparent illumination from the left), these step edges appear as dark or bright stripes. The step edges run along the close packed (110) directions thus allowing to determine the rotation of the xenon structure: (a) The hexagonal xenon phase is rotated by about 30° with respect to the Pt(110) directions. The “black holes” in the structure are probably due to vacancies (missing xenon atoms) in the xenon film. Scan width 160×160 Å², tip bias −400 mV, I = 2 nA. (b) Larger scale image showing a periodic modulation of the dense xenon monolayer of about 24 Å. Note that the contrast is enhanced with respect to (a) in order to emphasize the long-range modulation. Scan width 220×220 Å², tip bias −400 mV, I = 2 nA.

III. EXPERIMENTAL RESULTS

Rare gases physisorbed on single crystal surfaces can be considered model systems in surface science and have been investigated using various experimental techniques. In the case of xenon physisorbed on Pt(111) most of the structural aspects have been determined using reciprocal space methods, such as thermal energy He diffraction. As a result, the phase diagram of xenon on Pt(111) is now well established.14 Until now, however, only little was known on the nucleation and growth of the xenon layers or the role of surface defects and impurities on the xenon structure. One of the main reasons for this has been that real space images were not available for this system.

With the advent of low-temperature STMs this situation has changed. The first STM images of xenon on Pt(111) at low temperature (4 K) were reported in Ref. 15. This investigation, however, was focused on the low coverage regime and, due to the inherent restrictions of the fixed-temperature instruments mentioned above, it was not possible to study the influence of the surface temperature on the xenon structure and growth.

Here we present images of xenon on Pt(111) recorded with our new variable-temperature STM. A detailed discussion of the experimental results is beyond the scope of this article and will be presented in a forthcoming publication.16

Figure 3 shows the evolution of the morphology of about 0.75 ML of xenon on Pt(111) from adsorption at 17 K to beyond desorption at around 90 K. In (a) the structure of xenon right after adsorption is shown. Many small (two-dimensional) xenon islands dispersed all over the platinum terraces can be seen. Afterwards the sample was annealed at different temperatures (increasing in steps of about 2 K). The sample temperature was raised at about 1 K/s up to the desired temperature. During this heating cycle the tip was retracted several hundred Å from the surface. The sample was then immediately quenched again to 17 K and subsequently the xenon layer was imaged with the STM. The six images in Fig. 3 were chosen from a larger series of images all recorded at the same location on the sample. A detailed analysis of the complete series of images indicates a well-defined onset of the surface thermal mobility of xenon at about 27 K. Island ripening is evident in (b), which was taken just above the 27 K threshold. In (c) through (e) the xenon structures continue to grow larger in size until in (f) the xenon is completely desorbed.

Figure 4 presents atomically resolved images of a full monolayer of xenon on Pt(111) taken at 13 K. Both images are taken in the vicinity of vacancy islands of the underlying platinum substrate. The step edges of these vacancy islands coincide with the close packed (110) directions of the platinum substrate and thus allow one to determine the rotation of the xenon structure. As expected from earlier TEAS measurements,17 we find a hexagonal incommensurately rotated (HIR) phase oriented at about 30±3° with respect to these close packed directions (a). The periodic modulation of the dense xenon monolayer of about 24 Å (b) is also consistent with the TEAS results.14,17

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2Armstrong, Düsseldorf, Germany.
7EBL type 2, 1/2 in. long, 1/4 in. o.d., with nickel electrodes, by Staveley Sensors, East Hartford, CT 06108.
8EPO-TEK H35–175 (silver filled conducting epoxy), and EPO-TEK H377 (nonconducting) by Epoxy Technology Inc., Billerica, MA 01821.
9Kapton coated copper wires by Underwriter Laboratories/Detakta, Norderstedt, Germany.
11Corning Glass Works, Corning, NY 14830.
13Eurotherm type 818, Eurotherm Ltd., Durrington, Worthing, West Sussex, BN13 3PL, Great Britain.
16S. Horch, P. Zeppenfeld, and G. Comsa (to be published).